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ABSTRACT

This report reviews the problems of sulfur oxide emissions and nitrogen oxide emissions from stationary sources. The first part of the report discusses the adverse consequences to health from combustion of sulfur-containing fossil fuels. The health problem is discussed by relating sulfur oxide levels and respirable particulate matter with morbidity and mortality figures. Present ambient air quality standards are analyzed in terms of existing data. In addition to health effects, the report analyzes emission effects on agriculture, forestry, ecosystems, and building materials. The second part of the report deals with strategies for controlling sulfur-related power plant emissions. Attention is focused on electric utility generation, relationship of emissions to ambient air quality, efficient pricing, improving conversion efficiency, fuel shifting, and fuel preparation, flue gas desulfurization, and tall stacks and intermittent control systems. The second part also contains recommendations for abatement strategies. The formula involves a comparison of marginal cost with marginal benefit, the harmful effects of emissions, and the relationships between point source emissions and patterns of dispersion and deposition as it applies to each power plant. The final part of the report assesses nitrogen oxides in terms of sources of emissions and techniques of emissions control. (MR)

94th Congress
1st Session

COMMITTEE PRINT

AIR QUALITY AND STATIONARY SOURCE EMISSION CONTROL

A REPORT BY THE

COMMISSION ON NATURAL RESOURCES
NATIONAL ACADEMY OF SCIENCES
NATIONAL ACADEMY OF ENGINEERING
NATIONAL RESEARCH COUNCIL

PREPARED FOR THE

COMMITTEE ON PUBLIC WORKS
UNITED STATES SENATE

PURSUANT TO

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APPROVED AUGUST 2, 1973

MARCH 1975

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Printed for the use of the Committee on Public Works

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NOTICE

The project that is the subject of this report was approved by the Governing Board of the National Research Council, acting in behalf of the National Academy of Sciences. Such approval reflects the Board's judgment that the project is of national importance and appropriate with respect to both the purposes and resources of the National Research Council.

The members of the groups selected to undertake this project and prepare this report were chosen for their individual scholarly competence and judgment with due consideration for the balance and breadth of disciplines appropriate to the project. Responsibility for all aspects of this report rests with those groups, to whom sincere appreciation is hereby expressed.

Although the reports of our study committees are not submitted for approval to the Academies' membership or to the Board, each report is reviewed by a second group of scientists according to procedures established and monitored by the Academies' Report Review Committee. Such reviews are intended to determine, among other things, whether the major questions and relevant points of view have been addressed and whether the reported findings, conclusions, and recommendations arose from the available data and information. Distribution of the report is approved by the President only after satisfactory completion of this review process.

COMMISSION ON
NATURAL RESOURCES

NATIONAL ACADEMY OF SCIENCES

OFFICE OF THE PRESIDENT
2101 CONSTITUTION AVENUE
WASHINGTON, D. C. 20418

March 3, 1975

The Honorable Jennings Randolph
Chairman
Committee on Public Works
United States Senate
Washington, D. C. 20510

Dear Mr. Chairman:

I have the honor to transmit the report entitled "Air Quality and Stationary Source Emission Control" which was prepared for the Committee on Public Works of the U.S. Senate, pursuant to the request in your letter of 18 September 1974.

This study was organized by the Commission on Natural Resources in cooperation with other major units of our National Research Council. As noted in the Introduction, individual chapters were first prepared by specific individuals or groups. Each chapter was carefully reviewed by the parent unit, e.g., the Assembly of Life Sciences or the Assembly of Engineering and by the Commission; the latter's review guided preparation of the final chapter drafts, particularly the chapter entitled "Part Two in Brief, Strategies for Controlling Sulfur-Related Power Plant Emission." From the report proper, the Commission prepared the Introduction and Summary; each statement also carries the approval of the cognizant unit. In addition, the report was reviewed by an independent panel appointed by our standing Report Review Committee and the report is now compatible with the recommendations of these reviewers.

The report deals with yet another instance of great concern for protection of the environment and of the public health in which the body of available, reliable, pertinent, information is less clear in compelling conclusion than national decision-makers may reasonably require. Perhaps the principal contribution of the report is to reveal what is and is not known and, in this way, guide both the decisions which must be taken in the face of uncertainty and the future research program necessary to reduce that uncertainty.

Sincerely yours,

Philip Handler
President

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INTRODUCTION AND SUMMARY

In September 1974, the National Research Council presented the report Air Quality and Automobile Emission Control to the U.S. Senate Committee on Public Works. That report examined the health effects of a number of pollutants (carbon monoxide, nitrogen oxides, oxidants, sulfur oxides, and particulates), reviewed the state of knowledge about the relationship between emissions of pollutants from automobiles and ambient air quality, and presented a discussion and analysis of the costs and benefits of automobile emission control. At hearings held by the Senate Committee, Senator Jennings Randolph (D., W.Va.) indicated a need for a similar review of sulfur-related pollutants from stationary sources. The Senator formally requested such a review in a letter of September 18, 1974 to Dr. Philip Handler, President of the National Academy of Sciences (see p. xiv). Dr. Handler replied on October 24 (see pp. xv-xvi). Although Senator Randolph's request related only to sulfur oxides and particulates, it seemed useful and appropriate to include in the report a review of the extent of the problem of nitrogen oxide emissions from stationary sources and the techniques available for abatement of these emissions as well.

The Environmental Protection Agency has established ambient air quality standards for certain air pollutants. A table of currently mandated air quality standards can be found on p. xliii. The states were to devise implementation

United States Senate
COMMITTEE ON PUBLIC WORKS
WASHINGTON, D.C. 20510

September 18, 1974

Dr. Philip Handler
President
National Academy of Sciences
2101 Constitution Avenue, Northwest
Washington, D. C. 20418

Dear Dr. Handler:

Pursuant to discussions between us and our staffs, I request preparation of additional information to supplement the Academy Report to the Committee on Public Works on the health effects of air pollutants.

First, in order to assist the Committee in understanding the implications of current information regarding the health effects of sulfur oxides, the Committee would appreciate a more complete summary of information assembled for the submitted report and an analysis and validation of the available data on sulfates.

Second, the Committee would appreciate receiving an analysis of the ambient air quality impact of various strategies proposed for reduction of sulfur oxide and particulate emissions. We are interested in the impact of such strategies on both urban and rural sulfur dioxide and sulfate concentrations and on the relationship between the strategies proposed and the time in which air quality protective of public health can be achieved.

The Committee recognizes the constraints under which the Academy will operate to deliver the study by March 1, 1975, with the use of existing scientific and air quality data. You may also limit the area for the second study to that geographic region identified by EPA as having a significant sulfur oxide-related ambient air quality problem.

Because of their concern and their legislative responsibilities in this area, I am sending copies of this letter to Senators Baker, Muskie and Buckley.

I earnestly hope the National Academy of Sciences will be able to act expeditiously on the request which will conclude the current contracts with the Committee on Public Works and utilize the funds which have not been expended.

With best regards, I am

Truly,

Jennings Randolph
Jennings Randolph
Chairman

cc: Senator Howard H. Baker
Senator Edmund S. Muskie
Senator James L. Buckley

NATIONAL ACADEMY OF SCIENCES

OFFICE OF THE PRESIDENT
2101 CONSTITUTION AVENUE
WASHINGTON, D.C. 20418

October 24, 1974

The Honorable Jennings Randolph
Chairman
Committee on Public Works
United States Senate
Washington, D. C. 20510

Dear Mr. Chairman:

In response to your letter of September 18 and as discussed in meetings between the Senate Committee and National Academy of Sciences staff, we are prepared to undertake, for the Senate Public Works Committee, a study which would examine certain aspects of the control of sulfur oxide and particulate emissions. This study would be a logical sequel to those studies already completed. Although the Academy operates under certain constraints which make difficult a response in a very short time, we recognize that the legislative needs of the Committee are urgent. A useful review of (1) the state of knowledge of the health effects of sulfur oxides and particulates (much of which has already been supplied to the Committee), (2) the impact of power plant emission control strategies on ambient air quality, and (3) the more general consequences of those control strategies can be attempted within the time specified in your letter--that is, by March 1, 1975. However, it will be necessary that this study build upon and make use of certain already ongoing NAS activities as described in this letter.

One part of the study will evaluate and expand the data on the health effects of sulfur dioxide, particulates, and sulfates which were used in the preparation of the recent report for the U. S. Senate Committee on Public Works. The discussion will indicate which studies provide useful information in the following categories: animal studies, human laboratory studies, and epidemiological investigations. The methods being developed for making size and chemical characterizations of sulfates, as well as the data base presently available from epidemiological studies, will be included. In addition, the possible role of the automotive catalyst in increasing levels of sulfate will be considered. Also, an examination will be made of the known ecological impacts--acid rains, vegetation damage, etc.--of sulfur oxide and particulate emissions.

The Honorable Jennings Randolph
October 24, 1974
Page Two

The study will also examine what is now known about strategies for the reduction of emissions of particulate matter and sulfur oxides. In particular, an investigation of air pollution problems associated with electric utility generation by coal and oil-fired power plants will be undertaken. The geographical area to be emphasized in the study is the Eastern part of the United States, where widespread sulfur oxide-related ambient air quality problems exist. The investigation will center about the analysis of strategies and combinations of strategies to meet: (1) present national ambient air quality standards for particulate matter and sulfur oxides, and (2) other important health/welfare-related goals, for example, possible standards for sulfates and fine particulates.

Control strategies and combinations of strategies will be analyzed with the following parameters in mind:

-- time when air quality standards and goals are to be met;

-- impacts of implementing the various strategies;

-- expectations for electrical energy generation and demand within the time period studies;

-- present technological control capabilities as well as anticipated capabilities within the time period.

Available information on the significance of nitrogen oxides from stationary sources and control technologies for these emissions will be evaluated.

Briefly, this investigation will review the technical aspects of policy options (as they have been developed in the available literature, presentations, and reports) for protecting public health and controlling air pollution problems from coal and oil-fired power plants. Existing scientific, technological and air quality data will be used; where uncertainties exist in the data base and in known effects, these uncertainties will be delineated.

The Academy is pleased to be of assistance to the Committee in these matters.

Sincerely,

PHandler

Philip Handler
President

plans which would reduce emissions so as to achieve these ambient air quality standards. EPA has also established standards of performance for new stationary sources; the standards for fossil fuel-fired steam generators can be found on p. xliv.

The control of sulfur oxide emissions from stationary sources is clearly a controversial topic. A glance at the daily newspapers demonstrates the extent of the controversy. Full page advertisements debate the virtues and deficiencies of systems of control, and the debate is made all the more immediate by urgent problems of energy supply and demand.

The task of the National Research Council has been to examine and evaluate the existing data. The goal has been to present to the Public Works Committee scientific judgments concerning the state of knowledge of the effects of emissions from stationary sources and of the techniques and strategies available for their control.

Information concerning neither the magnitude of the deleterious effects of sulfur oxide emissions, nor the atmospheric chemistry of sulfur oxides, nor the control of emissions has been found to be sufficiently reliable and extensive to permit resolution of the attendant controversies with a high degree of confidence. This report indicates that there are considerable uncertainties concerning the extent of the harmful effects of sulfur oxide emissions, the molecular species responsible for the effects, and the specific relationships between point source emissions of sulfur dioxide and regional patterns of formation, dispersion, and deposition of sulfates. Based upon the Community Health and Environmental Surveillance System studies conducted by the U.S. Environmental Protection Agency, there appears to be an association between a level of sulfate of 8 to 12 micrograms/cubic meter in the ambient air and adverse health effects in elderly persons with heart and lung disease and persons with asthma. Somewhat higher levels of sulfate (13 to 15 micrograms/cubic meter) appear to be associated with increased prevalence of chronic bronchitis in adults, increased acute lower respiratory disease and decreased lung function in children, and increased frequency of acute respiratory disease. Within these broad categories practically every individual would be included, inasmuch as most persons suffer from symptoms of respiratory disease at some time. The studies upon which these statements are based need further verification.

Although recent information concerning the performance of scrubbers intended to remove sulfur dioxide from flue gases is encouraging, some additional experience, with opportunity for design improvement, would greatly enhance public and private confidence in the large and costly decisions which must be made. Nevertheless, there are strong suggestions that the benefits of abating emissions of sulfur oxides would be substantial and, for certain plants which affect areas where there already are high ambient concentrations of sulfur dioxide and suspended sulfate, substantial abatement costs would appear justifiable. Accordingly, Part Two of this report recommends that high priority should be given to emission abatement for power plants in and close upwind of these areas and that lower priority should be given to power plants far upwind from these areas.

This report suggests that there is a need for flexibility of response which can take into account specific conditions at each power plant. Innovative instruments of policy should be considered with particular attention to their application in conjunction with various technologies for control of pollutants. For example, an emissions charge on sulfur oxides might be contemplated as an immediate incentive to undertake control activities.

The studies and findings in this report on Air Quality and Stationary Source Emission Control taken with the earlier examination of Air Quality and Automobile Emission Control emphasize the need for an integrated study of air pollutants, their sources, and their effects. Synergistic effects among pollutants were discussed in the Automobile Emission Report which noted the need to understand the health effects of various combinations of pollutants. The present report also indicates the need for a better understanding of the effects of both sulfur oxides and nitrogen oxides on the acidity of precipitation, and it notes the dependence of atmospheric chemical reactions of sulfur-related pollutants upon the concentration of other pollutants (such as oxidants and nitrogen oxides) in the ambient air.

The major emphasis in this report is upon the effects and control of sulfur oxide pollutants, and useful and important conclusions can be drawn from the analyses. But the report would be remiss if it did not remind the reader that examination of part of a larger problem may lead to partial, perhaps even incorrect, solutions. It may be necessary to implement partial solutions, but ultimately the effects of all pollutants and the techniques for their abatement, individually and in combination, must be examined so that a coherent program for the control of air pollution may be developed.

SCOPE OF THE REPORT

The Commission on Natural Resources has reviewed all of the contributions to this report and is responsible for the findings and recommendations recorded in the summary.

Part One: Health and Ecological Effects of Sulfur Dioxide and Sulfates (Chapters 1 through 5) was prepared under the auspices of the Assembly of Life Sciences. This part of the report updates work included in the September 1974 report on <u>Air Quality and Automobile Emission Control</u> (Chapter IV of Volume 2) which examined effects of airborne particles and sulfur oxides on health. Chapters 1 through 4 were prepared for the ALS by Bernard Goldstein of the New York University Medical School; Chapter 5 was contributed by Ian Nisbet of the Massachusetts Audubon Society. John Redmond, Jr., served as staff officer to the Assembly of Life Sciences.

Part Two: Strategies for Controlling Sulfur-Related Power Plant Emissions (Chapters 6 through 13) was developed under the auspices of the Committee on Public Engineering Policy, Assembly of Engineering. To prepare this part, COPEP established a Review Committee on Air

Quality and Power Plant Emissions (see Part Two) under the chairmanship of Donald Katz of the University of Michigan. Strategies for the control of sulfur oxides and particulates are examined in detail in this part of the report. The impact of such strategies on both urban and rural sulfur dioxide and sulfate concentrations is emphasized along with the timing and costs of implementing them. Part Two focuses on air pollution problems associated with electric utility generation, particularly in the northeastern United States, the geographical area most affected by coal-burning power plants. Sections of this part are based on the work of individual consultants or staff members; the members of the Committee reviewed the working drafts and provided overall guidance. The discussion of the relationship of emissions to ambient air quality in Chapter 6 is based on the work of John Trijonis, of TRW, Inc. The acid rain discussion in Chapter 7 is the work of Ian Nisbet, who was also a member of the Review Committee. Chapter 8 on efficient pricing is drawn from work by Alfred Kahn, also a Review Committee member. Elias Gyftopoulos of MIT and T.F. Widmer of Thermo Electron Corporation drafted Chapter 9. Chapter 10 on improving conversion efficiency, fuel shifting, and fuel preparation is based on the work of Harry Perry of Resources for the Future. Chapter 11, flue gas desulfurization, draws on the work of Leigh Short of the University of Massachusetts and Arthur Squires, a Review Committee member. The discussion of tall stacks and intermittent control systems in Chapter 12 was prepared by Robert Dunlap of Carnegie Mellon University; and the discussion of alternative control strategies in Chapter 13 draws on analyses carried out for the Review Committee by D. Warner North and Miley Merkhofer of the Stanford Research Institute. These analyses were used by the Review Committee in drafting the section entitled "Part Two in Brief" and in forming its conclusions; and because of their scholary merit and pertinence to the subject matter of this study, the contributions of the various consultants and committee members were considered by the Review Committee to be essential inclusions in the report. Laurence I. Moss and Ronald J. Tipton served as staff officers to the Review Committee.

Part Three: Control of Nitrogen Oxides from Stationary Sources (Chapters 14 and 15) was prepared under the direction of the Commission on Natural Resources. It examines the relative contribution of various sources of nitrogen oxide emissions and reviews the techniques of nitrogen oxide emission control. (The health effects of nitrogen oxides and their reaction products were discussed in the September 1974 report on Air Quality and Automobile Emission Control and are not considered in this report on stationary source control. Trends in nitrogen oxide and oxidant air quality were also discussed in the earlier report and are not repeated here.) The discussions of nitrogen oxide sources in Chapter 14 and of tall stacks and intermittent control for nitrogen oxides in Chapter 15 are based on analyses by John Spengler, Anthony Cortese, and Douglas Dockery of the Harvard School of Public Health. The examination of control techniques in Chapter 15 is based on the work of Adel Sarofim and Richard Flagan of the Massachusetts Institute of Technology. Raphael G. Kasper served as staff officer to the Commission on Natural Resources.

SUMMARY

This summary highlights the principal findings and conclusions of the report. The reader is encouraged to examine the basis for each of the findings and conclusions as it is presented in detail in the body of the report.

SUMMARY OF PART ONE: HEALTH AND ECOLOGICAL EFFECTS OF SULFUR DIOXIDE AND SULFATES

(1) Adverse consequences to health from combustion of sulfur-containing fossil fuels cannot be simply ascribed to any one sulfur oxide acting alone. (The term sulfur oxide is used to mean the family of compounds including sulfur dioxide, sulfur trioxide, sulfuric acid, and various sulfate salts. Sulfur dioxide is the main sulfur oxide directly emitted by fossil fuel combustion.) Sulfur dioxide itself appears unlikely to be the direct cause of excess morbidity and mortality associated with stationary source fossil fuel combustion. However, levels of sulfur dioxide close to the current ambient air quality standards may be responsible for deleterious effects on health when inhaled in combination with respirable (very small) particulate matter or the oxidant air pollutant ozone. Oxidation products of sulfur dioxide, including sulfuric acid and suspended particulate sulfates, are more toxic than the parent compound and appear likely to be responsible for a substantial portion of adverse effects on health associated with stationary source combustion of fossil fuels. (See Chapter 3.)

(2) The processes governing the conversion in the atmosphere of sulfur dioxide to sulfuric acid and suspended sulfates are complex and

incompletely understood. It is clear, however, that the oxidation of sulfur dioxide is accelerated in the presence of other pollutants, particularly trace metals derived mainly from stationary source fossil fuel combustion, and components of photochemical smog derived primarily from automotive emissions. (See Chapter 2.)

(3) The specific chemical species responsible for toxicity have not been identified, and the levels of pollutants necessary to cause toxic effects have not been determined. This hampers the exact determination of the morbidity and mortality resulting from sulfur oxides. The use in epidemiological correlations of monitoring data for total suspended particulates and sulfur dioxide has undoubtedly led to imprecision inasmuch as these two measurements do not directly assay the causative agents. It is possible that the use of these indicators may have led to underestimation or overestimation of the health consequences of sulfur oxide or respirable particulate matter, but most likely underestimation. Particles in the respirable size range undoubtedly play a role in morbidity. (See Chapter 2.)

(4) In the next few years additional information will probably be available that will permit establishment of rational air quality standards for respirable particulate matter and suspended sulfates in order to protect the public. This will require substantial advances in monitoring and analytical techniques as well as improved assessment of health hazards. (See Chapter 2.)

(5) Review of the available data suggests that it is reasonable to predict that an increase in morbidity and mortality of susceptible individuals would result from an increase in ambient sulfur dioxide to levels appreciably above the current air quality standard. (At present, levels generally do not exceed this standard.) Susceptible groups represent a substantial fraction of the U.S.

population and include children, the elderly, asthmatics, and individuals with chronic cardiovascular and pulmonary disease. (See Chapter 4.)

(6) In view of the societal and economic impact of controlling sulfur oxide emissions, it is of major importance to quantify the health effects associated with sulfur oxides and the benefits to be gained by avoiding those effects. A number of attempts to do so have been made, and they are reviewed in this report. Analysis of data developed from the CHESS studies of the Environmental Protection Agency provides some guidance in defining the limits of the problem, but these and other currently available data on the health effects of sulfur oxides should be viewed with caution. (See Chapter 4.)

(7) Definition of a no-effect level (threshold) for the acute effects of sulfur oxides is difficult. It is not clear that there is any level of these pollutants above background that will not have an acute effect on the most susceptible individuals. However, it is beyond present knowledge to state whether acute responses to low levels are non-injurious adaptive responses or are responses which, often repeated, might lead to eventual respiratory impairment. High priority should be given to research evaluating the long-term physiological effects of sulfur oxide air pollution. (See Chapter 2.)

(8) Sulfur oxide emissions from catalyst-equipped automobiles may take the form of sulfuric acid mist or suspended sulfates. Since the emissions are at ground level, they are likely to be encountered by individuals. If an increasing proportion of the automobile fleet is catalyst equipped, these sulfur oxide emissions may constitute an appreciable part of the total sulfur oxides inhaled and a significant proportion of the threat to health from these compounds. As yet there are no direct experimental data on the health effects of the particular sulfur oxides emitted from automobiles. In the absence of experience and

sufficient data, we are unprepared to compare quantitatively the possible deleterious health effects arising from catalyst operation with the health benefits to be gained by the oxidation of hydrocarbons and carbon monoxide in automotive emissions. (See Chapter 2.)

(9) The visible direct effects of high concentrations of sulfur dioxide on susceptible species of plants have been recognized for many years. Knowledge of whether lower concentrations decrease productivity of natural flora and fauna is very meager. The ecological consequences of the increasing acidity of precipitation deserve thorough study. The increase in acidity of precipitation appears to be related to increased emissions of sulfur dioxide and nitrogen dioxide although the exact relationship needs further study. (See Chapter 5.)

(10) Identifiable effects of acid precipitation include acidification of soils, reduction in forest productivity, and depletion of fresh water fish populations. Materials, buildings, and various substances are degraded by sulfur oxides in the air and by acid precipitation. The full impact of these effects may not be felt for a number of years. Rough estimates of their likely magnitude suggest that they are relatively modest in economic terms, but they may also involve loss of recreational opportunity and aesthetic values. The possibility of additional effects, such as reduction in agricultural productivity or extensive injury to valuable ornamental plants, cannot be dismissed. (See Chapter 5.)

(11) Atmospheric hazes, attributable in large part to fine particulates including sulfates, are widespread in the eastern United States during the summer; their frequency appears to be increasing as emissions increase, and they may have effects on weather and climate. (See Chapter 5.)

(12) With the protection of human health as the goal, it is desirable to limit the atmospheric emission of sulfur oxides and respirable particulate matter. (See Chapter 4.)

SUMMARY OF PART TWO: STRATEGIES FOR CONTROLLING SULFUR-RELATED POWER PLANT EMISSIONS

Sulfur Oxide Emissions and Abatement Techniques

Reasons for Reducing Emissions of Sulfur Oxides

(1) Part One of this report points out that adverse consequences to health from combustion of sulfur-containing fossil fuels cannot be ascribed to any single sulfur oxide acting alone, and that sulfur dioxide itself appears unlikely to be the sole cause of excess morbidity and mortality associated with pollution in the form of sulfur oxides and suspended particulate matter. In particular, certain oxidation products of sulfur dioxide, including sulfuric acid and certain suspended particulate sulfates, are more toxic than the parent compound and may be responsible for a substantial portion of the adverse health effects associated with sulfur oxides and particulate pollution, a portion of which arises from stationary source combustion of fossil fuels. In the study of control strategies in Part Two of this report, the concentration of airborne particulate sulfates has been used as an index of pollution hazard. (See Part I.)

(2) Within a large region such as the northeastern United States, particulate sulfate concentrations in the atmosphere are related to regional emissions of sulfur dioxide, which is converted to sulfates after emission. Because sulfur dioxide and sulfates may be transported long distances before being removed from the atmosphere, and because during the transport period there is conversion of sulfur dioxide to sulfates, there is not always a close relationship between ambient concentrations of sulfates and emissions of sulfur dioxide in the

immediate vicinity. For example, in some rural areas in the Northeast where there are comparatively low sulfur dioxide emissions and low ambient sulfur dioxide levels, ambient sulfate concentrations are substantially above background levels. (See Chapter 6.)

(3) Concentrations of sulfates in airborne particulate matter are difficult to measure but appear to have increased. The amounts of sulfates deposited in rainfall have been increasing in parallel with the increase of emissions of sulfur dioxide. The acidity of precipitation in the eastern states has also been increasing; this is attributable to an increase in emissions of both sulfur dioxide and nitrogen oxides. (See Chapters 6 and 7.)

(4) In addition to the adverse effects on health from these pollutants, there are damages to materials, decreases in property values, and impairment of agriculture, forestry, and ecosystems. These effects point to the desirability of controlling the amount of man-produced sulfur compounds emitted into the atmosphere. (See Chapters 5 and 7, and Appendix 13-E.)

Power Plants a Major Source of Sulfur Oxide Emissions

(5) Steam electric generating plants that burn coal are major sources of sulfur oxide emissions, especially in the northeastern U.S. In this region, the quantity of anthropogenic sulfur oxides almost certainly exceeds the amount emitted from natural sources such as decaying vegetation. On a nationwide basis, more than 50 percent of total anthropogenic sulfur oxide emissions are produced through combustion of coal in power plants; in some regions the percentage is much higher. Other sources of sulfur oxides include space heaters, smelters, and industrial boilers. Information recently brought to the Committee's attention suggests that oil-fired combustion, particularly in small combustion devices such as home

furnaces, appears to be an important source of sulfuric acid emissions. Coal fired electrical generation may increase as much as 100 percent, possibly more, over the next decade. The resulting power plant emissions, if not controlled, may lead to increases in ambient sulfate levels in urban areas on the order of 10 to 40 percent as well as to increases in ambient sulfur dioxide levels and in the acidity of precipitation. (See Chapters 6 and 7, and Appendix 11-A.)

Emissions from Power Plants Depend in Part on the Use of Electricity, Which Depends on Many Factors.

(6) The Review Committee recognizes the national determination to decrease U.S. dependence upon imported petroleum fuels. Achievement of this goal is likely to entail a substantial increase in the use of coal for the generation of electricity. There are two reasons for this: more electricity will be required to permit reductions in the use of oil, and a larger fraction of the electricity produced will be generated from coal. The shortage of natural gas will also result in increased use of coal for the same reasons. The consequence of these changes could be a large increase in sulfur dioxide emissions. (See Chapter 6.)

(7) A major current use of oil and gas is for domestic and office space heating. Electrical energy could replace the direct use of oil and gas for space heating, but this could result in additonal burning of coal and accompanying emissions of sulfur oxides. Therefore, if such a change were contemplated, it would be desirable to employ space heating technology which would make efficient use of electricity. A shift to the use of electrical resistance heating would increase the overall consumption of fuel; however, there would be little, if any, increase in the overall consumption of fuel if the shift were, instead, to electrically-powered heat pumps, where local

weather conditions make this technically possible. Moreover, it is estimated that the total capital investment (at the point of generation as well as at the point of use) required for the heat pump system is less than that for the electrical resistance heating system. The alternative of producing combustible gas from coal to serve the heating market also appears to require substantially less capital and less total fuel consumption than electrical resistance heating. There are additional options for meeting some of the nation's space and water heating requirements, e.g., solar energy and possible new domestic petroleum and gas resources. Although each of these options will probably take at least three to five years before it can add significantly to incremental supplies, an examination of them should be included in a comprehensive analysis. (See Chapter 9.)

(8) Conservation and improved fuel use can substantially reduce the rate of growth in demand for electrical energy and in this way reduce the amount of coal used and the sulfur dioxide emitted in its generation. Improvements in fuel use can be accomplished by: (1) improved effectiveness of electrical apparatus used in the residential, commercial, and industrial sectors; and (2) on-site generation of electricity as a by-product of certain industrial processes. (See Chapter 9.)

(9) In a market economy, where consumption decisions are made by individual purchasers, economic efficiency requires that the price reflect the incremental cost of supply to society. This means, in an expanding industry, that price should equal the cost of obtaining new or replacement supplies of the resource, including social and environmental costs. If such prices are not charged, then either too much or too little of the resource will be used in relation to other resources. The rate at which fuel-saving technology will actually be applied depends upon the rate of pay-back for capital invested in such technology. One critical factor currently retarding the appli-

cation of new and more efficient technology is the manner in which fuel and electricity costs are determined. The price a user pays for energy is generally based on average costs of production rather than on incremental costs, which tend to be much higher. Similarly, the higher cost of supplying electricity during hours of peak system demand is not sufficiently reflected in rate structures. A fuller application to electricity rates of these incremental cost pricing principles could make a substantial contribution to conservation and thereby to reducing sulfur oxide emissions. (See Chapter 8.)

Flue Gas Desulfurization (FGD) Technology

(10) For power plants that burn low-sulfur (less than 1 percent sulfur) coal, either lime or limestone scrubbing is the most effective method now available for reducing emissions of sulfur dioxide in flue gases. Emissions of sulfur dioxide from such sources can be reduced by at least 90 percent with these methods. Successful operation has been demonstrated on commercial scale modules of 115 Mw for lime scrubbing and 170 Mw for limestone scrubbing. (See Chapter 11.)

(11) For power plants that burn medium or high-sulfur coal, lime scrubbing is the most effective near-term method for reducing emissions of sulfur dioxide in flue gases. Reductions of at least 90 percent have been achieved with this method. Successful operation of a lime scrubber in the desirable closed loop mode (i.e., with no release to the environment of water from the process) in a power plant burning medium-sulfur, low-chlorine coal has been demonstrated on a commercial scale. The demonstration power plant is a peaking unit producing flue gas equivalent to about 100 Mw capacity. Although it ordinarily operates intermittently, in these trials it has operated continuously, following changes in load typical of many utility boilers. The 100 Mw size is typical of the module that designers may provide

in multiples for dealing with the flue gas from boilers of much larger capacity. The chlorine content of coal affects successful scrubber operations for at least three reasons: (1) it contributes to the acidity of sludge slurries; (2) it retards the rate of oxidation of calcium sulfite to calcium sulfate in slurries; and (3) it leads to materials corrosion in stack equipment as a result of hydrogen chloride formation. Successful operation of a lime scrubber on flue gases arising from the burning of a high-sulfur coal of medium or high chlorine content has been observed on a bench and pilot-plant scale (1.0 and 10 Mw equivalent capacity), also in a closed-loop mode. Experience on a commercial scale can and should be obtained quickly for medium- and high-sulfur coals containing chlorine beyond 0.04 percent. This experience is necessary to resolve the question of the commercial availability of lime scrubbing technology for all coals. That this question will be resolved favorably is a matter for engineering judgment in light of the available chemical knowledge and performance comparisons. Some of the Committee members judge the probability to be 90 percent, while one member judges it to be 70 percent. (See Chapter 11.)

(12) The Committee is well aware of the problems which have been experienced in early installations of flue gas desulfurization processes. This discouraging experience has caused many in the electric utility industry, as well as others, to doubt the feasibility of the technology. The Committee finds, however, that there has been rapid advance in the understanding and application of scrubbing technology especially in the past year, and urges those who are skeptical to review this recent experience. If scrubbers ordered today are to operate as reliably as other components of the power generating system, they will require very careful engineering and initial "trouble shooting" operating procedures which may be extensive. There is a reasonable expectation that, in the near future, scrubbers will be available for purchase as routine components of power systems covering a wide

range of specific conditions, provided a vigorous development program is pursued. (See Chapter 11.)

(13) The large quantity of waste by-product associated with lime and limestone scrubbing is a disadvantage of these processes. Sludge storage using lined ponds has been successful for lime and limestone flue gas desulfurization installations. The availability of space near the power plant is an important factor in determining the feasibility of sludge storage using lined ponds and may substantially increase the cost of retrofitting plants already located in urban areas because of the need for transportation of sludge. The problem of ultimate disposal is being attacked by fixation of sludge to produce a material of low permeability and leachability, suitable for landfill and as a base for roads. Tests on commercial-scale modules are now underway in several locations. The results are reported to be promising. (See Chapter 11.)

(14) Installation of a lime scrubbing process for a new power plant burning high-sulfur eastern coal will require an added capital investment of about $100/kw (all costs in 1974 dollars). The investment could be as low as $60/kw and as high as $130/kw. Added operating costs, including capital charges, will range from 2 to 5 mills/kwh plus a cost of about 1 mill/kwh for energy loss and capacity derating. This is 15 to 30 percent of the total cost of generating power at a new plant. Sludge fixation, or sludge transportation, if needed, will add to these costs. (See Chapters 11 and 13.)

(15) A longer term approach to the problem of sulfur emissions involves conversion of the sulfur dioxide into elemental sulfur and regeneration of the scrubbing liquor. This would eliminate the problem of disposal of large quantities of sulfites and sulfates of lime and, instead, provide easily stored and commercially valuable elemental sulfur. There is successful commercial operating experience with regenerable

flue gas desulfurization processes on stack gases arising from the combustion of oil. There is no commercial operating experience using a regenerable process to treat stack gases arising from combustion of high-sulfur coal. A few such plants are under construction, and large-scale plant testing of such systems is now underway. Because the potential advantages of regenerative processes are significant, a careful evaluation of the desirability of increasing the funds being devoted to their development is warranted. (See Chapter 11.)

(16) About 70 percent of existing plant capacity may be retrofitted with lime or limestone FGD systems. The cost of retrofitting is normally 25 to 30 percent higher than the cost of installing scrubbers on a new plant of equal capacity. Older, smaller existing plants are more difficult and costly to retrofit than newer, larger existing plants. (See Chapter 11.)

Use of Low-Sulfur Coal and Washing of High-Sulfur Coal

(17) Most of the coal from eastern resources has a high sulfur content. Of the more limited eastern low-sulfur coal, a large fraction is held by owners who have dedicated these resources to metallurgical use. Washing of the high-sulfur eastern coals could, on average, reduce their sulfur content by about 40 percent (i.e., from a sulfur content of 3.5 percent to 2.1 percent). The amount of such reduction depends on the characteristics of the particular coal used. Coal washing will be of some benefit in reducing emissions of sulfur oxides. Conventional physical cleaning will reduce the total sulfur emitted by power plants by significant amounts (of the order of 40 percent overall) but it will not, in general, result in coals with a sulfur content meeting the New Source Performance Standards for SOx. However, coal preparation can reduce the sulfur content to levels that are permitted by the State

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Implementation Plans for existing plants in some rural areas. (See Chapter 10.)

(18) Large quantities of low-sulfur coal are found in the western U.S. Although new plants can be designed to burn these coals efficiently, in existing plants the boilers must be derated (i.e., operate at less than design capacity) to operate satisfactorily. Moreover, the transport of low-sulfur western coals to eastern markets will require construction of major additonal transport facilities. (See Chapter 10.)

(19) By shifting available low-sulfur coal away from plants that could burn higher sulfur coal and still meet ambient sulfur dioxide standards to plants in regions not meeting ambient standards for sulfur dioxide, some improvement in compliance with applicable ambient air quality standards for sulfur dioxide could be achieved. However, such shifting of low-sulfur coal will not reduce the total amount of sulfur in the atmosphere; therefore, this strategy should be considered at best an interim measure only. The potential for increasing ambient sulfate concentrations in downwind areas should be carefully assessed before any such strategy is implemented. (See Chapters 6, 7, 10, and 13.)

(20) Caution must be exercised in the substitution of low-sulfur coal in existing power plants, since there is a resultant risk of increasing emissions and ambient concentrations of particulate matter. The efficiency of devices for the removal of particulate matter, especially electrostatic precipitators, is decreased for fly ash from low-sulfur coals. Hence, a decrease in sulfur dioxide by fuel substitution may result in greater emissions of particulate matter, including any trace metals present in the coal. Such problems could be minimized by appropriate modification of equipment or process conditions or both. (See Chapter 10.)

Tall Stacks and Intermittent Control Systems (ICS)

(21) Tall stacks and/or intermittent control systems make it possible to meet ambient sulfur dioxide standards in carefully defined situations. The attractiveness of these systems lies in their low operating and capital costs, estimated to be 0.15 to 0.4 mill/kwh and $4 to $10/kw, respectively. The Committee does not recommend their use unless it is for carefully defined situations for an interim period until other strategies (e.g., flue gas desulfurization, low-sulfur fuel) can be implemented, or until further data are accumulated which would justify making them permanent. The application of tall stacks and/or intermittent control systems will not reduce total emissions of sulfur oxides to any significant degree; thus, this strategy does not decrease the total amount of sulfate in the regional atmosphere. The potential for increasing ambient sulfate concentrations in downwind areas should be carefully considered in advance, and effects on ambient concentrations monitored, if such a strategy is implemented. (See Chapter 12.)

Recommended Decisions and Decision Processes for Abatement Strategies

(22) Methodology is available for analyzing decisions among abatement alternatives: the deleterious consequences of sulfur oxide emissions to human health, ecological systems, material property, and aesthetic values should be evaluated and compared (for each power plant in a region) with the additional cost imposed on the generation of its electricity by the abatement methods. The calculation thus involves a comparison of marginal cost with marginal benefit for each power plant within the regional system of electric generation and air quality. However, conclusions drawn from the application of this methodology must of necessity reflect the adequacy of the information available at the time of application; at the present time uncertainty in

many critical variables and relationships severely limits the conclusions that can be drawn as to the best strategy alternatives for controlling sulfur oxide emissions. By assessing in probabilistic terms the respective costs and benefits for various alternative strategies, the methodology can indicate where, on the basis of the limited information available, stringent control is desirable and where more information would be advisable before a commitment to a particular emissions control strategy is made. (See Chapter 13.)

(23) There are considerable uncertainties concerning the extent of the harmful effects of sulfur oxide emissions, and concerning the specific relationships between point source emissions of sulfur dioxide and regional patterns of formation, dispersion, and deposition of sulfates. Any policy adopted now, therefore, should be reviewed periodically in the future and may have to be changed as a result of new findings. Nevertheless, the calculations shown in Chapter 13 suggest that the benefits of abating emissions of sulfur oxide may exceed the costs substantially for plants which affect areas where there are already high ambient concentrations of sulfur dioxide and suspended sulfates, such as urban areas in the Northeast. In addition, the Committee places importance on considerations of prudence; the consequences of an error in judgment which led to substantial damage to human health would be more serious than an error which led to an economic misallocation. Accordingly, the Committee recommends that high priority should be given to emission abatement from power plants in and close upwind of urban areas. Although the analysis in Chapter 13 indicates that lower priority should be given to power plants far (of the order of 300 miles) upwind from major cities, it also indicates that external costs imposed by emissions from these plants may be substantial. Since the capacity for installing flue gas desulfurization systems is limited, there will be a continuing opportunity to review the costs and benefits of

emission controls for plants now assigned to a low priority. (See Chapter 13.)

(24) National capacity to produce stack gas scrubbing equipment is limited. Further advances in applicable technology are expected to occur in the next few years. Scrubbing equipment should be installed first in those situations where its additional benefits in emissions abatement are judged to be highest with respect to its additional costs. All new plants, including those able to meet New Source Performance Standards without the use of scrubbers, should at least be constructed so as to permit subsequent retrofitting of flue gas desulfurization systems, since the cost of allocating space for that purpose is low. In time, the increase in coal use and further information on the effects of sulfur oxide emissions may indicate a need for a greater degree of emissions reduction. (See Chapter 13.)

The Value of Resolving Uncertainties
on the Effects of Sulfur Oxide Emissions

(25) Decisions about control strategies depend upon the information available at the time the decisions are made. A better understanding of the effects of suspended sulfates on health and of the chemistry of the atmospheric conversion of sulfur dioxide to sulfate could have a significant effect upon future decisions about sulfur oxide emissions abatement. Improving the available information about these aspects of sulfur emissions has an expected value on the order of hundreds of millions of dollars a year, which is at least ten times greater than the cost of a research program to resolve these uncertainties in approximately five years. (See Chapter 13.)

(26) Current assessments of the benefits of sulfur oxide emissions reduction for human health, ecological systems, materials, and aesthetic values could be greatly improved. Substantial efforts should be made to develop

improved models and data for use on a case-by-case basis to improve decisionmaking on emissions control strategy alternatives. There is also a need to investigate the distribution of costs and benefits among different individuals within society, and the effects of emissions controls and pricing policy on this distribution. (See Chapter 13.)

SUMMARY OF PART THREE: CONTROL OF NITROGEN OXIDES FROM STATIONARY SOURCES

(1) The quantity of nitrogen oxide produced by human activity throughout the world is on the order of 10 percent of the nitrogen oxide produced from natural sources. However, the anthropogenic nitrogen oxide emissions are concentrated in populated areas and are thus of concern in pollution control programs. (See Chapter 14.)

(2) National nitrogen oxide emissions have grown at an average rate of over 4 percent per year for the last three decades. (See Chapter 14.)

(3) At present, stationary source fuel combustion accounts for about half of all U.S. nitrogen oxide emissions, and electric power generation represents 24 percent of U.S. nitrogen oxide emissions. (See Chapter 14.)

(4) The nitrogen oxide emission rate per unit heat produced is greater from coal than from either oil or natural gas. Therefore, conversion of existing plants to permit the burning of coal and use of new coal-fired electric generating plants would increase nitrogen oxide emissions at a greater rate than that projected from historic trends. (See Chapter 14.)

(5) Transportation is the second largest source category, contributing 35.4 percent of the U.S. total nitrogen oxide emissions. (See Chapter 14.)

(6) Projections of future nitrogen oxide emissions demonstrate that, if present statutory standards are adhered to, stationary sources will contribute an increasing percentage of total nitrogen oxide emissions through 1990. (See Chapter 14.)

(7) There are geographical differences in nitrogen oxide emissions which reflect the distribution of industry, electric power generation, and population. Fifty-six percent of the national nitrogen oxide emissions are produced in the northeast states (EPA Regions I, II, III, and V). (See Chapter 14.)

(8) Thirty-nine percent of all U.S. nitrogen oxide emissions are generated in the 10 largest urban areas. In fact, 25 percent of the total U.S. emissions are produced in the five largest urban areas. This reflects the dominance of stationary fuel combustion and industrial process emissions. Only 22 percent of the nation's transportation-related nitrogen oxide is emitted from the 10 largest urban areas. Thus, in many urban areas, nitrogen oxide emissions from stationary sources are the dominant factor in determining ambient concentrations of this pollutant. (See Chapter 14.)

(9) There are considerable uncertainties in the 1972 nitrogen oxide emissions data as reported by the National Emissions Data System (NEDS). For example, examination of the data indicates that industrial process losses are probably significantly underestimated. (See Chapter 14.)

(10) Typically, within combustors nitrogen oxide is formed in localized, high-temperature regions by the oxidation of both atmospheric nitrogen (thermal NOx) and nitrogen that may be contained in the fuel (fuel NOx). (See Chapter 15.)

(11) The formation of nitrogen oxide in combustion systems can be suppressed, with varying degrees of success, by reducing the

oxygen content and temperature in the localized regions of the furnace contributing to emissions, usually in the vicinity of the flame. Reductions in the oxygen content in the flame zone reduce the emissions of both fuel and thermal NOx; reductions in temperature, however, produce significant reductions in only the thermal NOx. (See Chapter 15.)

(12) Methods that have been used to reduce the temperature in the combustor include : (a) injection of cooled combustion products, steam, or water into the flame volume; (b) reduction of the temperature to which combustion air is preheated; and (c) extraction of heat from the flame volume. (See Chapter 15.)

(13) Methods for reducing the oxygen content in the flame zone involve lowering the volume of air supplied to the burners by reducing the overall air/fuel ratio to the combustor (low-excess-air firing) or by reducing the air/fuel ratio for some burners without reducing the overall air/fuel ratio (staged combustion). (See Chapter 15.)

(14) Low-excess-air-firing, staged combustion, flue-gas recirculation, water injection, and reduced air preheat are control techniques that have been successfully demonstrated on utility boilers. The latter two methods, however, have an associated, usually unacceptable, penalty in thermal efficiency. Using combinations of the techniques listed above, an average reduction in nitrogen oxide emissions of 60 percent has been achieved for gas-fired utility boilers, 48 percent for oil-fired boilers, and 37 percent for coal-fired boilers. (See Chapter 15.)

(15) The applicability of combustion process modification to existing furnaces must be evaluated on a case-by-case basis. In general, boilers can be adapted for low-excess-air firing and staged combustion without major modification. Flue-gas recirculation may be

impractical on some existing units. (See Chapter 15.)

(16) The capital costs of nitrogen oxide emission reduction in utility boilers vary widely with specific installation size and design. They range from under $0.50/kw for staged combustion to $6.00/kw for flue-gas recirculation on existing units, and from near zero for staged combustion to $4.00/kw for flue gas recirculation on new units. (See Chapter 15.)

(17) The level of control achievable on industrial boilers is close to but not as great as that attainable with utility boilers. (See Chapter 15.)

(18) Reduction in nitrogen oxide emissions from stationary engines is possible, although such reduction is often accompanied by significant increases in fuel consumption. New engine designs may produce substantial reductions in nitrogen oxide emissions without increasing fuel consumption, but futher development of such designs is required. (See Chapter 15.)

(19) Fluidized bed combustion of coal provides a potential alternative to current utility boiler design. Tests on laboratory and pilot-scale fluidized bed combustors have yielded emissions that meet the current standards for new coal-fired units. Tests on larger scale units are needed to establish practical emission levels for commercial applications. (See Chapter 15.)

(20) The only intermittent control strategy that appears practical for NOx emission reduction is load switching of electric power generation. Load switching has limited applicability because of the variability in the contribution of electric power generation to local emissions. (See Chapter 15.)

(21) The advantages of tall stack release of sulfur dioxide to reduce ground level

concentrations do not apply for nitric oxide. Tall stacks potentially reduce ground level nitric oxide concentrations. However, nitric oxide converts to nitric acid and nitrates faster than sulfur dioxide converts to sulfuric acid and sulfates; and since the reaction products precipitate, there is a greater potential for local impact. (See Chapter 15.)

(22) There is considerable uncertainty about the effects of nitrogen oxide release from tall stacks on the formation of photochemical oxidants and on the ground level concentrations of oxidants and nitrogen dioxide. (See Chapter 15.)

National Primary and Secondary Ambient Air Quality Standards

Pollutant	Type of standard	Averaging time	Frequency parameter	Concentration µg/m³	Concentration ppm
Carbon monoxide	Primary and secondary	1 hr	Annual maximum[a]	40,000	35
		8 hr	Annual maximum	10,000	9
Hydrocarbons (nonmethane)	Primary and secondary	3 hr (6 to 9 a.m.)	Annual maximum	160[b]	0.24[b]
Nitrogen dioxide	Primary and secondary	1 yr	Arithmetic mean	100	0.05
Photochemical oxidants	Primary and secondary	1 hr	Annual maximum	160	0.08
Particulate matter	Primary	24 hr	Annual maximum	260	--
		24 hr	Annual geometric mean	75	--
	Secondary	24 hr	Annual maximum	150	--
		24 hr	Annual geometric mean	60[c]	--
Sulfur dioxide	Primary	24 hr	Annual maximum	365	0.14
		1 yr	Arithmetic mean	80	0.03
	Secondary	3 hr	Annual maximum	1,300	0.5

[a]Not to be exceeded more than once per year.
[b]As a guide in devising implementation plans for achieving oxidant standards.
[c]As a guide to be used in assessing implementation plans for achieving the annual maximum 24-hour standard.

Source: EPA Regulations 40 CFR 50; and Commerce Clearing House, Inc. Pollution Control Guide 1974.

New Source Standards of Performance for Fossil Fuel-Fired Steam Generators

Pollutant	Standard
Particulate matter	0.10 lb. per million BTU heat input, maximum two hour average
	20 percent opacity (except that 40 percent opacity is permissible for not more than two minutes in any hour)
Sulfur dioxide	0.80 lb. per million BTU heat input, maximum two hour average when liquid fossil fuel is burned
	1.2 lbs. per million BTU heat input, maximum two hour average when solid fuel is burned
Nitrogen oxides	0.20 lb. per million BTU heat input, maximum two hour average, expressed as NO_2, when gaseous fossil fuel is burned
	0.30 lb. per million BTU heat input, maximum two hour average, expressed as NO_2, when liquid fossil fuel is burned
	0.70 lb. per million BTU heat input, maximum two hour average, expressed as NO_2, when solid fossil fuel (except lignite) is burned

Source: EPA Regulations 40 CFR 60.42 to 40 CFR 60.44

PART ONE
HEALTH AND ECOLOGICAL EFFECTS OF SULFUR DIOXIDE AND SULFATES

Part One was prepared under the direction of the Assembly of Life Sciences of the National Research Council. Chapters 1 through 4 were written by Dr. Bernard Goldstein of the New York University Medical School. Chapter 5 is the work of Ian C. T. Nisbet of the Massachusetts Audubon Society. We are also indebted to the anonymous scientific reviewers who have contributed materially to the final form of Part One.

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CHAPTER 1
INTRODUCTION

Air quality standards for selected pollutants were mandated by the U.S. Clean Air Amendments of 1970, which aimed primarily at the protection of human health against atmospheric contaminants. The act states that among its purposes is "to protect and enhance the quality of the nation's air resources so as to promote the public health and welfare and the productive capacity of its population." The stimulus for establishing a sulfur dioxide standard was a significant body of evidence indicating that ambient sulfur dioxide was associated with adverse health effects at the high concentrations measured during recognized air pollution diasters and with less severe increase in morbidity and mortality at lower concentrations.

Much additional information has been obtained since the original air quality criteria document for sulfur oxides was published in early 1969 (USDHEW 1970). In general, epidemiologic studies have tended to confirm the association of adverse human health effects with ambient sulfur dioxide concentrations around the primary standard of 0.03 ppm (80 ug/m³)† annual arithmetic mean. The maximum 24 hour standard, not to be exceeded more than once yearly, is -.14ppm (365 ug/m³). It is generally recognized, and should be emphasized that this epidemiologic association does not necessarily imply causality. In fact, exposure of humans to sulfur dioxide in chambers has shown clearly

† Parts of this report were prepared by a computer-assisted text editing process for which the high-speed line printer does not yet provide subscripts or certain special symbols. Two conventions have been adopted as necessary throughout Parts One and Two: (a) for µ read u; (b) numerical subscripts in formulas are indicated by underscoring (e.g., SO4).

that sulfur dioxide by itself is incapable of producing an acute response in man even at concentrations considerably higher than 0.03 ppm.

This apparent discrepancy between epidemiologic observation and the results of controlled experiments is explainable by the presence in polluted air of sulfur dioxide oxidation products (such as sulfuric acid and particulate sulfates), which are potentially more toxic, and possibly by the synergistic effects of sulfur dioxide and other pollutants, including particles and ozone, within the respiratory system. It would therefore be inappropriate to use the failure to observe a human response to sulfur dioxide during controlled exposure as a reason to allow an increase in the emission of sulfur dioxide.

The literature concerning the health effects of sulfur oxides has recently been reviewed by scientific panels in the United States and elsewhere (NAS 1974, Rall 1974, Holland 1972,y NATO 1972). Those reviews have generally been organized by scientific subdiscipline, which provides a useful framework for consideration of specific points and integration of available information. They have been prepared by competent and thorough panels and are in the public record. The present document is organized by human disease processes that are believed to be related to the effects of sulfur oxides, and both animal experiments and controlled human exposures will be discussed according to disease process. It is hoped that this format will prove more intelligible to nonscientists, particularly those involved in decisions concerning clean air strategy.

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CHAPTER TWO

GENERAL CONSIDERATIONS

ATMOSPHERIC SOURCES, INTERACTIONS AND SINKS

An understanding of the sources, atmospheric reactions, and eventual sinks of sulfur oxides and particles would be of enormous value in studying the health effects of these pollutants and devising appropriate control strategies. Despite a substantial effort, which has been reviewed by a number of authors (Charlson 1974, Goldstein and Nelson 1974, Whitby 1974, Urone and Schroeder, 1969), our knowledge of these processes is far from complete.

Sulfur exists in the atmosphere generally in three forms: hydrogen sulfide, sulfur dioxide, and particulate sulfate. Hydrogen sulfide is almost completely a product of natural sources, such as anaerobic bacteria (Grey and Jensen 1972), but is also to a small extent derived from industrial activity. This gas is relatively rapidly oxidized to sulfur dioxide and, on a global basis, may be the major source of atmospheric sulfur dioxide. However, man-made sources of sulfur dioxide, particularly fossil-fuel combustion, predominate in industrialized areas and their surroundings. Sulfate in the atmosphere mainly results from the oxidation of sulfur dioxide or is derived from sea spray in the form of sea-salt particles. The latter are believed to be

somewhat larger than the oxidation products of sulfur dioxide (Kellogg et al. 1972). If so, sea-salt sulfate particles should be less important toxicologically, because they will tend to settle out of the atmosphere more quickly and will not penetrate as deeply into the respiratory tract. Almost all the sulfur oxide emitted from the stationary combustion of fossil fuels is in the form of the dioxide, but about 2-5 percent is directly released as sulfuric acid.
Fossil fuels used in automobiles generally contain relatively small amounts of sulfur and are thus insignificant as sulfur dioxide sources. However, the advent of catalytic converters will result in the oxidation of gasoline to sulfuric acid which will be released in the exhaust. This could conceivably lead to a considerable ground level of sulfuric acid, particularly in heavily traveled city streets which are often poorly ventilated. This potential problem requires careful consideration before mandating the use of catalytic converters (USEPA 1975).

A number of authors have assessed the total atmospheric sulfur balance, including sources, atmospheric residence time, and eventual sinks. At present, these are estimates, inasmuch as important pertinent information is lacking (Hill 1973). Of particular relevance to the problem of sulfur oxide control is the extent to which natural and man-made sources contribute to airborne sulfur dioxide and sulfate concentrations in the United States. It appears that the bulk of airborne sulfur dioxide and sulfate in industrialized and heavily populated areas, such as the northeastern United States, is derived from fossil-fuel combustion (Altshuller 1973). However, further refinement of the data is necessary.

The atmospheric residence times of the various sulfur oxides are not precisely known. Estimates range from hours to weeks for sulfur dioxide and longer for fine particulate sulfates. Residence time depends on such factors as rainfall, windspeed, and temperature. It should be noted that climatologic conditions affect the deposition of gaseous sulfur dioxide and of particulate sulfates differently. Furthermore, particle size and density, which also depend in part on weather, will affect the rate at which particles settle out of the atmosphere. Presumably, the height at which pollutants are emitted into the atmosphere will also modify sulfur oxide residence times. Although the exact residence times have not been established, there is ample evidence from European and American studies to indicate that sulfur dioxide and its oxidation products may be distributed up to hundreds of miles downwind from emission sources. With the possible exception of unusual atmospheric conditions in highly polluted areas, the average residence time of sulfuric acid is presumably very short.

The eventual sinks for sulfur oxides are not fully evaluated. There is some evidence that the oceans are major endpoints for the increased global output of sulfur compounds resulting from modern industrialization (Spedding 1972). Rain plays an important role in removing atmospheric sulfur oxides, but the resulting increase in raindrop acidity is believed to have affected vegetation in the United States and northern Europe adversely. Sulfur oxides returned to the soil may be reduced to hydrogen sulfide by anaerobic bacteria and released again into the atmosphere (Abeles and Craker 1971). It is therefore inappropriate to ascribe to natural sources all the sulfur dioxide produced by atmospheric oxidation of hydrogen sulfide until further information is available concerning the extent to which fossil-fuel sulfur oxide deposited in the soil is recycled. This would most likely be inconsequential on a global basis, but it might have some impact in regional air quality considerations.

Sulfur dioxide in the atmosphere is subject to a complex series of chemical reactions. For the purpose of discussion of potential health effects, a simplified reaction scheme can be written:

$$SO_2 + (O) \longrightarrow SO_3 \qquad (1a)$$

$$SO_3 + H_2O \longrightarrow H_2SO_4 \qquad (1b)$$

$$H_2SO_4 + X^{++} \longrightarrow XSO_4 = 2H^+ \qquad (2)$$

$$H_2O + SO_2 \longrightarrow H_2SO_3 \longrightarrow H^+ + HSO_3^- \qquad (3)$$
$$\longrightarrow 2H^+ + SO_3^=$$

In reaction 1a, sulfur dioxide, SO_2 is oxidized to sulfur trioxide, SO_3 the anhydrous form of sulfuric acid, H_2SO_4. Inasmuch as gaseous sulfur trioxide reacts almost instantaneously with water vapor to form sulfuric acid aerosol (reaction 1b), reactions 1a and 1b can be considered to be a single reaction in which sulfur dioxide is oxidized to sulfuric acid. This process occurs naturally in clean air and sunlight at the low rate of about 0.1 percent/hr. However, the rate is greatly increased by the presence of other air contaminants through two general mechanisms: catalysis by-trace air ions, particularly metals; and a photochemical oxidation process initiated by light in which hydrocarbons, nitrogen oxides, and activated forms of oxygen play major roles. These two processes will be discussed in more detail below. Once formed, sulfuric acid may react further to form particulate sulfates, as indicated in reaction 2, in which X represents metals or ammonium. This simplified reaction scheme indicates the three forms of sulfur oxides that are usually considered to have potential health effects: sulfur dioxide, sulfuric acid, and particulate sulfates. However, it does not indicate apparent differences in health consequences between various particles, and it ignores other potential sulfur oxide pathogens, such as organic sulfates and particulate sulfites, and possible synergistic effects between sulfur oxides and other pollutants. Furthermore, X-ray photoelectron spectroscopy has reportedly identified seven separate chemical species of sulfur in pollutant aerosols in California.

Sulfur dioxide is highly soluble in water. In the absence of processes leading to its oxidation, sulfur dioxide forms the weakly acidic sulfurous acid, H_2SO_3, which dissociates to bisulfite ion, HSO_3^-, or sulfite ion, $SO_3^=$, depending on the pH of the aqueous solution (reaction 3). Two major types of analytic procedures have been used to measure sulfur dioxide in conjunction with the epidemiologic studies to be described below. Earlier studies usually measured the sulfation rate of a lead peroxide candle over relatively long periods. This procedure is somewhat at the mercy of meteorologic conditions and may to some extent include other sulfur oxides. More recent studies have used volumetric measurements of sulfur dioxide that are generally accurate and reliable.

The catalytic oxidation of sulfur dioxide occurs in water droplets or on suspended solid particles that contain absorbed sulfur dioxide and cations capable of accelerating the oxidation of sulfur dioxide to sulfuric acid (Chun and Quon 1973, Cheng et al. 1971, Foster 1969, Gartrell et al. 1963, Johnstone and Moll 1960, Johnstone and Coughanowr 1958, Junge and Ryan 1958). The metallic cations implicated in this process include vanadium, iron, manganese, copper, lead, and aluminum; many of these are present in effluents with sulfur dioxide. Among the numerous variables that affect this complex reaction are the atomospheric concentrations of sulfur dioxide and the participating cations, the efficiency of the catalyst, the adsorption rate of sulfur dioxide and its diffusion within the aerosol, and the size and pH of the aerosol. These are in turn affected by weather factors, including relative humidity (which has a major influence on droplet size) and temperature (which controls the solubility of sulfur dioxide). Under uniform conditions, the reaction tends to turn itself off, in that it is inhibited by the lower pH resulting from the formation of sulfuric acid, which decreases the solubility of sulfur dioxide. However, at lower pH, water vapor is more likely to add on to the aerosol, thereby decreasing the hydrogen ion concentration (increasing the pH) and allowing the reaction to proceed. In this respect, the presence of alkaline compounds, such as metal oxides and ammonia, is also important, in that they will increase the reaction rate by buffering droplet acidity. Elucidation of the catalytic oxidation of sulfur dioxide may be important to an understanding of the potentially toxic reactions that occur within the respiratory tract, where the relative humidity is high.

The photochemical oxidation of sulfur dioxide is also a highly complex process that is incompletely understood (James et al. 1973, Atkins et al. 1972). The requirement for sunlight limits this reaction to daylight hours, which is not the case for the catalytic oxidation of sulfur dioxide. Information concerning atmospheric photochemical reactions has been derived mainly from studies of the formation of ozone from oxygen. Superficially, the oxidation of sulfur dioxide to sulfur trioxide appears similar, in that it is predominantly a gas-phase reaction in which free-radical intermediates and excited states of oxygen are central. These reactive species are produced mainly by the action of light on nitrogen oxides in the presence of hydrocarbons. Resulting hydrocarbon radicals, singlet oxygen, and oxygen atoms have all been implicated in sulfur dioxide oxidation. One possible complication is that the formation of sulfuric acid aerosol conceivably decreases sunlight and thereby lessens the rate at which further sulfuric acid is formed. Ozone itself can also oxidize sulfur dioxide in the presence of water droplets, but this reaction occurs very slowly in the gas phase (Cox and Penkett 1972). It has recently been observed that photochemical reactions occur in a power-plant plume, resulting in the production of both ozone and sulfuric acid (Davis et al. 1974). A synergistic effect of ozone and sulfur dioxide on human pulmonary function has also been observed.

The relative contributions of the catalytic and chemical processes to sulfuric acid formation in polluted air are unknown. The concentration and chemical composition of the nonsulfur components of the pollutant mixture seem important. A theoretical approach to this problem has been presented by Judeikis and Siegel (1973).

Much of the available information on rate constants of the individual reactions was obtained from air-chamber studies, and some caution is necessary in extrapolating the results to ambient conditions. The few studies performed in ambient air have led to the estimate of well over 2 percent/hr for the rate of oxidation of sulfur dioxide in polluted air. A number of chamber and ambient studies are tabulated in more detail in the Rall report (1974).

These considerations illustrate the enormous complexity of atmospheric sulfur oxide interactions and highlight the potential importance to air pollution control of ascertaining the extent to which the various contaminants participate in producing adverse health effects or forming the offending agents. Two types of information are obviously necessary: the relative toxicities of the various airborne sulfur oxides, and the sources and atmospheric processes that result in their formation. These subjects are under study. However, available evidence indicates that further elucidation of these processes will not result in the identification of a single sulfur oxide derivative that is both solely responsible for health effects and uniquely controllable. Therefore, although further study is important, the most likely outcome pertinent to air pollution control will probably be the confirmation of the need to decrease atmospheric emission of the precursor compound, sulfur dioxide.

The natural background concentrations of sulfur oxides are difficult to ascertain, because of the widespread dissemination of these compounds from fossil-fuel combustion. For sulfur dioxide, the natural background concentration appears to be less than 0.005 ppm. In comparison, short-term concentrations well over 1 ppm have been reported during severe air pollution episodes. The U.S. standard of 0.03 ppm (annual arithmetic mean) has recently been achieved in most urban areas with the use of low-sulfur fuels. The maximal allowable 24-hr concentration is 0.14 ppm, not to be exceeded more than once a year.

Very little information is available on ambient sulfuric acid concentration, because there has been no adequate analytic method. On the basis of indirect methods, sulfuric acid has been reported to account for 12-60 percent of total atmospheric sulfate. Promising new techniques have recently been developed (Charlson et al. 1974, West et al. 1974). Presumably, the natural background concentration is negligible.

Particulate sulfate concentrations have been measured in a number of localities for more than a decade, so more information is available. But there is some question concerning the validity of the monitoring technique. One problem is the possibility that sulfates detected on the filter paper of a high-volume sampler have been formed *in situ* by catalytic oxidation of sulfur dioxide drawn through the apparatus (Forrest and Newman 1973). Although this has undoubtedly been a source of error (which it is hoped will be avoided by newer sampling techniques), evaluation of the data suggests that the estimation of the sulfate burden in various communities has been reasonably accurate qualitatively, and perhaps quantitatively. In particular, that possible source of error would not account for the consistently observed higher ratio of sulfates to sulfur dioxide in rural air. The rather significant rural sulfate burden, despite minimal sulfur dioxide content, has been especially evident in the Northeast and has led the Environmental Protection Agency to suggest that long-range transport of sulfur oxides produced by the relatively prevalent fossil-fuel combustion in this area has produced a regional decrease in air quality. Representative data from the study of Altshuller (1973) indicate that the annual average sulfate concentration in eastern urban areas is about 13.5 $\mu g/m^3$, compared with 6.4 $\mu g/m^3$ in western urban areas. Nonurban sulfate concentration in the East averaged 8.1 $\mu g/m^3$, and in the West, 2.6 $\mu g/m^3$. Owing to the generally higher sulfur dioxide concentration in eastern urban areas, the ratio of sulfur dioxide to particulate sulfate was higher in eastern than in western cities. Altshuller found that particulate sulfate and sulfur dioxide concentrations are linearly related only at lower sulfur dioxide concentrations. At higher sulfur dioxide concentrations, sulfate concentrations appear to flatten out. If validated with improved analytic techniques, this observation might indicate a limitation in the oxidation of sulfur dioxide within urban boundaries.

It must be emphasized that particulate sulfates constitute a group of compounds and that there is evidence of a difference in inherent toxicity between individual components of the group. Accordingly, in addition to measuring total sulfates, it is necessary to determine accurately the composition of this particulate fraction. The pH of particulate sulfates also appears to be important, in that acid sulfates have been found to have greater pulmonary effects in guinea pigs than neutral sulfates. The role of the weakly basic ammonia in forming acid sulfates has been evaluated in a few controlled chamber studies; atmospheric ammonia in the parts-per-billion range may have a significant impact on the formation of acid sulfates (McKay 1971, Van Den Heuvel and Mason 1963).

Of utmost importance to sulfur oxide toxicity is the observation that the overwhelming majority of sulfate compounds derived from fossil fuels are in the submicrometer range and can therefore penetrate deeply into the respiratory tract. It is generally accepted that only these smaller particles are responsible for the health effects associated with suspended particles. Although sulfates make up only 10-25 percent of total suspended particles in many areas, they undoutedly form a larger fraction of the small particles responsible for health effects.

Particles are derived from many sources, both natural and man-made. Stationary combustion of fossil fuel is a major source of particles in urban air, both because of its sulfur content and because of the presence of trace elements. However, the exact sources and inventory of particles in polluted air are not known and presumably depend heavily on local factors.

Most of the studies of adverse health effects discussed below have used one of two methods to determine particles. The preferred method in the United States has been the use of a high-volume sampler to draw a standard amount of air through filter paper; the trapped material is weighed and reported as total suspended particles. Europeans have generally used a procedure differing in a number of respects, including the final analysis, which depends on the darkness of the material on the filter paper, referred to as smoke. Both are reported in micrograms per cubic meter ($\mu g/m^3$) of air. Concentrations of total suspended particles tend to be about 1.5-2.5 times higher than concentrations of smoke shade when the same air is tested, although the difference is smaller at high concentrations (Commins and Waller 1967).

The total-suspended-particles method appears to be preferable, because it does not depend on the color of the particles, but both procedures have serious limitations from the point of view of determining health effects. It is clear that only particles smaller than about 2-3 μm in diameter can penetrate deeply into the lung. Furthermore, the exact composition of the particles may be very important, with respect to production of respiratory tract damage. The available data indicate that, for a given concentration of total suspended particles, there may be a wide range in the fraction that is respirable or that is of a specific form, such as sulfates or nitrates. Accordingly, the total suspended particulate or smoke shade procedures result in only an indirect measurement of those particles in polluted air which are injurious. Furthermore, the cation content of respirable particulates has generally not been measured, nor have the potential health effects of such cations been explored in detail.

METHODS OF STUDY OF SULFUR OXIDE HEALTH EFFECTS

In Vitro Studies

Test-tube studies in which cells or cellular constituents are exposed to pollutants have as their major goal the delineation of the biochemical mechanism of pollution action. These studies are also valuable in ascertaining characteristics that may be useful in further studies of animals or humans exposed to pollutants. It must be emphasized that dose-response data obtained from such studies cannot be readily generalized to humans breathing polluted air. Therefore, although this approach is a necessary part of evaluating pollutant effects, it does not directly provide information concerning acceptable concentrations for population exposure. Whether *in vitro* studies indicate that low or high doses of a pollutant produce an effect may be meaningless, unless the actual dose delivered *in vivo* is known. There are a number of mechanisms by which pollutants can be prevented from reaching a given target site or alternatively concentrated to high levels in that area. However, *in vitro* studies can indicate which of many potential toxic effects might predominate in a given tissue.

Animal Experiments

The exposure of animals to defined concentrations of pollutants is an important and necessary toxicologic approach. Compared with experimental human exposure, animal studies have the significant advantage of allowing the investigator to obtain samples and use concentrations that would otherwise be precluded by ethical considerations. They are generally simpler and much less expensive. In addition, animal studies provide the only reliable method of ascertaining the long-term effects of controlled pollutant exposures. In the case of sulfur oxide toxicity, important information has been obtained from animal studies that has had pertinent input into the design and understanding of both human-exposure and epidemiologic approaches. The difficulties in extrapolating animal data to man are derived to a large extent from species differences. This is particularly true for pulmonary effects, in view of the relatively marked difference in respiratory tract anatomy and physiology between man and most laboratory animals. A further difficulty is the inability of the laboratory animal to communicate such subtle effects as discomfort and irritation. It should be noted that the usual conservative toxicologic approach is to assume that any effect observed in animals at a given dose can also be observed in man. In addition, in the extrapolation of standards from animal studies to man, it has generally been considered prudent to use a substantial safety margin below the minimal effective dose in animals. However, a smaller safety factor is required in situations where adequate human dose-response data are available.

Controlled Human-Exposure Experiments

Controlled human-exposure studies in which volunteers inhale defined concentrations of air pollutants, either singly or in mixtures, have provided significant information concerning the effects of sulfur oxides. This approach allows determination of a doseresponse curve and in some cases the establishment of the minimal effective dose.

However, controlled human-exposure experiments have some limitations. As pointed out above, this approach is often cumbersome and costly and is restricted in the measurable response characteristics. The pollutant dose is delivered in an artificial manner, in that the pollutant mixture usually present in ambient air is not duplicated. Accordingly, unless the investigator is specifically aware of possible synergistic pollutant interaction, controlled human exposure to an individual pollutant may provide misleading results. In the case of sulfur dioxide, the failure of usual ambient concentrations to produce effects in normal man has led to the erroneous interpretation that this compound is without harm at these concentrations.

Another limitation of human-exposure experiments is that generally healthy people are studied, rather than sensitive population groups, because the latter are often too sick to be subjected to these studies. A no-effect exposure of healthy volunteers to pollutants may not be pertinent to susceptible subjects. Furthermore, human-exposure studies usually measure only acute responses to pollutants and have rarely provided any insight into the possible effects of chronic exposure. Inasmuch as pollutant concentrations that produce chronic disease after many decades of exposure do not necessarily result in a measurable acute response, one cannot with complete safety use no-effect concentrations in studies of human acute responses as the sole basis for air quality standards.

Epidemiologic Studies

In comparison with other fields of scientific endeavor, the epidemiologic approach used for the study of air pollutant effects is relatively crude and expensive. However, when properly performed, it has provided very useful information. Furthermore, epidemiologic studies are absolutely necessary to ascertain the minimal concentrations of air pollutants that produce deleterious health effects in human populations.

The ideal information to be extracted from epidemiologic studies would be an accurate dose-response curve for health effects. A major problem pertinent to investigation of sulfur oxides is the great difficulty involved in quantitating dose. The bulk of available epidemiologic studies evaluating sulfur oxides are seriously limited by the lack of accurate dose information for sulfuric acid and respirable particulate sulfates. In most earlier studies, measurements of only sulfur dioxide and total suspended particles are available. However, the evidence indicates that neither of these measurements directly reflects potential health effects, in that oxidation products of sulfur dioxide are probably more responsible for health effects than sulfur dioxide alone and because, of the total suspended particles, only the smaller ones can penetrate into the respiratory tract.

Another frequent difficulty with dosage data in epidemiologic studies is that the number of measuring stations is too small to identify pollutant concentrations over a large area. The reports of Corn and I. Goldstein (Goldstein 1974, Corn 1970) indicate the problems associated with such an approach. However, even if sufficient monitoring stations are used in an area to assess ambient concentrations accurately, the problem is compounded by microenvironmental variations. Exposure of an individual varies greatly, depending on his exact location and activity. Workers might spend a large portion of the day in completely different pollution environments than those who stay at home. There is also a marked difference between indoor sulfur dioxide and outdoor sulfur dioxide, and this is modified further by the exact location (Benson et al. 1972, Wilson 1968, Biersteker et al. 1965). Indoor sulfur dioxide concentrations depend on building material, wall and floor coverings, and the nature of heating and cooling systems. Outdoors, it has been shown that greenery is able to remove substantial amounts of sulfur dioxide (Martin and Barber 1970); thus, there would presumably be a difference in individual exposure, depending on whether a shrub were upwind or downwind from a pollution source. These considerations point out the potential usefulness of developing a personal monitor for epidemiologic studies.

Recent epidemiologic studies of acute health effects have begun to measure concentrations of suspended sulfates and to quantitate more accurately the size distribution and chemical characteristics of particles, so more accurate dose data should be forthcoming. However, the relative lack of such information in the past hampers determination of the role of sulfuric acid and particulate sulfates in causing or aggravating chronic respiratory disease, which presumably reflects the cumulative effects of inhalation of pollutants over a period of decades. Long-term sulfate monitoring data are available for various sites, and reasonably valid assumptions can be made by extrapolating present monitoring and emission information to the past, so that epidemiologic studies of the prevalence of chronic disease in relation to individual sulfur oxides are not totally precluded.

In terms of the response side of the dose-response curve for sulfur oxides, a relatively large mass of information is available on both acute and chronic effects. Numerous studies have evaluated the effects of short-term exposures on acute responses, such as mortality, and on various indicators of respiratory morbidity. For such studies to provide valid information, other factors must also be evaluated. Unfortunately, variables like temperature and influenza epidemics have often not been taken into account. Furthermore, different analytic and statistical approaches have been used to evaluate the available data, which makes it difficult to compare studies performed by different investigators. The most thorough studies will, of course, develop a highly complex set of data, which will require great sophistication to handle and analyze. In such situations, interpretation of the applicability of the results can be difficult. For instance, the finding that one particular response is statistically related to a particular dose of a pollutant at the 5 percent level of confidence ($p = 0.05$) --i.e., there is a 1-in-20 possibility that the result is due solely to chance -- would have questionable value in a study in which 20 sets of response-dose combinations were evaluated.

Another point should be noted on the subject of the relevance of epidemiologic studies of acute nonlethal responses in setting air quality standards: The question of what constitutes a disease effect and what can be considered a normal nonharmful response. As an extreme, it could be argued that there is no need to protect an entire population against a pollutant concentration that causes a few susceptible people to cough once a day. To the extent that measured acute responses can be considered completely reversible, their importance is open to question. However, although a cough itself may be merely a transient symptom, it may also indicate an acute irreversible effect on the respiratory tract. It is also conceivable that repeated reversible adaptive changes over a period of many decades may lead inexorably to a chronic disease process.

The resolution of this question must ultimately come from epidemiologic studies of the chronic effects of exposure to pollutants. Such studeis have often taken the approach of determining the prevalence of respiratory disease in communities with different degrees of pollution. The prevalence of respiratory disease has usually been measured by administration of a questionnaire, sometimes in conjunction with pulmonary-function tests. Major requirements for the successful completion of such studies include the use of a valid questionnaire, the determination of pulmonary function with equipment that gives reproducible results, and, most important, the standardization and minimization of observer variation. Unfortunately, such requirements have not been fully met in all studies. There is also the problem of using communities that differ only in their pollution. Occupational exposures, socioeconomic and ethnic differences, and time of residence in the area may blur the findings. Cigarette-smoking, the most important factor in respiratory disease, must be carefully evaluated. Recent studies have even suggested that cigarette-smoking by parents may increase the frequency of respiratory disease in their children (Colley 1974, Nelson et al. 1974). Subtle differences in life style may be important and may account for the observed higher prevalence of respiratory disease in urban than in rural areas. To the extent that exercise improves cardiopulmonary function, the rural resident may have an advantage over the sedentary urban resident. In addition, it is possible that some people with chronic respiratory disease move into the city in search of better medical care or out of the city to escape pollution.

Another approach to the determination of the effect of air pollution on the prevalence of respiratory disease is to study communities repetitively as their degree of pollution changes. This has the distinct advantage of allowing individual communities to serve as their own controls. In retrospect, it is unfortunate that intensive community studies were not started in the middle of the last decade and continued through succeeding years in cities that experienced a marked decline in sulfur dioxide concentration owing to pollution control measures. Recent fluctuations in the availability of lowsulfur fuel may provide an opportunity to perform such studies.

This discussion has focused generally on the difficulties in performing and interpreting epidemiologic studies related to the effects of sulfur oxides. It must be emphasized that careful epidemiologic studies can yield excellent information concerning the health effects of air pollutants. Such studies have clearly indicated a causal role of atmospheric sulfur oxides in human disease. At the present time, the critical considerations discussed above are significant mainly in determining which sulfur oxide contaminant at what concentration produces how much disease.

NORMAL LUNG FUNCTION IN RELATION TO SULFUR OXIDE HEALTH EFFECTS

Respiration is a complex, finely tuned process that results in the delivery of oxygen to the bloodstream and the removal of carbon dioxide. Air is brought into the respiratory system through either of two external passageways, the nose and the mouth. These join at the back of the throat and lead through the larynx into the trachea. In the upper part of the chest, the trachea subdivides into right and left mainstem bronchi, which lead to the lungs. The bronchi further subdivide about 26 times, becoming progressively smaller in diameter until reaching the alveoli. The alveoli are sac-like bags surrounded by blood-containing capillaries in which gases are exchanged between blood and the air brought down the bronchial tree. Optimal gas exchange, the major function of the lungs, requires an appropriate balance between blood flow and the volume of air reaching the alveoli. Both are influenced by a host of neuronal, physical, and humoral factors. Sulfur dioxide seems to affect the delivery of gases to and from the alveoli, rather than blood flow itself or the intra-alveolar gas-exchange process. However, there is some evidence that inhalation of aerosols of sulfuric acid, and perhaps particulate sulfates, which are small enough to penetrate far into the lung, may alter alveolar gas exchange. In addition, the susceptibility of patients with cardiac disease to air pollution episodes and the apparent reflex connections between the heart and lungs make it at least possible that sulfur oxide inhalation secondarily affects cardiopulmonary hemodynamics.

The respiratory consequences of the inhalation of sulfur oxides and particles depend greatly on which external orifices are in use. The convoluted and moist nasal passageways are extremely efficient filters for highly soluble gases, such as sulfur dioxide, and for particles (Vaughan et al. 1969, Frank et al. 1967, Frank and Speizer 1965, Dalhamn and Strandberg 1963). A number of studies have indicated that quiet breathing through the nose results in almost total clearance of sulfur dioxide and of larger particles. However, it must be emphasized that there is evidence suggesting a nasal-bronchial reflex whereby stimulation of the nasal mucosa results in constriction of the bronchi. It has been hypothesized that the nasal mucosal concentration of sulfur dioxide required to produce bronchoconstriction may be lower than the bronchial concentration required (Andersen et al. 1974). Although there is some evidence to support the latter hypothesis, bronchoconstriction in response to nasal sulfur dioxide has been observed only at relatively high pollutant concentrations. It is therefore doubtful whether inhalation of gaseous sulfur dioxide solely through the nose at ordinary ambient concentrations has in itself any direct untoward effect in man. Whether the discharge of nasal receptors at concentrations below the threshold to produce bronchoconstriction has an additive effect with other pollutant and nonpollutant bronchoconstrictor factors is unknown.

The mouth is a far less efficient filter than the nose and permits a much higher concentration of inhaled gaseous or particulate agents to reach the upper airways. A propensity to breathe through the mouth would therefore be expected to lead to an increased rish of pollutant effect in the respiratory tract. Among the causes of increased mouth breathing are exercise, emotional stress, and blockage of the nasal passageways, the latter usually occurring as a consequence of viral upper respiratory infection. The relative susceptibility to air pollutants of children, who frequently mouth breath, and perhaps of chronically ill individuals with upperrespiratory infections may be explainable on this basis.

Although less efficient than the nose, the mouth, pharynx, and larynx would still be the site of deposition of an appreciable amount of inhaled sulfur dioxide and aerosols. The efficiency of removal would be expected to depend to some extent on the rate of air flow into the lung. During exercise, pulmonary ventilation can increase by about a factor of 10; and at high inspiratory flow rates, more pollutants would be expected not only to be brought into the upper airways, but also to penetrate more deeply into the lung before eventual deposition. This may be another reason for the apparent sensitivity to pollution of children, who generally exercise far more often than adults.

An important aspect of pulmonary function is the defense against foreign material inhaled into the trachea and more distal portions of the lungs. The mucoid secretion of cells lining the trachea and larger parts of the bronchi plays a major role in protecting lung parenchyma from exogenous agents. Under normal conditions, respiratory tract mucus can be considered to be a renewable disposable lining that acts to mask underlying structural elements. Fine hair-like structures called cilia, which line the airways, impart the major propulsive force to move mucus up toward the throat, where it is swallowed. Unfortunately, relatively little is known about the basic biologic mechanisms responsible for normal mucociliary transport. Of particular pertinence would be additional information on ability of mucus to buffer inhaled acid aerosols (Dalhamn 1956) and on the reactions of multiple pollutants -- e.g., sulfur dioxide and ozone -- within this layer.

As with many other defense mechanisms, exaggeration of the normal response is associated with disease processes. Thus, increased mucus production is a hallmark of chronic bronchitis. Difficulty in the clearance of mucus from airways, resulting in interference with air flow, is a complicating process in a number of respiratory disorders, including chronic obstructive pulmonary disease. This is a problem particularly during expiration when the bronchi are normally relatively constricted.

Very high concentrations of sulfur dioxide have been shown in short-term animal studies to interfere with mucociliary transport (Ferin and Leach 1973, Spiegelman et al. 1968, Dalhamn and Strandberg 1963). Chronic exposure to exceedingly high concentrations has been reported to produce an increase in the number of mucus-secreting cells in the tracheobronchial tree similar to that observed in human chronic bronchitis (Lamb and Reid 1968). However, there is no direct evidence of an effect of sulfur oxides, in concentrations approaching ambient conditions, on mucociliary function in the lower respiratory tract.

Alveolar macrophages are also important in pulmonary defense. These phagocytic mononuclear cells play a major role in destroying inhaled pathogenic microorganisms, and they are also able to phagocytize inhaled particles. Although products of stationary fossil-fuel combustion have been epidemiologically associated with an increased incidence of respiratory infection, very little is known about the effect or realistic concentrations of sulfur oxides or particles on alveolar macrophage function *in vivo*. The possibility that ingested particles interfere with the bactericidal effects of alveolar macrophages appears worthy of study.

INDIVIDUAL VARIATIONS IN RESPONSE

The air pollution literature contains a refrain citing the enormous individual variability in response to air pollutants. This has been observed not only in epidemiologic studies and controlled human-exposure experiments, but also in investigations involving laboratory animals with similar genetic and environmental backgrounds. Variability in individual response has obvious implications for air pollution control strategy. If the goal, as implied in the Clean Air Act, is to protect every free-living individual in our society, the obviously air quality standards based solely on the response of the average person will not protect hypersensitive groups. The following discussion considers some susceptible groups and the possible underlying basis for their sensitivity.

Groups that have been suggested as more sensitive to the effects of air pollution include young children (possibly because of their higher inspiratory flow rates, more mouth-breathing, and frequent respiratory tract infections), the aged (perhaps because of decreased cardiopulmonary adaptive capacity), people with preexisting chronic bronchitis and emphysema (at least partly because of impaired respiratory tract defense mechanisms), and asthmatics (who tend to hyperrespond to inhaled irritants). Obviously, a substantial portion of the entire population fits into these categories.

There is excellent evidence that ethnic groups are inherently different in their response to respiratory irritants. Such differences might explain some of the wide variation in international chronic bronchitis prevalence rates, although cigarette-smoking, environmental factors, and patterns of medical diagnosis undobtedly play a large role (Holland et al. 1965, Mork 1964, Olsen and Gilson 1960). In Great Britain, there is some suggestion that the Welsh may be inherently more susceptible to environmental respiratory stressors.

Differences between the white and nonwhite populations in the United States have been observed. Groups with pulmonary-function values lower than usual include blacks, Asians, and Puerto Ricans (Stebbings 1973, Oscherwitz et al. 1972, Stebbings 1972, Densen et al. 1967). There is also fairly good evidence that blacks are less responsive than whites to inhaled toxic agents. Densen et al. evaluated more than 12,000 male New York City postal and transit workers and found that, although the prevalence of chronic bronchitis in nonsmokers was slightly greater in the nonwhite population, the prevalence among smokers was much greater in the nonwhite group (Densen et al. 1967). For any given degree of cigarette consumption, nonwhites were less symptomatic than whites. Similar findings have been observed in other studies (Finklea et al. 1974, Massaro et al. 1965). Review of the U.S. chronic respiratory disease mortality data also tends to support a greater susceptibility to pulmonary irritants among the white population. Although the male death rates for bronchitis and emphysema were similar in whites and nonwhites 25 years ago, the recent steep increase in deaths from these disorders has been much greater in whites, so there is now a substantial difference between the two (HEW 1974).

Differences between the sexes have also been observed, with women appearing to be far less responsive to cigarette-smoking or air pollution. Although a few questionnaire studies have shown equal or greater prevalence rates of respiratory symptoms in women, this is probably because (as pointed out by Ferris) women have filled out the questionnaires for entire families and are more cognizant of their own symptoms (Ferris 1969).

Determination of ethnic differnces may be very important in the interpretation of the many excellent and comprehensive Japanese epidemiologic air pollution studies. In general, these studies have tended to yield evidence of an effect of much lower concentrations of pollutants than have American or European studies. This has been true for photochemical oxidants and nitrogen dioxide, as well as for sulfur dioxide and particles. In addition to possible ethnic variations in response, this apparent susceptibility might result from some difference in methodologic approach or from synergistic effects due to the presence of significant amounts of both photochemical and stationary-source pollutants in the same areas. Accordingly, it is believed that it would be inappropriate to generalize Japanese dose-response data to the American population until proper comparative studies are performed. These are urgently needed.

The respiratory tract has a complex series of defense mechanisms that allow it to rid itself of inhaled toxic agents. Therefore, it can be predicted that any disease process that interferes with this self-cleansing will sensitize the lungs to the effects of air pollutants. This has been amply demonstrated in a number of population studies in which acute morbidity in response to daily air pollution variations was most readily observed in people who had preexisting lung disease. Similarly, an acute respiratory tract infection might render a person transiently susceptible to pollution (Creasia et al. 1973). Chronic cigarette-smoking also tends to result in a sensitivity to air pollution, and the effects appear to be independent and roughly additive (Finklea et al. 1974, Lambert and Reid 1970).

All those factors, however, do not account completely for the large variability in response to air pollutants among healthy people. Undoubtedly, genetic factors are important. Evidence on this point includes the study of Cammer et al., in which the rate of tracheobronchial clearance of an aerosol was found to differ markedly among normal people, but to be highly similar in monozygotic (identical) twins and somewhat less similar in dizygotic (fraternal) twins (Camner 1972). A familial tendency toward chronic bronchitis has also been suggested, and inherited factors might be responsible for differences in the metabolism of pollutants and in the bronchoconstrictive response to inhaled irritants.

In addition to disease states and smoking, other, more subtle environmental factors might play a role in conditioning the response to sulfur oxides. Cohen et al. (1973) have reported that the administration of tungsten to rats on a low-molybdenum diet produced a loss in sulfite oxidase activity and an increase in sensitivity to the systemic effects of high sulfur dioxide concentrations. It is unlikely that such gross alterations will occur in normal humans, but the possibility that nutritional or pharmaceutical agents can modify the response to air pollutants cannot be overlooked.

Another mechanism by which responses to air pollutants may be altered is adaption. There is excellent evidence from controlled human-exposure studies and the occupational hygiene literature that the acute response to sulfur dioxide decrease in many if not all people during continued or repetitive exposure. A number of epidemiologic studies have also indirectly suggested that adaption to sulfur oxides and particles may occur during the course of a winter exposure period. Although adaption to ambient concentrations of sulfur oxides and particles is still open to question, two points about this process are worthy of mention: First, adaption to the acute effects of a toxic agent does not necessarily imply protection against chronic effects; in fact, over a long period, the adaptive response may itself be harmful. Second, if adaption does exist, it is conceivable that a lessening in the response of individuals to the pollutant will be observed during a period of gradually decreasing atmospheric concentration; however, once a new concentration has become stable and is maintained long enough for the adaptive response to wear off, variations around the lower concentration might again produce acute responses. The implications of the latter supposition for the recent period of decreasing air pollution are obvious. Accordingly, more information on the extent and duration of human adaption to sulfur oxides would be of great value.

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CHAPTER 3

BASIC BIOMEDICAL EFFECTS OF SULFUR OXIDES

BIOCHEMICAL MECHANISMS OF SULFUR OXIDE TOXICITY

Sulfur dioxide is a weak acid anhydride that is highly soluble in aqueous solutions, although its exact solubility at low concentrations has not been determined. At usual physiologic pH, sulfur dioxide in solution forms a mixture of sulfite, $SO_3^{=}$, and bisulfite, HSO_3^{-}, ions. These ions are rapidly oxidized to sulfate, $SO_4^{=}$, by a widely distributed enzyme, sulfite oxidase. Accordingly, one can consider the biochemical mechanism of sulfur dioxide toxicity in terms of its weak acidity, of the action of sulfite-bisulfite ions, or of the effects of sulfate ions.

The receptors for a number of neurochemical reflexes appear to be highly sensitive to changes in hydrogen ion concentration, and it is therefore conceivable that a major mechanism of sulfur dioxide toxicity is expressed through reflex bronchoconstrictive responses to a slight decrease in pH. Similarly, equivalent concentrations of the stronger sulfuric acid should be even more likely to produce a bronchoconstrictive response, and acid sulfate particles could activate the same pathways.

Relatively little is known about the distribution, metabolism, and eventual fate of sulfite and bisulfite ions in man. Experiments using radioactively labeled sulfur dioxide, $^{35}SO_2$, have confirmed that the bulk of inhaled sulfur dioxide is absorbed and distributed throughout the body (Yokoyama et al. 1971, Balchum et al. 1960).

However, the final chemical form of this sulfur is unknown; it is most likely to be sulfate inasmuch as almost all the radioactivity is excreted as sulfate (Yokoyama et al. 1971). No information is available on the sites and rates of oxidation of sulfur dioxide to sulfate within the lungs or peripheral blood. The studies of Gunnison and his colleagues (Gunnison and Palmes 1974, Gunnison and Palmes 1973, Gunnison and Benton 1971) have demonstrated that rabbits exposed to sulfur dioxide develop measurable plasma concentrations of thiosulfonates that persist for days. Humans were found to have comparatively lower blood thiosulfonate concentrations after controlled exposure to sulfur dioxide at 0.3-6.0 ppm (Gunnison and Palmes 1974). Thiosulfonates are formed by the addition of sulfur dioxide across protein disulfide bonds. Although they have been identified thus far only in blood, the formation of lung thiosulfonates could be a mechanism of sulfur dioxide toxicity. The activity of enzymes containing integral disulfide bonds, such as ribonuclease, might be affected by this action of sulfur dioxide. The disruption of disulfide bonds is also believed to be the basis for <u>in vitro</u> potentiation of the red cell membrane effects of the indirect pathway of complement (DeSandre et al. 1970), the complement pathway reportedly responsible for the histamine release that is induced by allergen-reagin and results in asthma attacks (Malley et al. 1973). However, there is no direct evidence to link pulmonary or systemic thiosulfonates with sulfur dioxide toxicity. Furthermore, it should be noted that acetyclysteine, a common aerosol component used in the treatment of bronchitic disorders, is a sulfhydryl compound that is believed to act therapeutically by disrupting disulfide bonds in sputum.

Another potential biochemical mechanism of sulfite toxicity is the formation of free radicals. These reactive chemical species are presumably responsible for a number of the effects of sulfite observed during <u>in vitro</u> incubation with biologic compounds, including reduced pyridine nucleotides, tryptophan, methionine, indol-3-acetic acid, vitamin K, and thiamine (Shih and

Petering 1973, Yang 1973, Yang and Saleh 1973, Yang 1970, Klebanoff 1961). Because only micromolar amounts of sulfite are required for many of these reactions, it is at least conceivable that some occur in the lungs. The exact free-radical species responsible for these effects are not known and may depend on the concentrations of trace metals and other interacting components. Superoxide anion (McCord and Fridovich 1969), hydroxyl, and sulfite radicals have been implicated in many disease processes, including cancer and aging. However, by analogy with similar processes, it is likely that the enzymatic oxidation of sulfite to sulfate by sulfite oxidase occurs in a tightly controlled milieu, thereby preventing the release of damaging free radicals. It is obviously important to obtain further information on the concentration of sulfite oxidase and on free-radical scavenging processes in the lungs.

Sulfite has also been shown to form adducts with flavin compounds and with folic acid, although in the latter case a large excess of sulfite is needed (Muller and Massey 1969, Vonderschmitt et al. 1967).

As will be discussed in more detail later, sulfite and bisulfite ions can react with deoxyribonucleic acid (DNA) to produce the deamination of cytosine. The ribonucleic acid (RNA) component uridine has also been shown to form an unstable intermediate on reaction with sulfite-bisulfite ions (Shapiro and Brauerman 1972). Neither reaction has been demonstrated in vivo; but the reaction with uridine occurs at lower concentrations and at normal pH, so it is more likely to be toxicologically significant. Modification of RNA would be expected to interfere with protein synthesis, and this has been demonstrated after incubation of bacteria with sulfite-bisulfite ions (Shapiro and Brauerman 1972). However, incubation of algae with bisulfite appears to interfere with DNA synthesis, rather than protein synthesis (Das and Runeckles 1974).

Other effects of sulfite-bisulfite ions observed at high concentrations in vitro include alterations of platelet function and interference in the metabolic formation of red-cell 2,3-diphosphoglyceric acid, an important intermediate in the regulation of oxygen delivery to the tissues (Kikugawa and Hzuka 1972). However, free sulfite ions have not been detected in the blood of rabbits after exposure to sulfur dioxide at 25 ppm (Gunnison and Benton 1971).

There is little to suggest that sulfate ions formed as a result of sulfur dioxide inhalation play any role in toxicity. Sulfate is a normal body constituent in sufficient quantities so that ambient sulfur oxides would add little to total body concentrations. It is, of course, possible that a local increase in slulfate concentration within the lungs could be significant. This might alter sulfation rates of mucopolysaccharides, which are important extracellular lung components. In this respect, decreased concentrations of sulfated mucosubstances were observed by histochemical techniques.in the bronchial surfaces of dogs chronically exposed to sulfur dioxide at 500-600 ppm (Spicer et al. 1974). Additional biochemical measurements revealed an increase in the activity of tracheobronchial glycotransferases. One of the few other studies to measure enzymatic changes after sulfur dioxide exposure (300 ppm, 6 hr/day for 10 days) reported increased activity of acid hydrolase, but not of other lysosomal enzymes, in alveolar macrophages (Barry and Mawdesley-Thomas 1970).

In summary, the biochemical mechanisms by which ambient concentrations of sulfur oxides produce effects in the lung are unknown. It it is accepted that the major physiologic consequence of sulfur oxide inhalation is bronchoconstriction, it is conceivable that effects are totally explainable by a decrease in pH. Hyperplasia of mucus-secreting cells, which may result from prolonged sulfur oxide exposure, might also be a response solely to acidity. However, this conservative approach to an explanation of sulfur oxide toxicity is most likely an oversimplification. Further studies using sensitive biochemical techniques and reasonable sulfur oxide exposures would be of value.

PHYSIOLOGIC AND ANATOMIC EFFECTS OF SULFUR OXIDES

The literature concerning animal respiratory physiologic response to the inhalation of sulfur oxides, particularly sulfur dioxide, is relatively voluminous. Much of the work has originated in the laboratory of Amdur, who has evaluated in depth the bronchoconstrictive response of guinea pigs to various ceoncentrations of sulfur dioxide, particles, and sulfuric acid, alone and in combination. Extensive studies of guinea pigs and other animals have also been done by others and this subject has been reviewed by Amdur (1971), Riley (1974), Alarie and Palmes (1974), and Dubois (1969).

The following discussion will concentrate on studies that have provided insight into the possible mechanisms of toxic action of the various sulfur oxides and that appear to be pertinent to an understanding of human response.

The major quantitated physiologic indicator of response to sulfur dioxide has been an increase in resistance to pulmonary flow, which has usually, but not always, (Davis et al, 1967) been related to bronchoconstriction. This has been observed in a number of animal species (Corn et al. 1972, Frank and Speizer 1965, Nadel et al. 1965, Swann et al. 1965, Salem and Aviado 1961, Balchum et al. 1960) and in man. In general, the acute response to sulfur dioxide in animals has been short-lived and has not persisted after cessation of exposure. In some instances, a return toward normal pulmonary flow resistance has been observed during continous sulfur dioxide exposure, indicating some degree of adaption. The apparent threshold for this acute response to sulfur dioxide is very high, relative to usual ambient concentrattions.

A number of ingenious pharmacologic and surgical techniques have been used to explore the mechanism by which increased respiratory flow resistance occurs, the site of the sulfur dioxide receptor, and the specific airway locale that is most affected. The major overall impression is that multiple potential receptors and effectors in various parts of the respiratory tract can increase flow resistance (Frank and Speizer 1965) locally or reflexively in other parts of the airway. Inasmuch as sulfur dioxide inhaled in clean air is almost totally absorbed in the upper airways (Andersen et al. 1974, Frank et al. 1969, Vaughan et al. 1969, Frank et al. 1967, Frank and Speizer 1965, Dalhamn and Strandberg 1961), its effects on bronchi lower in the respiratory tract must be mediated by some indirect pathway. The studies of Widdicombe, Nadel, and their co-workers have demonstrated a number of receptors in the upper respiratory tract that, when stimulated, result in cough, local airway narrowing, or constriction of other respiratory tract areas (Nadel et al. 1965, Nadel and Widdicombe 1962, Widdicombe et al. (1962). Although airway narrowing due to sulfur dioxide generally appears to result from reflexes mediated through the vagus nerve, humeral pathways acting on smooth muscle may also be operative (Nadel et al. 1965, Salem and Aviado 1961). It is also possible that sulfur dioxide absorbed in the upper respiratory tract may be carried in the pulmonary circulation to the lower airways, thereby providing another possible mechanism of bronchoconstriction (Frank et al. 1967).

Another important point that is obvious from animal experiments is the substantial and consistently observed variability in the response of different animals of the same species. Laboratory animals of the same species tend to have more homegeneous genetic and environmental backgrounds than does man, so an even greater variability in human response would be expected.

An interesting study was presented by Islam et al. (1972), in which dogs were exposed sequentially and repetitively to an aerosol of the bronchoconstrictive agent acetylcholine and then to sulfur dioxide for 1 hr. After sulfur dioxide exposure, marked potentiation of the bronchoconstrictive response to acetylcholine was observed; it appeared to be linear in response to sulfur dioxide at 1-5 ppm, but declined somewhat at 10 ppm. The pertinence of this study is its suggestion that sulfur dioxide may potentiate the response to neuropharmacologic agents that normally act to produce bronchoconstriction after vagus nerve stimulation.

The studies of the acute response to sulfur dioxide alone have provided important information, but more pertinent data have been obtained from experiments in which animals were exposed to combinations of sulfur dioxide and particles or to other sulfur oxides. Amdur originally demonstrated a synergistic effect of sulfur dioxide aerosols of respirable size (Andur 1957). However, a synergistic interaction of sodium chloride particles and sulfur dioxide could not be confirmed in casts (Corn et al. 1972) or man. The recent work of McJilton et al. (1973) has provided additional insight into this process by demonstrating the importance of relative humidity: A synergistic response in the pulmonary flow resistance of guinea pigs exposed to a submicrometer aerosol of sodium chloride (900-1,000 ug/m³) and sulfur dioxide (1.1 ppm) was observed only at a relative humidity of greater than 80 percent; no effect on pulmonary flow resistance was observed when the relative humidity was less than 40 percent, nor when the animals were exposed to sulfur dioxide or the aerosol alone. The authors hypothesize that the deliquescense of sodium chloride, which occurs at about 70 percent relative humidity, leads to an increased loading of sulfur dioxide onto the aerosol and therefore to a greater delivery of sulfur dioxide to the lower part of the lungs. With high relative humidity, the pH of the aerosol is low, owing presumably to the formation of bisulfite ions (no sulfate was detected).

Two points should be made. First, the relative humidity of the respiratory tract is close to 100 percent, so deliquescence of inhaled dry sodium chloride will occur in any event; however, the authors suggest that the bulk of sulfur dioxide will be removed within the guinea pig nose and that there is insufficient time for both the deliquescence of dry sodium chloride within the nose and the absorption of sulfur dioxide onto the aerosol before the sulfur dioxide is scrubbed out by the nasal mucosa. Second, the studies of Amdur demonstrating synergism in this same nosebreathing animal were performed at about 50 percent relative humidity, which is below the deliquescence point of sodium chloride; this might be explainable by the fact that sulfur dioxide can still be absorbed onto dry particles, although to a lesser extent. Inasmuch as Amdur generally used higher aerosol concentrations, absorption onto the dry particles may have been sufficient to account for the findings of synergism.

Other studies by Amdur and her colleagues in the gunea pig, evaluating a series of different inert aerosols inhaled in conjuction with sulfur dioxide, have indicated that particle size is of great importance (the smaller the particle, the greater the effect); that potentiation is observed only with soluble aerosols (the greater the solubility, the greater the effect); that soluble metallic aerosols produce a greater effect than inert aerosols, presumably because of the catalytic formation of sulfuric acid; and that the effect of aerosol-sulfur dioxide mixtures tends to persist, whereas that of sulfur dioxide alone disappears rapidly (Amdur and Underhill 1968, Amdur 1959, 1957).

This guinea pig model has also been used to study the effects of other sulfur oxides. Aerosols of sulfuric acid are far more potent than sulfur dioxide in increasing pulmonary flow resistance, and the response is more rapid and prolonged (Amdur 1971, 1969, 1959, 1958). Sulfuric acid is also more potent than the combined effects of sulfur dioxide and sodium chloride aerosol, particularly at low concentrations. The aerosol size of sulfuric acid influenced the degree of the increase in pulmonary flow resistance, the course of the response, and the extent of airway obstruction. Synergism between sulfur dioxide and sulfuric acid aerosol on pulmonary flow resistance in guinea pigs has also been reported (Amdur 1957).

Short-term inhalation of aerosols of various particulate sulfates were found to increase pulmonary flow resistance at concentrations at which inert aerosols have no effect (Amdur and Corn 1963). Zinc ammonium sulfate was more potent than either zinc sulfate or ammonium sulfate; ~~this~~ suggests that the acidity of the aerosol may be important. However, ammonium sulfate had a smaller effect than zinc sulfate. Again, toxicity was inversely related to particle size. A 20 percent increase in airway resistance was noted during exposure to zinc ammonium sulfate at 200 ug/m³ (particle size, 0.3 um). The rationale for the study of these compounds was the retrospective observation of Hemeon (1955) that zinc ammonium sulfate and zinc sulfate were present in substantial concentrations during the 1948 Donora air pollution disaster. Ferric sulfate inhaled alone was found to increase pulmonary flow resistance, but ferrous sulfate had no effect (Amdur and Underhill 1968).

A recent study by Alarie et al. (1973) evaluated mice exposed to sodium sulfite and sodium metabisulfite. Only the latter compound produced sensory irritation, with effects similar to those of sulfur dioxide. The authors hypothesize that the formation of bisulfite in the slightly alkaline nasal mucosa is a central mechanism of effect.

Amdur has summarized her studies in guinea pigs and prepared dose-response curves comparing the effects of the various sulfur oxides (Amdur 1971). The data clearly indicate that sulfur dioxide is far less irritating than equivalent concentrations of sulfuric acid and zinc ammonium sulfate. The relative toxicity of the latter two compounds depends on particle size. In some cases, zinc ammonium sulfate appears to be more potent than sulfuric acid. These findings emphasize the need for accurate determination of the composition and size of particles in polluted air. In general, the data indicate that atmospheric oxidation of a relatively small fraction of sulfur dioxide will produce particulate aerosols more toxic than the parent gas. Furthermore, the effects of sulfur dioxide and its products are ~~probably~~ additive and perhaps synergistic.

Man has been the subject of a number of controlled experimental studies of the acute effects of sulfur dioxide. Increased airway resistance, due presumably to bronchoconstriction, is the major feature of the response to sulfur dioxide (Melville 1970, Nadel et al. 1965, Frank et al. 1964, 1962). Recent studies by Andersen et al. demonstrating that sulfur dioxide concentrations resulting in an increased forced expiratory volume do not affect airway closing volume indicate that only the relatively larger parts of the bronchi are affected (Andersen et al. 1974). Increased airway flow resistance is also observed as a consequence of exposure to very high concentrations of particles (Dubois and Dautrebande 1958). Only healthy subjects have been used in these experiments, and, although marked variability has been observed, responses to sulfur dioxide concentrations as low as 0.75 ppm have only rarely been reported (Bates and Hazucha 1973). This effect of sulfur dioxide is rapidly reversible and, as in animals, appears to be mediated through vagus nerve relfex arcs (Nadel et al. 1965a, 1965b). Sulfur dioxide at 1-5 ppm also may produce an increase in pulse rates and respiration, as well as a decreased tidal volume, although this is not consistently observed (Amdur et al. 1953). Inhalation through the nose appears to produce a smaller response than inhalation by mouth, but again there is some variability in results. In some subjects, the response to sulfur dioxide appears to decrease on prolonged or later exposure.

An interesting series of studies have been presented by Weir and Bromberg (1973, 1972). Normal subjects had an increased airway resistance during exposure to sulfur dioxide at 3 ppm for 5 days, which persisted for less than 2 days after removal from the chamber. Exposure of patients with preexisting respiratory disease to 0.3-3.0 ppm for a week resulted in highly variable responses, but produced no consistent effect.

Attempts to demonstrate a synergistic effect of sulfur dioxide and sodium chloride aerosol in man have been unsuccessful (Burton et al. 1969, Frank 1964, Frank et al. 1964). However, Snell and Luchsinger (1969) observed synergism of 0.5-ppm sulfur dioxide and a submicrometer distilled water aerosol on maximal expiratory flow rate in one experiment. An additive effect of combined exposure to sulfur dioxide (5 ppm) and nitrogen dioxide (5 ppm) has also been reported (Abe 1967).

Of great interest is a recent study by Bates and Hazucha (1973) which demonstrated a synergistic effect of sulfur dioxide and ozone in healthy men. No changes in pulmonary function were observed in volunteers who breathed 0.37-ppm sulfur dioxide alone for 2 hr. However, inhalation of 0.37-ppm ozone and 0.37-ppm sulfur dioxide produced a definite decrement in maximal midepiratory flow rate. Ozone alone at this concentration had a much smaller effect. It is of further interest that the volunteers underwent intermittent exercise in the exposure chamber and that sulfur dioxide alone at 0.75 ppm had a small but significant effect. Because this is the lowest concentration of sulfur dioxide reported to have an effect in man, the results suggest that exercise potentiates the action of sulfur dioxide, perhaps by increasing mouth-breathing and inspiratory flow rates.

There are a number of possible explanations for the observed synergism between ozone and sulfur dioxide. A likely one is that sulfuric acid was formed by the reaction of these two agents in the presence of water vapor or mucous in the respiratory tract. It is also possible that, by decreasing the pH of the mucous lining, sulfur dioxide increases the solubility of ozone and thereby potentiates an ozone effect. Further studies of this important observation are needed, particularly to rule out the possibility that sulfuric acid aerosol was formed in the chamber.

Pattle and his colleagues (Sim and Pattle 1957, Pattle and Cullumbine 1956) have exposed healthy volunteers to sulfuric acid. At the high concentrations used in their experiments (5-10 ppm, 20-40 mg/m^3), a marked increase in airway resistance was observed, as were chest discomfort, rales, cough, and lacrimation. Two subjects who were repetitively exposed developed bronchitits symptoms. The presence of water vapor potentiated the observed responses, but addition of ammonia vapor decreased the effect. The latter finding suggests that ammonium sulfate is not as toxic as sulfuric acid.

A number of investigators have studied the effects of short-term exposure to sulfur dioxide on various aspects of mucociliary transport. Decreased ciliary activity has been reported in the tracheas of rabbits after inhalation (100-200 ppm) or direct tracheal exposure (10 ppm) (Dalhamn and Strandberg 1961, Calley 1942). This effect was not potentiated when carbon particles capable of absorbing sulfur dioxide were inhaled with it (Dalhamn and Strandberg 1963). The clearance of particles from the lower respiratory tract has been evaluated in donkeys, in which inhibitory effects were observed at 300 ppm (Spiegelman et al. 1968), and in rats, in which sulfur dioxide had a slight stimulatory effect at 0.1 ppm, but resulted in a decrease in clearance at 1 ppm for 170 hr. (Ferin and Leach 1973). In the latter study, short term high-dose exposures had a smaller effect than long-term low-dose exposures. Andersen et al. (1974) recently reported a study in which healthy human volunteers were shown to have a decrease in nasal mucous flow rate during 6 hr of exposure to sulfur dioxide at 1.5 and 25 ppm, although the findings at 1 ppm were not statistically significant. Subjects who initially had the lowest nasal flow rates were most affected by sulfur dioxide. The decrease in flow rate was most prominent in the anterior part of the nose, which absorbed the bulk of the sulfur dioxide. Increased nasal flow resistance was also observed in all subjects. This had previously been reported as an inconstant effect by Speizer and Frank (1966).

Studies of the long-term effects of sulfur dioxide in animals have generally demonstrated no deleterious effects at concentrations within the range of usual ambient conditions. Chronic inhalation of concentrations greater than 100 ppm has resulted in hyperplasia of mucus-secreting cells and metaplasia of the bronchial epithelium similar to that observed in chronic bronchitits in man (Spicer et al. 1974, Asmundsson et al. 1973, Goldring et al. 1970, Lamb and Reid 1968, Pattle and Cullumbine 1956). However, in a study of cynomolgus monkeys, Alarie _et al_. (1972) observed no changes in sensitive pulmonary-function tests, lung histology, or blood chemistry during 78 weeks of uninterrupted exposure to sulfur dioxide at 0.14 or 1.28 ppm. Other studies by the same group demonstrated no effect of sulfur dioxide on guinea pigs exposed for a year to similar concentrations (Alarie et al. 1970). In addition, no adverse effects were observed in guinea pigs and monkeys exposed to fly ash (obtained from the effluent of coal-burning plants and having a mass median diameter of 2.6-3.4 um) inhaled with sulfur dioxide in the same concentrations and durations as in the above experiments (Alarie et al. 1973). Inhalation of sulfur dioxide (35 ppm) and corn starch and corn dust particles (particle size, > 2 um) has been reported to have effects on pigs similar to that of sulfur dioxide alone (Martin and Willoughby 1972). Rabbits inhaling sulfur dioxide at 10 ppm for 16 hr/day initially had an increased rate of clearance of inhaled particles (3-6 um), but this diminished after 6 weeks of exposure (Holma 1967).

These studies were performed in healthy animals and might not mimic the long-term effects of sulfur dioxide on people with chronic lung disease. However, production of lung disease by treatment of hamsters with papain or exposure of dogs to nitrogen dioxide has not been found to potentiate the response to later exposure to sulfur dioxide at very high concentrations (Lewis et al. 1973, Goldring et al. 1970). In addition, animals chronically exposed to Los Angeles air responded similarly to clean air controls when acutely exposed to sulfur dioxide (Emik et al. 1971).

Fewer studies of long-term exposure to sulfuric acid have been performed. Thomas et al. (1958) reported that exposure of guinea pigs to 2,000 ug/m³ for 3 months produced little effect. At higher concentrations, the pathologic findings depended on particle size. Alarie et al. (1973) observed no effects on lung function or histology of guinea pigs exposed for a year to sulfuric acid at 80 ug/m³ (particle size, 0.84 um) or 100 ug/m³ (particle size, 2.8 um). However, adverse effects were observed in cynomolgus monkeys exposed to various concentrations of sulfuric acid for 78 weeks. The changes were slight but detectable at 380 ug/m³ (particle size, 2.2 um) and 480 ug/m³ (particle size, 0.5 um) and were more prominent at higher concentrations. Major observations included alterations in the distribution of ventilation (as measured by nitrogen washout), hyperplasia of the bronchial epithelium, and thickening of the bronchiolar walls. Particle size appeared to have little relation to eventual damage, although there was a tendency toward less adverse effects with particles in the smaller (submicrometer) range.

Lewis and his colleagues (Lewis et al. 1974, 1973, Bloch et al. 1972, Lewis et al. 1969, Vaughan et al. 1969) have performed a series of studies in beagles that included individual or combined exposure to sulfur dioxide and sulfuric acid for up to 5 years. A mixture of sulfur dioxide (13.4 mg/m³, 5 ppm) and sulfuric acid (900 ug/m³) resulted in physiologic changes that suggested both lung parenchymal disease and airway obstruction. Sulfuric acid alone produced a decrease in lung diffusing capacity; sulfur dioxide alone resulted in increased airway resistance and alterations of lung compliance similar to those reported in other animals. In one study, beagles that had previously been chronically exposed to nitrogen dioxide tended to have a somewhat smaller response to sulfur oxides; this suggests that the preexisting lung alterations were protective. Additional studies by this group have provided suggestive, but not conclusive, evidence that inhalation of sulfur dioxide-sulfuric acid mixtures potentiates the pulmonary hyperinflation produced by chronic inhalation of raw auto exhaust and may lead to some cardiac abnormalities. One technical problem concerning sulfuric acid explosures is that ammonia produced by animal excreta might buffer the sulfuric acid and produce ammonium sulfate aerosol.

Among the most important results of the animal and controlled human exposure experiements cited above is the insight into potential mechanisms of sulfur oxide toxicity at ambient concentrations. Sulfur dioxide acting alone is unlikely to be physiologically important, except during periods of severe inversion coupled with grossly inadequate emission control. However, sulfur oxide concentrations that are within the range of ambient concentrations not infrequently experienced in polluted areas of the United States may produce effects by one or more of the following mechanisms:

1. A direct effect of ambient sulfuric acid.
2. A direct effect of ambient suspended particulate sulfates.
3. An effect of sulfur dioxide potentiated by adsorption onto inert particles of respirable size.
4. An effect of sulfur dioxide potentiated by metallic particles that catalyze its oxidation in the airway.
5. An effect of sulfur dioxide potentiated by simultaneous inhalation of ozone.

Inasmuch as the various sulfur oxides tend to be present together in polluted air, the effects described above may well occur simultaneously in an additive, or perhaps synergistic, manner.

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CHAPTER 4

HEALTH EFFECTS OF SULFUR OXIDES

CHILDREN

A number of studies have focused on the effects of air pollutants in children. This is a useful group to study, in that their lesser mobility produces a more homogeneous pollution exposure and the effects of cigarette-smoking and occupational exposure are minimized. Furthermore, children appear to be at greater risk than healthy adults, owing presumably to their greater degree of mouth-breathing, relatively greater tidal volumes, and higher frequency of respiratory tract infections. The effects of air pollutants in children have been reviewed by a committee of the American Academy of Pediatrics (1970) and more recently by Wehrle and Hammer (1974).

An increase in the death rate of the very young has been noted in some, but not all, studies of air pollution episodes. Logan (1953) noted an increased neonatal and infant mortality in the December 1952 London fog episode, but Greenberg et al. (1967) did not observe any change in death rate in this age group during a less severe 2-week episode in New York in 1963. Lave and Seskin (1970), in their broad statistical analysis of the effects of air pollution in the United States, have suggested an association with neonatal mortality.

A number of large-scale British studies have evaluated the effects of air pollution on the health of children. Colley and Reid (1970) surveyed respiratory disease prevalence in 1966 in over 10,000 children 6-10 years old in England and Wales with a questionnaire and measured peak expiratory flow rates. A social-class gradient was observed for chest disease, but a pronounced increase in prevalence was observed in each social class in association with greater pollution. This was particularly true in the children of unskilled and semiskilled workers. An unexplained excess in prevalence in Wales may represent an ethnic difference. Small differences in peak expiratory flow rate were found when the results were calculated independently for those with and without a history of respiratory disease or symptoms. The authors point out that differences may have been obscured by difficulties in standardizing the observers and the 70 peak flow meters used in the study.

Another British study of over 10,000 children was performed at about the same time by Holland et al. (1969) in different areas of Kent. They observed independent and additive effects of social class, family size, history of significant respiratory disease, and area of residence on pulmonary function, as measured by peak expiratory flow rate. Residential area appeared to have the greatest influence on pulmonary function; this suggested an effect of air pollution. However, the authors note that all four factors together accounted for only about 10-15 percent of the total variation; other determinants, possibly including host factors, are probably also important. Only minimal pollutant monitoring data are presented that describe the monthly average smoke concentrations in three of the four areas studied for November 1966 to March 1967. In the most polluted area, which had the lowest peak expiratory flow rates, the highest recorded monthly average smoke shade was 96 ug/m³.

A study by Douglas and Waller (1966) evaluated over 3,000 children 15 years old who had been born in the first week of March 1946 and were followed regularly as part of a comprehensive health survey. Domestic coal consumption was used to divide areas into four pollution classes. The frequency and severity of lower respiratory tract infections were clearly related to air pollution. However, no consistent effect on upper respiratory infections, otitis, or tonsillitis was observed. The following factors demonstrated a gradient of air pollution effect, in that there was an increase at each succeeding stage of pollution severity: first lower respiratory tract infection before the age of 9 months and before age of 2 years; more than one attack in first 2 years for both boys and girls; hospital admission for bronchitis in first 5 years; pulmonary rales or rhonchi noted on two or more chest examinations or by the age of 15; excess episodes of school absences; and long periods of absence from primary school for bronchitis. Only small differences in social class among the air pollution categories were noted, and this factor was not evaluated further. Nor were other possible variables, such as crowding, assessed. Some data are given in the appendix concerning air pollution in 1962-1963. As indicated by the authors, this must be approached with caution, particularly as pollution was probably worse in earlier years. Taken as a whole, the data give an impression of linearity of response (lower respiratory infection) in children to air pollution beginning at somewhat higher than present U.S. standards for total suspended particles and sulfur dioxide.

This cohort of children born in 1946 was reevaluated at the age of 20 (Colley and Reid 1970). The most noticeable finding was the dominant effect of cigarette-smoking on the respiratory symptoms of these young adults. A history of chest disease before the age of 2 also had a significant, but smaller, effect. After adjustment for these two variables, air pollution had only a small and not statistically significant effect. The same was true for socioeconomic class. There was a tendency for the air pollution effect to be greatest in the lower socioeconomic classes; this suggests an interaction between these two variables in the production of respiratory symptoms. As pointed out by the authors, the nonresponders to the follow-up questionnaire were overrepresented in the high-pollution category, which might tend to bias the results. However, the results are consistent with those of other studies and indicate that the effects of air pollution on young adults are minimal at most, particularly in relation to cigarette-smoking.

Lunn et al. (1970, 1967) evaluated children in Sheffield, England, in 1965 and were able to repeat the study on some members of the same group 4 years later, when air pollution had decreased. This type of study, in which a cohort is reevaluated after a change in the environment, has also been performed in Berlin, New Hampshire, and is of great value in ascertaining the effects of air pollution. At the time of the original study, there was a distinct gradient in air pollution in the four districts under evaluation. In 1964, the cleanest area had a mean daily sulfur dioxide concentration of 123 ug/m³ and mean daily smoke of 97 ug/m³. The mean daily sulfur dioxide and smoke values in the other areas were 183 and 230, 219 and 262, and 275 and 301 ug/m³. Questionnaires were sent to the parents of 5-year-olds living in the vicinity of the school. The children were then examined, and pulmonary function tests were performed. Socioeconomic class, number of children at home, and number of persons sharing the same bedroom all had some effect on the findings. However, when the data were evaluated within each social class, a distinct increase in respiratory morbidity for both upper and lower respiratory diseases was observed. For the lower socioeconomic classes, a history of persistent cough was obtained in 23.9 percent of the children in the cleanest area, and there were stepwise increments in from each area as the pollution increased--39.6 percent, 43.8 percent, and 60.9 percent. Lower-than-predicted forced expiratory volume (0.75 sec) and forced ventilatory capacity were observed only in the most polluted community. The 11-year-olds examined at the same time had similar, but less marked, differences between areas.

When the original 5-year-old group was reexamined 4 years later, in 1969, the differences between areas had narrowed and were not statistically significant. A major decrease in air pollution had occurred; in 1968, mean daily sulfur dioxide and smoke concentrations for the four areas were 94 and 48, 166 and 41, 186 and 118, and 253 and 169 ug/m³. There was also less respiratory disease in this 9-yearold group than in the 11-year-olds evaluated 4 years earlier. It should be emphasized that this lessening in a gradient of respiratory disease was associated with about a 45-80 percent decrease in particles, as measured by smoke, with only approximately a 10-25 percent decline in sulfur dioxide. Furthermore, during the latter part of the study, the smoke and sulfur dioxide concentrations were still appreciably above American air quality standards. Unfortunately, no data on ambient concentrations or gradients for sulfate are available.

Biersteker and van Leeuwen (1970) evaluated peak flow rates in about 1,000 schoolchildren in two districts of Rotterdam: a wealthy suburban area in which winter mean smoke concentration was about 50 ug/m³ and winter mean sulfur dioxide concentration was 200 ug/m³, and a poorer urban area with about 50 percent higher pollutant concentrations. Air pollution monitoring techniques are not described. Although the children in the cleaner area generally had higher peak flow rates, this could be accounted for by differences in height and weight. In fact, following multiple regression analysis, there was a slight but statistically significant increase in peak flow rates in the polluted area. The authors also observed a statistically significant increase in the prevalence of childhood chronic bronchitis in the more polluted area (5.5 percent), compared with the cleaner area (1.0 percent). A smaller, nonsignificant increase in asthma rate was also found (2.2 percent vs. 1.7 percent).

A number of Japanese investigators have also reported an association of sulfur dioxide and particles with prevalence of respiratory morbidity and a decrease in pulmonary function in children (Kagawa et al. 1974, Toyama 1964).

Responses appear to occur at relatively low pollutant concentrations. However, as discussed earlier, there is some question as to whether the results can be extrapolated to the U.S. population.

As part of the Nashville air pollution study, Sprague and Hagstrom (1969) observed that sulfation rates correlated with middle-class deaths in the age range of 1-11 months and that dustfall was associated with neonatal deaths. However, the authors found it difficult to remove socioeconomic factors, which varied with air pollution, as well as with neonatal and infant mortality. Furthermore, their assignment of arbitrary linear numbers to different socioeconomic classes is questionable.

Collins et al. (1971) studied environmental factors affecting child mortality in England and Wales during 1958-1964. Included in this analysis were indexes of domestic pollution, industrial pollution, proximity of a power station, social class, overcrowding, population density, and education. Each of these factors except power-station proximity was significantly correlated ($p<0.001$) with the mortality of infants less than a year old. Because all the social indexes were highly intercorrelated, it was difficult to disentangle their individual effects. However, partial correlation analysis showed that the bulk of the increased mortality could be attributed to domestic and industrial pollution. Unfortunately, the data are not presented in such a way as to permit assessment of a possible dose-response relationship.

As part of the Berlin, New Hampshire, study (described in more detail later), Ferris (1970) investigated the relation of air pollution to school absences and pulmonary function. First- and second-graders from seven schools were studied in 1966-1967. No consistent effect of air pollution on total school absence rates or school absences due to respiratory illnesses were observed. A relationship may have been obscured by the fact that not all children lived close to their schools. There were some statistically significant differences in pulmonary function, particularly peak expiratory flow rate, that suggested an effect of air pollution. The worst school (school A) in terms of pulmonary function was subjected to much higher amounts of dustfall, but not of sulfur dioxide (as measured by lead peroxide candle), which suggests that the effect was due mainly to particles. However, school A's pupils were of a lower socioeconomic class than those of most of the other schools. The only other school with pupils of a similarly low socioeconomic class (school J) was in an unpolluted area, and, although pupils in school A tended to have lower pulmonary function than those in school J, the difference was not reported to be statistically significant.

Anderson and Larsen (1966), in a similar study of first-graders in British Columbia, were also unable to show an effect of air pollution on school absences. However, there was a statistically significant decrease in peak expiratory flow rate in the two towns affected by a Kraft pulp mill, compared with that in the clean town. This may have been related to ethnic differences, which were not studied. Air monitoring data are not presented.

A recent study by Mostardi and Leonard (1974) reported a decrease in pulmonary function in high-school students in Barberton, Ohio, an industrialized area, compared with that in Revere, Ohio, a more rural community. These students had initially been evaluated in 1970 and were restudied in 1973. Socioeconomic factors were said to be similar. The Barberton students had a statistically significant and similar decrease in vital capacity in both periods: a decrease in 1-sec forced expiratory volume in 1970, but not 1973, and in maximal aerobic capacity on exercise (Vo_2 max) when tested in 1973. No difference was observed in maximal midexpiratory flow rate, an index of small-airway disease. The finding of a difference in Vo_2 max is of interest, in that this test is usually not performed in air pollution studies and has potential health significance. However, this result could conceivably be accounted for by a relative lack of routine exercise in the urban group, although participation in organized athletics was similar. The air monitoring data show that

Revere cannot be considered a low-pollution community, in that yearly mean concentration of total suspended particles, which decreased from 83 to 71 ug/m³ from 1969 to 1973, was generally greater than the U.S. air quality standard. Furthermore, the sulfur dioxide concentration (by lead peroxide candle, 0.96 mg of sulfur trioxide per 100 cm² per day in 1971) was much higher than that recorded in Berlin, New Hampshire, although there is an unexplained decrease to 0.36 mg/100 cm² per day in 1972. The air monitoring data for Barberton show a peak yearly mean 24-hr concentration of total suspended particles of 109 ug/m³ in 1969, which decreased to 77 ug/m³ in 1973, and peak yearly sulfur dioxide of 1.11 mg/100 cm² per day. This study tends to support an association between a decrement in childhood pulmonary function with air pollution slightly above the current U.S. standards for total suspended particles and sulfur dioxide. The data, although suggestive, do not clearly indicate whether improvement in air quality had a positive effect. Potential criticism of the study includes the relatively small numbers of students tested and the lack of presented data on cigarette-smoking, although this question was apparently asked. In addition, a contemporary urban-rural difference in the use of nontobacco cigarettes is conceivable.

The Community Health and Environmental Surveillance System (CHESS) program has conducted a number of extensive studies of the health of children (EPA 1974). In the Salt Lake area, four communities were selected for a retrospective study of lower respiratory infection on the basis of an expected gradient for sulfur dioxide and suspended sulfates (Nelson et al. 1974). Pollutant concentrations for the period before 1970 were estimated from monitoring and emission data, as well as meteorologic factors. Information about lower respiratory infection in children 5-12 years old was obtained by questionnaires mailed to parents and distributed through the schools. Almost 9,000 nonasthmatic students were studied; the results indicated an increased incidence of total lower respiratory infection, croup, and bronchitis, but not of pneumonia or hospitalization, in the most polluted community. This high-pollution community had an estimated mean sulfur dioxide concentration in 1967-1970 of 91 μg/m³, compated with 33 μg/m³ in the next highest area, and an estimated mean suspended sulfate concentration of 15 μg/m³, compared with 9 μg/m³. The concentration of total suspended particles in the high-pollution community in 1967-1970 was 63 μg/m³, which was lower than the concentrations in two of the other communities. The concentrations of suspended nitrates were similarly low in all the communities. Other factors evaluated--including the educational status of the father, mean family size, and parental smoking habits--did not appear to account for the increased incidences of croup and bronchitis in the high-pollution community. Investigation of physicians' records tended to validate the questionnaire findings and did not reveal any difference in diagnostic patterns between communities. An analysis of the duration of residence in each area showed that the increased attack rates for croup and bronchitis in the high-pollution community were observed solely in children who had lived there for 3 years or longer. In this subgroup, the data on croup, which had the largest gradient, showed a 3-year attack rate in the high-pollution area of 26.4 percent, compared with 14.5-16.9 percent in the other communities. The 3-year attack rate for bronchitis was 23.6 percent, compared with 16.5-17.1 percent in the cleaner communities. These attack rates were adjusted for age, sex, and socioeconomic status.

It is noteworthy that people who had lived in the high pollution community for less than 3 years had much lower attack rates than short-term residents of the other communities. The explanations advanced for this finding are at best tentative. There were fewer short-term than long-term residents, so these anomalous findings decreased only slightly the overall gradient in lower respiratory infection attack rate for all residents. The questionnaire response rate was relatively low, 67 percent, and one-sixth of them had to be discarded because of inadequate information. Although a small-scale study appeared to show no difference between respondents and non-respondents, this may have been insufficient to rule out bias. The authors also state that there was a difference in attack rate between the community with mean suspended sulfates of 9 ug/m³ and the two communities with lower suspended sulfates but higher sulfur dioxide concentrations. This statement does not appear to be completely warranted by the data.

In general, the data do appear to substantiate an increase in lower respiratory disease in children exposed for a few years to sulfur dioxide at 90-100 ug/m³ and suspended sulfates at 15 ug/m³ in the presence of total suspended particles at concentrations below the air quality standard. No firm conclusions can be drawn, but the findings raise the suspicion of an effect of suspended sulfates.

A similar retrospective study was performed in almost 6,000 children living in five Rocky Mountain communities (Finklea et al. 1974). The gradient in sulfur dioxide and total suspended particles was much greater than in the Salt Lake Basin, and there was also a difference in suspended sulfates. Unfortunately, the analysis of the data was complicated by distinct differences in socioeconomic status and parental smoking, which varied with air pollution. After adjustment for age, sex, and socioeconomic status, an increase in the attack rate for lower respiratory infections in nonasthmatics was observed in pooled data from the two high-pollution communities. (In 1968-1970, those two communities had sulfur dioxide concentrations of 177 and 375 ug/m³, total suspended particle concentrations of 63 and 102 ug/m³, and suspended sulfate concentrations of 7.2 and 11.3 ug/m³; of the three low-pollution communities, two had relatively negligible concentrations of each pollutant, and the third, "low III" had a mean sulfur dioxide concentration of 67 ug/m³, a total suspended particle concentration of 115 ug/m³, and a suspended sulfate concentration of 7.3 ug/m³.) However, the results in nonasthmatics tended to be significant only when calculated for people who had two or more episodes and only in the subgroup that had lived in the same community for 3 years or longer. As in the Salt Lake study, croup rates correlated better with pollution than did bronchitis. There was also a tendency toward an increase in pneumonia and hospitalization rates in the high-pollution communities, but this was not statistically significant. Again, the finding of less disease in newcomers to the high-pollution areas than in newcomers to the low-pollution areas was puzzling. A more clearcut increase in the attack rate for lower respiratory illness was observed in asthmatics, but this was a smaller group and the findings were not adjusted for socioeconomic status, etc. Accordingly, the asthma data are inadequate to support firmly the investigators' contention of a pollutant effect in the "low III" community.

The findings are not as impressive as in the Salt Lake Basin, particularly in view of the larger gradient of sulfur dioxide and suspended particles (although not of suspended sulfates). Although comparison of these two similar studies is somewhat impeded by the larger difference in socioeconomic status between communities in the Rocky Mountain area, it should be noted that the relatively less polluted Salt Lake Basin area had higher adjusted attack rates--particularly multiple attack rates--of lower respiratory infection. Whether this represents physician diagnostic patterns, other local factors, or a difference in the administration of the two studies is unclear.

Somewhat different studies were performed in Chicago and New York in which the attack rate of acute respiratory disease in family members was obtained by telephone interview of mothers. In Chicago, data were obtained in 1969-1970 on over 2,500 families that had at least one child in nursery school (Finklea et al. 1974). No information about those who did not volunteer for the study is given. Families were assigned to pollutant categories by place of residence with reference to nearby monitoring stations. The three exposure categories were defined as Intermediate, High, and Highest, and the 1969-1970 monitoring data on sulfur dioxide and total suspended particles were 57 and 111 $\mu g/m^3$ for the Intermediate category, 51 and 126 $\mu g/m^3$ for the High category, and 106 and 151 ug/m³ for the Highest category. Pollution concentrations had been much higher previously. Socioeconomic and other factors investigated were relatively similar in the three groups. Among families that had resided in the same area for 3 years or longer, there was a 48 percent increase in the risk of lower respiratory infection in the nursery-school children in the Highest category, compared with those in the Intermediate category. Older siblings had a 16 percent increase in the risk of lower respiratory infection, and younger siblings had a 9 percent increase. An increase in the risk of upper respiratory infection in children was observed only in the younger siblings. Increased illness rates in the highest-pollution community were also observed during an influenza epidemic. Relatively little information is presented for the High-pollution community.

A similar telephone survey was performed in New York in 1970-1971 (Love et al. 1974). Volunteers were obtained from two communities within New York City--Howard Beach in Queens and the Westchester area of the Bronx--that were categorized as having intermediate pollution and from Riverhead, Long Island, which was chosen as the low-pollution community. In the years preceding the study, both New York City communities had sulfur dioxide and total suspended particle concentrations well above air quality standards. However, at the time of the study, annual mean sulfur dioxide concentrations were 51-63 ug/m³, and the concentrations of total suspended particles were at or somewhat above the air quality standard of 75 ug/m³. In Riverhead, pollutant concentrations had generally been low; in 1971, the sulfur dioxide concentration was 23 ug/m³, and the total suspended particle concentration was 34 ug/m³. Suspended sulfate concentrations in 1971 were 13.2 and 14.3 ug/m³ in the two New York City communities and 10.2 ug/m³ in Riverhead. This less distinctive gradient presumably represents atmospheric oxidation of sulfur dioxide en route to Riverhead from the New York City area.

Only a slight increase in lower respiratory tract illness in New York City children, compared with those in Riverhead, was observed. This was of borderline statistical significance ($p=0.07$). There was no difference in the attack rate for upper respiratory illness between the communities. Data are presented that suggest that respiratory illness was more severe in the intermediate-pollution communities. This was calculated by means of an arbitrary severity score whose assumed linearity is open to question. The problems with the New York City data include the proximity of the Howard Beach community to Kennedy Airport, possible differences in medical use patterns among the communities, and the lack of any trend in repeated lower respiratory tract infections in children, compared with those in other CHESS studies.

A conservative interpretation of the New York and Chicago CHESS family studies would be that they tend to add further weight to the reported association of childhood lower respiratory illness with concentrations of sulfur dioxide and total suspended particles above the current U.S. air quality standards. By themselves, however, they are not conclusive. Data on adults obtained in these studies are discussed below.

Hammer et al. (1974) have recently presented a paper on a further CHESS study in the New York area. Questionnaires concerning lower respiratory infection rates in children 1-12 years old were distributed to schoolchildren in the first through sixth grades to be filled out at home. The three New York communities previously studied and one additional area, Sheepshead Bay in Brooklyn, were evaluated. The past pollutant concentrations in Sheepshead Bay were estimated from the Howard Beach data, although the two communities are about 6 miles apart. Response rates for the questionnaire were about 80 percent. The 3-year lower respiratory infection attack rates were adjusted for the sex of the child and the educational status of the head of the household and are tabulated for the age groups 1-4 year, 5-8 years, and 9-12 years. White children in the Riverhead community tended to have lower rates for total lower respiratory illness, croup, and (in children 5-12 years old) bronchitis, compared with the New York City communities. However, the largest gradients were observed for pneumonia and hospitalization rates in the group 1-4 years old; in these categories, Riverhead reported a much greater incidence than the New York City communities. Black children had an even greater increase than white children in lower respiratory infection, croup, and bronchitis associated with pollution. Again, there was a markedly reversed gradient for hospitalization: black children 1-8 years old in Riverhead were hospitalized with a frequency 6 times greater than that of black children in New York City. The statistical significance of these findings is difficult to determine from the unpublished manuscript.

The anomalous data on pneumonia are attributed to two physicians who were responsible for the care of a large part of the Riverhead community, although the extent to which this factor contributed to the findings is not indicated. Pneumonia rates were relatively low and therefore perhaps somewhat less reliable. It is also conceivable that hospitalization rates were affected by the availability of hospital beds. If the findings on pneumonia and hospitalization are ignored, the data show a slight but inconsistent effect of sulfur dioxide and total suspended sulfate concentrations higher than air quality standards (in conjunction with a gradient for suspended sulfates) on the lower respiratory infection rate in children. However, more information about this unpublished study would be useful.

Additional CHESS studies have evaluated pulmonary function in children. Forced expiratory volume (0.75 sec) was measured in over 2,000 schoolchildren 5-13 years old living in the three New York areas described above during the 1970-1971 school year (Shy et al., 1974). Boys 9-13 years old living in Riverhead had a slightly but significantly higher forced expiratory volume than those living in New York City. A similar nonsignificant trend for girls in this age group was also noted. In contrast, no differences in the pulmonary function of the group 5-8 years old were observed. The authors suggest that the finding of an effect only in the older children is due to the recent improvement in New York City air quality, so that only those old enough to have been exposed to the much higher pollution of the past are affected.

In addition to some anomalous findings and possible unadjusted differences between the urban and suburban children (e.g., in cigarette-smoking), the major difficulty with this study appears to be the pulmonary function equipment, which has since been replaced because of instrument drift. This latter problem is perhaps overcome by the large numbers tested, but the findings require replication, particularly in view of the great importance of determining whether the costly control measures used in cities (such as New York) have led to a decrease in the adverse health effects of air pollution.

An earlier study in Cincinnati evaluated the forced expiratory volume of second-grade schoolchildren in six different schools paired for pollution (Shy et al. 1974), socioeconomic, and ethnic factors. Pulmonary function was measured four times a month in November 1967. February 1968, and May 1968 with the Stead-Wells spirometer, a standard but somewhat cumbersome instrument. Mean sulfur dioxide concentrations were 50 ug/m³ or less. The major pollution gradients were observed for total suspended particles, which averaged 96-114 ug/m³ in the high-pollution areas and 77-82 ug/m³ in the low-pollution areas; suspended sulfates, which averaged 8.9-9.5 ug/m³ in the high-pollution areas and 8.2-8.8 ug/m³ in the low-pollution areas; and suspended nitrates, which averaged 2.7-3.2 ug/m³ in the high-pollution areas and 2.4-2.6 ug/m³ in the low-pollution areas. Among white children, forced expiratory volume was lower in the more polluted areas, and this finding correlated best with suspended sulfates. Pulmonary function was not correlated with pollution in black children. Although the forced expiratory volume tended to be lowest in the winter months, when pollution was high, the results did not directly correlate with pollutant measurements immediately preceding the pulmonary function measurement.

Both the New York and Cincinnari pulmonary function studies (Shy et al. 1973) have been criticized by Higgins and Ferris (1974), who, after pointing out inconsistencies in the data and the relatively small observed differences, assert that "random scatter seems to be a much more likely explanation than air pollution."

Recently, Chapman et al. (1974) have presented a further CHESS study in which ventilatory function was evaluated in children living in Birmingham, Alabama, and Charlotte, North Carolina. These two cities are of interest, in that their sulfur dioxide concentrations are negligible and their particle concentrations tend to be high. In Charlotte, geometric mean total suspended particles had previously exceeded the air quality standard, but had declined; the concentration in 1972 was 70 $\mu g/m^3$. Total suspended particle concentrations in Birmingham were appreciably higher; in 1972, the concentration was 127 $\mu g/m^3$. A difference in suspended sulfates was also apparent; in 1972, Birmingham had a relatively high arithmetic mean concentration of 13.3 $\mu g/m^3$, and Charlotte had a concentration of 9.7 $\mu g/m^3$. Forced expiratory volume (0.75 sec) was measured in the fall, winter, and spring of the 1971-1972 school year in almost 8,000 children 5-13 years old in these two cities. In both black and white students and in both boys and girls, the forced expiratory volume was less in the more polluted city of Birmingham than in Charlotte.

As is true of other CHESS studies, the major positive finding is the consistency of an effect of air pollution, despite a number of experimental shortcomings. These include the necessity of changing the spirometer in the middle of the year because of equipment difficulties, the higher educational status of the Charlotte schoolchildren's parents, the greater degree of school busing in Charlotte, and the use of different personnel to perform the testing in the two cities (it is hoped that this was overcome by supervisory personnel). Unexpected findings include an inverse relationship between pulmonary function and socioeconomic class in Charlotte, a greater difference in females than in males (contrary to most other studies), and a decrease in the mean forced expiratory volume during the school year in Birmingham. It is not clear whether the anomalous data represent minor quirks of the population under study or indicate intercity variations in the performance of the pulmonary function tests. Continued study of these same children, thereby allowing each population to serve as its own control during changes in air pollution in the next few years, should provide important information.

Another recent study is that of Lebowitz et al. (1974), who noted a decrease in forced expiratory volume and maximal midexpiratory flow immediately after outdoor exercise in children living in a smelter community. This may have been related to pollution, but monitoring data are not given.

The studies described above support the contention that children are at risk when subjected to urban air pollution. This is particularly true for lower respiratory tract infections and for asthma, which is discussed in the next section. The findings further suggest that the current air quality standards for sulfur dioxide and suspended particles do not have appreciable safety factors in regard to this effect. Therefore, it should be assumed that concentrations of sulfur dioxide or total suspended particles above the present standards will result in an increase in childhood lower respiratory tract infection. There is a distinct possibility that suspended sulfates are directly related to this effect.

One crucial question concerning the benefits of further air pollution control cannot be definitely answered at present: whether repeated childhood respiratory infections result in an increase in respiratory morbidity in adulthood. Obviously, the association of air pollution with a transient minor childhood respiratory infection is of less potential public health and societal significance if such illnesses have no sequelae in adulthood. This is a controversial subject, with literature supporting both sides of the question (Colley and

Reid 1970, Holland et al. 1969, Reid 1969, Harnett and Mair 1963, Rosenbaum 1961). However, it must be emphasized that the advent of antibiotics may have greatly changed the impact of childhood bacterial respiratory illnesses while having little effect on viral illnesses. If it is accepted that the pulmonary consequences of bacterial infection may be more serious than those of the usual viral illness, then the fact that people born at the time that penicillin first became generally available are now only in their third decade makes it difficult to establish whether present recurrent childhood respiratory illnesses will lead to increased chronic respiratory disease in later adult life. Although, from the medical point of view, the available evidence indicates that it is prudent to assume that recurrent childhood respiratory infections do play a role in the eventual development of disabling chronic respiratory disease, there is at present no absolute scientific evidence to support this assumption.

BRONCHIAL ASTHMA

Bronchial asthma is a disease characterized by attacks of marked hyperconstriction of the trachea and bronchi, leading to wheezing and gasping for breath. Treatment of an uncomplicated attack is generally aimed at bronchodilatation by pharmacologic means. As with the other respiratory disease states under discussion, it is difficult to provide a clearcut definition of this disorder without either being too restrictive or avoiding overlap with other pulmonary pathology.

In its most obvious form, bronchial asthma begins in childhood, and there is a seasonal incidence of attacks that appear to be related to an allergic precipitating factor. Between attacks, respiratory function is relatively normal, although an increased susceptibility to bronchoconstrictive agents has been noted. Bronchial asthma in adults may arise *de novo* or may represent childhood asthma that has failed to remit. Clinically, asthma has often been divided into extrinsic and intrinsic types. In the former, exogenous allergens are known or suspected; the latter has no obvious cause or is associated with respiratory infections. Asthma in adults tends to be in the latter classification and, particularly when associated with respiratory infection, often has a peak incidence of attacks during the winter, rather than summer and fall, as observed with allergic individuals. It is particularly difficult in adults to formulate criteria that will distinguish true bronchial asthma from cases of bronchitis in which bronchoconstriction is more prominent than usual; such bronchitis is often characterized by the term "asthmatic bronchitis."

The reported prevalence rates for asthma vary widely, with perhaps 3-5 percent being a reasonable estimate. The asthma mortality for all age groups in the United States in the last decade was around 2-3/100,000. This disease accounts for a reported 23 percent of all school-day absences due to chronic conditions (Gordis 1973).

Patients with allergic asthma are notably difficult to study, in view of their marked variability of response. One or more specific allergens are usually required to elicit an asthmatic attack, but many asthmatic people appear to undergo a marked hyperreactivity of the bronchial tract, so that a host of exogenous and endogenous factors, including psychologic stress, significantly interact in the genesis of an acute attack. Because asthmatics may represent an extreme on the scale of hypersusceptibility to bronchoconstriction, they appear to be ideal subjects for epidemiologic study of the acute response to air pollutants. However, the multiplicity of factors producing a response might also tend to obscure any specific reaction to pollutants. Furthermore, the seasonal nature of asthmatic attacks presumably reflects meteorologic factors, as well as variations in specific allergen sources. The extent to which factors that affect allergen concentrations are synchronous with factors that affect pollutant concentrations will have a great impact on any observed association between asthma and air pollution. These considerations suggest that epidemiologic studies of the association between air pollution and asthmatic attacks should include relatively large numbers of asthmatics and pay careful attention to meteorologic conditions.

An association of asthma attacks with air pollution has been reported by a number of investigators and has been reviewed by Zweiman et al. (1972) and by Thomas and McGovern (1971). Retrospective questioning of residents of Donora after the 1948 air pollution disaster revealed that 87.6 percent of the asthmatics reported respiratory symptoms during this episode, compared with 77.2 percent of people with cardiac disease and 42.7 percent of the general population (Schrenk et al. 1949).

Zeidberg et al. (1961), as part of the Nashville air pollution study, evaluated 49 adults and 34 children with bronchial asthma over a 1-year period. Diaries obtained from each subject were used to calculate daily attack rates, which were then correlated with mean annual and daily sulfation rates according to place of residence. There was almost a doubling in daily asthma attack rates in areas with annual sulfation rates of 0.350 mg/100 cm² per day or greater, compared with those in areas with sulfation rates of less than 0.150mg/100 cm² per day. This was due almost totally to more than a fivefold increase in adult male attack rates in the more polluted area. The 30 days with the highest sulfation rates had a 22 percent increase in asthma attacks, compared with the 46 days with the lowest pollution. When daily sulfation rates were analyzed according to asthma attack rates on the following day, the association was reported to be even more statistically significant, implying a delay in the manifestation of toxicity. The sulfation rates for the more polluted days are not specified, but other data make it probable that the high concentrations were near or below an equivalent 24-hr sulfur dioxide concentration of 0.14 ppm, which is the current standard. Linear correlation coefficients between monthly asthma attack rates and various meteorologic conditions were calculated. These were interpreted by the authors as not showing any consistent association of asthma attack rate with temperature. However, the tabulated data show that in four of 12 months there was a statistically significant inverse correlation of attack rate with temperature. Although not clearly stated, the lack of reported extensive analysis of the association of total suspended particles with asthma attack rates suggests that this was not believed to be a significant factor.

Panels of asthmatic subjects have also been evaluated as part of the ongoing CHESS studies in the New York City and Salt Lake City areas. The Salt Lake Basin was chosen, because a large part of its pollution is due to a single smelter source (Finklea et al. 1974). This results in appreciable concentrations of sulfur dioxide and suspended sulfates in association with relatively low concentrations of total suspended particles, nitrates, nitrogen dioxide, and other usual urban pollutants that might tend to obscure the effect of sulfur oxides. Four communities with different pollution concentrations were evaluated. In contrast with the other Salt Lake Basin CHESS studies, the data in the asthma study were related to daily air pollution within each community. Approximately 50 panelists in each community, about half of whom were over 16 years old and who lived within 2 miles of the monitoring station, filled out a weekly diary from March to September 1971. The data evaluated were daily attack rates, 24-hr average pollution concentrations, and minimum temperatures. It should be noted that these communities were well within the annual average air quality standards for sulfur dioxide and total suspended particles during the time of the study. Only rarely were 24-hr standards exceeded. Between 16 and 33 percent of the original panel subjects withdrew from the study and were replaced at variable rates in the different communities.

The data were subjected to a complex series of statistical evaluations, and the final results were interpreted as demonstrating a definite association of relatively low concentrations of suspended sulfates with the asthma attack rate when the minimal temperature was above 49 F. Simple correlation coefficients showed that the strongest association with asthma attack rates was the inverse of daily minimum temperature. In this analysis, a significant statistical association of suspended sulfates with asthma attacks was observed only in the high-pollution community; the low-pollution community had a negative association of asthma attack rates with suspended sulfates at all temperatures. Multiple regression analysis, which evaluated the effect of air pollutants after first considering that of minimum temperature, again showed a highly significant statistical association of sulfates with daily asthma attack rates in the high-pollution community. The data for all four communities were then combined to derive temperature-specific relative-risk models for the various concentration ranges of each pollutant. The base rate for the computation of excess risk was derived from the asthma attack rate on warm days on which there were low concentrations of the specific pollutant. Again, the most significant effect was noted for sulfates on days when the minimum temperature was greater than 49 F. In this temperature range, with the base asthma attack rate calculated for days in which 24-hr suspended sulfate concentrations were 6 ug/m³ or less, there was a 17 percent increase in risk associated with sulfate concentrations of 6.1-8.0 ug/m³, a 35 percent increase in risk associated with concentrations of 8.1-10.0 ug/m³, and a 50 percent increase in risk associated with concentrations greater than 10 ug/m³. Much smaller increases in risk were also reported in the same temperature range when the concentration of either total suspended particles or sulfur dioxide was above 60 ug/m³.

These data were also used to extrapolate expected risks for pollution concentrations at the 24-hr standard (not to be exceeded more than once a year). For sulfur dioxide, this was 254 percent, and for total suspended particles, 170 percent of the baseline on days when the minimum temperature was greater than 49 F.

Approximate "thresholds" for an effect on asthma attack rates were also calculated. As stated in the summary of the 1970-1971 CHESS document (Finklea et al. 1974), these are worst-case estimates and are not to be taken as definitive findings. The estimated threshold for suspended sulfates is 1.4 ug/m³; for sulfur dioxide, 23 ug/m³; and for total suspended particles, 71 ug/m³. Those are all for days when the minimal temperature is greater than 49 F.

In the New York area, asthma panels were selected in three communities, two of which were designated as having intermediate pollution, and one, low pollution (Finklea et al. 1974). There were initially about 50 subjects in each panel who lived within 1/2 miles of a monitoring station, and the methods of data gathering and analysis were similar to those in the Salt Lake Basin study. All communities were consistently within the 24-hr standards for sulfur dioxide and total suspended particles at the time of the study. The study was performed from October 1970 to May 1971; that is a colder time of year than in the Salt Lake area and would miss part of the high-pollen season. The overall response rate was less than that for the Salt Lake Basin. Only 75 percent of the mailed diaries were usable in the analysis.

No consistent simple correlations were found between attack rates and individual pollutants or temperature. With multiple regression analysis after adjustment for temperature, some association of asthma attack rates with suspended sulfates was observed in two of the communities, and with sulfur dioxide and suspended nitrates in one community each. Temperature-specific relative-risk models were calculated for daily minimum temperatures of less than 30 F, 30-50 F, and greater than 50 F. Compared with those of the Salt Lake Basin studies, increases in relative risk were small. A 13 percent increase was noted for total suspended particles at 76-260 ug/m³ on days when the minimum temperature was 30-50 F. In the same temperature range for suspended sulfates, there was a 9 percent increase at 8.1-10.0 ug/m³ and an 8 percent increase when the 24-hr concentration was above 10 ug/m³. A 10 percent increase in asthma attack rate was also noted when the minimum temperature was above 50 F and the suspended sulfate concentration was greater than 10 ug/m³. No consistent effect of sulfur dioxide on asthma attack rates was observed. Fitting of threshold functions resulted in an estimate for suspended sulfates of 12 ug/m³ when minimum daily temperature was 30-50 F and 7.3 ug/m³ for minimal temperature of less than 30 F. The threshold for total suspended particles was estimated at 56 ug/m³ when minimum daily temperature was 30-50 F; it was further calculated that, at 260 ug/m³, the 24-hr standard, there would be a 22 percent increase in asthma attack rate.

The 1970-1971 CHESS studies have received a good deal of criticism, although it must be noted that many of the potential problems are discussed in detail in the CHESS document (EPA 1974). Specific problems with the two asthma studies described above include the relatively poor response rate and high turnover, which may be inherent in diary studies of patients with this disorder; the presence of anomalous data, such as a decrease in the temperature-specific risk of asthma attacks associated with increasing sulfate concentrations on days when the minimum temperature was 30-50 F in the Salt Lake area; the failure to consider temperature change, rather than absolute temperatures; the lack of information on atmospheric allergens, which conceivably vary with pollution concentrations; and the tendency toward overanalysis and overinterpretation of the available data. Reasonable preliminary conclusions from these studies are that they do provide support for the association of air pollution with asthma attacks and that they further implicate suspended sulfates as an important deleterious component of polluted air. It does not appear to be necessary or reasonable at this time to draw firm conclusions concerning the 1970-1971 CHESS asthma studies, inasmuch as data have been collected and analyzed for later years and the reports will be forthcoming soon. If the more recent studies are able to replicate the 1970-1971 findings, this will provide much firmer support for the interpretations advanced by the CHESS investigators.

Other CHESS studies evaluating lower respiratory infection in children have also studied asthmatics. Increased attack rates of croup and bronchitis in this group were observed in both the Rocky Mountain and Salt Lake Basin study areas (Finklea et al. 1974, Nelson et al. 1974).

Investigators at the U.S. Environmental Protection Agency have also studied a panel of asthmatics in New Cumberland, West Virginia (Cohen et al. 1972). This small town was the location of a low-stack power plant that used high sulfur coal with no control measures. Of the 43 asthmatics identified in the community, the records of 20, mostly adults, were suitable for study. These people filled out weekly diaries for 7 months, which were used to calculate asthma attack rates in association with meteorologic factors and pollutant concentrations. Temperature was strongly and negatively associated with attack rates. After multivariate adjustment for the effect of temperature, all the pollutants measured--including total suspended particles, sulfur dioxide, suspended sulfates, and suspended nitrates--were also significantly correlated with asthma attack rate. However, after temperature and any single pollutant were taken into account, no significant relation with any of the other pollutants remained. Therefore, the data do not provide information on which, if any, single pollutant was responsible for the observed association. After adjustment for temperature, suspended sulfate was the pollutant most highly correlated with asthma attack rate; but, as pointed out by the authors, this may have been due to the exceptionally high correlation of suspended sulfates with temperature. Furthermore, the attack rate on days when the suspended sulfate concentration was higher than 20 $\mu g/m^3$ was not significantly greater than that on days when it was lower than 20 $\mu g/m^3$. In this high-low analysis, significant increases in attack rate were observed for daily total suspended particles and for daily sulfur dioxide when the points of demarcation were 150 $\mu g/m^3$ and 0.07 ppm, respectively.

Other facets of this study include a commendable attempt to validate the diaries by physician visits and a relatively extensive monitoring system focused in a small area. In addition, it is one of the few point-source studies that have evaluated the effects of pollutants that originated in a power plant, rather than a smelter or pulp mill. Unfortunately, the results are handicapped by the very small number of asthmatics who were studied. In view of the great variability in the response of asthmatics, the justification for many of the highly sophisticated statistical techniques used in this study is perhaps open to question. Nevertheless, the general findings appear to support the contention that power-plant effluents are instrumental in potentiating asthma attacks.

Sultz et al. (1970) evaluated the incidence of hospitalization in Erie County, New York, for asthma, eczema (an allergic skin disorder), and diabetes mellitus in children under 15 years old, in comparison with total suspended particle concentrations and socioeconomic status. The study used the data of Winkelstein et al. (1967), who, on the basis of monitoring for 1961-1963, had previously divided Erie County into four areas of annual mean concentration of total suspended particles; less than 80, 80-100, 100-135, and more than 135 $\mu g/m^3$. Four categories of social class were obtained by dividing the socioeconomic data for census tracts into quartiles. Asthma hospitalization data were obtained for 1956-1961, and eczema hospitalization data for 1951-1961. Standardized morbidity ratios were obtained by dividing the observed rates for given pollutant ranges or social classes by the total rates in the entire population. The morbidity ratios for asthma hospitalization rose steadily, from 86 percent for low pollution to 114 percent for high pollution. No consistent effect for social class was observed. There was an even steeper gradient for eczema, with morbidity ratios of 53 percent for low pollution and 130 percent for high pollution. However, there was also a wide difference for social class. The disparity between asthma and eczema hospitalization rates

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in the high-pollution area, compared with the low-pollution area, was even greater when the analysis was restricted to males under the age of 5. Diabetes mellitus hospitalization rates did not appear to be associated with air pollution. No attempt was made to evaluate the effects of sulfur dioxide, nor were meteorologic conditions considered.

It is difficult to interpret the finding of an association of air pollution with an allergic skin condition. Although earlier authors had suggested that fossil-fuel combustion products, particularly sulfur dioxide, might be allergenic (Pirila et al. 1963, Pirila 1954), there is inadequate evidence to support this hypothesis. People with childhood eczema tend to develop allergic respiratory disease, but this would not account for their hospitalization with a skin condition. One possible conclusion is that this study illustrates a potential pitfall in epidemiologic studies of allergic asthma: that meteorologic conditions conducive to a distribution of pollutants in a given area may also predispose to a similar distribution of allergenic organic material.

Chiaramonte et al. (1970) studied emergency-room visits at a Brooklyn hospital during a 3-week period in November 1966 in which a sharp increase in sulfur dioxide concentrations occurred in the middle week. The highest 24-hr sulfur dioxide concentration was 0.8 ppm. A statistically significant increase in visits for all respiratory disease and for asthma was observed 3 days after the sulfur dioxide peak. This delay may have been complicated by the Thanksgiving holidays. No data on weather or on other pollutants are given.

A more recent study of the relation of asthma emergency-room visits to air pollution in New York City has been reported by I. Goldstein and Block (1974). They evaluated data from hospitals in Harlem and in Brooklyn near the Bedford-Stuyvesant area for the period of September-December 1970. Air monitoring information was obtained from nearby stations. With a linear multiple regression model, using asthma visits as the dependent variable and daily sulfur dioxide and average temperature as the independent variables, they calculated multiple and partial correlation coefficients that demonstrated that, when temperature was held constant, there was a positive correlation between asthma visits and sulfur dioxide in Brooklyn, but not in Harlem. In both areas, the results were similar for younger and older groups and for different hospitals. Repeat analysis for the group under 13 years old in September-December 1971 at Kings County Hospital in Brooklyn replicated the 1970 findings. Negative correlations of asthma visits with temperature were observed, but these were not as strong as the positive correlations for sulfur dioxide. The tabulated data show a 50-90 percent increase in asthma emergency-room visits in the group under 13 years old on the 12 high-pollution days, when sulfur dioxide concentrations averaged 0.108 ppm. Mean daily sulfur dioxide in Harlem was similar to or slightly higher than that in Brooklyn.

Temporally, this study at least partially overlapped two other New York studies: the CHESS diary study, in which one of the panel areas was not too far from Brooklyn, and the study of Rao et al. (1973), which also used Kings County Hospital pediatric emergency-room asthma visits, including the period covered by I. Goldstein and Block (1974). As discussed below, the latter study, which used no temperature data and less sophisticated statistical techniques, did not find a correlation of asthma with sulfur dioxide concentration. The finding of strong correlations in Brooklyn but not in Harlem among similar ethnic and socioeconomic groups is unexplained. As suggested by the authors, this may indicate the presence of a causative agent that is distributed with sulfur dioxide in Brooklyn, but not in Harlem. In this respect, it should be noted that air pollution derived from the industrial areas of northern New Jersey has a tendency to pass over Manhattan before fumigating Brooklyn. Further analysis of these data for longer periods and with additional variables is underway.

Some studies have failed to show a positive correlation of air pollution with asthma attacks. In an investigation of nighttime emergency visits to a hospital in Brisbane, Australia, Derrick (1970) reported that there was, if anything, a negative correlation between smoke shade concentration and asthma. In this area, the weather factors associated with an increase in asthma attacks were completely different from those resulting in high smoke shade concentrations.

Greenburg et al. (1964) noted a sharp increase in New York City hospital emergency-clinic visits in September that was related to the onset of cold weather, and not to air pollution. No statistically significant increase in symptoms in asthmatics in relation to air pollution was noted in a Chicago study by Carnow et al. (1969), although there was a slight tendency toward heightened morbidity on the day after increased sulfur dioxide concentrations. Meteorologic data are not presented.

Rao et al. (1973) also reported a negative correlation of smoke shade with pediatric emergency-room visits in Kings County Hospital in Brooklyn. No association with sulfur dioxide was observed. It is of interest that the period of this study, October 1970-March 1971, was also included in the CHESS investigation cited above. However, the report of Rao et al. has a number of serious deficiencies, including the lack of temperature data and the fact that the hospital district includes areas at an appreciable distance from the monitoring station. In addition, emergency-room visits may not constitute as sensitive an indicator as individual diaries. Glasser and Greenburg (1967), in a study of the November 1966 New York air pollution episode, noted an increase in emergency-clinic visits in only three of seven hospitals studied. This may, however, have been affected by the intervening Thanksgiving Day holiday.

Air pollution has also been postulated to be causally related to periodic outbreaks of asthma in New Orleans (Weill et al. 1964) and to the asthma observed in American servicemen stationed in the Yokohama (Phelps 1965) area after World War II. However, in neither case has a specific etiology been established, and the role of products of stationary fossil-fuel combustion is in doubt.

Taken as a whole, the studies described here suggest that sulfur oxides and/or particles potentiate asthma attacks in susceptible people. Such effects might be reasonably expected to occur at pollutant concentrations somewhat above the present air quality standards in conjunction with specific meteorologic factors. The data also indicate a possible role of suspended sulfates. The degree of variability observed in these studies is probably due in part to inherent difficulties in the study of asthmatics. Whether these difficulties lead to a net underestimation of the relation of air pollution to asthma is not clear.

A puzzling facet of the association of asthma attacks with sulfur oxides is the apparent delay in the onset of the attack after peak exposures to sulfur dioxide reported in a number of studies. If particulate sulfates are the offending agents, it is possible that this lag reflects the time required for oxidation of sulfur dioxide. However, most asthma attacks occur late at night, when people tend to be indoors and at rest and when the air pollution burden is presumably at its lowest. Furthermore, the bronchoconstrictive effect of sulfur oxides observed in animals and man is usually relatively rapid. It is therefore difficult to envision the mechanisms for pollutant-induced asthma attacks on the basis of the available knowledge of sulfur oxide effects. Perhaps future studies should pay more attention to the exact time of attack and additional information should be obtained concerning the possibility that sulfur oxide exposure sensitizes the respiratory tract to the later action of other bronchoconstrictive agents.

CHRONIC BRONCHITIS AND EMPHYSEMA

Definition and Classification

Chronic lung disease may be divided into a number of classifications on the basis of such characteristics as etiology, pathologic anatomy, and symptomatology. For some pulmonary disorders, there is generally good agreement as to the diagnostic criteria necessary to define the entity. However, most people with chronic respiratory disease have a disorder, or group of disorders, variously known as chronic bronchitis, emphysema, chronic obstructive pulmonary disease, or chronic nonspecific respiratory disease. A major reason for the multiplicity of terms and the differing diagnostic criteria among physicians is that most patients have a combination of the pathophysiologic findings usually ascribed to different types. The following simplified discussion of chronic lung disease is presented mainly for the purpose of assisting laymen to understand the later discussion of the health effects of air pollution. (Asthma has already been discussed separately.)

Chronic bronchitis is best typified as a disorder characterized by the continuous hypersecretion of mucus into the airways, leading to problems associated with keeping the bronchial airways open. Any interference with mucociliary transport or any further stimulation of mucus production, both of which may be consequences of sulfur oxide inhalation, would have deleterious effects. Long-term exposure of animals to very high concentrations of sulfur dioxide has been found to produce an increased number of mucus-secreting cells and a pathologic picture resembling chronic bronchitis.

Emphysema, in its purest form, refers to destruction and dilation of alveoli, the air sacs where gas exchange occurs, and hyperinflation of the lung. In the United States, the term "chronic obstructive pulmonary disease" has often been used, and this can be divided into two types: Patients with type A, in which alveolar destruction predominates, tend to have severe shortness of breath, but little cough or sputum production. Patients with type B have mostly brochitic findings--they experience pronounced cough and sputum production with less shortness of breath. Although it is more common in type B disease, patients with both types often develop secondary heart disease (cor pulmonale). These types appear to represent the extremes of a continuum, with most patients having a mixture of alveolar destruction and bronchial hypersecretion. In general, longstanding chronic bronchitis leads to emphysema, although emphysema is occasionally observed in the absence of bronchial hypersecretion. With respect to the effects of sulfur oxides, it is apparent that pollutants that tend to be absorbed in the upper airway, such as gaseous sulfur dioxide, are more likely to be associated with chronic bronchitis, whereas those which reach the alveoli, such as fine particles, might produce emphysema. Conversely, the presence of chronic respiratory disease could interfere with the clearance and disposal of inhaled pollutants, thereby potentiating their effects.

Obstruction to air flow during expiration is an important part of the clinical picture in patients with chronic bronchitis and emphysema. Acute bronchoconstriction, which is a feature of the response to inhaled irritants in laboratory animals and man, would be expected to worsen any preexisting obstructive component materially. Population studies of the effects of air pollutants have often used pulmonary function tests, such as peak expiratory flow rate and forced expiratory volumes, to assess airway obstruction. However, although these are excellent screening devices, more sophisticated measures of pulmonary function are required to assess accurately the pathologic processes and airway sites of respiratory obstruction. The term "chronic nonspecific respiratory disease" has been used by a number of epidemiologists in recent years and appears to be more appropriate for air pollution studies, in that the available information is usually obtained by questionnaires or simple pulmonary function testing and is therefore not sufficient to distinguish clearly between possible types of chronic respiratory disease.

There have been substantial international differences in the reported prevalence rates for chronic bronchitis (Reid 1962, Christensen and Wood 1958). This may be due in part to ethnic factors, extent of cigarette consumption, or presence of air pollution, but to a large extent it represents differences in diagnostic criteria. Most of the studies about to be discussed are from the American or British literature, so it should be noted that the term "chronic bronchitis" is used far more often by the British. This does not negate the appropriateness of the British findings to chronic respiratory disease observed in the United States.

A discussion of the effects of air pollution in chronic respiratory disease would be incomplete without mention of two major determinants of lung function: cigarette-smoking and aging. Cigarette consumption has been clearly and consistently related to a loss of lung function and to a prevalence of chronic respiratory disease. Among those who smoke, it is a much larger determining factor in their disease than is exposure to usual concentrations of urban air pollutants. Of course, cigarette-smoking is voluntary, and breathing is not. A significant decrease in adult lung function occurs with age, so most indexes of pulmonary function are standardized for a given age range. Thus, a 30-year-old with pulmonary function in the normal range for a 70-year-old would be considered to have respiratory disease. The processes involved in "normal" aging are not understood, but, to the extent that they may represent an accumulation of minor pathologic insults, the inhalation of irritant air pollutants over many decades conceivably accelerates lung aging. This is, of course, speculative.

The classification and epidemiology of chronic respiratory disease in relation to air pollution have been reviewed by Ferris (1969, 1973) Higgins (1974), and Holland (1972).

Mortality Studies

There is an extensive literature dealing with the adverse health effects of industrial effluents. Epidemiologic studies relating respiratory morbidity and mortality to air pollution have been a fixture of the medical literature for quite some time. An early example of the epidemiologic approach in which a geographic comparison of death rates is used to ascertain the effect of air pollution was reported by Ramazzini in 1713 (Wright tr. 1964):

"A few years ago a violent dispute arose between a citizen of Finale, a town in the dominion of Modena, and a certain business man, a Modenese, who owned a huge laboratory at Finale where he manufactured sublimate. The citizen of Finale brought a lawsuit against this manufacturer and demanded that he should move his workshop outside the town or to some other place, on the ground that he poisoned the whole neighborhood whenever his workmen roasted vitriol in the furnace to make sublimate. To prove the truth of his accusation he produced the sworn testimony of the doctor of Finale and also the parish register of deaths, from which it appeared that many more persons died annually in that quarter and in the immediate neighborhood of the laboratory than in other localities. Moreover, the doctor gave evidence that the residents of that neighborhood usually died of wasting disease and diseases of the chest; this he ascribed to the fumes given off by the vitriol, which so tainted the air near by that it was rendered unhealthy and dangerous for the lungs. Dr. Bernardino Corradi, the commissioner of ordnance in the Duchy of Este, defended the manufacturer, while Dr. Casina Stabe, then the town-physician, spoke for the plaintiff. Various cleverly worded documents were published by both sides, and this dispute which was literally 'about the shadow of smoke', as the saying is, was hotly argued. In the end the jury sustained the manufacturer, and vitriol was found not guilty. Whether in this case the legal expert gave a correct verdict, I leave to the decision of those who are experts in natural science."

Studies of mortality depend heavily on the accurate filing of death certificates. Unfortunately, this is not a highly reliable source of data, particularly if it is used to sample the total amount and types of respiratory disease mortality in a given area. In patients with chronic pulmonary disease, the immediate cause of death might be an otherwise minor respiratory infection or cardiac failure secondary to the pulmonary disease. Furthermore, the cause of death may not be directly related. The extent to which specific chronic respiratory diseases are listed as immediate causes of death or as underlying conditions depends on the familiarity of the certifying physicians with the individual cases, their diagnostic acumen, and local medical usage. In addition, death certificates are often filed according to place of death, which may not be where the person lived for most of his life.

Despite these possible complicating factors, the substantial rise in recent years of chronic respiratory disease mortality clearly indicates that this disorder is an increasingly important factor in the well-being of the general population (HEW 1974). Age-adjusted death rates for bronchitis, emphysema, and asthma combined in the United States in 1958 were 6.9/100,000 population (males, 11.4; females, 2.9) in 1958 and 12.1/100,000 (males, 21.4; females, 4.7) in 1967, on the basis of approximately the same classification of disease. A closer look at the data reveals that, despite this increase, there has been an absolute decline in reported asthma deaths, which formerly accounted for 60 percent of total chronic respiratory disease deaths and now account for less than 10 percent. This great increase in bronchitis and emphysema mortality is unlikely to be due simply to changes in diagnostic criteria. The increase in chronic respiratory disease death rate is more notable in white males than in nonwhite males; this is consistent with the apparent greater effect of pulmonary irritants on whites. During this period, death rates for influenza and pneumonia combined were relatively stable; they are currently about double the death rate for chronic respiratory disease.

The causal relation of severe air pollution episodes to increased mortality is firmly established (Glasser and Greenburg 1971, Greenburg et al. 1962, Logan 1953, Schrenk et al. 1949, Firket 1931). Older people with preexisting heart and lung disease are particularly at risk, although increased mortality in all age groups was observed during the disastrous London fog of 1952. An important question concerning these severe episodes is whether the life span is shortened only for those who die immediately or for a larger portion--perhaps all--of the population. If these episodes only shorten slightly the life span of the severely ill, then one would expect to observe fewer than the usual number of deaths in the period after an episode. This has been observed on occasion, but never to the equivalent extent of the excess of deaths during an episode. However, the variable nature of mortality data does not permit the determination of whether life span is shortened only in relatively unhealthy people, who will then die in the days and weeks after exposure, or in a much larger segment of the population.

An extensive literature concerning the association of air pollution with respiratory disease mortality in Great Britain is available. In 1964, Martin reviewed a number of these studies, evaluating variations in mortality in relation to air pollution. For the London area, it appeared that there were critical concentrations of smoke (2,000 $\mu g/m^3$) and sulfur dioxide (0.4 ppm) above which a marked immediate excess in mortality occurred during 1954-1957. Further study of the winters of 1958-1959 and 1959-1960 showed high correlations of death rates (computed as 15-day moving averages to remove seasonal trends) with both sulfur dioxide and smoke. The effect appeared to be a relatively immediate one, and it was difficult to determine a threshold from the plotted data. Pemberton and Goldberg (1954) evaluated the 1950-1952 mortality rates in county boroughs of England and Wales. A significant correlation with sulfur dioxide (sulfation rate) was observed for bronchitis mortality in males 45 years old and over. Much smaller associations of bronchitis death rates were observed with sulfation in females and with particles in either sex. Stocks (1959) noted an association of bronchitis mortality with air pollution for 1950-1953 in the county boroughs of England and Wales after adjustment for the effects of population density. Evaluation of a smaller area in which more comprehensive air monitoring data were available showed a correlation of bronchitis mortality with dust deposit, but not with smoke shade, whereas the opposite was true for lung cancer.

Daly (1959) also reported a high correlation between fuel consumption and bronchitis mortality in middle-aged men residing in large towns in England and Wales for 1950-1952. After adjustment for social class, the correlation was somewhat smaller. Pneumonia mortality was only slightly related to fuel consumption.

During a similar period, 1950-[illegible], two studies were reported by Gorham ([illegible] 1958) that are of particular interest. I[illegible] he found that bronchitis mortality in [illegible]d, Scotland, and Wales was strongly i[illegible]ly correlated with the pH of winter precipitation. At a pH of 4 or lower in urban areas, the bronchitis mortality was 100/100,000; at a pH of 6 or higher, it was 60/100,000. He also noted that the rural bronchitis mortality was 46/100,000, which implies that rain acidity is not the only factor involved. A smaller, but still significant, correlation of bronchitis mortality with winter sulfation rate (a measure of sulfur dioxide) was also observed. A partial regression equation was calculated in which bronchitis mortality per 100,000 population is equal to 192(0.838)a, where a is the pH of winter precipitation.

Gore and Shaddick (1958) evaluated standardized mortality ratios for deaths in a 2-year period (1954-1956) in different areas of London and compared them with pollution, social class, and percentage of people born in London. Smoke was directly related to bronchitis deaths, but sulfur dioxide was not. However, in association with length of residence, both these pollutants were significantly correlated with bronchitis mortality. Similarly, Hewitt (1956) observed a significant correlation of London bronchitis deaths, and deaths from all causes, with a composite index of sulfur dioxide and percentage born in London. These studies suggest that the duration of exposure to London pollution was important in eventual bronchitis deaths and indicate a long-term effect of pollution on mortality. Similar studies have not been performed in the more peripatetic American population.

Burn and Pemberton (1963) divided the heavily polluted area of Salford, Great Britain, into five districts on the basis of smoke and sulfur dioxide concentrations. Deaths from all causes, bronchitis, and, to a lesser extent, lung cancer were related to air pollution in 1958-1959. Deaths from arteriosclerotic heart disease and strokes were not associated with pollution. The positive findings may be due in part to social-class differences, which could not be evaluated.

More pertinent to current considerations of air pollution control strategy is the question of whether daily variations in pollution concentrations at or somewhat above current U.S. air quality standards have any short-term effect on mortality. To evaluate this question, it is necessary to assess carefully the variations in daily mortality not due to air pollution. Of particular importance are factors that correlate with both air pollution and death rates, such as meteorologic conditions and influenza epidemics, the latter tending to occur during the winter heating season.

Among the studies of the effect of air pollution on short-term mortality in Great Britain is that of Boyd (1960), who related weekly temperature and pollution concentrations to mortality in the succeeding week. Both smoke and sulfur dioxide concentrations correlated with deaths due to respiratory disease, and the effects were most apparent at lower temperatures. One relatively unusual finding among British investigators was a better correlation with sulfur dioxide than with smoke. The sulfur dioxide data were tabulated as mean weekly sulfur dioxide concentrations of less than 0.1, 0.1-0.15, and more than 0.15 ppm. An increase in respiratory disease mortality was generally observed in each temperature range as sulfur dioxide concentration increased.

Numerous American studies have related mortality to air pollution. Those dealing with acute episodes have been cited elsewhere in this document. An interesting follow-up of the 1948

Donora disaster was performed by Ciocco and Thompson in 1957 (1961). People who reported illness during the acute episode later had an increased prevalence of illness and higher mortality rates than those who did not report being affected in 1948. This still held true, although to a lesser extent, when the analysis was restricted to those who had been in good health before the 1948 episode. The findings may be interpreted as indicating that air pollution shortens the life span of those who survive an acute episode or that those who respond to environmental stress are more susceptible to serious diseases. Most likely, the correct interpretation is a combination of the two.

Some relatively early U.S. Public Health Service studies were described by Rumford in 1961. Four pairs of Chicago health districts were selected for differences in industrial activity but similarity in socioeconomic status. Evaluation of white females showed an increased mortality from all heart and lung disease in the industrial areas. However, in Philadelphia, only deaths from chronic rheumatic heart disease could be related to dustfall. Even earlier studies by Mills (1952, 1948, 1943) had indicated a relation of sootfall to respiratory mortality in Chicago, Pittsburgh, Cincinnati, and Detroit.

An extensive series of studies, including mortality evaluations, were performed by the U.S. Public Health Service in Nashville, Tennessee, beginning in 1957 (Zeidberg et al. 1967). The area was divided into nine categories on the basis of socioeconomic class and pollution. The latter was determined by a very extensive monitoring system that obtained data on sulfation rate, soiling index, and dustfall. There was also direct monitoring of sulfur dioxide, which confirmed that Nashville had relatively low concentrations of this contaminant. The cutoff point for the high-pollution area, selected to be one standard deviation above the mean for the entire city, was based on a geometric mean sulfur dioxide concentration of 0.013 ppm and a sulfation rate of 0.4 mg/100 cm^2 per day. The authors conclude that the data indicated a direct relation between age-specific respiratory disease mortality and sulfation rate. However, there are a number of problems with their analysis. In an attempt to control for the large inverse effect of socioeconomic class on respiratory disease mortality, they studied the middle socioeconomic class. Although this group showed a direct relation of all respiratory disease mortality with both sulfation rate and soiling, no association with deaths due to bronchitis and emphysema was observed. Furthermore, the middle class was very broadly defined; this raises the possibility that the elimination of a socioeconomic effect was incomplete. Cardiovascular disease mortality was reported to have a small and inconsistent association with the soiling index, particularly in females (Zeidberg et al. 1967).

Wilkelstein and his colleagues (1968, 1967), performed a series of studies in Erie County (Buffalo), New York, in which monitoring data were used to divide the area into pollution categories. Mortality from all causes and mortality from chronic respiratory disease in white males 50-69 years old were found to be associated with concentrations of suspended particles. The standardized mortality ratios for chronic respiratory disease mortality in this group were 76 in air pollution category 1 (total mean suspended particles, less than 80 $\underline{u}g/m^3$), 98 in category 2 (80-100 $\underline{u}g/m^3$), 112 in category 3 (100-135 $\underline{u}g/m^3$), and 137 in the highest air pollution category (over 135 $\underline{u}g/m^3$). These data were apparently of great influence in the establishment of the U.S. air quality standard for total suspended particles, 75 $\underline{u}g/m^3$ (annual mean). However, socioeconomic class was inversely related to deaths from chronic respiratory disease, and the areas with greater pollution tended to be less affluent. The authors suggest that the air pollution effect is at least partially independent of the socioeconomic-class effect. This contention is supported by reasoning based on inspection of standardized data for various subgroups, rather than more detailed multiple regression analysis or assessment of statistical significance. In a review of this work, Winkelstein (1970) in effect argues that these more sophisticated statistical tests are unwarranted, in view of the relative crudeness of the data obtained in epidemiologic air pollution studies. This is a crucial point, particularly because, without such further statistical analysis, it is difficult to obtain precise dose-response information or to assess minimal no-effect concentrations.

Evaluation of the Erie County mortality data in relation to sulfation rates did not reveal any association, nor was there any evidence of a synergistic effect with total suspended particles. However, the gradient for sulfation rate was not as pronounced as that for particles, and that may have affected the analysis.

It should be noted that both the Buffalo and Nashville mortality studies have been criticized on the basis of their lack of data on cigarette-smoking. Obviously, a difference in cigarette consumption in the local areas could account totally for the effect ascribed to air pollution. However, to the extent that lung cancer can be considered to be an excellent biologic indicator of cigarette-smoking, the failure to observe any difference in lung cancer rates related to air pollution in these two studies implies that the data are not confounded by a gradient in cigarette consumption.

New York City has been the site of a number of epidemiologic studies relating air pollution to mortality. Excess mortality during acute air pollution episodes was reported in a series of studies by Greenburg and his colleagues (Glasser et al. 1967, Greenburg et al. 1967, Greenburg et al. 1962). McCarroll and Bradley (1966) demonstrated that less severe air pollution episodes were also associated with increased mortality. However, of more pertinence to present questions concerning the required extent of air pollution control are studies that evaluated pollution concentrations and mortality on a daily basis over a long period.

Hodgson (1970) studied daily mortality in New York City from November 1962 to March 1965. Death rates were related to pollutant concentrations measured at one monitoring station in

Manhattan and to a measure of low temperature. Air pollution variables were highly related to mortality from respiratory and cardiac disease. Particle concentration was a better predictor of the effect than was sulfur dioxide concentration. Mortality from other causes was not associated with air pollution. Additional reported findings include the lack of an apparent threshold for an effect of pollutants on mortality and the fact that the environmental influence on respiratory and cardiac deaths is exerted only by pollution concentrations on the day of death and, to a lesser extent, on the preceding day. Excess mortality due to air pollution was calculated for average monthly increases in particles and sulfur dioxide, but the definition of the pollutant units is not clear. Possible criticisms might be related to inadequate adjustments of mortality data for meteorologic factors, influenza epidemics, and seasonal influences. In particular, a source of bias may have been introduced by the use of three winters and only two summers.

Glasser and Greenburg (1971) studied the effects of air pollution on mortality in New York City in 1960-1964 solely for the months October - March. Adjustments for remaining seasonal factors were made by use of a moving average, and deaths were recorded as deviations from the normal 5-year mean. These were related to smoke shade (COHS) and sulfur dioxide measurements from one New York City monitoring station. Temperature was recorded in terms of deviation from the expected daily normal. When deviations in daily deaths were compared with daily sulfur dioxide concentrations, a sharp increase in mortality was noted for the days when the sulfur dioxide concentrations were greater than 0.3 ppm. A slight gradual non-significant increase was also observed at lower concentrations of sulfur dioxide. The relation of smoke shade to deaths was more variable at lower concentrations, and a sharp increase in excess mortality was present only on the most polluted days. These findings were uncorrected for temperature. However, when regression analyses with meteorologic factors were performed, sulfur dioxide was still found to be positively correlated with the deviation in daily mortality, and this association tended to be confirmed by a further analysis in which days with similar weather conditions were compared. The major increase in deaths was again associated with days when sulfur dioxide concentrations were above 0.3 ppm, although a gradual increase below this concentration was also observed. This study tends to support an association of deaths in New York City with daily sulfur dioxide concentrations higher than permitted (U.S. air quality standard, 0.14 ppm). However, it can be questioned whether the data have been sufficiently adjusted for more subtle meteorologic effects on the death rate, as well as for the effect of influenza epidemics.

Two more recent studies have used highly sophisticated statistical techniques to evaluate New York City mortality rates; although both have shown an association with air pollution, their results have been markedly different. Buechley et al. (1973) assessed daily variations in the death rate in the entire New York metropolitan area for 1962-1966 in relation to air monitoring data from one station in upper Manhattan. They also assessed a number of variables that were shown to affect the death rate, including temperature, day of the week, holidays, and influenza. The effects of temperature were handled in a very complex fashion in which death rates were adjusted for seasonal cycles, extreme heat, and daily and weekly temperature differences. After regression analysis and serial elimination of the other variables, sulfur dioxide was found to be highly correlated with the residual mortality. Mortality was 2 percent more than expected on days when sulfur dioxide concentrations were above 500 $\mu g/m^3$ (0.19 ppm), and 1.5 percent less than expected on days when sulfur dioxide concentrations were less than 30 $\mu g/m^3$ (0.01 ppm). Graphic portrayal shows a tendency toward a stepwise progression in mortality with increasing sulfur dioxide concentration and gives the impression that there is no appreciable threshold for this effect. However, the differences at the lower concentrations of sulfur dioxide are not statistically significant. The authors state that particles do as well as sulfur dioxide in predicting death and that a similar but weaker effect was observed in the Philadelphia area. However, the data are not presented.

Schimmel and Greenburg (1972) reported a much higher estimate of deaths associated with sulfur dioxide and smoke shade. Their study used data from the same monitoring station as that of Buechley et al. (1973), but was limited to deaths in 1963-1968 solely within New York City. They also considered the effect of pollutant concentrations on the days preceding death, as well as on the day of death, and included the cause of death in their analysis. The highest estimate of deaths due to smoke shade and sulfur dioxide were obtained when the regression was performed for pollution alone. This estimate was lower after adjustment for other variables, including mean daily temperature, seasonal and annual trends, and day of the week. Estimates of the percentage of deaths immediately related to air pollution depended on the analysis and ranged from 8 to 16 percent. The authors discuss why the various approaches might overestimate or underestimate the true effects and conclude that 12 percent is a reasonable assumption. Mortality from all causes, except diseases of infants, was related to pollution. When the excess deaths were partitioned between sulfur dioxide and smoke shade, 80 percent of them appeared to be associated with smoke shade.

The reasons for the large contrast between these two papers are difficult to disentangle. The much lower association observed by Buechley et al. may be due to more intensive adjustment for temperature variables and to the consideration of the effect of influenza. The higher estimate in the Schimmel and Greenburg paper might be related to their consideration of the contribution of pollutant concentrations on the days preceding the day of death. However, Lebowitz (1973), using a different statistical approach, found an almost immediate effect of pollution on mortality in New York and other cities. To the extent that it is inappropriate to use one monitoring station to characterize a large area, the approach of Buechley et al. is more vulnerable, in that they used mortality data from the entire metropolitan area, rather than New York City alone. In this respect, the study of Blade and Ferrand (1969) showed a reasonably good correlation between the many New York monitoring stations. However, a report evaluating more recent data from the same network did not find as close a relationship (Goldstein et al. 1974). Another pertinent point is that, if, in fact, 12 percent of New York City residents had their life expectancy decreased by the short-term effect of pollution in the middle of the last decade, then air pollution was a far more serious public health hazard than most people believed.

Despite their individual differences, the four studies of New York City mortality in relation to daily pollution concentrations have relatively similar overall findings. Each has found that, before the institution of recent control measures, air pollution in New York City was associated with an increase in daily mortality. In addition, there is the suggestion that this effect may not have an appreciable threshold. The lack of agreement as to whether particles or sulfur dioxide correlated best with mortality may constitute indirect evidence that the measurements are only indicators of the pollutants, related or unrelated, that are actually responsible for toxicity.

These studies also illustrate that epidemiology is a relatively imprecise science. An observed association becomes far more credible when it is replicated by different scientists using different approaches, particularly in a field as complex as the health effects of air pollution. Although the results are in the same direction, the fact that they vary widely in magnitude indicates further the difficulty in determining precisely what degree of air pollution produces how much illness in how many people.

Both Buechley and Schimmel are extending their observations to more recent years, in which sulfur dioxide concentrations have been greatly reduced. This reduction has been caused by the use of low-sulfur fuel, but it has been associated with only a relatively small decrease in particle concentrations. A tentative and incomplete analysis of the more recent mortality data has been presented by Schimmel et al. (1974); there appears to be no change in the finding that 12 percent of deaths are associated with air pollution. If that is true, it implies that control measures have thus far had little effect. However, this finding must be approached with caution, until both groups have had a chance to complete their analyses and there is an opportunity to review their results in detail.

Morbidity

It is reasonable to assume that, for every person who dies during an air pollution episode, many more will be made sick. Furthermore, relatively minor daily variations in atmospheric pollution would be expected to have a greater effect on morbidity than on the mortality of the population at risk. Accordingly, one would predict that morbidity would be a better indicator of air pollution effects than would mortality. However, it is apparent from the studies to be described that the evidence of an association of relatively low pollutant concentrations with respiratory tract morbidity is at most slightly better than that for mortality. This may be a function of the measurement of response. Death is a readily quantifiable all-or-none phenomenon; the gradation between health and illness can be very subtle, particularly when large populations are evaluated. Epidemiologic studies of respiratory tract morbidity have used a number of measuring devices, including daily diaries, questionnaires, work and school absence rates, hospitalization rates, clinic visits, and the testing of pulmonary function. All these have some difficulties that might obscure an association with air pollution. In addition, as discussed earlier, some variables, such as meteorologic factors, are related to pollutant concen-• trations and have independent effects on respiratory disease. As is the case with air pollution, such a factor as temperature would be expected to have more subtle effects on morbidity than on mortality. It is therefore necessary to assess carefully these possibly confounding variables in the study of the relationship of respiratory disease to air pollution.

Many of the studies that have suggested an association of air pollution with chronic respiratory disease are based on the higher prevalence rates in urban, compared with rural, areas. Although the increased prevalence of chronic respiratory disease in urban adults is conceivably due in part to the population density and greater mobility--leading to a higher incidence of endemic and epidemic respiratory tract infections, compared with that in more isolated rural areas--this would not explain the major differences observed. Furthermore, comparisons of cities with similar population densities but different degrees of air pollution have tended to confirm an association of air pollution with chronic respiratory disease. As discussed earlier, studies that compare prevalence rates of chronic respiratory disease among different areas are of value only if careful attention is paid to standardization of observers, observation methods, and the populations at risk.

Higgins et al. (1958, 1956) conducted a series of studies in rural areas of Scotland and Wales and the industrial town of Leigh, in which the prevalence of respiratory symptoms and the forced expiratory volume (0.75 sec) were measured in a random sample of men and women 55-64 years old. Slight changes consistent with an urban effect were observed, but these were minor, compared with the effect of a past or present history of working in coal mines. Reid and Fairbairn (1958) studied about 500 British postmen who had retired because of chronic bronchitis. They noted that those over 44 years old living in more polluted areas had a higher attack rate of illness and that, after treatment, those living in more polluted areas died of their disease faster. Air monitoring data are not given for these studies, which were conducted in the 1950's. In 1960-1961, Holland and Reid (1965) noted an increase in the frequency of respiratory symptoms and a decrease in pulmonary function in workmen living in London, compared with those in more rural areas of southern England. A comprehensive study by the College of General Practitioners (1961) in Great Britain confirmed the presence of a significant urban-rural difference in bronchitis rates. A comparative study of England and the United States has been reported by Holland et al. (1965).

Fairbairn and Reid (1958) have also evaluated sickness absence rates and retirements because of bronchitis in British civil servants in relation to an indirect measure of air pollution. In 1948-1954, total sickness rates in postmen correlated significantly with both population density and pollution; however, bronchitis disability was significantly related only to pollution. The distribution of bronchitis disability was similar to that of bronchitis mortality for the entire British population. In a 3 percent sample of the total civil service during 1946-1953, the sickness absence rates because of various respiratory tract illnesses were calculated for postmen, men working indoors, and women working indoors. In all three groups, after standardization for population density and domestic crowding, there was a positive correlation of pollution with bronchitis that approached statistical significance. Upper respiratory tract infections and influenza did not correlate with polution, although influenza in men was strongly associated with crowding.

Sickness absence rates due to bronchitis were also evaluated by Cornwall and Raffle (1961) in London transport workers in 1952-1956. Absences of 4 days or longer were related to the area in or around London in which they worked, largely in buses or trolleys. The incidence of bronchitis in the total group correlated closely with dense London fogs. Those working in the more rural areas around London had lower bronchitis absence rates than those within London. When the bronchitis rates in those working in different areas of the countryside around London were compared, the highest rates were observed in the direction in which prevailing winds would be expected to transport London air pollution. Reid reported a similar observation in London postmen in a more restricted periurban area (1956).

The concept of an interaction between cigarette-smoking and air pollution effects is supported by the study of Lambert and Reid (1970), a postal survey of nearly 10,000 britons 35-69 years old. In nonsmokers, the prevalence rates of respiratory symptoms were relatively unaffected by air pollution. However, air pollution had a definite and independent effect on prevalence rates in smokers. The authors constructed symptom prevalence ratios standardized for age and smoking in relation to atmospheric smoke and sulfur dioxide concentrations. Men residing in areas with mean annual smoke concentrations of less than 100 $\mu g/m^3$ had symptom prevalence ratios of 97, compared with 112 for those residing in areas with smoke concentrations of 100-150 $\mu g/m^3$. Higher smoke concentrations were associated with greater increases in prevalence ratios. The data on men for sulfur dioxide were similar but not as dramatic. When annual mean sulfur dioxide concentration was less than 100 $\mu g/m^3$, the symptom prevalence ratio was 87; at 100150 $\mu g/m^3$, it was 96. However, there were only 11 male responders in the areas with low sulfur dioxide. For women, the trends were smaller and less consistent, in keeping with the smaller urban-rural differences for chronic bronchitis mortality and morbidity in women. The data as a whole further support the dominant influence of cigarette-smoking, compared with air pollution, on respiratory disease.

A study that apparently had a significant impact on the original U.S. air quality standard for sulfur dioxide (0.03 ppm, annual arithmetic mean) was performed in Genoa by Petrilli et al. (1966). The frequency of respiratory disease, as measured by an undescribed slight modification of the British Medical Research Council questionnaire, was determined in seven districts of Genoa in an indigent population receiving free medical care and in non-smoking women over 60 years old who were said to be long-time residents of the areas. Sulfur dioxide was measured by a volumetric procedure, which is unfortunately not detailed. Total dustfall, suspended particles, and sulfation rate (by the lead peroxide candle method) were also measured in each district. Chronic bronchitis, tonsillitis, rhinitis, and influenza tended to be increased in the polluted areas, but were not as linearly related to sulfur dioxide as was the bronchitis rate. Mean temperatures were somewhat lower in the more polluted areas, but this did not totally explain the observed effect on bronchitis.

There are some points about the data that are unclear, including the rationale for arithmetically averaging the bronchitis rates of the groups 15-64 years old and 65 and over, whether the data were corrected for the ages of the subjects examined, and whether there were differences in cigarette consumption.

One point of interest in the study of Petrilli et al. (1966) concerns the three least polluted districts, all of which had annual average sulfur dioxide concentrations of 0.025 ppm. One of the districts, Molo, is reported as having a sulfation rate of 0.68 mg of sulfur trioxide per 100 cm^2 per day; the other two districts both had rates of only 0.17 mg/100 cm^2 per day. Molo does not significantly differ in suspended matter, total dustfall, or mean winter temperature, and its influenza rate is intermediate between those of the other two districts low in sulfur dioxide. However, this district, with a fourfold higher sulfation rate, had a substantially higher bronchitis prevalence rate (6.0 percent vs. 2.1 percent and 3.1 percent) and chronic bronchitis rate (9.8 percent vs. 2.1 percent and 2.5 percent) than the other two districts. This increase does not appear to be accounted for by the somewhat greater number of older people examined in Molo.

These findings could be interpreted as indicating that other atmospheric sulfur compounds (in addition to sulfur dioxide) that were measured by the lead peroxide candle method were in part responsible for the observed differences. However, the earlier volumetric sulfur dioxide measurement methods were relatively inaccurate at low concentrations, and it is conceivable that these three communities differed in sulfur dioxide concentration. With the conversion factor obtained in the Nashville study (Zeidberg et al. 1964), the sulfur dioxide concentrations based on the sulfation rate would be close to the 0.025 ppm reported for Molo, but would be lower for the other two districts. This could indicate the possibility of a geographically related difference in respiratory morbidity associated with sulfur dioxide concentrations below the current U.S. standard. In view of the difficulties with the study, of Petrilli et al., such an interpretation must be viewed with caution.

The Nashville study described above analyzed morbidity in relation to air pollution. A questionnaire was given to over 3,000 domiciles; the housewife was usually the respondent for the entire family. Illness rates were inversely correlated with socioeconomic class. When the middle class was evaluated, a direct relation between all illnesses and pollution, as measured by sulfur dioxide and soiling index, was observed for persons 55 years old or older, but not for younger people. Total illness rates of nonworking females, who presumably spent more time in the air pollution category to which they were assigned by area of residence, correlated better with air pollution than did the illness rates of more mobile working women. Problems with the study include the broad definition of the middle class, a lack of relation of air pollution to respiratory illness, and the absence of information concerning cigarette-smoking. Again, it should be emphasized that the Nashville area was relatively unpolluted at the time of study.

Ishikawa et al. (1969) evaluated 600 lungs obtained through autopsy in Winnipeg and St. Louis in 1960-1966. On the basis of emission data, St. Louis was clearly more polluted than Winnipeg; however, the autopsy sets were reasonably well matched in terms of sex, age, and race. Lungs from people with known histories of occupational exposure were excluded. There was a marked increase in emphysema in St. Louis in all smoking categories of males, but not females. The authors report that cigarette-smoking and air pollution acted synergistically in the production of severe grades of emphysema. However, review of the data shows that the relative amounts of total emphysema and mild and moderate grades of emphysema fit an additive model more closely than a synergistic model.

A series of studies performed by Ferris and his colleagues (Ferris and Anderson 1962) have evaluated respiratory disease prevalence in Berlin, New Hampshire, at different times and in comparison with another small city, Chilliwack, British Columbia. At the time of their original study in Berlin in 1961, the sulfur dioxide concentrations were probably about the current standard, in that the mean lead peroxide candle sulfation rate was recorded as 0.731 mg of sulfur trioxide per 100 cm^2 per day. However, the mean 24-hr total suspended particles were 180 $\mu g/m^3$ or, as indicated by the authors, perhaps somewhat higher. This study demonstrated that cigarette-smoking and aging were the major determinants of chronic respiratory disease prevalence and of pulmonary function. The possibility that air pollution might have some effect was suggested by an almost twofold increase in prevalence in each disease category in nonsmoking men resident in the most polluted areas of Berlin. However, this difference was not statistically significant and, as pointed out by the authors, the possible influences of ethnic, social, and economic differences could not be established.

In 1963, the same investigators (Ferris and Anderson 1964) performed a similar study, although with a slightly different questionnaire, in Chilliwack, British Columbia, a community with essentially clean air, and compared the results with those from Berlin. Using expected age-specific prevalence rates for chronic bronchitis and irreversible chronic obstructive lung disease calculated for both populations combined, it was found that nonsmoking females in Chilliwack had about a 6-8 percent decrease in prevalence of chronic respiratory disease. The prevalence rates for nonsmoking men were the same in both communities. From the Berlin data, multiple regression equations were calculated for forced expiratory volume (1 sec) and peak expiratory flow rate. Applying these equations to the Chilliwack data revealed that in all smoking categories the pulmonary function was about 8 percent higher than predicted in Chilliwack. Although these findings are consistent with an effect of air pollution, ethnic differences or subtle variations in life style might also explain the results.

A further study in Berlin was performed in 1967 (Ferris et al. 1973). At that time, there had been a decrease in sulfation rate to 0.469 mg of sulfur trioxide per 100 cm^2 per day and a decrease in particle concentration to 131 $\mu g/m^3$. When the same subjects were reevaluated, there was a lower prevalence of chronic nonspecific respiratory disease and better pulmonary function than expected after taking into account the effect of aging and changes in cigarette-smoking. On the basis of prediction equations derived from the 1967 Berlin population, the 1961 population had about a 5 percent decrease in forced vital capacity and peak expiratory flow rate in men and a 5 percent decrease in forced vital capacity in women, all of which were statistically significant. No difference in forced expiratory volume was observed. This improvement in respiratory function, which is consistent with a beneficial effect of air pollution control, is not as great as the observed beneficial effects of cessation of cigarette-smoking in the same study.

Stebbings (1971), in a series of studies of chronic respiratory disease in Hagerstown, Maryland, has evaluated the effects of past urban residence on the peak expiratory flow rate and forced expiratory volume (1 sec) of over 400 white nonsmoking men 35-64 years old. None of these men had lived in Hagerstown during the preceding 20 years or more. A positive association of childhood urban residence with peak expiratory flow rate was observed, and this effect persisted until about the age of 28. In contrast, those who had lived for longer than their first 28 years in an urban area had a relative decrease in peak expiratory flow rate. No consistent effect on forced expiratory volume was observed. The data were not explainable on the basis of socioeconomic factors, and there were too few men with significant chronic respiratory disease to suggest that selective migration was a factor. The prevalence of respiratory symptoms was positively correlated with long-term urban residence, although the numbers were small. The author's tentative explanation for the increase in peak expiratory flow rate observed in nonsmokers who had spent their childhood in urban areas is that an urban factor, perhaps air pollution, might lead to a compensatory increase in pulmonary function. However, if such an exposure continues long enough, a decrement in function will occur. If, in fact, childhood exposure to air pollution is the factor that leads to an increased peak expiratory flow rate, this would tend to contradict the results of a number of shorter-term studies described above.

Intercommunity differences in the prevalence of respiratory disease have also been assessed as part of the CHESS program (Goldberg et al. 1974, Hayes et al. 1974, House et al. 1974). Studies in the Salt Lake Basin, Rocky Mountain, and New York areas were based on questionnaires filled out generally by the mothers of schoolchildren concerning the health of family members. The format of the studies and the pollution concentrations of the various communities have already been described in some detail above. The prevalence rates of bronchitis (defined as cough or phlegm on most days for at least 3 months each year) were calculated independently for smokers, ex-smokers, and nonsmokers among mothers and fathers who had lived in the area for 2 years or more and who were not occupationally exposed to presumed respiratory pathogens. In the Salt Lake Basin area (House et al. 1974), the high-pollution community had the highest bronchitis prevalence rates in all six categories tested, and the difference was statistically significant. A tendency toward an increased prevalence rate was also noted in the second-most-polluted community, compared with the other two, but this was not statistically significant. The bronchitis prevalence rates were about 100 percent higher in nonsmokers and about 40 percent higher in smokers in the high-pollution community, compared with the other communities. In the Rocky Mountain study (Hayes et al. 1974), an increased prevalence of bronchitis was again noted in the high-pollution communities in each smoking category. However, the data were not as clearcut, and the differences were relatively small, except for the nonsmoking group in which the prevalence rate was greater by twofold to threefold in the high-pollution communities. Socioeconomic factors may have been a problem in this area. In the Rocky Mountain and Salt Lake areas, studies of subjects who were occupationally exposed demonstrated that this factor had a substantially greater effect on bronchitis prevalence than did air pollution. However, the effects of cigarette-smoking were even greater and ranged up to 10 times that attributed to air pollution.

Studies in the New York area (Goldberg et al. 1974) again demonstrated a higher bronchitis prevalence rate in the two intermediate communities than in the low-pollution community, and in general the gradient was steeper than that observed in the Salt Lake Basin and Rocky Mountain areas. Cigarette-smoking was again the major factor in bronchitis prevalence rates.

Although these studies have been criticized on the grounds of anomalous data in each area and among the various areas and on the basis of possible biases involved in self-administered questionnaires, the studies taken together do tend to support an effect of air pollution on chronic bronchitis prevalence rates. In addition, it should be noted that the subjects of these studies do not represent a particularly susceptible population group. However, in terms of extrapolating these data to questions concerning the appropriateness of present air quality standards, the reported increases in chronic bronchitis prevalence rates presumably reflect in part past exposure to air pollution, and all the high-pollution communities in the New York study (Goldberg et al. 1974) had previously been well in excess of the air quality standards. It is hoped that these studies will serve as a baseline for the future evaluation of whether, and to what extent, present emission controls have produced an amelioration of chronic bronchitis rates.

An additional respiratory disease prevalence study, performed by the CHESS (Finklea et al. 1974) group in young military recruits undergoing examination in the Chicago induction center, found only a small and inconsistent relation of respiratory symptoms with area of origin, which, although tending to be in the direction expected for an effect of air pollution, was not statistically significant.

Among the studies that have purported to demonstrate an association of morbidity with sulfur dioxide concentrations below the current standard is the work of Sterling et al. (1969), which studied hospitalization rates and length of hospital stay in Los Angeles from March to October 1961. After day of the week was taken into account, various pollutants, including sulfur dioxide and ozone, were found to correlate with hospital admission for diseases thought to be related to air pollution. Comparison of the one-third of the days with highest sulfur dioxide concentrations with the one-third of the days with lowest sulfur dioxide concentrations showed that the former had 13 percent more admissions for infectious disease, 10 percent more for bronchities, and 10 percent more for acute upper respiratory infections. Little effect was observed on diseases not believed to be related to air pollution. A slight positive correlation of sulfur dioxide concentration with length of hospital admission for relevant diseases was also noted. Detailed air monitoring data are not reported; however, the mean daily sufur dioxide concentrations for the period was about 0.013 ppm, in keeping with the generally low concentrations observed in southern California. It appears unlikely that sulfur dioxide was the sole pollutant responsible for the observed findings, which, if valid, presumably reflect the role of sulfur dioxide as an indicator of specific meteorologic conditions associated with the buildup of pollutants.

Gregory (1970) evaluated records of sickness absence because of bronchities in workmen with chronic bronchitis employed in an urban steel factor for the period 1955-1961 in England. With multiple regression analysis, the monthly bronchitis incidence rates and prevalence rates were found to correlate best with temperature ($r = 0.75$). Positive correlations with smoke and sulfur dioxide were not as great and may have been temperature-dependent. Because the winter of 1957-58 was noted to have particularly high bronchitis absence rates, this period was chosen for further study. When weekly averages were computed for bronchitis incidence, prevalance, pollution, and meteorologic factors, a slight nonsignificant correlation was noted among the various illness and environmental factors in the same week. However, bronchitis incidence was significantly correlated ($r = 0.48$) with the mean temperature ($r = 0.11$) and sulfur dioxide concentration ($r = 0.34$) of the preceding week. In contrast, the bronchitis prevalence rate correlated best with temperature ($r = 0.41$) and only minimally with mean pollution concentrations of the preceding week. When maximal weekly meteorologic and pollutant data were evaluated, the only statistically significant association was between maximal smoke concentrations and the bronchities incidence rate of the following week ($r = 0.50$). The author points out that the delay in the increased incidence of bronchitis may be due to the lack of work absence data for Saturdays and Sundays and the tendency of workmen to finish out the week before going off sick. The greater effect of temperature on prevalence rates may reflect the reluctance of recuperating people to return to work on a colder day. From a graph of the data, it appears that a 100-$\mu g/m^3$ increase in mean weekly smoke concentration would be associated with approximately a 0.6 percent absolute increase in the incidence of bronchities in workers already identified as having chronic bronchitis. With respect to extrapolation of the findings, it should be emphasized that these workmen represented less than 10 percent of the total work force and that these data are for only one winter of six.

Another approach to evaluating the effects of pollution on chronic respiratory disease is to follow a cohort of people. Lawther and colleagues (1970) have presented a series of studies in which bronchitic patients attending chest clinics have been asked to fill out diaries in terms of whether they felt better, the same, or worse on any given day. In their original study, about 180 London patients were followed during the winter of 1955-1956, and a significant correlation of worsening of symptoms with smoke and sulfur dioxide was observed. An increase in illness appeared to be related to an increase in pollution, rather than to an absolute measure of pollution. Meteorologic factors had relatively small and insignificant effects on illness rates. A further study of 248 bronchitic patients in the London area in the winter of 1957-1958 also showed a correlation between pollution episodes and illness. Again, little relation to temperature or humidity was observed; however, it was noted that the patients tended to become more symptomatic as the winter progressed. A similar relation of pollution to bronchitis was observed in the immobile populations, but not in bronchitics living in unpolluted areas.

A larger study was performed in the winter of 1959-1960 in over 1,000 chest-clinic patients, with a slightly modified questionnaire. A worsening of symptoms was observed each time either smoke or sulfur dioxide exceeded 1,000 $\mu g/m^3$. The lowest pollutant concentration associated with any definite decrease in well-being was about 600 $\mu g/m^3$. At the time of the next large study by the same group, in 1964-1965, there had been a significant abatement in London smoke concentrations, with little change in sulfur dioxide. The mean winter concentrations of smoke were 342 $\mu g/m^3$ in 1959-1960 and 129 $\mu g/m^3$ in 1964-1965; those of sulfur dioxide were 296 $\mu g/m^3$ in 1959-1960 and 264 $\mu g/m^3$ in 1964-1965. A mean symptom score was calculated by assigning one, two, or three points to the subject's daily description of whether he felt better, the same, or worse. When these were tabulated for individual winter months and correlated with monthly pollutant concentrations, statistically significant associations were observed with both smoke and sulfur dioxide for the 1959-1960 period. However, in 1964-1965, the positive correlations that were still observed tended not to be statistically significant. When pollution episodes for both periods were evaluated in reference to days with peak mean symptom scores, definite relationships were obseved, although occasionally these were out of phase by a day. The minimal daily pollution concentrations leading to a significant response appeared to be about 500 $\mu g/m^3$ for sulfur dioxide in conjunction with about 250 $\mu g/m^3$ for particles.

The authors identified 87 people who in 1964-1965 were the most sensitive to changes in the degree of pollution, and 50 of these presumably susceptible people were restudied in 1967-1968, by which time mean winter smoke concentration had decreased to 68 $\mu g/m^3$ and mean winter sulfur dioxide concentration was somewhat lower, at 204 $\mu g/m^3$. The correlations of mean symptom scores for this group of subjects in 1964-1965 were 0.39 with smoke and with sulfur dioxide; in 1967-1968, the correlation with smoke was 0.31, and that with sulfur dioxide was 0.28. Although the correlations in 1967-1968 were somewhat lower, they were still statistically significant. Also statistically significant was a negative correlation with temperature in both periods, and, although the correlations were lower than those with the pollutants, the data do not appear to have been adjusted for this meteorologic variable. Of great interest is the finding that sulfuric acid concentration in 1964-1965 had a high correlation of 0.51 with mean symptom score; but in 1967-1968, a correlation of only 0.26.

The authors note that their findings may perhaps be best associated with peak pollutant concentrations during the day, rather than average 24-hr concentrations, although data to support this assertion are not presented. Also noted is that daily pollution concentrations appear to have more of an effect than daily temperature, particularly early in the study, when relatively severe pollution episodes were frequent. Of further interest is the repeated observation that, as the winter progressed, the patients appeared to be less responsive to similar pollution episodes. The possibility of individual adaptation that this suggests has been discussed in some detail above.

Lawther et al. (1974), have also recently reported a series of studies in which pulmonary function in a small group of normal subjects and two bronchitics in the same hospital laboratory in London was measured frequently. The results were highly variable, and, although a relationship with daily and hourly sulfur dioxide concentrations was observed in some subjects, the major determinant appeared to be respiratory infection.

An additional study that suggests that pollution control measures in the London area have led to a smaller association of respiratory dysfunction with air pollution was reproted by Emerson (1973). Eighteen patients with various degrees of chronic bronchitis and asthma living relatively close to an air monitoring station were repetitively studied with a battery of pulmonary-function tests for up to 82 weeks, beginning in November 1969. Pulmonary function was correlated with a 5-day average of smoke and sulfur dioxide concentrations, including the day of testing and the 4 preceding days. A statistically significant correlation, presumably negative, between sulfur dioxide concentration and forced expiratory volume (1 sec) was observed in only one patient, and between sulfur dioxide concentration and maximal expiratory flow rate in only two patients. In no case was a significant correlation with smoke concentration reported; however, more frequent significant correlations with meterologic factors were observed. Mean concentrations in 1969-1970 were 193 ug/m³ for sulfur dioxide and 44 ug/m³ for smoke; those in 1970-1971 were 187 ug/m³ for sulfur dioxide and 41 ug/m3 for smoke. The rationale for the use of a 5-day average for pollutants is not given, nor is it clear to what extent disability may have kept these patients indoors. Fletcher et al. (1968) have also reported a general decline in sputum production of bronchitics in association with a decrease in London pollution. This was not as great as that observed in patients who had discontinued smoking. More recently, Howard (1974) noted that patients with obstructive airway disease studied in Sheffield between 1966-1972 had less respiratory dysfunction than a group of the same age and smoking habits studied between 1960-65 when pollution levels had been higher.

The applicability to the United States of the recent relative inability of British investigators to detect an effect of air pollution, compared with the past, appears worthy of further comment. British pollution abatement measures have let to a considerable reduction in concentrations of particles, with relatively less change in sulfur dioxide. In contrast, the major effect of air pollution control measures in the United States has been to decrease sulfur dioxide, rather than particles. This raises the possibility that respirable particles are more of a health hazard than is sulfur dioxide. Although this may be true, it is a somewhat facile explanation and definitely cannot be used as an argument against the control of sulfur dioxide. A number of differences between the British experience and ours should be kept in mind: The content of particles in London differed greatly from that usually observed in American cities and may have played a role in the reported health effects. This may be particularly important with respect to acid sulfates; if the data are available, a comparison would be of interest. Furthermore, it is conceivable that the British have adapted to their high particle concentrations and that further effects will not be observed until concentrations have stabilized again. Another point is that there appears to have been a much smaller decrease in particulate sulfate in U.S. urban areas than in sulfur dioxide, possibly because of vagaries of the oxidation process within urban atmospheres. However, the reduction in sulfur dioxide emission in the cities might still lead to a decrease in sulfate in the countryside. It is not clear whether the kinetics in the air of British urban areas is similar.

Two studies in Chicago have evaluated the symptoms of patients with chronic respiratory disease with refernce to daily environmental factors. Burrows et al. (1968) used diaries to study 115 Chicago area clinic patients for the 2-year period 1963-1964. Data on symptoms, which were available for about 50 percent of the days from all patients, were compared with various pollutant measurements (excluding particles) obtained in the Chicago Loop and with meteorologic information. When a symptom severity score was calculated from data on cough, sputum production, and shortness of breath, the correlation with daily sulfur dioxide concentrations was a relatively high 0.55. However, a better correlation with mean daily temperature was observed (r = 0.07). Furthermore, when the data were calculated solely for the months of December, January, and February, no relation between symptom severity and sulfur dioxide was obsered. In fact, multiple regression analysis revealed that, after temperature was held constant, there was a statistically significant negative partial correlation (r = 0.20) between symptoms and sulfur dioxide during the winter months. Introduction of lag periods of up to 5 days did not result in any significant correlations with sulfur dioxide. Selection of 7 days when there was considerable variability in suspended particle concentration in different areas of Chicago and analysis of symptom severity by area of residence resulted in a slightly negative correlation with particle concentration. Mean pollutant data are not given, but apparently the peak daily sulfur dioxide concentration was around 0.8 ppm, and Chicago undoubtedly exceeded current U.S. air quality standards during this period. The data indicate that, for this group of patients, low temperature is the dominant environmental factor in the severity of chronic respiratory disease during a Chicago winter. In view of the high positive correlation with sulfur dioxide for the entire year, one wonders whether an air pollution effect would have been observed in a detailed analysis during a period of milder weather. This possibility will be discussed in more detail later.

Another study in Chicago used a similar approach and found a correlation of pollution concentrations on the same or previous day with the morbidity of patients with chronic respiratory disease who were 55 years old or older. Carnow (Carnow et al. 1969) evaluated over 500 patients throughout Chicago who were part of a bronchopulmonary disease registry with a diary in which was recorded dialy the presence or absence of chest illness defined in terms of the patinet's usual status. The data were subdivided by an age classification and by the degree of bronchitis. The days of acute respiratory illness were related in each of these groups to sulfur dioxide concentrations by area of residence determined in a network of monitoring stations. In patients with greater degrees of bronchitis, there appeared to be a gradient of illness in relation to daily sulfur dioxide concentration that was present for all age groups and was also noted for 1- or 2-day lags of the pollution concentration. The effect on acute illness was most marked in those 55 years old or older a day after exposure. For this group the chest illness rates were 26.5 percent a day after a sulfur dioxide concentration of more than 0.3 ppm and 13.6 percent a day after a concentration of 0.0-0.04 ppm. In those with lesser degrees of bronchitis, no effect was observed in the older group, but there was a slight tendency toward a response in the younger group. A further analysis was performed for October to December 1967, with each patient as his own control; a comparison was made of the sulfur dioxide concentrations on the days when the patient reported illness or no illness, or on the previous days. Again, the best correlations were with sulfur dioxide concentrations on the days preceding illness for those with more severe grades of bronchitis who were 55 years old or older.

It is of interest to contrast these two Chicago area studies, which have reported opposite results. The analysis of Carnow et al. (1969) appears to be more likely to detect an effect of air pollution on the morbidity of patients with respiratory disease, in that the population was subdivided, allowing detection of a particularly susceptible group, and the sulfur dioxide concentrations were evaluated in relation to an extensive monitoring network, rather than an individual station. However, these advantages seem to be more than overshadowed by the lack of assessment of meteorologic factors, which were shown by Burrows et al. (1968) as well as other investigators, to have a significant effect on respiratory tract morbidity.

Another point can be made that is speculative, but nevertheless intriguing. The Carnow et al. analysis was performed for October-December, whereas that of Borrows et al. evaluated the colder months of December-February. It is possible that increases in sulfur dioxide in the fall are associated with more respiratory tract effects than similar increases occuring in the winter or early spring. This is suggested by the sutdies of Lawther at al. (1970) discussed earlier and by the data of Buechley at al. (1972), in which sulfur dioxide correlated well with New York City area mortality in the fall, but not in the winter. Although these are likely to be random observations of no real significance, three possible explanations can be suggested: (1) An overwhelming effect of low temperature makes it technically difficult to detect lesser effects due to air pollution. (2) People with chronic bronchitis may spend more of their time indoors during the colder winter months and therefore be somewhat protected from ambient pollution. (3) Adaption to the effects of sulfur dioxide or other pollutants occurs, whereby initial peaks early in the home heating season result in more of an acute effect than do similar or higher peaks later in the winter. The possibility of adapation to short-term effects of sulfur dioxide has been suggested in animal and human controlled exposure experiments, and relatively long-term adaption appears to occur in occupationally exposed people. Whether such an effect occurs over a period of months in usual ambient conditions is, of course, unknown. Furthermore, adaption to the acute effects of a toxic agent does not necessarily imply the prevention of chronic long-term effects.

Spicer and Kerr (1966) evaluated selected groups of patients with moderate degrees of obstructive airway disease who lived near an air monitoring station in Baltimore. In their first study, airway resistance was measured in seven patients 5 days/week for 14 weeks; in their second study, 14 patients were evaluated for 47 consecutive days. In each study, it was observed that the airway resistance of each patient tended to rise and fall together, implying the existence of an environmental factor, although it was not possible to separate an individual pollutant or meteorologic variable. During the course of the work, 24-hr particle concentrations were as high as 500 $\mu g/m^3$, and 2-hr sulfur dioxide concentrations were as high as about 0.13 ppm. However, detailed air monitoring data are not given. A further serial study by these authors (1970) on healthy students revealed little correlation of respiratory function with air pollution.

Winkelstein and Kantor (1969) evaluated respiratory symptoms reported in a questionnaire of white women in the Buffalo area in 1961-1963. Unfortunately, the questionnaire had not been designed for the study of respiratory disease, and there was poor reproducibility of results when a small subsample was retested. With the same air pollution districts as in the Erie County study described before, it was found that cough and phlegm in nonsmokers over 45 years old correlated with particle concentration, although the results were not statistically significant. No gradient for sulfur dioxide was observed. Some influence of residential mobility on the results for smokers was discerned.

The CHESS program evaluated panels of elderly people with and without cardiopulmonary symptoms in the New York area (Goldberg et al. 1974). In general, this was the least successful of the CHESS efforts. Among the problems were a very low participation rate and a high dropout rate during the course of the study, which introduced frequent interactions with the staff and a distinct possibility of bias. Furthermore, members of the panel in the low-pollution community were healthier than those in the higher-pollution communities, although this problem was avoided somewhat by using each subject as his own control. The analyzed data show an association of morbidity with pollutants. Suspended sulfates appear to have the strongest effect. However, the additional sophisticated analyses reported in the study appear unwarranted, in view of the difficulties described.

Although most of the studies cited have noted an association of air pollution with respiratory disease, this is not always the case. In a series of three related studies, Comstock et al. (1973) were unable to detect an effect of air pollution on pulmonary function or respiratory symptoms of American and Japanese male telephone workers. The first and second studies were performed in 1962-1963 and 1967-1969 in four locations in the United States: Washington, Baltimore, Manhattan (New York City), and a rural part of Westchester County, New York. The third study was performed in Japanese working in Tokyo, but living in areas with differing pollution concentrations. A standardized questionnaire and 1-sec forced expiratory volume were the source of the data, which were analyzed in relation to cigarette-smoking, place of birth, areas of past and present residence, and area of workplace. There was a clearcut effect of age and cigarette-smoking, but no effect of area consistent with a response to air pollution was observed. The American data can be criticized, in that only Manhattan would be expected to have appreciable air pollution concentrations, and the cooperation rates were lowest in this gorup, which was also tested at periods somewhat different from those of the other areas. However, the data largely support an absence of an appreciable response to air pollution in healthy people working in a rather strenuous occupation. The findings are, of course, not pertinent to more susceptible population groups.

Another study that failed to show an effect of air pollution is that of Hrubec et al. (1973) who analyzed over 4,000 pairs of male twins 41-51 years old by means of a mailed questionnaire. The air pollution data for the area of residence are not fully described, but were apparently calculated from U.S. air monitoring and emission data, as well as from meteorologic factors. These were used to obtain estimates of suflur dioxide, particles, and carbon monoxide, which were then averaged together. Comparison of twin pairs revealed that cigarette-smoking and alcohol consumption were directly related to bronchitis and prolonged cough. A slight relation with urban residence was also present, which had previously been reported in a study of twins in Sweden, (Cederlof 1966), but there was no association with the estimates of air pollution. The major question concerning the study of Hrubec et al. is the validity of the air monitoring data, particularly in view of the use of carbon monoxide in the estimate.

Briersteker et al. (1969) were unable to observe any relation between time of residence in the polluted city of Rotterdam and respiratory symptoms or pulmonary function in a study of municipal employees.

In summary, an effect of suflur oxides and particles on the prevalence rate, morbidity, and mortality of chronic bronchitis and emphysema is apparent and will almost certainly occur at pollutant concentrations only somewhat above present U.S. air quality standards. Determination of the lower limits of this effect will require continued epidemiologic study, with measurements of sulfuric acid and individual particulate sulfates, as well as other repirable particles. This will be greatly aided by animal studies aimed at determining which of the sulfur oxides, alone or in combination, is responsbile for health effects. It appears that a particularly crucial subject for epidemiologic study is the extent of mortality associated with daily variations in sulfur oxides and particles, including evaluation of the suggestion that there is no threshold for this effect. If this is, in fact, true, or if a relatively high fraction of daily mortality is associated with present pollutant concentrations, then it is likely that the morbidity data presented here substantially underestimate the effects of suflur oxides and particles.

Respiratory Tract Infections

Infections of the respiratory tract are among the most common diseases in man, accounting for a significant part of total morbidity and mortality in the general population. Respiratory tract infections account for a very high percentage of time lost from work or school, and their economic impact includes substantial costs for drugs, doctors' fees, and hospitalization.

These infections can be conveniently divided by anatomic location. Those affecting the upper respiratory tract -- which is usually considered to include the nose, mouth, pharynx, and larynx -- tend to have less severe health consequences to normal adults, particularly in this antibiotic era; however, they have an enormous economic impact on society as a whole. Those affecting the lower respiratory tract, including bronchitis and pneumonia, generally have more immediate and longer-term health consequences. Respiratory tract infections may also be classified by etiologic agent. Viral infections are very common and include mild upper respiratory tract infections usually referred to as colds. However, influenza can have severe consequences, particularly in the aged and those with preexisting cadiopulmonary disease; and viral pneumonias do occur. Acute bacterial infections of the lower respiratory tract tend to have more serious implications, particularly because they may leave residual damage, which acts as a focus for recurrent disease. Other microbiologic agents producing diseases such as fungi, Mycobacterium tuberculosis, and Mycoplasma -- will not be discussed here, because they do not appear to be related to air pollution. It should be stressed that the division between upper and lower respiratory tract infections and between viral and bacterial agents is somewhat arbitrary, particularly inasmuch as lower respiratory bacterial infection is often a sequelae of acute viral upper respiratory infections.

Many of the studies that have evaluated the daily well-being of people with chronic respiratory disease in relation to pollutant concentrations can be considered to have indirectly assessed respiratory tract infections, in that these play a major role in the progression of chronic bronchitis. What would be a minor respiratory tract infection to an otherwise healthy person may have serious consequences in someone with underlying lung disease. Such people may also be at more risk of development of pulmonary disability.

Most of the evidence associating respiratory tract infections with products of stationary fossil-fuel combustion has been obtained in studies of children, which were described earlier. To summarize, there is reasonably good evidence that concentrations of sulfur dioxide and particles above U.S. air quality standards are associated with an increased incidence of lower respiratory tract infection, particularly croup and bronchitis, in children. The findings have also suggested a possible role for atmospheric suspended sulfates. The association with pneumonia is inconsistent and probably not real. In addition, there is little evidence that upper respiratory infections in childhood are related to sulfur dioxide and particle concentrations. A variable association of pullution with ear infections, which are often sequelae of upper respiratory tract infections in childhood, has been reproted.

The following discussion will evaluate mainly respiratory tract infections in adults not identified as having chronic lung disease and will also describe animal studies related to this topic.

Fairchild and his colleagues (1972) used a mouse influenza model to study the effects of sulfur dioxide. Combined exposure to relatively high concentrations of sulfur dioxide and to influenza virus was found to produce a higher incidence of pneumonia at higher concentrations but less pneumonia at lower concentrations than was observed in control animals.

In addition to producing an increase in the incidence of infection, it is conceivable that pollutants increase the morbidity of a preexisting infection, perhaps by interfering with the mechanisms by which the respiratory tract acts to contain and combat infectious foci. This was studied in mice exposed to sulfur dioxide at 20 ppm for a week after inhaling an infective dose of influenza virus. These animals developed more evidence of pneumonia than mice that received the same viral dose but were not exposed to sulfur dioxide (Fairchild et al. 1972). The results were similar when sulfur dioxide exposure (25 ppm) preceded the viral infection. However, if the mice were exposed to sulfur dioxide at 2-5 ppm for a week after the influenza dose, a decrease in the extent of pneumonia was observed. Sulfur dioxide exposure did not appear to affect the growth kinetics of influenza virus. Not was it clear whether the observed pneumonia represented viral infection of the lung or a secondary bacterial process. These results may perhaps be explained by studies that indicate that lower doses or shorter exposures to sulfur dioxide tend to increase mucociliary clearance, whereas higher doses or longer exposures inhibit mucociliary flow (Ferin and Leach 1973, Holma 1967). The authors also mention unpublished findings that sulfur dioxide at 6-20 ppm results ~~in~~ inhibition of influenza viral growth in the nasal cavities of mice.

In another study of mice, Giddens and Fairchild noted that inhalation of sulfur dioxide at 10 ppm for 24-72 hr had much greater pathologic effects on the nasal mucosa of mice with mild upper respiratory tract disease than on those of a disease-free group (Giddens 1972). This could also be interpreted as indicating potentiation of a respiratory tract infection by sulfur dioxide, but the converse is equally possible. The basic mechanisms postulated for either assumption are similar, in that any insult to the respiratory tract is liable to interfere with the normal defense against other insults.

Studies that evaluated the effects of air pollutants on the pulmonary clearance of bacteria were reported by Rylander (1969). After inhalation of either live or dead Escherichia coli, groups of guinea pigs were exposed to sulfur dioxide at 5-10 ppm, to manganese dioxide aerosol at 5,900 ug/m³ (90 percent of particles smaller than 0.5 um), or to both for 3 hr (Rylander et al. 1971). Negligible effects were observed with either manganese dioxide or sulfur dioxide alone. However, combined exposure led to a statistically significant decrease in the removal of either the live or dead bacteria from the lung. It should be noted that removal of dead bacteria presumably reflects mucociliary clearance, whereas removal of live bacteria also measures lung bacteriacidal capability. Also of interest in this study is that manganese dioxide is relatively insoluble, and earlier studies by Amdur and Underhill had failed to show a synergistic effect of sulfur dioxide and manganese dioxide on guinea pig bronchoconstriction (Amdur and Underhill 1968). Rylander et al. suggest that the combination of these two agents produces a decrease in pH that accounts for the biologic effects. However, previous studies by Tylander had similarly demonstrated a synergistic effect of suflur dioxide and coal dust in this same system, and it is not clear whether this could be due to pH (Rylander 1970, 1969). It is of interest that viral infections have been shown to retard the clearance of dust particles; this suggests that infectious disease might potentiate an effect of atmospheric particles on lung clearance, leading in a sense to a vicious circle (Creasis et al. 1973).

Battigelli et al. (1969) were unable to demonstrate any effect of sulfur dioxide in combination with graphite dust on the bacterial microflora of rats. Animals were exposed to graphite dust at 1,000 μg/m³ or to graphite dust and sulfur dioxide at 1 ppm for 12 hr/day for 4 months and compared with nonexposed controls. Bacterial samples form the nasal turbinates, the main stem bronchi, and the lungs at weekly intervals revealed no significant differences among the three groups.

A number of studies have also suggested that prolonged exposure to sulfur dioxide results in an alteration of immunologic response (Ardelean et al. 1966, Zarkower 1972, Zavrotskii 1959). Antibody formation and cellular immunity play a role in preventing lung infection, so any decrement in these responses would presumably potentiate a deleterious response to microorganisms. A study by Goldring (1967) in which hamsters were repetitively exposed to sulfur dioxide at 650 ppm showed no additive effect of influenza virus. There was, in fact, some evidence that this concentration of sulfur dioxide may have killed the virus. A recent sudy by Zarkower has evaluated a number of characteristics of immune response in mice exposed to sulfur dixide at 2.0 ppm, to carbon particles at 558 ug/m³ (particle diamter, 1.8-2.2 um), or to both for up to 200 days (Zarkower 1972). Variable responses, depending on the duration of exposure, were observed; but, by the end of the experimental period, the animals exposed to sulfur dioxide or carbon alone had a reduction in serum hemagglutination titers to killed *Escherichia coli* administered as an aerosol. Combined exposure to sulfur dioxide and carbon produced less than an additive response. In contrast, there was an enhancement of the number of antibody-forming cells in the mediastinal lymph nodes with either carbon or sulfur dioxide after 135 days of exposure that was no longer observed after 192 days of exposure. Mice exposed to both agents had a synergistic increase in mediastinal lymph node antibody production after 135 days that was still present after 192 days. The major implication of these findings is that long exposure to relatively low concentrations of sulfur dioxide produces a lateration in the immune system of the mouse. As pointed out by the author, the mechanism of this effect is puzzling. Further studies are indicated before these results can be extrapolated to humans breathing ambient polluted air, particularly because infectious disease has not been noted as a problem in humans occupationally exposed to the sulfur dioxide concentrations studied or to higher concentrations.

An interesting *in vitro* study that has apparently not been folowed up is that of Lawther et al. (1969). They noted that aqueous extracts of particles collected from London air had a stimulatory effect on the growth of *Haemophilus influenzae*, a bacteria that has been implicated in the progression of chronic bronchitis and often recovered from the sputum of chronic bronchitis patients.

Many of the epidemiologic studies of the association of respiratory tract illness with air pollution have evaluated illness absences among large populations of workers. Angel et al. studied the weekly attack rate and prevalence of respiratory illness in men working in a post office savings bank and an engineering works in London who lived within 2 miles of an air monitoring station (Angel et al. 1965). The group consisted of 85 men, representing about one-fourth of the work population, who were apparently selected to some extent on the basis of frequency of chest illness. They were seen by a physician at least once every 3 weeks from October 1962 to May 1963. The attack rate of minor respiratory illness was associated equally with weekly means smoke and sulfur dioxide concentrations. After standardization for pollutant effects, no correlation with temperature was observed. The prevalence rate was found to be more strongly associated with smoke than with either sulfur dioxide or temperature. These results are somewhat different from those reported by Gregory for bronchitics (discussed above). This may be due in part to differences in the extent of underlying illness, although it is not clear from the data of Angel et al. (1965) whether their population group contained subjects with chronic bronchitis.

Dohan and Taylor assessed the incidence of illness absences of women working in manufacturing plants of a large U.S. corporation during 1955, 1957, and 1958 (Dohan 1961, Dohan and Taylor 1960). Mean annual monitoring data for the eight eastern areas that had plants with more than 900 women workers were correlated with respiratory illness absences lasting more than 7 consecutive days. There was a fivefold difference in the incidence of respiratory illness between the factories with the most and the fewest illnesses. A remarkably high correlation ($r = .964$; $p \leq 0.001$) was observed with the mean annual suspended particulate sulfate concentration, and the rank order of the eight plants was consistent in each of the 3 years studied. Furthermore, in the year of a major influenza epidemic, the areas with higher particulate sulfate concentrations had greater increases in respiratory illness absence rates. The four areas with the highest illness absence rates had mean annual suspended sulfate concentrations of 13.2-19.8 ug/m³. Of the four lowest areas, sulfate data were available for only one (7.4 ug/m³), but sulfate concentrations were estimated to be very low in another. The communities with the two highest respiratory illness absence rates had the two highest concentrations of total suspended particles (173 and 188 ug/m³); however, the gradient for this pollutant was otherwise unrelated to respiratory illness. Some tendency toward as association of airbone copper, nickel, and vanadium was also observed. When the data were evaluated for types of respiratory disease, an association with suspended sulfates was observed for influenza and bronchitis, but not for pneumonia. These findings did not appear to be related to interplant differences in age, weather, crowding, occupational exposures, or the number of children at home. As pointed out by Dohan, the observed effect may have been due to a prolongation of the duration of respiratory tract illness to 8 days, rather than to an increase in the incidence of disease. One problem with the study is that mean annual sulfate concentrations were based on only 21-25 determinations in four of the five areas for which these data are available. Nevertheless, the observed correlation is remarkably high for a study of this sort and strongly suggests an effect of suspended sulfates.

Ipsen et al. (1969) evaluated the relationship between industrial absences, air pollution, and meteorologic factors in plants in Philadelphia and Camden, New Jersey, in 1960-1963. Total suspended particles, particulate sulfate, and soiling index were measured. Morbidity, defined as incidence and prevalence of respiratory disease, was associated with weather factors and with pollutants. Of the pollutants measured, the immediate correlation with suspended sulfates was the strongest. However, this almost disappeared when partial correlation coefficients were calculated in which each variable was adjusted for the others. This analysis demonstrated that weather had the major effect on respiratory morbidity, although there was a significant positive correaltion of an additive index of all three pollutants with the prevalance of respiratory disease on the same day or a week later. The actual monitoring data are not presented and presumably are rather limited, with respect to the number of sites available. The place of residence of the workers is also not taken into consideration.

Verma et al. studied respiratory and non-respiratory illness absence data for 1965-1967 on white-collar workers, 16-64 years old and employed at a New York City insurance company (Verma et al. 1969). Respiratory disease absence rates per 1,000 employees per day were calculated, and the data were expressed as deviations from the average respiratory disease absence rate in relation to daily air pollution concentrations. On days when the temperature was in the 16-50 range, there were 4.50 more respiratory disease absences per 1,000 employees than average when the 24-hr sulfur dioxide concentration was 0.25 ppm or greater and 0.65 more per 1,000 employees when the concentration was less than 0.25 ppm. Again, with sulfur dioxide at 0.25 ppm as the dividing line, smaller differences in the respiratory disease absence rates were observed at higher temperatures. When the data were calculated for smoke shade, with 1.6 COHS as the dividing line, the gradient was not as steep in the temperture ranges of 16-50 F and 77-103 F, but was greater than that for sulfur dioxide in the temperature range of 51-76 F. No consistent effect of carbon monoxide was observed, and nonrespiratory illnesses did not correalte well with pollution.

Further analysis revealed a close association of respiratory illness with pollution and meteorologic factors for lag periods as long as 7 days. The data were strongly influenced by seasonal cycles, and removal of time trends greatly decreased the positive relationship between respiratory disease illness absences and pollution. A linear air pollution model was found to account for about 20 percent of the observed variability in illness absence rates. The authors conclude that, although no causal relation can be inferred, there is an association between air pollution, meteorologic variables, and respiratory illness absences from one period to the next. It should be noted that New York City at present rarely exceeds a 24-hr sulfur dioxide concentration of 0.25 ppm, and it is not clear from this paper whether an effect might be observed at lower sulfur dioxide concentrations. A similar study in the New York area with more extensive air monitoring data would be valuable.

A number of studies have used cough to assess the effects of air pollution. Coughing is, of course, a nonspecific respiratory tract response. However, in the absence of chronic respiratory disease or an obvious atmospheric irritant, cough is usually ascribed to acute respiratory infections.

Loudon et al. (1969) assessed prescription rates for exempt narcotic anticough medicines in relation to environmental factors in Dallas. A negative correlation with temperature, but little (if any) effect of air pollution was observed. Monitoring data are not given.

McCarroll et al. (1967), as part of a series of extensive studies of a New York City population living close to a monitoring-station, assessed cough and eye irritation, but a distinct difference was noted when lag periods were studied. The pollutant effect on eye irritation represented by suflur dioxide concentration was almost immediate, whereas the maximal effect on cough occurred 1 or 2 days later. Particles were not consistently related to eye irritation, but were correlated to some extent with cough. There is some periodicity of the data that is not explained. The air monitoring data are not described, and meteorologic conditions were apparently not examined.

The same group of investigators also noted some correlation of respiatory tract illnesses with air pollution episodes. Of interest was an analysis of their data that attempted to define a subpopulation of persons who were particularly sensitve to environmental conditions and could therefore be used as monitors (Lebowitz et al. 1972). A specific subset of people were identified who appeared to develop a higher incidence of upper respiratory tract infection when subjected to meteorologic extremes or high pollution concentrations. An additional analysis of this group focused on the complex interactions of weather and pollution with regard to symtoms (Cassell et al. 1969). An increased incidence of upper respiratory infection was also observed in four New York City old-age homes during a 1962 pollution episode in which there was no apparent effect on mortality or hospital admit visits for upper respiratory infection or asthma (Greenburg et al. 1963).

Prospective studies of respiratory tract illness in the general community have also been performed as part of the CHESS studies in New York and Chicago (Finklea et al. 1974, Love et al. 1974). These have already been described. In both areas, an increased incidence of lower respiratory tract infection was obseved in mothers and fathers living in the more polluted communities. In Chicago, there was a slight tendency toward an association of upper respiratory tract infection with pollution, but the opposite was observed in New York. In all communities, mothers had higher illness rates than fathers; this probably represents a bias due to the mothers' filling out the questionnaire. The data were interpreted by the authors as indicating that air pollution in the two New York City communities might be responsible for 5 extra days of restricted activity and one extra physician visit a year for an average family of four. The findings must be interpreted cautiously, particularly in view of some inconsistencies in the data, the possible effects of extraneous variables (such as indoor pollution), and the rather low questionnaire response rates. However, by and large, these studies support the conclusions discussed below.

The data cited indicate relatively clearly an effect of products of stationary fossil-fuel combustion on the incidence of lower respiratory tract infections (not including pneumonia). The evidence does not support an association of these pollutants with upper respiratory tract infection, except perhaps in a particularly susceptible population. A no-effect concentration has not been established and may be below the pollution concentrations associated with the present air quality standards for sulfur dioxide and particles. However, the no-effect concentration appears more likely to be at or somewhat above the present standards. A role of suspended particulates in the production of lower respiratory tract illness is strongly suggested, but not proved. This association with suspended sulfates clearly deserves further evaluation, which may result in the establishment of an air quality standard more directly related to protecting the public against pollutant-induced lower respiratory tract infection.

Lung Cancer

There is no substantial evidence that directly implicates sulfur oxides in the causation of lung cancer. However, some observations indirectly suggest a relationship and clearly indicate that more study of this subject is required.

In vitro studies have indicated that sulfur dioxide can react with the deoxyribonucleic acid (DNA) cytosine, a component of chromosomes, which carry genetic information. Incubation of cytosine with bisulfite, a hydrated form of sulfur dioxide, results in the formation of the unstable intermediate dihydrocytosine-6-sulfonate, which deaminates to form uracil (Hayatsu et al. 1970, 1970, Shapiro et al. 1970). The conversion of cytosine to uracil after incubation with bisulfite has been observed in viral and bacterial DNA, yeast ribonucleic acid (RNA), and synthetic nucleic acid polymers. Theoretically, a modification of nucleic acid constitutents in a molecule containing genetic information is potentially mutagenic. An increased frequency of reversion of transition mutants of *Escherichia coli* consistent with mutagenesis has been observed by Mukai et al. (1970), and two scientific groups have observed bisulfite mutagenesis in bacteriophage (Summers and Drake 1971, Hayatsu and Miura 1970). However, it should be noted that optimal conversion of cytosine to uracil occurs at a pH of 5.5, with little or no reaction at the usual body pH of 7.4. All three studies of bisulfite microorganism mutagenesis detected results at low pH; in the study of Mukai at al. (1970), no effect was observed in the physiologic pH range. An *in vitro* study in which phytohemagglutinin-stimulated human lymphocyte cultures were bubbled with 100 ml of sulfur dioxide at 5.7 ppm demonstrated chromosomal abnormalities, as well as a decrease in DNA synthesis and mitosis (Schneider and Clakins 1970). Chromosomal abnormalities have been observed in pollen exposed to less than 0.1 ppm sulfur dioxide (ma et al. 1973). A lethal effect of sulfur dioxide (25 ppm) on tissue-culture cell lines has also been reported. Of interest in this study is that sulfite salts were more toxic to cell cultures than were equivalent concentrations of sulfate (Thompson and Pace 1962). An alternative mechanism of sulfite-induced damage to DNA might occur in which free radicals, developed during sulfite oxidation at physiologic pH, alter nucleic acid constituents in a manner presumably similar to that of radiation. Again, there is no evidence that any of these reactions occur *in vivo* after inhalation of sulfur oxides.

The epidemiologic evidence suggesting an association of sulfur oxides with cancer is at best indirect. For the most part, it is based on the generally observed higher lung cancer incidence in urban than in rural areas, which appears to be unrelated to cigarette-smoking. This urban effect is still open to question, as is its relation to sulfur oxides. Stocks (1960) and Burn and Pemberton (1963) have found positive correlations of smoke pollution and lung cancer. However, Ashley (1967) reported slight negative correlations of both smoke and sulfur dioxide with lung cancer, despite high positive correlations of these two pollutants with bronchitis mortality in residential areas of Great Britain. If there is an urban effect and it is related to air pollution, aromatic hydrocarbons are more likely to be causative pollutants (Carnow and Meier 1973, Stocks 1960). Higgins (1974) has noted a decrease in the trend of lung cancer in Britain in recent years that cannot be totally accounted for by changes in cigarette-smoking and that appears to follow the decrease in particulate air pollution. Studies in Nashville (Hagstrom and Sprague 1967) and in Erie County, New York (Winkelstein and Kantor 1969), which evaluated the relation of cancer rates within the same urban area to different degrees of air pollution, have found some association of sulfur dioxide or total suspended particles with some nonpulmonary tumors, but none with cancer of the lung, which would be the presumed site of action of inhaled sulfur oxide irritants.

Lee and Fraumeni demonstrated a greatly increased risk of lung cancer in smelter workers exposed to arsenic in the presence of relatively high concentrations of sulfur dioxide (Lee and Fraumeni 1969). A smaller lung cancer gradient for sulfur dioxide exposure was observed, and it was technically difficult to separate the effects of these two agents. Although an independent role of sulfur dioxide is possible, the data are probably best interpretable as representing a promoting action by sulfur dioxide on arsenic carcinogenesis, perhaps analoguous to the findings of Laskin et al. (1970) discussed below.

Some of the most impressive evidence consistent with the possibility that sulfur oxides are at least partly responsible for an increased incidence of urban lung cancer has been obtained in animal studies. Peacock and Spence (1967) exposed a strain of mice that has a high incidence of spontaneous pulmonary adenoma to sulfur dioxide at 500 ppm for 5 hr/day, for 300 or more days. An increase in adenomas and what is described as carcinoma was noted in the sulfur dioxide-exposed group. In the ongoing sutdies of Laskin et al. (1970), animals have been exposed individually, simultaneously, and sequentially to benzopyrene, and aromatic hydrocarbon air pollutant, and to sulfur dioxide (3.5 ppm). Lung squamous cell carcinomas have been found in the groups inhaling both benzopyrene and sulfur dioxide. Although benzopyrene is a highly potent carcinogen in many systems, lung cancer had previously been observed only after tracheal instillation of this agent, and not during inhalation. These interesting findings must be interpreted with caution, especially because high pollutant concentrations were used in the initial study. In addition, it is not clear whether the findings represent an independent carcinogenic effect of sulfur dioxide in addition to benzopyrene or a potentiation of benzopyrene carcinogenesis by an otherwise unrelated consequence of sulfur dioxide exposure. Recent preliminary studies by this group appear to indicate that combined exposure to nitrogen dioxide and benzopyrene also results in lung carcinogenesis. If this is confirmed, it would tend to support the hypothesis that sulfur dioxide promotes benzopyrene carcinogenesis through its action as a nospecific irritant. However, whatever biomedical mechanisms are involved, further animal inhalation studies with lower pollutant concentrations are definitely warranted.

Despite the biochemical and animal inhalation studies cited, there appears to be insufficient epidemiologic evidence to assign a definite risk to the possibility that ambient sulfur oxides are a factor in the production of human lung cancer.

Quantitation

There have been a number of attempts to quantify the damage caused by air pollutants. To do so, it is necessary to assign some numerical value to the expected health effects in relation to given degrees of air pollution. In the preceding sections of this review, where permitted by the data, quantitative estimates of morbidity and mortality in association with air pollution have been cited from individual papers. It would be possible to graph these estimates and analyze them statistically to arrive at some quantitative estimate of the amount of morbidity and mortality associated with measured concentrations of individual pollutants. However, such an exercise would be grossly misleading and would undoubtedly lead to erroneous conclusions. Each of the studies cited earlier must be considered in relation to the population at risk, the nature of the measured and unmeasured pollutants present, and the limitations in the gathering and analysis of data.

Furthermore, it should be clearly understood that the assessment of dollar costs related to air pollution health effects is a numbers game. As in any game, there are some basic rules that must be accepted. The major rule in this game is that illness can be fully quantified in terms of dollars. That this premise is unacceptable and perhaps unethical must be kept in mind during any discussion of this topic.

A major problem in any quantitative approach to air pollution health effects is in defining a completely safe level of exposure; so-called threshold or no-effect levels. The difficulties with this concept have been ably discussed by different authors in the recent National Academy of Sciences report on "Air Quality and Automobile Emission Control" (NAS 1974). The last paragraph of the statement prepared by Palmes for the NAS-NRC Committee on Sulfur Oxides is as follows:

It should be recognized at the outset that there is no value other than zero that will carry with it assurance of obsolute safety, or zero risk. There are, however, finite concentrations that, in the light of present understanding, would reasonably be expected to produce a very small risk of adverse effects. These adverse effects can range from minor and transient irritation to serious chronic diseases, such as emphysema. Depending on the benefits of the process(es) that introduces the contaminant into the environment, this risk could be judged acceptable or unacceptable. Obviously, as the toxicologic data base is increased, the calculated risk should be changed accordingly. Thus, a given concentration of a specific material could be acceptable at one time and not acceptable later. Similarly, the benefits would necessarily be reevaluated as the technology changes. In summary, the acceptability of a degree of contamination would depend on the risk-benefit appraisal at a particular time.

An interesting introduction to the methodology for quantifying air pollution costs is given in a symposium edited by Wolozin (1966). In general, the participants point out the difficulties in applying economic approaches to this field and apperar rather dubious about obtaining reasonably accurate estimates. Ridker (1967), however, has described a number of methods to evaluate air pollution costs. He estimated the cost of air pollution in 1958 at $360-400 million of the total $2 billion cost of respiratory disease. The figure of 18-20 percent of pollution costs due to air pollution is derived from studies of urban-rural differences in respiratory and lung cancer mortality rates.

A thorough analysis of pollution costs due to disease is presented by Lave and Sesking (1970). They derive their estimate of the impact of air pollution on various diseases by extrapolation form the literature and by extensive analysis of the urban factor in disease. Multiple regression analysis is used for such factors as population density, race, sex, and socioeconomic and age variables, as well as air pollution. They conclude that a 50 percent reduction in urban air pollution would account for 25-50 percent of the excess urban mortality and morbidity from bronchitis, 25 percent of lung cancers, 25 percent of respiratory disease, 10 percent of cardiovascular morbidity and mortality, and 15 percent of cancer in general. On the basis of these figures, a total savings of $2.08 billion in health costs would be associated with a reduction in air pollution by 50 percent. This reduction would bring more polluted cities into the category of urban areas with relatively clean air. They further estimate that such an abatement would lead to a 4.5 percent reduction in the economic costs associated with all morbidity and mortality.

Waddell (1973) has reevaluated these data to assess the amount of money to be saved if the reduction in air pollution reached 1970 standards and has derived a figure of $3.73 billion in 1970. He also discusses an EPA analysis in progress that used data from the

CHESS studies to derive an estimate of the health costs of sulfur oxides and particles of $0.9-3.2 billion. Barrett and Waddell (1973) also reevaluated the Lave and Sesking data to take into account indirect costs not included in the original analysis; by using the figure of 4.5 percent of total health costs due to air pollution, they arrive at a figure of $4.3 billion.

Many of the more recent studies of air pollution health costs have used Lave and Seskin's data (1970) as the baseline for further analysis and have generally derived much higher figures, ranging up to $15 billion, although this is due partially to inflation. Some of these studies have been reviewed by Waddell (1973), Babcock and Nagda (1973), and Williams and Justus (1974). The later authors are highly critical of other studies of this subject and in their own analysis report the figure of $62-311 million as the yearly cost of air pollution to health. Their major objections to the higher figures are that these are based on overestimates of the contribution of air pollution to urban respiratory disease and that there has been a misinterpretation of the Lave and Seskin data to which the additional costs have been added. The first point is discussed below and seems to constitute a valid objection. The second point appears to be partly correct, in that Lave and Seskin were evaluating the major part of the urban air pollution effect and that analyses that have restricted the Lave and Seskin data to sulfur oxides and particles and then added costs for other pollutants have therefore been counting effects twice. However, although the analysis of the air pollutant contribution to urban disease by Lave and Seskin probably did include the effects of nitrogen oxides, it did not consider problems usually associated with carbon monoxide. The criticism by Williams and Justus (1974), concerning the doubling of Lave and Seskin's estimate of $2.08 billion to derive a figure for the total costs of air pollution is probably inaccurate, in that other invetigators appera to be using the $4.3 billion estimate of Barrett and Waddell (1973). In addition, the Williams and Justus analysis probably is in error, in that they assert that cigarette-smoking and air pollution have synergistic effects; that is contrary to most studies, which show an additive relation.

In an interesting report by Chapman et al. (1970), projected emission data are used to estimate the excess adverse affects to be expected if standards are not met in the future. A preliminary assessment of the same areas has also been presented by the Brookhaven National Laboratory (Hamilton, Ed. 1974). Recently, Jaksch and Stoevener (1974) reported a study of outpatient medical costs in relation to air pollution in Portland, Oregon, in which it was estimated that an increase in total suspended particles from 60 ug/m³ to 80 ug/m³ would result in a 3.5 percent increase in outpatient medical costs per contact with the medical system for respiratory disease.

The Ford Energy Policy Project has also evaluated the health costs of stationary fossil-fuel combustion, but its model is concerned mostly with factors not related to community air pollution. However, a panel of the American Public Health Association (APHA) estimated health effects associated with energy use as part of the input into the Ford Energy Policy Project. The original APHA document developed a model to determine numerically the adverse health consequences expected from a partial conversion of energy souces from oil to coal. The authors strongly biased their results by selecting from among the most apparent examples of air pollution health effects in the literature and then overinterpreted the specific findings. This resulted in an extrapolation that indicated that the partial conversion of energy sources from oil to coal would result in severe health effects to a large population. Following a review process, the document was revised, and the results were restricted to more limited circumstances with less of an impact (Carnow et al. 1974). Unfortunately, the original unpublished extrapolations were included in a press release from the American Public Health Association which was widely reported and is still being quoted.

The above discussion does not include non-disease costs of air pollution. These are considered in some detail in a number of the references. Some of these costs are more or less quantifiable, including effects on agriculture, damage to materials, and loss in property values. Others, such as aesthetic effects and annoyance, cannot readily be expressed in numbers. Even the quantifiable effects may have hidden values not easily put into dollar terms. For instance, the monetary costs of covering up the rapidly deteriorating marble facade of New York's City hall is a matter of record.. But one cannot place a number on the resulting cost to the eye of the casual stroller.

It must be emphasized that it is inappropriate merely to balance the health dollar costs with the price of air pollution control. There is an increment above the economic costs that each person would be willing to pay to avoid being ill. Obviously, this increment will vary with the degree of suffering or life-shortening involved, the absolute dollar costs, and financial circumstances of the individual. Most people, for example, would be willing to pay 25 cents if they could somehow magically avoid a painful cut for which the sole economic cost is 1 cent in bandages. Few would be willing or able to pay $25,000 to avoid an appendectomy costing $1,000.

Although it would seem reasonable to approach a cost-benefit analysis by dividing the air pollution control costs on a per capita basis, this may not be appropriate for the benefits. That is because of the marked variability in individual response to pollution and because most members of the population do not now know whether they belong to a hypersusceptible group that will in the future be severely affected by pollution. A more valid economic approach might be to inquire whether the per capita air pollution control cost is a reasonable price for each individual to pay as a form of insurance against the possibility of being significantly and adversely affected by the absence of air pollution control.

The following estimation of the health costs of sulfur oxides is presented with great reluctance due to reservations concerning its validity and usefulness. The analysis is restricted to the effects of sulfur oxides that appear to be reasonably justified on the basis of the foregoing review and is related to estimated health effects within a reasonable range of present pollution conditions. The goal is to provide judgment estimates of possible use to economists interested in costing out the health effects of sulfur oxides. The inexactitude of both the health effects and the economic data appears to justify the use of round numbers and broad estimates. Within these contraints, the bias will be to overestimate the effects of sulfur oxides, and the figures can be considered as upper limits based on available information.

The most apparent effect of sulfur oxide air pollutants is on increasing the morbidity and mortality associated with chronic respiratory disease, particularly chronic bronchitis and emphysema. One approach to assessing the dollar costs for this effect is to estimate the fraction due to air pollution nationwide. Review of the literature reveals that, in addition to air pollution, there are a number of factors associated with the prevalence of chronic respiratory disease, including cigarette-smoking, occupational exposure, constitutional characteristics, and the vicissitudes of aging. Meteorologic factors, particularly low temperature, play an improtant role in acute exacerbations of disease and in mortality. Cigarette-smoking is undoubtedly the major factor in causation of disease, so it would be useful to compare its effects with those of air pollution. This can probably best be done by use of the CHESS studies (EPA 1974), which have carefully and consistently looked at this problem in a number of communities, and which are unlikely to have underestimated the effects of air pollution. In the CHESS summary (Finklea et al. 1974), it is stated that the relative contribution of air pollution to chronic bronchitis prevalence is one-third to one-seventh as strong as that of cigarette-smoking, except for males in New York City, in whom the contribution of air pollution was slightly larger than that of smoking. This is due to the remarkably high prevalence of bronchitis in nonsmokers in New York, ranging up to 5 times as high as that observed for males in other polluted CHESS communities. As pointed out by the CHESS investigators, this is "a finding difficult to accept in the light of other evidence." Furthermore, the relative effect of air pollution was only one-eleventh that of cigarette-smoking in the Chicago military recruit study. The data of Lamber and Reid (1970), obtained during periods of very high pollution in Great Britain, also suggest that air pollution has at most one-third of the effect of cigarettes.

Accordingly, it seems that a reasonable estimate is that air pollution is responsible for at most one-fourth of the effect of cigarette-smoking on bronchitis prevalence, and presumably eventual bronchities mortality, among those who both smoke and live in urban areas. However, assuming that 20 percent of chronic respiratory disease is due to air pollution nationwide is inappropriate, in that not everyone is a cigarette-smoker and some fraction of the problem is related to constitutional factors and occupational exposure. An even greater dilution will occur when a factor is added for the fraction of the population exposed to significant concentrations of products of stationary fossil-fuel combustion. In addition, some degree of chronic bronchitis prevalence and mortality may be related to nitrogen oxides, which are only partially derived from this source. Thus, it seems reasonable to assign no more than 10 percent of chronic bronchitis prevalence in the general population to the effects of sulfur oxides.

Alternatively, if one accepts the figure of 70 percent as the amount that cigarette-smoking contributes to chronic respiratory disease, a figure of 10 percent for sulfur oxides appears reasonable after the effects of occupational exposure, constitutional factors, etc., are considered.

With respect to acute morbidity occuring during the course of chronic respiratory disease, two major factors are the community incidence of upper respiratory infections and temperature variations. After consideration of the dilution factors mentioned above and the epidemiologic observations of this problem in relation to current air pollution, it again appears reasonable to attribute to sulfur oxides no more than 10 percent of the acute morbidity in patients with chronic respiratory disease.

The effect of air pollution on acute respiratory infection is difficult to estimate. There is little evidence of any outstanding effect of sulfur oxides on the incidence of upper respiratory infections. However, in view of some findings that suggest such an effect, particularly in susceptible populations, it seems appropriate to set a figure of 1 percent for the effect of sulfur oxides nationwide. There is much better evidence of an association with lower respiratory tract infections. One approach to assessing the economic costs is to use the figures provided in the CHESS New York City (Love et al. 1974) study, which showed a substantial effect of pollution on lower respiratory tract infections. It was estimated that the average family of four would expect to have 5 extra days of restricted activity and one extra physician visit per year due to air pollution. Because no work time is lost by children, not all fathers and mothers are employed, and people do not work every day, the activity-restricting effect could be roughly translated into 1-2 working days lost per year per employed person in the population at risk as a result of sulfur oxides. However, until this CHESS study is replicated, these figures should be viewed with caution.

Estimates of the effects of sulfur oxides and particles on asthma attack rate have also been provided by the CHESS document. Specific risks appear to increase by 10-50 percent on days with increased pollution and in conjunction with specific meteorologic factors. Making a rough attempt to account for days when the air is clean or the climate is not conducive again suggests that a reasonable estimate from the data might be a 10 percent yearly increase in attack rate in polluted areas, or perhaps 5 percent nationwide.

As discussed earlier, there is insufficient evidence that sulfur oxides participate in the causation of lung cancer or other cancers to assign any risk to this problem.

Separate estimation of mortality due to air pollution is difficult inasmuch as this is due mainly to chronic respiratory disease, which has already been considered. However, one can use the Buechley data (Buechley et al. 1973) for the New York area, which indicate that 2 percent of deaths in 1962-1966 were associated with sulfur dioxide and particles and then arbitrarily extrapolate from the number of deaths for chronic pulmonary disease at that time to assess the total nonrespiratory deaths.

A factor for increased morbidity in patients with cardiopulmonary disease should also be used. However, the bulk of the effects in chronic bronchitis has already been considered. The effects of air pollution in patients with cardiac disease can be roughly estimated from the CHESS (Goldberg et al. 1974) study as being in the range of 10-30 percent reporting more symptoms. The extrapolation of this figure to medical care costs is difficult.

A number of points deserve emphasis. The estimated health costs given above are based on the assumption that the major question concerns the benefits of air pollution control measures in addition to those already in use. The figures are not applicable to discussion of the relaxation of currently operative controls. Although no firm dose-response curves can be given, it is clear that an increase in sulfur oxides above the present concentrations would produce more adverse health effects than would be prevented by a decrease in pollutant concentrations of an equivalent amount. A possible source of underestimation of the health costs is the imprecision of current pollutant monitors. It is conceivable that improved quantification of sulfuric acid or a particular respirable sulfate would result in the observation of a much greater association of sulfur oxides with health effects than is now evident.

In response to the request of the U.S. Senate Committee on Public Works, a panel of the National Academy of Sciences (NAS) is endeavoring to delineate the effects of various control strategies on atmospheric sulfate concentrations in relation to their monetary costs. To quantify the potential benefits of air pollution control measures, it would be extremely useful to have available dose-response curves indicating the extent of adverse health effects produced by given concentrations of a specific pollutant. On the basis of the CHESS studies, the EPA has estimated dose-response curves for suspended sulfates. As pointed out by the CHESS investigators, their data represent first approximations that require further replication. The dose-response curves provided by CHESS may have underestimated the true effects by a factor of 2 or overestimated them by a factor of 10. This judgment is provided solely in the interest of suggesting a framework for the NAS analysis of the impact of various control strategies and in recognition of the urgency of making decisions concerning these strategies.

It must be kept in mind that suspended sulfates are merely an indicator of the health effects of sulfur oxides. Although, on the basis of the scientific literature, it is reasonable to assume that suspended sulfates are a better indicator of sulfur oxide toxicity than is sulfur dioxide, there is clearly a difference in relative potency between various forms of sulfate, and there is little likelihood that one oxide of sulfur is solely responsible for the observed health effects of all oxides of sulfur. The use of suspended sulfates as the basis for an analysis of control strategies has the advantage of indicating the importance of the atmospheric oxidation of sulfur dioxide. However, because the relative potencies of the various sulfur oxides in polluted air have not been ascertained, it might be preferable to relate health effects to units of sulfur emitted into the atmosphere. Furthermore, in view of the uncertainties and possibly inappropriateness of presenting human suffering in terms of dollar costs, a numerical estimation of illnesses might be a more relevant basis for quantifying atmospheric sulfur emission.

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CHAPTER 5

ECOLOGICAL EFFECTS

INTRODUCTION

There is an extensive literature on ecological effects of sulfur oxides, and a number of reviews have been published (Heggestad and Heck 1971, Brandt and Heck 1968, Wood 1968, Webster 1967, Smith 1974, Naegele 1973, Rennie and Halstead 1973, USDHEW 1969, Hindawi 1970, Halstead and Rennie 1973, Delisle and Schmidt 1973, Stokinger and Coffin 1968, Hobbs et al. 1974), including some preliminary attempts to assess economic losses (Rennie and Halstead 1973, Waddell 1974, USDA 1965, Benedict et al. 1971). In the limited time available, no attempt has been made to repeat these views. The primary purpose of this survey is to assess which of the various reported effects is likely to be sufficiently important to influence the choice of a strategy for emission control.

The procedure for decision analysis outlined in Figure 1 of Chapter 13 includes the specification of models for ecological damage and ecological costs associated with various levels and patterns of occurence of sulfur oxides. The purpose of these models is to estimate the marginal costs imposed through ecological effects by additional emissions of sulfur oxides. Although Chapter 13 suggests that such estimates should be made separately for each power plant, this survey will consider only the two extreme cases: nationwide adoption of either (a) an emissions control strategy, or (b) a dispersal strategy (burning high sulfur fuel in rural areas and using tall stacks and/or intermittent control to meet ambient SO_2 air quality standards). Under the emissions control strategy, it is assumed that ambient levels of SO_2 would not increase on the average, and would probably continue to decrease in urban areas. Under the dispersal strategy, the analysis in Chapters 6 and 7 suggests that there would be a small general increase in ambient SO_2 levels in urban areas, a substantial increase in ambient SO_2 levels in rural areas,* and a larger (40-175 percent) increase in the acidity of precipitation.

To place the problem in economic perspective, the dispersal strategy would result in an increase in annual emissions of about 20 10^9 pounds of sulfur (19 10^6 tons SO_x) in the U.S., according to Table V in Chapter 6. Accordingly, an effect which leads to environmental costs of about $200 million per year would correspond to an average incremental cost of 1 cent per pound of sulfur emitted. Since other effects (e.g. those on human health and materials) are thought to involve costs substantially larger than 1 cent per pound of sulfur emitted (Chapter 13), the only <u>tangible</u> ecological effects that merit detailed consideration are those that involve nationwide economic costs of at least this amount.** To anticipate the results of this survey, none of the known tangible effects would involve costs of more than a few cents per pound of sulfur. However, some weight should be given

*It is assumed that a well-conducted dispersal strategy would relieve the damage now being caused by existing low-level point sources, but would lead to a general increase in ambient concentrations, roughly in proportion to the increase in emissions (i.e. by about 65 percent according to Table V in Chapter 6).

**This statement is not intended to deny the importance of intangible effects, such as effects on aesthetics, recreational opportunities, or unique natural assets; such effects need to be weighed together with tangible costs and benefits. Nor is it intended to deny the importance of local damage around individual point sources, which may justify specific local measures to limit emissions or to provide compensation.

to certain hypothetical effects which would impose significantly larger costs if they actually occur.

There are five major obstacles to assessing the ecological costs of a change in sulfur oxide emissions:

(a) Most investigations and reviews have been concerned exclusively with direct effects of sulfur dioxide (Heggestad and Heck 1971, Brandt and Heck 1968, Wood 1968, Webster 1967, Smith 1974, Naegele 1973, Rennie and Halstead 1973, USDHEW 1969, Hindawi, 1970); effects of acid rain have received attention only very recently (Bolin et al. 1971, Likens and Bormann 1974), and there is extremely little information on effects of suspended particulate sulfates.

(b) Most experimental studies of effects of SO_2 on plants have investigated only acute effects at relatively high exposures; effects on growth and productivity at lower exposure levels have been reported but have not been fully investigated (Heggestad and Heck 1971, Brandt and Heck 1968, Wood 1968, Webster 1967, Smith 1974, Naegele 1973, Rennie and Halstead 1973, USDHEW 1969).

(c) Other sublethal effects of air pollutants on plants included enhancement of nutrient stresses, increased susceptibility to insect attack or disease, and effects on soil microorganisms; some experts consider that these are potentially much more important than acute injury (Smith 1974).

(d) Synergistic effects of SO_2 and ozone on various plants have been observed and may occur at pollutant concentrations well below levels that are of concern for human health (Tingley et al. 1973, Menser and Heggestad 1966).

(e) Sublethal effects, such as reduced growth or increased susceptibility to disease, are very difficult to measure in agricultural crops or wild plant populations because of the simultaneous effects of other variables such as weather and other air pollutants.

EFFECTS OF SULFUR DIOXIDE ON VEGETATION

After reviewing studies of direct effects of air pollution on vegetation, Waddell (1974) adopted a figure of about \$200 million as a best estimate of its annual cost in the U.S.; of this, however, only about 5 percent was attributed to effects of sulfur dioxide. These figures, derived primarily from the work of Benedict et al. (1971) are likely to be low, for several reasons: (a) losses resulting from reduction in yield were largely ignored; (b) ornamental plants were under-valued, in that only replacement costs were used as a proxy for aesthetic values; (c) some of the damage attributed exclusively to oxidants may well have been caused by synergism between oxidants and SO_2; (d) no figures appear to have been included for damage to pines, which are very sensitive to SO_2 and to SO_2/ozone combinations and have been extensively damaged around a number of point sources (Rennie and Halstead 1973, USEPA 1971, Costonis 1971, Linzon 1971).

Whatever value is assigned to pines and ornamental vegetation, damage to them is likely to be reduced by any emissions control strategy designed to achieve compliance with ambient air quality air standards for SO_2. There is some evidence that white pines are damaged even at SO_2 levels below the U.S. primary annual standard (Linzon 1971); such damage may continue to be of local significance and may justify more stringent control in areas where pines are of economic importance. However, it is unlikely that the total impact of this damage will be increased above the present level. Accordingly, unless there are large effects of SO_2 on crops at levels a little higher than those now prevailing in non-urban areas, the effects of the projected increase in sulfur emissions by 1980 are likely to be small.

However, it should be noted that one expert in the field has recently expressed a contrary opinion (Heck 1973, pp. 128-129):

> "The potential effect of an increase in oxidant and/or sulfur dioxide concentration is difficult to forecast. At some level the genetic resistance within a species is not sufficient to cope with a pollution insult. This level varies for both native and cul-

tivated species. Once a given pollution level is reached, the effect may increase rapidly with only slight increases in pollution. An educated guess suggests that a doubling of present pollution concentrations on the East Coast could, under otherwise favorable environmental conditions, produce from 25 to 100 percent loss of many agronomic and horticultural crops and severe injury to many native species. Several growing seasons at these higher pollution levels could result in the temporary or permanent loss of native species and major changes in many ecosystems. We are not far from pollution levels which could cause precipitous effects on agricultural production in the more humid areas of the U.S. However, an important variable must be considered in making any predictions based on increased pollution levels. This is that the capability of the living organism to respond and adapt to changes in its environment, within specific ranges of an adverse insult, has not been adequatley determined."

Accordingly the possibility of large adverse effects cannot be dismissed.

EFFECTS OF ACID PRECIPITATION ON TREES AND FOREST PRODUCTIVITY

A number of studies in Scandinavia have suggested a progressive adverse effect of acid precipitation on the growth of coniferous forest trees (Bolin et al. 1971, Jonsson and Sundberg 1972, Marlmer 1973, Overrein 1972, Dahl and Skre 1971). In much of the glaciated parts of northern Europe, soils are naturally acid and deficient in nutrients. The productivity of forest land is closely correlated with the soil levels of calcium, which is subject to leaching by acid precipitation. A study in Sweden showed a significant reduction in growth between 1945 and 1965 in the stands most subject to soil acidification (Jonsson and Sundberg 1972). The extent of the reduction in growth has been estimated to be of the order of 0.3 percent per year in Sweden (Bolin et al. 1971), but perhaps as much as 1 percent per year in Norway (Dahl and Skre 1971). Projecting ahead on the assumption of a continued increase of SO_2 emissions in western Europe, it has been estimated that the overall reduction in growth in Swedish commerical forests might be as much as 15 percent by the year 2000 (Bolin et al. 1971).

In North America, the principal commercial forestry based on coniferous trees in the North-east is in Maine, Ontario, southern Quebec and the Maritime Provinces: these lie in the same geographical relationship (500-1500 km down-wind) to the major SO_2 emitting regions in the U.S. as the Scandinavian forests to the major emitting regions in western Europe. No measurements of the acidity of rain have been traced for these areas of Canada, but high rates of sulfate deposition and acidic precipitation have been recorded in northern Maine (Chapter 7). Since the forest types and soils in this region are generally similar to those in Scandinavia, similar effects on forest growth would be anticipated.

In a study of a hardwood forest in New Hampshire, Whittaker et al. (1974) found that an "abrupt and striking" decrease in volume growth and productivity had taken place about 1960. The change (an 18 percent decrease) was unprecedented in the history of wood volume growth for the forest; the authors tentatively suggested that it might be related to the effects of acid rain and/or drought. The change is not necessarily comparable with that observed in Scandinavian coniferous forests, because soil nutrients were maintained at a fairly high level (Likens et al. 1971); however, leaching of nutrients from the leaves was measured (Eaton et al. 1973). Foliar leaching has been observed in experimental studies with birches exposed to acid mists at pH 4.0, and tissue damage was observed at pH 3.0, but not at pH 3.3 (Wood and Bormann 1975, Wood and Bormann 1974). Growth reduction was not significant even at pH 3.0, however (Wood and Bormann 1974).

Growth abnormalities and tissue damage in various species of pines have been associated with acid rain with pH in the range 4.0-4.5 under field conditions (USEPA 1971, Gordon 1972, Hindawi and Ratsch 1974, Gordon 1974) and with simulated acid rain at pH 3.3 and 4.0 under experimental conditions (Gordon 1974, Shriner and Decot 1974). However, the results have been disputed (Wood 1975) and details of the experiments have not yet been published. Pines are especially sensitive to air pollutants (Heggestad and Heck 1971, Brandt and Heck 1968, Wood 1968, Webster 1967, Smith 1974, Naegele 1973, Rennie and Halstead 1973, USDHEW 1969, Hindawi 1970, Gordon 1972), and the results cannot necessarily be extended even to other coniferous trees.

In the present early stage of investigation, with many studies still unpublished, it is difficult to assess the significance of the various effects summarized above. However, the weight of evidence suggests a strong possibility that acid rain at present and projected concentrations may have widespread effects upon trees in the northeastern U.S. and eastern Canada. The industry at risk is substantial: the total stumpage value of forest harvested annually in Ontario, Quebec, and the Maritime Provinces is about $600 million, and the total value of shipments (lumber, paper, etc.) is 4-5 times larger than this (Rennie 1974, Env. Canada 1973). Except in northern Maine, the forest industry in the northeastern U.S. is relatively small, but there is another substantial industry (annual cut over $800 million) based on pines from eastern Virginia and southern Tennessee southwards (Stat. Abstract U.S. 1973), which would be affected if the recent southward extension of the area subject to acid rainfall (Chapter 7) is continued. Without more direct evidence for the nature and magnitude of damage it would be idle to speculate further, but clearly the problem is not negligible and needs further investigation.

In several areas of the Northeast, especially near the larger cities, the average pH of rain is now around or below 4.0, and locally the average pH of summer rains is as low as 3.5 (Chapter 7). The projected increase in acidity of precipitation if emissions are uncontrolled (Chapter 7) would bring these levels close to those (3.3 and lower) at which acute injury to plants has been recorded under experimental conditions. Accordingly there is a substantial possibility that major damage to vegetation may take place in the most affected regions, especially in urban and suburban areas. The primary concern would probably be for ornamental and garden plants, which are a major recreational resource and contribute substantially to residential property values. The apparent sensitivity of pines is of special significance in this regard, because of their importance in ornamental plantings and for screening.

EFFECTS OF ACID RAIN ON AGRICULTURE

There are very few studies of the effects of acid rain on crop plants, and apparently none has investigated the pH range of greatest interest, 3.5-4.5. Seedlings of kidney beans and soy beans are reported to have shown signs of acute injury when exposed to simulated acid rain at pH 3.2 (Shriner and Decot 1974). A variety of physiological effects was observed in beans exposed to sulfuric acid mist at pH 3.0, but not at pH 3.5 (Ferenbauch 1974). Indirect effects of acid rain on plants include effects on soil micro-organisms, especially those responsible for nitrogen fixation (Rennie and Halstead 1973, Bolin et al. 1971, Shriner 1974), interactions with bacterial and fungal pathogens (Shriner 1974), effects on reproduction (Kratky), possible interactions with herbicides (Gordon 1974), and enhancement of the uptake of soil cadmium (Andersson and Nilsson 1974). None of the studies reported to date indicates a substantial effect on a crop plant at the level of acidity now encountered in precipitation. However, more investigation is needed, because even a small reduction in growth of a major agricultural crop, for example, could have a major economic impact.

The only clearly identifiable effect of acid rain on agricultural systems is the acidification of soils (Bolin et al. 1971). Present-day agricultural practices already tend to acidify soils, so that lime is routinely used to maintain soil pH. The deposition of sulfuric acid and acid sulfates would require use of additional lime; direct absorption of SO_2 would have a similar effect because it is oxidized to sulfuric acid in the soil and similarly contributes to acidification. If it is assumed, as a rough approximation, that half the sulfur oxides emitted are deposited as acid-forming substances on agricultural land, the projected emission of 19 million tons additional SO_2 in 1980 would require the use of about 12 million tons additional lime. At a current cost of $14-18 per ton, including apreading, this would involve additional costs approaching $200 million annually.* Probably only part of this cost would be felt immediately, because some of the soils involved are now reasonably well buffered.

EFFECTS OF ACID RAIN ON FISH AND AQUATIC ECOSYSTEMS

Investigators in Scandinavia have reported major changes in the flora and fauna of acidified lakes and streams (Bolin et al. 1971, Almer et al. 1974, Jensen and Snekvik 1972, Grahn et al. 1973, Johansson et al. 1973). The most striking effects were those on fresh-water fish, expecially salmon and trout, which are progressively eliminated as the pH of ~~the water~~ falls to 5 and below (Almer et al. 1974, Jensen and Snekvik 1972). A similar phenomenon has

*The offsetting value of the sulfur as a nutrient (Grennard and Ross 1974) is probably of negligible significance in the northeastern U.S., where soil sulfur concentrations are generally adequate to maintain productivity and current rates of deposition are already very high (see Chapter 7).

been recorded in acidified lakes in Ontario, where most fish species failed to reproduce after the pH fell into the range 4.7-5.2 (Beamish 1974, Beamish et al.). Disappearance of the fish followed cessation of reproduction, which involved failure of the females to spawn (Beamish et al.) and failure of eggs to hatch (Johansson et al. 1973).

As discussed in Chapter 1, continuation or increase in current emissions of sulfur oxides is expected to lead to similar pheonomena in the northeastern U.S. and in eastern Canada. The rate of acidification is difficult to predict in individual cases and it may take years or decades before some marginal lakes and streams become critically acidified. However in Massachusetts, for example, many lakes and streams already show peaks of acidity in early spring (with pH often in the range 4.5-5.5) and stocking of trout has to be delayed for 2-8 weeks (Cronin and Dixon). The problem can be ameliorated somewhat by addition of lime to managed ponds, but this does not always work, is expensive or impracticable for running waters, and is not recommended as a long-term solution (Bolin et al. 1971, Cronin and Dixon). The primary consequence of losses in fresh water fish populations would be restriction of recreational opportunity, but the economic aspects should not be ignored: there are some 12 million fresh water fisherman in the northeastern U.S., and their annual expenditure on their sport is of the order of $150-400 per capita (Stat. Abstract U.S. 1973, USDI 1967, Bridges and Sendak 1968: figures prorated to 1974).

In addition to effects on economically important fish, changes in populations of a wide variety of aquatic plants and animals have been recorded in acidified lakes (Almer et al. 1974, Grahn et al. 1973). In general, the biological productivity of lakes and marshes is related to concentrations of bicarbonates and calcium in the water. For example, Patterson (1974) has found a close relationship between the reproductive success of waterfowl and the concentrations of calcium and bicarbonates in the ponds where they breed. Continued acidification is therefore likely to lead to a general reduction in biological productivity, except in fresh waters that are well supplied with calcium.

ECOSYSTEM EFFECTS

In a broad sense, human health and welfare are dependent ultimately on the maintenance of the functioning of natural ecosystems. Direct effects of sulfur oxides on human health have been considered in Chapters 1-4; indirect effects are difficult to assess except by consideration of specific components of natural ecosystems, as in the preceding paragraphs. Woodwell (1970) has raised the possibility that the prolonged occurrence of acid rain in the northeastern U.S. may have long-term adverse effects at the ecosystem level. Woodwell showed that the general effect of physical and chemical stresses is to impair the structure and functioning of ecosystems. A specific relevant example is the profound changes induced in fresh water lake ecosystems by acidification (Almer et al. 1974, Grahn et al. 1973). If such broad effects were starting to take place in terrestial ecosystems they would ultimately have major effects on human welfare and would be difficult to reverse.

ATMOSPHERIC AEROSOLS

A recent study in St. Louis has shown that sulfates (sulfuric acid, ammonium sulfate and ammonium bisulfate) are the predominent constituents in the fine particulate aerosols which form visible, light-scattering hazes in eastern Missouri (Charlson et al. 1974). Such visible, turbid air is noted in summer through the eastern U.S. south and west to Arkansas and Kansas (Flowers et al. 1969), and only really disappears with massive intrusions of Canadian air in winter (Charlson et al. 1974). Munn (1973) reported a progressive increase in the frequency of summer hazes in the Canadian Atlantic Provinces in the period 1953-71: the hazes were associated with south and southwest winds and were attributed to transport of aerosols from the eastern U.S. A similar increase in haziness has been recorded in Ohio, Kentucky and Tennessee (Miller et al. 1972). The immediate effect of these changes in visibility is primarily aesthetic, but the possibility of an effect on weather and climate cannot be dismissed, since the aerosols interact with visible light and have the potential for influencing cloud processes (Hobbs et al. 1974).

SUMMARY AND CONCLUSION

(1) The direct effects of sulfur dioxide on vegetation are difficult to assess quantitatively and may have been under-estimated in past studies. However, they are probably unlikely to increase, and may decrease, if an increase in emissions is accompanied by well-managed dispersal.

(2) Effects of acid rain on natural systems are probably of greater consequence than effects of sulfur dioxide, especially if emissions are permitted to increase. However, studies of the effects of acid rain are in their infancy: several important studies are in progress or as yet unpublished.

(3) Identifiable effects of acid rain include acidification of soil, reduction in forest productivity, and depletion of fresh water fish populations. The full impact of these effects may be delayed for years or even decades. Rough estimates of the likely magnitude of these effects suggest that they are relatively modest in economic terms, each involving external costs of the order of 1 cent per pound of sulfur emitted. However, they may also involve loss of recreational opportunity and aesthetic values.

(4) The possibility of large additional effects, such as extensive injury to valuable ornamental plants or reduction in agricultural productivity, cannot be dismissed, especially if the acidity of precipitation is permitted to increase.

(5) Atmospheric hazes, attributable in

large part to fine sulfate particles, are wide-spread in summer in the eastern U.S. Their frequency appears to be increasing as emissions increase. The possibility of effects on weather and climate cannot be dismissed.

(6) Effects of sulfur oxides and acid rain on man-made materials have not been considered in this chapter.

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PART TWO
STRATEGIES FOR CONTROLLING SULFUR-RELATED POWER PLANT EMISSIONS

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PART TWO IN BRIEF: STRATEGIES FOR CONTROLLING SULFUR-RELATED POWER PLANT EMISSIONS

RELATION OF EMISSIONS TO AMBIENT AIR QUALITY AND CHEMISTRY OF PRECIPITATION

Man-made emissions of sulfur oxides in the United States have been increasing at about 4 percent per year and amounted to about 34 million tons in 1970 (see Table 6-2). The most important sources are electric power plants (57 percent), industrial processes (16 percent), and other stationary sources (22 percent) (see Table 6-2). The greatest density of emissions is in the northeastern states (east of the Mississippi and north of Alabama-South Carolina (see Figures 6-2;13-10). There are also substantial emissions (3-4 million tons per year) in parts of southeast Canada adjacent to the U.S. (see Table 7-2). At least in this region, man-made emissions greatly exceed natural emissions (see Table 7-2).

Most emissions are in the form of sulfur dioxide. After emission, sulfur dioxide mixes with the ambient air by diffusing both vertically and horizontally, and is transported (generally eastwards or northeastwards) by the wind. Some of the sulfur dioxide is oxidized to form sulfates, which may in turn form aerosols and travel long distances with the wind (see Chapter 6). The estimated rate of oxidation of sulfur dioxide to sulfates in the atmosphere varies considerably, from as low as 0.1 percent per hour to as high as 30 percent or more per hour, depending on local conditions such as the humidity and the relative concentrations of other air pollutants. This rate of conversion is typically more rapid in urban air than in rural air (see Chapters 6 and 7).

The principal means of removal of sulfur oxides from the atmosphere include absorption of gaseous sulfur dioxide by the ground or by vegetation, and deposition of sulfates in rain and snow (see Chapter 7). Surveys conducted in the northeastern United States suggest that roughly 33 percent of the sulfur oxides are eventually returned to earth as sulfates in precipitation (see Table 7-2). Figure 6-1 pictures the way sulfur oxides are transported after emission, transformed into sulfates, and ultimately returned to ground.

Because sulfur dioxide is absorbed fairly rapidly by the ground, emissions from stacks are probably more important than low level emissions as a source of sulfate aerosols downwind (see Chapters 6,7). For the same reason, ambient concentrations of sulfur dioxide measured at ground level are determined primarily by sources nearby and a short distance upwind; in contrast, ambient concentrations of sulfates are determined by sources further upwind (see Chapter 6). Accordingly, ambient concentrations of sulfur dioxide are generally greater than those of sulfates at urban stations, but are lower at some rural stations (see Chapter 6). Measurements of suspended sulfate aerosols (see Figure 6-4) and of sulfates in precipitation indicate that high levels of sulfate are dispersed very widely throughout the northeastern United States and eastern Canada (see Figure 7-1). The pattern of deposition suggests transport over distances of several hundred km downwind from the principal source areas (see Chapter 7, and Figure 7-1).

The acidity of the suspended sulfate aerosols has not been measured directly, but can be determined indirectly by measuring the acidity of precipitation (see Chapter 7). Acid precipitation is a regional phenomenon in the northeastern United States and eastern Canada, and its distribution covers roughly the same area as experiences the highest sulfate levels (see Figures 7-2, 7-4). The fraction of sulfates falling out as acid sulfates in precipitation is in some areas as much as 80 percent; its regional average is about 24 percent (see Table 7-3). About three-quarters

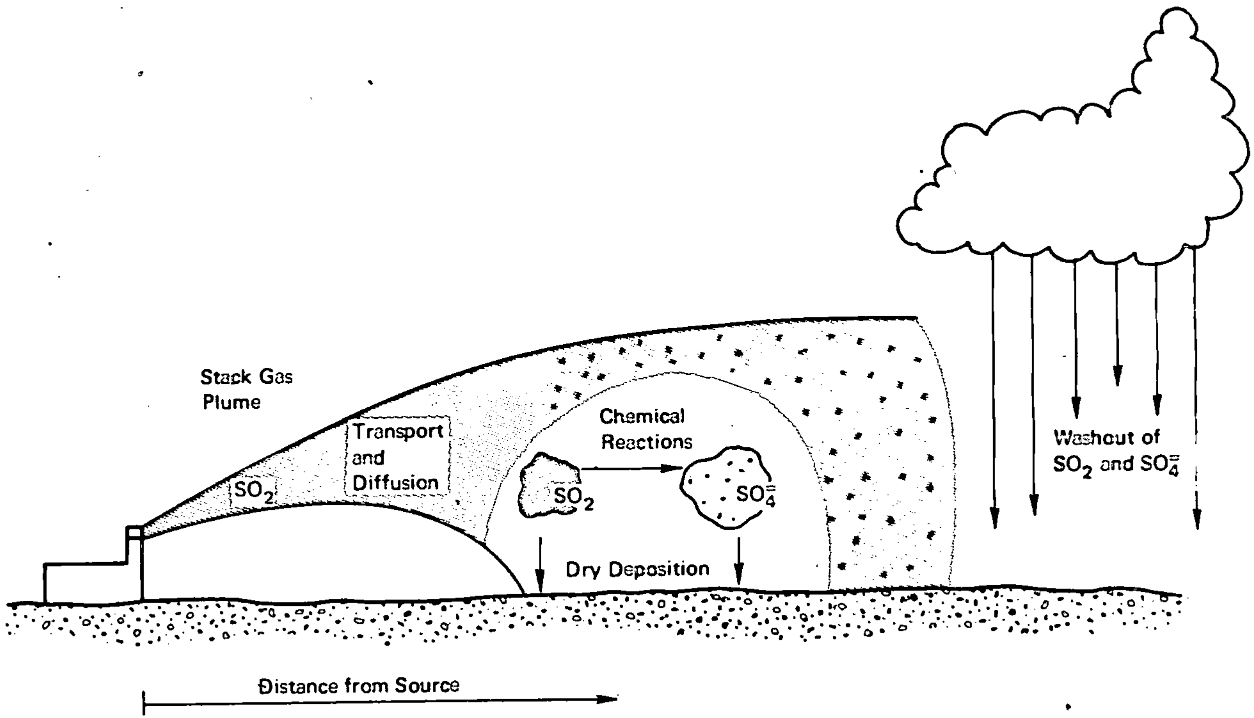

FIGURE 6-1: Processes Involved in the Relationship of Sulfur Oxide Emissions To Air Quality

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of the acidity of precipitation is in the form of sulfuric acid and is attributable to sulfur oxide emissions; most of the remainder consists of nitric acid and is attributable to nitrogen oxide emissions (see Chapter 7, and Table 7-3).

Between 1960 and 1970 total emissions of sulfur oxides in the United States increased by about 85 percent, primarily due to a near-doubling in emissions from electric power plants (see Table 6-2). However, as a result of limitations imposed on the amount of sulfur permitted in fuels, sulfur oxide emissions in urban areas were reduced substantially during that decade (while those in non-urban areas increased disproportionately: see Table 6-2). In consequence ambient concentrations of sulfur dioxide in urban areas decreased significantly. However, ambient concentrations of suspended sulfates in urban areas remained approximately constant throughout the decade and there is some evidence that ambient sulfate levels in non-urban areas have increased (see Chapter 6). This probably reflects the long-range dispersal of air-borne sulfates, and indicates that ambient sulfate concentrations are determined primarily by total <u>regional</u> emissions. The acidity of precipitation has increased more rapidly than total emissions, perhaps reflecting a depletion of neutralizing materials in the air (see Table 7-3). The area affected by acid precipitation has also expanded to include most of eastern North America (see Figure 7-1).

In the absence of emission controls, emissions of sulfur oxides from power plants are projected to double again in the decade 1970-80, and a small increase in SOx emissions from other sources is considered likely (see Chapter 6). Extrapolating from past trends and considering the relative importance of urban and non-urban sources, this growth in SOx emissions is projected to cause only a small increase (0 to 20 percent) in average urban sulfur dioxide concentrations, but a larger increase (18 to 42 percent) in average urban sulfate concentrations (see Chapter 6). The total acidity of precipitation is expected to increase by much larger factors (perhaps as much as threefold) and its distribution may extend over a wider area than previously was the case (see Table 7-4). Part of the increase in acidity of precipitation would be attributable to nitric acid and would not be averted by sulfur oxide emission controls (see Table 7-4, Chapter 7).

A highly simplified model is presented in Chapter 13 to predict the likely impact of a single source of sulfur dioxide on rural and urban air quality downwind of it. Applying reasonable average values for rates of diffusion, absorption and oxidation of sulfur dioxide, it is demonstrated that a single power plant of 600 MW burning 3 percent sulfur coal could cause an increase of the order of 0.15 ug/m^3 in the annual average level of suspended sulfates in an urban area approximately five hundred km (300 miles) downwind. (The range of uncertainty in this calcuation is wide: 0.03 to 0.3 ugm/m^3.) The figure of 0.15 ugm/m^3 is consistent with the analysis of the regional distribution of emissions and ambient air quality summarized in Chapter 6. The impact of a power plant on sulfate air quality far downwind is quite strongly dependent on the rate of oxidation of sulfur dioxide to sulfate, and hence, among other factors, on the amounts and distributions of other pollutants in the atmosphere.

EFFICIENT PRICING AND CONSERVATION

One important means of achieving the broad goals of our national energy policy is to promote conservation. Limiting the consumption of electricity or reducing its rate of growth would reduce emissions of sulfur oxides and other pollutants and thus the damages to health and the environment from the utilization of coal to produce electric power.

One essential device in limiting wasteful consumption of energy is to see to it that its price is equated to its marginal social cost of production. In fact, our pricing of electricity falls far short of this requirement in many ways.

Electric utilities are in most jurisdictions regulated on an original, or historic, cost basis. This means that their capital costs are measured by, and as a return on, the cost historically incurred. But the only measure of marginal cost that has any economic significance is current costs, or, as one sets rates for the future, the cost that will be incurred or saved during the period when those rates are in effect. In times of rapid inflation, those marginal costs tend, naturally, to rise relative to average company revenue requirements, when the latter are based heavily on historic costs. Basing all electricity rates on incremental costs in these circumstances would produce excessive revenues, by traditional regulatory standards. The requirements of economic efficiency could, however, be approximated by setting rates at incremental cost for those categories of demand that are particularly responsive to price. Alternatively, excess revenues could be rebated equally (or in some other manner not related to individual consumption) to all customers.

Second, and operating in the same direction as the use of average rather than marginal cost pricing, is the failure of electricity rates typically to reflect peak responsibility. The required level of investment in generating and transmitting capacity depends specifically on the level of demand at the times of system peak consumption. If the economically proper amount of capacity is to be constructed, therefore, it is that particular consumption, i.e., consumption at the time of peak utilization, that must be charged the full marginal costs at that time of making that capacity available. This could be done, for example, by installation of time-of-day meters. In contrast, consumption truly and inalterably off peak should not pay any capacity costs. It is clear, generally, that the failure of most electric utility pricing to reflect these peak responsibility principles, as well as to measure capacity costs in current rather than historical terms, gives rise to a greater demand for electricity and a consequent greater construction of plant than would otherwise occur.

While efficient pricing of energy is one very important means for reducing wasteful use of electricity and, therefore, for reducing sulfur dioxide emissions, it is by no means the only device, nor is it necessarily sufficient. Improved effectiveness of fuel utilization typically requires capital outlays either by individuals or businesses. Capital shortages and high interest rates both inhibit investment in energy-conserving fixed assets. These considerations argue for government assistance to individuals and businesses in such forms as tax rebates or low-interest loans for energy-conserving investments. At the minimum, non-economic institutional barriers which make it difficult for many to obtain financing, even at market rates of interest, should be overcome.

MODIFICATION OF DEMAND FOR ELECTRIC POWER

Modifications of demand for electricity, and therefore of emissions of sulfur oxides, can be brought about by improvements in effectiveness of fuel utilization in end-uses and by shifting from oil and gas to coal as primary fuel for space heating.

Demand for fuels for electricity generation can be reduced by increasing end-use effectiveness in two general ways: (1) generation of electricity as byproduct of certain industrial processes; and (2) improvement of efficiency of electrical apparatus used in the residential, commercial, and industrial sectors.

Generation of Byproduct Electricity in Industry

Process steam raising and direct combustion heating account for 74 percent of total industrial fuel consumption. If all process steam were to be raised in combination with electricity generation as much as 1270 billion kw-hrs of electricity could be generated at a net fuel saving of about 6.2 quads or 258 million tons of coal per year in 1985.

Similarly, if waste heat from industrial heating processes were to be recovered by bottoming cycle electric generators, as much as 182 billion kw-hrs of electricity could be generated at a net fuel saving of 1.82 quad or 76 million tons of coal per year in 1985.

Not all of the potential by-product electricity can be generated because some industrial plants are too small in scale or operate too few hours per year to justify the expense for the installation of the fuel saving equipment. The rate at which fuel-saving equipment will be installed will depend on fuel and electricity prices. If the price of electricity sold by a utility is determined by average capital costs of supplies rather than capital costs of new supplies, then an individual firm would likely prefer to purchase electricity rather than generate by-product electricity.

To illustrate the effect of pricing electricity based upon average rather than incremental costs, a comparison was made between on-site power generators and purchased electricity. This analysis showed that with present prices for electricity, the overall costs to an industrial user were about equal for purchased electricity vs. a bottoming cycle to generate by-product power from waste heat. However, the incremental power demand placed on electric utilities for purchased power results in the expenditure of almost 50 percent more of the nation's capital than would be required if the bottoming cycle were installed. These comparisons are made on the assumption that the industrial demands in question fall no less than the average of all other demands on the system peak, and so would be incrementally responsible for a propertionate share of the costs of providing the requisite capacity.

Improved Efficiency of Equipment and Processes

Significant opportunities exist for fuel saving through improvements of effectiveness of electrical equipment used in various applications and through re-optimization of electricity consuming industrial processes. Several examples are included here to illustrate the potential for modifications of demand for electricity.

Air-conditioning and refrigeration equipment accounts for over 20 percent of all U.S. electricity and 5.4 percent of fuel consumed for all purposes. The performance (Btu cooling per watt hour of electricity) of such equipment can readily be improved by 30 percent using known heat-transfer technology. The potential fuel saving would be 1.8 quads, or 75 million tons of coal per year in 1985.

A comprehensive study of all electricity consuming processes in industry is needed in order to establish the potential fuel saving. The aluminum electrolysis process serves as an illustration of what might be obtained. By operating the Hall-Process electrolysis cells at lower current density, which is equivalent to increasing capital cost by 22 percent, the electricity required per ton of primary aluminum can be decreased by 16 percent, namely by 2,500 kw-hrs per ton. The potential fuel saving would be 0.25 quads, or 10 million tons of coal per year in 1985. Still further savings in aluminum production may be realized through changes in the process itself such as that being investigated by Alcoa.

Applying recently published FEA guidelines on lighting, it is possible to reduce demand by 133 billion kw-hr in 1985. The fuel saving by electric power plants would be 75 million tons of coal per year.

Shift of Space Heating Load from Oil and Gas to Coal as Primary Fuel

The use of electricity, and the overall consumption of fuel, could increase enormously if a large-scale shift from the direct use of oil and gas to electrical resistance for space and water heating were to take place. Little, if any, increase in overall fuel consumption, and a more moderate increase in electricity demand would occur if the shift were to electrically-powered heat pumps. The heat pump also appears to require a lower overall capital investment for the combined fuel supply and heating equipment than does electrical resistance heating; this would clearly be so if widespread resort to electric heating results in the emergence of a winter peak, because the heat pump would, in that event, produce major savings in the required amount of generating capacity. The alternative of producing gas from coal to serve the heating market also appears to require substantially less capital and less total fuel consumption than electric resistance heating. Relative fuel requirements and capital investment needs, including equipment for both fuel supply and end-use, were estimated for each option to be as follows:

Comparison of Fuel and Capital Requirements for Alternatives to Electric Resistance Space Heating

	Fuel Consumption	Capital Requirements
Electric Resistance	1.0	1.0
Electric Heat Pump	0.55	0.78
Coal Gasification	0.78	0.57

Of course, other options are available for meeting the nation's space-heating requirements, for example, using solar energy or new oil and gas resources, and each of these should also be assessed.

FLUE GAS DESULFURIZATION (FGD)

Introduction

Are flue gas desulfurization systems reliable and operable for scrubbing stack gas effluents from the combustion of high sulfur coal of the eastern United States?

We have considered this question in light of the definition of industrial-scale reliability set forth by the National Academy of Engineering's panel in 1970--viz., (a) satisfactory operation on a 100-Mw or larger unit for more than 1 year and (b) availability of adequate technical and economic data for confident projection of commercial designs for specific local and regional conditions.

Our point of view has been that an operation at most locations in the eastern United States would not be able to discharge salty water. That is, the operation must work in the closed loop mode.

Only lime and limestone scrubbers have yet operated successfully on coal at the commercial scale for extended periods of time, and we have considered the foregoing question in detail only for these systems.

Although there is room for improvement, advances in knowledge of the mechanical design of scrubbers and of the selection of materials for their constructions are sufficient to provide reasonable assurance that a failure in a large-scale test will probably not be the result of a design failure that causes the test scrubber to fall far short of the best for the scrubber's type.

Accordingly, our analysis has concentrated upon the availability of chemical knowledge and the adequacy of performance comparisons among bench, pilot, and commercial scrubbers. Although an ideal technical base for scrubber design would include complete and detailed knowledge of the chemistry, we recognize that the base can fall somewhat short of this ideal if the performance comparisions at various sizes of equipment provide a good empirical knowledge to offset some ignorance in respect to scrubber chemistry. However, commercial experience at scrubber conditions that realistically represent those to be expected for a given design is a sine qua non. This is especially true for a design that is recognized to be difficult. We accept the NAE 1970 study's criterion of substantially 100-Mw capacity as necessary to provide commercial experience.

Until just a little more than 1-1/2 years ago, it was believed that a lime or limestone scrubber in the closed loop mode inevitably operated with a liquor that was supersaturated in respect to gypsum, and that an operation free of troubles from plugging and scaling would depend upon keeping the degree of supersaturation below a critical level beyond which scaling by deposits of gypsum become intolerable. It was recognized that the danger from scaling increased as the degree of oxidation of sulfur to the sulfate form became greater.

It has been learned recently that lime scrubbers can advantageously operate with a liquor that is unsaturated in respect to gypsum. The availability of this option depends, among other things, upon putting some limit upon the degree of oxidation of sulfur to sulfate.

Two lime scrubbers frequently cited as examples of outstanding commercial success are known to operate in the unsaturated mode. Performance comparisons among bench, pilot, and commercial scrubbers are available only for this mode. Accordingly, this mode must be considered the furthest along in its development and the nearest to readiness for wide application. We will treat this option first and in the greatest detail. We will follow with briefer treatments of lime scrubbing in the supersaturated mode, and of the limestone scrubbing in both supersaturated and unsaturated modes.

Lime Scrubbing in the Unsaturated Mode

The unsaturated mode of operation has been observed in the followin lime scrubbing systems:

1. Paddys Run Station of Louisville Gas & Electric. This is a 65-Mw coal-fired peaking unit. The longest operation has been 45 days, during which the unit followed typical load variations for a utility boiler. The unit uses about 80 percent excess air (an unusually high amount), and so the flue gas flow rate is roughly equivalent to 100-Mw; on this basis, the flue gas is equivalent to that from coal at 2 percent sulfur. The operation is closed loop.
2. Mitsui Miike industrial boiler in Japan. The flue gas flow is equivalent to about 160-MW electrical capacity. The coal is 2 percent sulfur. All scrubbing operations in Japan are open loop, including the Mitsui Miike unit. The operation is steady around the clock,

and the unit operated more than 2 years before its recent conversion to limestone scrubbing.

3. EPA's pilot lime scrubber at TVA's Shawnee Station. The flue gas flow is for about 10-Mw electrical capacity. The coal is 3.5 percent sulfur. The operation in the unsaturated mode lasted 17 days, and was shut down voluntarily. Heavy scale formed on the mist eliminators early in the run. The operation did not simulate load following.

4. TVA's bench lime scrubber at TVA's coal-fired Colbert Station. The flue gas flow is for about 1-MW electrical capacity. The coal is 3 percent sulfur. The operation in the unsaturated mode lasted three months. It did not simulate load following.

5. EPA's bench lime scrubber at Research Triangle Park, handling flue gas equivalent to 0.1-MW. The fuel is low-sulfur oil or gas, and sulfur dioxide is added. Sometimes fly ash is added to the scrubbing liquor together with lime. An operation at a given set of conditions is typically for five days.

Scrubbers 3, 4, and 5 operated in the closed loop mode.

If we could regard the operation at Paddys Run as typical of conditions to be encountered by designers of lime scrubbers for wide distribution in the eastern United States, the performance comparisions among the above-named scrubbers might provide an adequate technical base for design. Unfortunately, Paddys Run is not typical. The coal at Paddys Run is unusually low in chlorine content, and the lime used is a by-product of acetylene manufacture from calcium carbide.

By an unfortunate coincidence, Mitsui Miike's scrubber is unusual in just these same two respects.

Data obtained from operation of scrubber 5 has recently cast important new light upon the chemistry of lime scrubbing, especially in explaining some of the difficulties that scrubbing experiments have encountered. The data reveal that presence of chloride ion in the liquor makes operation in the unsaturated mode more difficult, while presence of magnesium ion makes such operation easier.

Although scrubbers 3 and 4 operated with liquors containing substantial quantities of both chloride and magnesium ions, we regard the absence of commercial-scale experience with a coal of moderate to high chlorine content to represent a serious deficiency in the technical base now available to the designer of a scrubber for medium to high sulfur coal. This judgment is reinforced by the fact that the degree to which sulfur is oxidized to the sulfate condition in the Paddys Run scrubber is remarkably low, between about 1.5 to 3 percent. Oxidation in the other scrubbers is higher, and sometimes markedly so. As noted earlier, operation in the unsaturated mode becomes more difficult the higher the degree of oxidation.

Experience can and should be obtained quickly for a commercial lime scrubber using liquor that contains chloride ion at a high level. For example, the Paddys Run scrubber could be operated with admixture of hydrogen chloride gas to the flue gas entering the scrubber. Any test of an operation with such a liquor should provide a realistic assessment of the problems of maintaining a strictly closed water loop.

Confidence in the technical base could be much enhanced by operating the Paddys Run scrubber with ordinary lime in place of the carbide lime that has been used in operations to date.

Confidence in the technical base could also be much enhanced if further experimentation at the bench or pilot scale can reveal what variables are important in promoting or minimizing oxidation of sulfur to sulfate in the scrubbing liquor. Quantitative relations connecting the variables to the oxidation level would be of great value. At present, we cannot be sure that we can write down a complete list of the important variables, and quantitative relationships are completely lacking.

We reiterate, however, that lime scrubbing in the unsaturated mode is furthest along in its development. A resolution of the question of its commercial viability can be acquired soon, through easily obtained commercial experience that will allow adequate performance comparisons among bench, pilot, and commercial units. The probability that the question will be answered favorably is a matter for engineering judgment in light of the now available chemical knowledge and performance comparisons. Some of us judge the probability to be 90 percent, while one member judges it at 70 percent.

Lime Scrubbing in the Supersaturated Mode

A large scrubber (of a novel, horizontal design) has operated in the supersaturated mode at Mohave Station of Southern California Edison Co. The coal is of unusually low sulfur content, and the system is yet untried on medium or high sulfur coals, for which design is more difficult.

EPA's 10-Mw lime scrubber at TVA's Shawnee Station has usually operated in the supersaturated mode, and the operators regard its performance as successful, although there has been trouble at the mist eliminators.

No recent commercial experience is available for comparison with the 10-Mw results. Although operation of the Fulham Station lime scrubbers in England in the late 1930s was reportedly successful, information on this operation is not sufficient to provide a basis for modern scrubber design.

Some workers take the view that operation far into the supersaturated mode, at a high degree of oxidation, will be possible even in the closed loop mode if the scrubber is designed physically to operate without trouble from gypsum deposits. Such a mode of operation would require a new path of development that might be said to have only barely been begun.

Limestone Scrubbing in the Supersaturated Mode

The sucessful operation of a large limestone scrubber at the Cholla Station of Arizona Public Service contributes relatively little to the technical base for design of scrubbers for medium or high sulfur coals, because of the low sulfur in the coal burned at Cholla Station.

The operation is closed loop, but experiences water loss by evaporation from the sludge pond.

Large limestone scrubbers (of similar design) are operating at Commonwealth Edison's Will County Station and at Kansas City Power & Light's LaCygne Station. Both systems operate in the open loop mode. Availability of the Will County scrubber has recently been good, but the operation now blends Western low sulfur coal into high sulfur Illinois coal, to provide a fuel at an average sulfur level of about 1.5 percent. The LaCygne operation is on coal containing 5.5 percent sulfur, and is subject to fouling and plugging that is dealt with by cleaning out each of seven scrubbing modules about once every five nights.

EPA's nominally 10-Mw pilot limestone scrubbing system (at TVA's Shawnee Station) has operated in the closed loop, supersaturated mode. The ratio of liquor flow to gas flow is considerably higher than in the Will County-LaCygne design. There is a higher content of solids in the liquor. A longer time is allowed for reaction of limestone with the spent liquor. The gas velocity in the scrubber is reduced, thereby derating the unit to about 7-Mw. These conditions are closely comparable to conditions reported to have been successful in operating limestone scrubbers in the closed loop mode at Fulham Station in England in the late 1930s.

No recent commercial experience is available for comparison with the 10-Mw results.

TVA's 1-Mw bench limestone scrubber at TVA's Colbert Station has operated successfully with a novel mist eliminator whose design prevents wash liquor (used to keep the mist eliminator clear) from entering the main flow of scrubbing liquor. Commercial experience for comparison with the 1-Mw results will be obtained at TVA's Widows Creek Station.

Limestone scrubbers in Japan commonly operate with high degrees of oxidation of sulfur to sulfate and high supersaturation of the scrubbing liquor. In these units, the objective is to produce gypsum for wallboard manufacture. The operations are open loop. The Mitsui Miike unit cited earlier has recently converted to limestone. Several other coal-fired units have recently begun to operate, or are about to operate, with limestone scrubbing systems.

As mentioned earlier in connection with lime scrubbing, some workers take the view that operation far into the supersaturated mode, at a high degree of oxidation, will be possible if the scrubber is designed to accomodate deposition of gypsum. The operation would be closed loop, unlike the systems in Japan. A new path of development would be required to provide the proposed mode of operation.

Limestone Scrubbing in the Unsaturated Mode

Limestone scrubbing in the unsaturated mode has been demonstrated in EPA's tiny bench unit at Research Triangle Park. There is general agreement that operation in the unsaturated mode will be much more difficult for limestone scrubbing than for a lime system. This option could not be taken seriously until it has been demonstrated on a scale larger than the 0.1-Mw bench unit.

Availability of Lime and Limestone Scrubber Technology

Commercial lime scrubbers for an Eastern United States utility coal medium in sulfur (1 to 3 percent) and low in chlorine (less than 0.04 percent) can be ordered today with reasonable confidence.

No commercial experience is available for a lime scrubber on commonly occurring coal both medium or high in sulfur and higher than 0.04 percent in chlorine. Chlorine is known to interfere with the lime scrubber chemistry and to make operation more difficult. Experience can and should be obtained quickly for medium and high sulfur coals containing chlorine beyond 0.04 percent. The experience would resolve the issue of commercial availability of lime scrubbers for these coals. The probability that the issue will be resolved favorably is a matter for engineering judgment in light of data now available from 1.0 and 10 Mw lime scrubbers.

Some engineer members of the Committee felt the probability to be 90 percent, while one member judged it at 70 percent.

No other scrubbing technology is available for order today with confidence for Eastern United States coals medium or high in sulfur. Other scrubbing techniques await commercial demonstration on such coals.

Regenerative Flue Gas Desulfurization Processes

The Wellman-Lord and magnesium oxide processes, both of which recover a useful by-product, are now being installed in high sulfur coal plants. Successful operation of these plants would represent a major advance in FGD technology, in that a proven regenerable process would now be available. In addition, there is a non-regenerable process, sodium carbonate, operating on low sulfur western coal.

There is substantial successful FGD experience in Japan, but it is not directly applicable because the installations are not coal fired and operate in an open loop mode which permits discharge of untreated water to the environment. Within the U.S. there are several Wellman-Lord processes installed in the petroleum/petrochemical industry for sulfur recovery. These are operating successfully.

Cost of Flue Gas Desulfurization

The installed cost of a lime scrubbing FGD system ordered today for a new coal fired plant will be about \$100/kw. The installation cost could be as low as \$60/kw or as high as \$130/kw. The estimated operating costs for lime FGD systems are 3 to 6 mills/kwh, including ponding of sludge, energy loss, and capacity derating. About one half this operating cost is capital charges. If expensive pond facilities (e.g., plastic lined) are required, as opposed to a clay lined pond, an additional charge of up to 0.7 mills/kwh would result. Chemical fixation, if used or needed, could raise the 3 to 6 mills/kwh by 0 to 1.0 mills/kwh. There is considerable uncertainty in fixation costs, because of limited experience with the processes. Other FGD processes, limestone, Wellman Lord, etc. will require about the same capital cost as lime. If the cost of sludge disposal becomes large enough, say about 2 mills/kwh, a regenerable process with by-product credit would have a potentially significant economic advantage.

Residuals Produced from Flue Gas Desulfurization

Particulate concentrations leaving a FGD system are typically .01 to .02 grains/SCF which is adequate to meet air quality criteria. Some very limited data suggests that about 40 percent of the particulates leaving a scrubber are sulfates (calcium, magnesium) and not fly ash.

A typical 1000 MW power plant will generate about 1,185,000 tons/year of fly ash and sludge for disposal (wet basis). This requires, over the lifetime of the plant, about 377 acres over the life of the plant for disposal compared with 108 acres for disposal of fly ash only. The 377 acres is of the same order of magnitude as that required for the power generation facilities. Chemical fixation is now being developed to produce a stabilized material with low permeability and leachability. Tests on commercial scale modules are now underway in several locations to further evaluate fixation processes and potential secondary pollution problems. Results are reported to be encouraging.

Capability of Vendors and Utility Companies

There are about 3 to 5 vendors who have substantial commercial experience in FGD installations and another 10 or so who have capabilities in this area. Estimates have been made that each vendor can design and install 3 to 5 FGD systems per year. The EPA has estimated that 90,000 MW of FGD systems are needed by 1980 to control sulfur dioxide emissions from coal fired plants. There are now

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17 systems under construction and 63 planned--a total of 37,000 MW of generating capacity. It will be difficult to meet the target of 90,000 MW by 1980.

There is also a potential scarcity of engineering manpower to design and construct the FGD systems. The utility companies for the most part do not have on their plant staffs skilled process engineers who can provide the technical service so necessary to make an FGD system, once installed, operate successfully.

CONTROL OF AMBIENT SULFUR DIOXIDE CONCENTRATIONS WITH TALL STACKS AND/OR INTERMITTENT CONTROL SYSTEMS

Temporal emission controls for reducing ambient sulfur dioxide levels, otherwise known as intermittent control systems (ICS), have been recognized as a viable air pollution control technique in this country only for certain limited situations, and have been the focus of a lively debate between various regulatory agencies and some parts of the electric utility industry. The term "ICS" refers to a system of control whereby the rate of emissions from a pollutant source is curtailed when meteorological conditions conducive to ground-level pollutant concentrations in excess of the ambient standard exist or are anticipated to occur. For power plants, two intermittent control strategies are potentially available: (1) fuel switching, i.e., burning a temporary supply of low sulfur fuel; or (b) load switching, i.e., switching a portion of the electrical load to an interconnected generating station with available capacity in excess of demand.

Tall stacks are inherently an integral part of ICS programs, since increased stack height can yield a decreased need for intermittent emission reductions. For example, at its Kingston Steam Plant, TVA suggests that anticipated average yearly requirements for ICS measures will drop from 55 days per year (with a 7-hour average duration) for the plant's current stack configuration (four 250-foot stacks and five 300-foot stacks) to zero days per year for two 1000-foot stacks.

Legal Background

The application of tall stack/ICS measures for dispersal of SOx pollutants can be effective in reducing ground-level sulfur dioxide concentrations in the vicinity of power plants burning high sulfur coals and oil. However, on the basis of long-term averages, such measures provide generally negligible reductions in the amount of pollutants emitted. It is principally for this reason that the Environmental Protection Agency considers constant emission reduction techniques far superior to dispersion strategies. EPA claims the dispersion concept is not compatible with the Clean Air Act requirement that constant emission limitations be enforced whenever possible. In the most significant legal decision to date on this subject, Natural Resources Defense Council v. EPA, the 5th Circuit Court of Appeals upheld this contention, ruling that there is an express Congressional mandate in the 1970 Clean Air Act amendments that emission reduction is the preferred method of meeting ambient pollutant standards. A case addressing this same issue (Tennessee Valley Authority v. EPA) is currently in the 6th Circuit Court of Appeals. More recent legislation, the Energy Supply and Environmental Coordination Act of 1974 (ESECA), provides additional insight into Congressional intent regarding implementation of tall stack-ICS technology. According to language contained in ESECA, the application of ICS technology is permitted only under certain circumstances; for example, these systems can sometimes be used to comply with interim control requirements when extensions of time are granted to utilities to meet emission standards.

Although recent court decisions, EPA regulations, and ESECA suggest otherwise, this evaluation of tall stack-ICS technology was undertaken under the assumption that this technique could become a legally permissible method of controlling sulfur dioxide concentrations. This is because:

(1) ICS measures could provide an alternate means of attaining and maintaining sulfur dioxide ambient air quality standards, independent of reliance on flue gas desulfurization (FGD) technology or use of low sulfur fuels.

(2) Implementation of these systems could result in attainment of existing ambient standards in a shorter period of time than would otherwise be possible.

(3) These measures could be important interim techniques, given limitations on availability of FGD systems and low sulfur fuels over the next few years, given the difficulty in retrofitting some facilities with FGD systems, and given distribution and allocation problems associated with low sulfur fuels.

Assessment of Intermittent Control Systems (ICS)

Assessments of four aspects of ICS technology are offered, including an analysis of system performance, potential implementation, cost, and secondary environmental impacts.

Performance

A number of different types of potential ICS applications exist, with individual situations requiring (a) various degrees of difficulty in air quality modeling and forecasting, (b) different problems in monitoring actual air quality, and (c) assorted frequencies and severities of required emission reductions. Operating tall stack-ICS systems have generally proven effective in reducing the number and extent of excess concentrations of sulfur dioxide in the vicinity of single SOx sources. However, for operating systems for which data are currently available, only the TVA Paradise Steam Plant ICS installation appears to be meeting all air quality objectives for the time period considered. This is due to the fact that the Paradise plant, according to indicators which measure the degree of difficulty associated with a given application of ICS technology, is located where relatively few problems would be anticipated in implementing ICS control.

Constraints on ICS Control Implementation

Availability of control by tall stack-ICS methods is a function of stack and emission parameters, as well as local meteorology and terrain. For nine TVA power plants under different sets of conditions, reduced emissions are called for between 0.1 to 4.4 percent of the time, on an hours per year basis. These decreases in emissions must be supplied either by load shifting or fuel shifting; a number of constraints apply concerning each of these techniques which limit the potential for emission reductions by these methods at various plants.

Costs

Costs for implementing the tall stack-ICS approach are significantly less than the costs involved in installing FGD systems, both in terms of capital and annual cost requirements. Based on TVA operating experience, with an expanded cost range to allow for the different conditions that may be confronted, costs for ICS control can be expected to be as follows:

Capital costs	=	\$4 - 10/kw
Operating costs, including annualized capital charges	=	0.15 - 0.4 mills/kwh

Agency costs for monitoring and enforcement of ICS systems represent a secondary cost of control, not included in the estimates above.

Secondary Environmental Impact

Much of the controversy surrounding implementation of the tall stack-ICS approach is associated with the impact of this technology on acid-sulfate particulate matter formation, and effects of these sulfates on health, welfare, and aesthetics. These matters are discussed elsewhere in this report. It is important to note here that Application of tall stack-ICS technology does not significantly reduce total emissions of sulfur oxides; hence, this strategy does not decrease ambient sulfate concentrations. so ambient sulfate concentrations will not be reduced.

Enforcement

An important issue regarding implementation of ICS technology concerns the legal enforceability of these systems. Recently proposed EPA regulations for ICS control place particular emphasis on enforcement of violations of emission limitations included within an approved operational manual for each system, as well as enforcement based on violations of ambient sulfur dioxide standards. Enforcement based on this dual concept, which was not done previously, should be more effective than enforcement based on violations of ambient standards alone. Many elements of the enforcement procedure can be established prior to approval of the ICS approach by the control agency.

Public Policy Toward Tall Stack-ICS Control

A potential compromise position regarding public policy toward tall stack-ICS technology is as follows: the technology should not yet be accepted as a permanent control technique because of the probability of substantial potential risks due to increased sulfur dioxide atmospheric loadings and hence of sulfates downwind. At the same time, the technology could be implemented as an interim control measure in carefully defined situations because it permits ambient standards to be met while continuous controls are implemented, and because it would help reduce the current clean fuels deficit.

OTHER TECHNIQUES FOR REDUCTION OF SULFUR IN THE ATMOSPHERE

The options immediately available to the utility industry for greatly reducing sulfur emissions to the atmosphere are severely limited. They are restricted to stack scrubbing and shifting to low sulfur fuels. A number of advanced technologies that will bear significantly on the problem are now under investigation, but they will not have reached the stage of commercial deployment until the early to mid 1980s. Several elements of our national energy position will tend to increase the amount of sulfur emitted to the atmosphere.

Existing and projected shortages of natural gas will reduce the amount of clean fuel (low in sulfur, low in particulates) available for electric utility use. Domestic petroleum production has not been able to meet domestic demand for a number of years and new national policies are aimed at further reducing the imports that have made up the deficit. As a result the electric utility industry will have even less petroleum to use in the future than it has in the past. These factors will tend to increase total sulfur emissions from power plants.

Nuclear capacity, which had been projected to rise sharply between now and 1985, will, as a result of recent decisions delaying its installation made by utility companies, fall far short of early projections. Estimates by the Atomic Energy Commission in 1972 placed 1985 nuclear capacity at approximately 250,000 Mwe. It now appears likely that actual capacity in that year will not exceed 120,000 Mwe. If the growth in total electrical demand does not decrease proportionately, this will throw an increased burden on coal to the degree that the decrease in nuclear capacity exceeds the decrease in electrical demand from earlier forecasts. Much of the delayed capacity is in the eastern part of the United States where low sulfur fuels are not readily available.

A range of actions are available to state and federal governments that could result in reversing the downward trend in expected additions to nuclear capacity and recapturing a substantial portion of the capacity that is now being delayed. This could result in a reduction in coal consumption of 75 million tons in 1980 and very substantial additional tonnage by 1985. If these actions are not taken, sulfur emissions may be expected to increase.

Eastern low-sulfur coal reserves are large but much of the reserves are held for metallurgical use, and are not available to the electric industry. Existing production capacity for these low-sulfur coals is inadequate and is likely to remain so. By shifting available low-sulfur coal to plants not meeting primary standards from plants which could burn higher sulfur coal and still meet the primary standards, some reduction in violations of existing ambient air quality standards could be achieved. It is estimated that a shift of about 36 million tons could be made to reduce the tonnage in violation by about 15 percent. Low sulfur western coals will be usable in new coal fired plants designed to burn them if transportation capacity is increased but their use in retrofitted plants will be limited.

A number of technologies may be available in future years that could favorably impact on sulfur emissions.

A low sulfur-low ash product can be produced from coal; commercial (solvent refining) plants to do so may be in operation by 1982-83. The products made from a coal with costs of 80 cents per million BTU are estimated to cost in the range of $1.75 to $2.00 per million BTU. Alternatively, a low BTU-low sulfur gas can be made from coal (80 cents per million BTU) with costs in approximately the same range. Commercial plants making gas may be operational in 1981 or 1982.

High sulfur coals may be burned directly in an environmentally acceptable way in a fluidized bed operated either at atmospheric or elevated pressure. A full scale unit designed for atmospheric pressure could be in operation by 1980 or 1981 and a pressurized unit approximately two years later. If fluidized bed combustion can be successfully developed, it should produce a clean fuel for boiler use that is less costly than either a low sulfur-low ash coal or low BTU-low sulfur gas produced from coal.

Proven conventional coal cleaning methods can reduce the sulfur content of coal significantly. However, unless the original sulfur content is already low enough to nearly meet the sulfur oxide emission standards, conventional coal cleaning methods will not bring most coals into compliance. Advanced coal cleaning methods may be able to increase the amount of sulfur and ash removed but all of the processes are in their early stages of development and many may be high cost for the extra sulfur removal that is accomplished.

Methods of generating electricity at increased efficiencies (in order to reduce the pollution load per unit of electricity generated) are not expected to come into widespread use until 1985 or later. As important as it is to continue R&D on these advanced power cycles, they offer no solution for reducing sulfur oxide or particulate emissions in the period between 1975 and 1985.

For many uses, particularly in new installations, either electricity or a pipeline gas made from coal could be used to supply energy requirements. The choice of which route to select should be based on supplying energy at the lowest marginal cost to the user.

Pipeline gas (of heat content of approximately 1000BTU/cu ft) from coal can be produced at the gasification plant by the early 1980s for approximately $2.50 to $3.00 per million BTU. Electricity produced at a new coal-fired base load plant (equipped with air and water pollution controls) would cost about 2.6 cents per kwh or $8.10 per million BTU. With resistance heating (100 percent conversion of electricity to useful heat) electricity costs would be approximately twice that of gas from coal (excluding transmission and distribution costs for both fuels). Under the same assumption, with a heat pump with a seasonal performance factor of 2, electricity would cost 30 to 60 percent more than gas.

Total costs to the consumer for electricity (after adjusting transmission and distribution costs upward by 50 percent over the 1968 costs) using resistance heating (at 100 percent efficiency) would be about $12.80 per million BTU, or from 100 percent to 120 percent more than gas made from coal. If a heat pump were used (with a seasonal performance factor of 2) the cost would be about $6.40 per million BTU, or about the same, for the same usable BTUs, as gas made from coal.

Solar heating of domestic hot water is technologically feasible and probably economic in many areas of the country now. If institutional barriers to its widespread deployment are overcome, it would reduce the use of coal for generation of electricity and release natural gas for other purposes and thus make a measurable contribution to air quality.

ANALYSIS OF ALTERNATIVE STRATEGIES

The basic question addressed in analyzing alternatives is whether the benefits from sulfur oxide removal by such means as flue gas desulfurization or switching to low-sulfur coal justify the additional increment of cost in electricity generation. This question is addressed by examining representative values of the ecónomic and emissions-related characteristics for different generic classes of power plants. The increase in cost of electricity is compared with the social benefit acheived through reduction in sulfur oxide emissions to find the value at which a switch in abatement techniques is advisable.

In this report a comprehensive cost-benefit formulation is proposed, illustrated by the diagram of Figure 13-1. The intention of this formulation is to provide a methodology in which the influence of the various factors on the decision is made readily apparent. This methodology is available for analyzing the decision among abatement strategies on a case by case basis in cost-benefit terms. The marginal increase in the cost of electricity is compared to the marginal benefit per pound of sulfur oxide emission reduction achieved, for each alternative strategy. This methodology can be

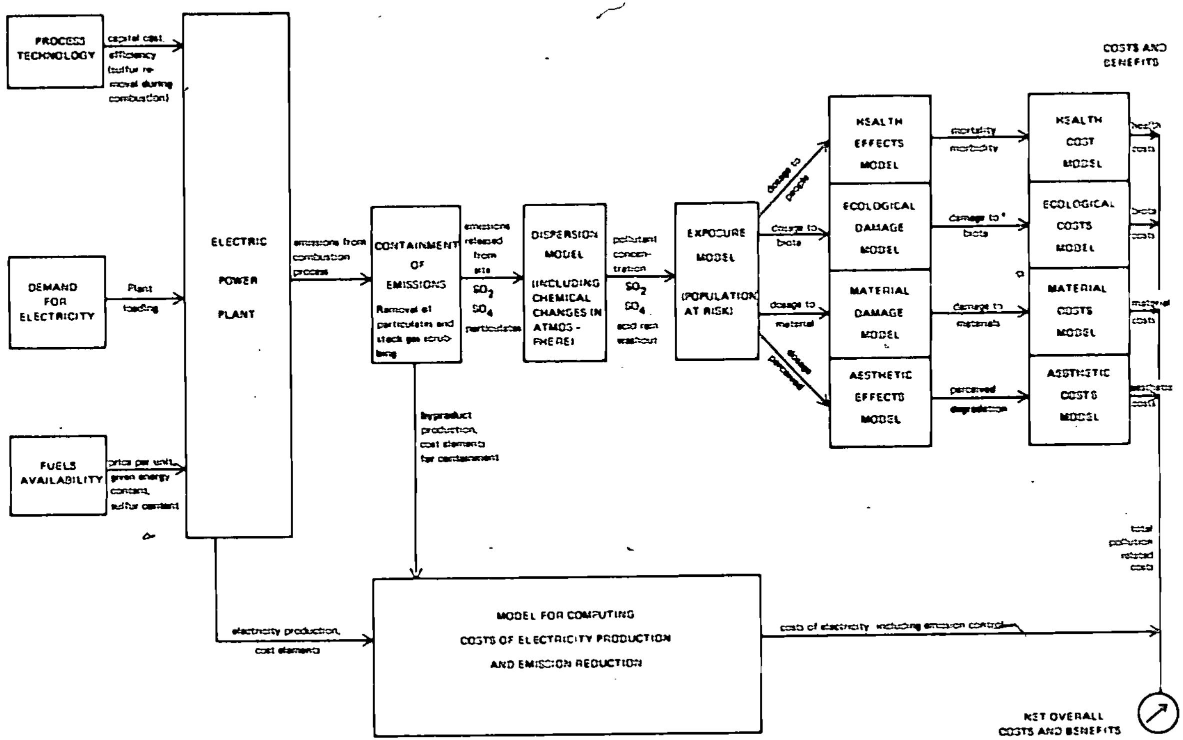

FIGURE 13-1: Overview: Sulfur Oxide and Fine Particulate Pollution From a Stationary Source

used to determine priorities for implementation of abatement technologies.

The accuracy of model studies depends on inclusion and accurate description of all important cause-and-effect relationships. The limited information presently available makes decisions among strategy alternatives difficult. The use of models is more to enhance understanding of the relative importances of the various factors that affect the decision than to determine with precision what is the optimum strategy. If changing a factor in the analysis within its range of uncertainty leads to a change in the preferred strategy alternative, then that uncertainty would clearly be worth something to resolve before a commitment to a particular strategy is taken. Likewise, it may be that despite uncertainties in many factors, one alternative appears sufficiently better than the others that a commitment now appears preferable to the costs incurred by deferring the decision. The purpose of the analysis is to enhance understanding of a complex problem, including the effects of the uncertainty upon the decision. While specific numbers are used to illustrate the calculations, it should be appreciated that many of the numbers and relationships in the model are uncertain, and subject to considerable change as additional information becomes available. The analysis should be taken as a method for organizing and trying to place in perspective the information presently available to serve as a basis for decision making on sulfur oxide emissions control.

The Need to Consider Decisions on a Case by Case Basis

For a specific power plant, differences in fuel availability and price, plant loading, plant age, and the economics of abatement strategies may differ considerably from the representative values used in the analysis. Hence the marginal cost of emissions reduction per pound of sulfur removed may differ appreciably from the values used in the calculations of Chapter 13. The calculation of marginal costs of sulfur oxide abatement strategies should be based on the particular economic and technical factors for each individual power plant.

The consequences of emissions may vary considerably from one power plant location to another, and these differences should also be taken into account in the decision among alternative strategies. The relation between emissions of sulfur oxides and ambient levels of sulfur dioxide and sulfates, and the potential for damage to human health, vegetation and other ecological systems, material property, and aesthetic values will in general differ as a result of regional and local factors. As a result, the value of reducing emissions should be assessed separately for different power plants. At some locations the value per pound of emissions reduction may be judged much higher than at other locations.

Another reason for examining the emissions control decision on a regional and local basis is that limited resources are available to implement quickly the widespread use of low sulfur coal or stack gas scrubbing. Scrubbing equipment should be installed first in those situations where the benefits of emissions abatement exceed the costs by the greatest amount, and thereafter should be installed on other plants where the benefits are judged to exceed the costs, as the equipment becomes available.

Assessing the Value of Sulfur Oxide Emissions Reduction

The assessment of the deleterious consequences of sulfur oxide emissions to human health, vegetation and living systems, material property and aesthetic values is difficult. Decisions on emissions control strategy require a trade-off between eliminating emissions and increasing the cost of producing electricity. It is better to have explicit judgments stated for these trade-offs so that emissions control decisions can be made on a consistent basis that takes into account variations in local and regional situations, and so that the affected parties (utilities, consumers of electricity, those impacted by the consequence of the emissions) are aware of the basis on which emissions control priorities are being set. Hence, it is highly advisable to carry out an assessment of pollution consequences in monetary terms, that is, per pound of sulfur oxides emitted. These assessments should reflect the price that society is willing to pay in increased electricity costs to reduce sulfur emissions.

For the analysis described in Chapter 13 three generic cases were used to illustrate how this methodology might be put into effect: a representative existing plant in a remote rural location (e.g., the Eastern Ohio-Western Pennsylvania-West Virginia area), a new plant to be constructed in the remote rural location, and an oil fired plant located in the vicinity of a major metropolitan area such as New York that might be converted back to coal. To the extent permitted by the limited time and resources allowed for the study, available literature and expert judgement were used to estimate human health, ecological, and materials damage, and aesthetic consequences of sulfur oxide ambient levels, and to assign monetary costs to these effects. To relate ambient levels to sulfur oxide emissions, a model for oxidation and dispersion was constructed. Together these steps allowed a pollution cost to be computed per pound of sulfur emitted from the power plant. Assessment of pollution consequences in terms of cost per pound of pollutant emitted permits the comparison of costs and removal efficiencies on an economic basis and helps to identify the most important factors that affect the choice among the alternative strategies.

The available information for carrying through the analysis is extremely limited, and the decisions among alternative strategies appear quite close. For the case of a plant in or near an urban area, some alternative for reducing total emissions (e.g., low sulfur eastern coal or flue gas desulfurization) appears clearly preferable to burning of high sulfur coal.

The main result of the analysis is to characterize of the uncertainties. The most important areas of uncertainty appear to be the sulfur oxide emissions to ambient sulfate relationship and the health effects of ambient sulfate. A simple model was used for the relationship between sulfur oxide emissions from a power plant and the incremental increase in ambient sulfate in urban areas several hundred miles downwind. The chemistry of the oxidation process for transforming sulfur dioxide into sulfate was identified as the most crucial area of uncertainty in determining the emission to ambient relationship. Variations in oxidation rate over a reasonable range of values could lead to changes in the increase of ambient sulfate levels from given sulfur emissions over the range of a factor of ten. Health effects were modeled using a very crude dose-response relation drawn mainly from the limited data of the CHESS studies. Chronic respiratory disease and aggravation of heart-lung disease symptoms appear to be the most important health effects. The range of uncertainty for the incidence of each health effect for a given change in ambient sulfate was judged to be a factor of twenty.

These uncertainties were roughly characterized in terms of probability distributions, and an overall probability distribution calculated for the pollution cost per pound of sulfur emitted, for the representative plant in a remote rural location and the representative plant in an urban region. These distributions indicate considerable uncertainty; new information on the emissions to ambient relationship or on health effects could easily change the estimated pollution cost by at least a factor of two or more in either direction from the nominal values.

The Marginal Cost of Reducing Sulfur Oxide Emissions

Analysis of the representative cases yields the following results. With 0.9 percent sulfur eastern coal priced at $32/ton ($1.33 MMBTU), 33 percent above a high (3 percent) sulfur coal available for $24/ton ($1.00 MMBTU), a switch from the high sulfur to low sulfur coal is advisable if sulfur emissions abatement is worth at least 19 cents per pound of sulfur removed from the stack gases. Under the assumption for a new plant of $100/kw capital cost for a lime scrubber, 17 percent amortization, 0.8 mills/kwh operating costs (including 0.3 mills/kwh for sludge disposal), and a 6 percent energy loss and capacity derating, flue gas desulfurization using lime scrubbing adds 4.5 mills/kwh to the cost of producing electricity. Flue gas desulfurization becomes preferred to the premium priced low-sulfur coal if sulfur removal is worth at least 37 cents per pound. If low-sulfur coal is not available, flue gas desulfurization is preferred to burning high sulfur coal if the value of emissions reduction is at least 23 cents per pound.

For an existing plant the cross-over value of sulfur removal for switching from high-sulfur coal to low-sulfur coal remains the same, 19 cents per pound of sulfur removed. With the assumptions for a retrofit installation of $125/kw capital cost, 17 percent amortization for the scrubber, 1.1 mills/kwh operating costs (including 0.5 mills/kwh for sludge disposal), and 6 percent energy loss and capacity derating, flue gas desulfurization using lime scrubbing adds 6.1 mills/kwh to the cost of producing electricity. It becomes preferred to premium priced low-sulfur coal only when sulfur removal is worth at least 53 cents per pound. If low-sulfur coal is not available, flue gas desulfurization is preferred to coal preparation if the value of emissions reduction exceeds 26 cents per pound of sulfur removed. In urban or near-urban areas where disposal of sludge is difficult, costs of sludge disposal may be of the order of 0.9 mills/kwh. Lime scrubbing would then not be preferred to low-sulfur coal unless sulfur abatement was worth 59 cents per pound. If low-sulfur coal is not available, lime scrubbing is advisable when sulfur removal is worth at least 28 cents per pound.

For the representative cases considered, low-sulfur eastern coal or flue gas desulfurization appears to be somewhat more cost effective per pound of sulfur removed than use of low-sulfur western coal and coal preparation. However, these latter alternatives may be cost effective in some situations. For new plants in the mideastern region low-sulfur western coal may be an attractive alternative, especially if its cost relative to eastern coal is reduced. Use of low-sulfur western coal in existing plants will generally not be economic because derating or expensive retrofitting will be required. Coal preparation appears barely competitive with flue gas desulfurization if low-sulfur coal is not available: sulfur removal can be accomplished by coal washing at a cost of about 25 cents per pound for an existing plant. Even with sludge disposal assumed to cost 0.5 mills/kwh, a lime scrubber retrofit will be preferred if sulfur removal is worth at least 26 cents per pound of sulfur. Lime scrubbing, moreover, reduces net emissions by about 90 percent compared to about 33 percent for coal washing. The costs and effectiveness of coal washing vary considerably depending on the type of coal. In some situations coal preparation may be an attractive strategy, especially as an interim measure.

The value of the cross-over points are sensitive to the emissions levels and costs of electricity given in Table 13-22. If different values are used the cross over points will change. The cross-over between low sulfur coal and flue gas desulfurization is particularly sensitive: A change of 1 mill per kilowatt-hour in the cost of flue gas desulfurization changes the cross over point by 23.2 cents for a new plant, and 19.8 cents for a retrofit installation. If the comparison is between high sulfur coal and flue gas desulfurization, the sensitivity is rather low: A change of 1 mill per kilowatt-hour in the cost of flue gas desulfurization causes a change in the cross-over point by 5.0 cents for a new plant and 4.3 cents for a retrofit installation. The cross-over point between high sulfur and low sulfur coal changes by 6.3 cents for a 1 mill increase in the cost for low sulfur coal for a new plant, and 5.5 cents for an existing plant. These are both equivalent to 0.57 cents increase in the cross-over point for a 1 cent per million Btu change in the price differential of low sulfur coal over high sulfur coal.

Results of the Analysis for Representative Plants

For both the new and existing plant in a remote rural location the nominal pollution cost is computed to be about 20 cents per pound of sulfur emitted. The range of uncertainty is at least 8 to 40 cents per pound. The decision for both new and existing plants is very close between the alternatives of burning high-sulfur coal without abatement measures and switching to low-sulfur eastern coal. The nominal value of the emissions reduction is just above the marginal increase in cost of electricity incurred by switching to the low-sulfur coal. This marginal cost was assessed as 19 cents per pound of sulfur removed. For the representative urban case, an existing urban plant to be reconverted to coal, the nominal pollution cost is computed to be about 55 cents per pound of sulfur, with a range of uncertainty of at least 19 to 110 cents per pound. For this case the decision very close for an existing plant between eastern low sulfur coal and flue gas desulfurization; the alternative of buring high-sulfur coal appears poor by comparison. If low-sulfur eastern coal is not available, flue gas desulfurization appears to be the best decision. The marginal cost of lime scrubbing (compared to burning high sulfur coal) is 28 cents per pound of sulfur removed with a sludge disposal cost of 0.9 mills/kwh.

An assumption of considerable importance is the value associated with health effects. Willingness by the individuals potentially affected to pay to avoid sickness may not be an adequate standard to judge the value of morbidity caused by air pollution. It is quite possible that health values considerably higher than those used in this analysis ($250 per case of chronic respiratory disease, $20 per day of aggravated heart-lung disease symptoms) will be judged appropriate as the basis for setting public policy on sulfur emissions control. If, for example, values four times those of our nominal assignments were used for the health effects, sensitivity analysis shows that the pollution cost is shifted up to a range where for both the representative rural and urban plants the best decision would be flue gas desulfurization.

The Value of Resolving Uncertainty

The decisions on control strategy depend on the adequacy of the information available at the time the decsion must be made. There is great value to improving our information about certain aspects of sulfur oxide pollution. The value of resolving uncertainty derives from the idea that better information might show, for example, that pollution costs are lower than was estimated, and costly abatement methods are not warranted. With some probability then, their extra cost might be saved. In particular, a better understanding of the health effects of sulfates and of the chemistry of the conversion of sulfur dioxide to atmospheric sulfates could have a significant effect on future decisions on control of sulfur oxides. A rough calculation of the value of resolving these uncertainties gives a value of about $2 million per year for the representative 600 MW plant in the remote rural location. If low-sulfur coal is not available, the value of resolving uncertainty drops to a little over $1 million per year. For the urban location, the value of resolving uncertainty on the sulfur oxide emission to ambient sulfate relationship and on the magnitude of the health effects is in the range of $1 million a year.

Extrapolating these values to the collection of eastern power plants that now or in the near future might burn high sulfur coal yields an estimate of the order of a quarter of a billion dollars per year. This is roughly 25 times the annual cost estimated by EPA for a research program to resolve these uncertainties.

Other areas of our knowledge on the effects of sulfate emissions should be greatly refined. Health effects not included in our analysis might prove far more serious than those identified so far: for example, sulfate might prove to have a causative role in chronic lung diseases such as emphysema, or in lung cancer. In addition to health effects, acid rain, decreases in visibility, materials damage, adverse effects on ecological systems, and possible climatic effects of sulfur emissions all deserve much more extensive investigation than has heretofore been undertaken.

Decisions to be made on sulfur oxide emission from power plants will involve tens of billions of dollars in electrical generation costs in the next decade and massive effects on human health and welfare. Greatly expanded efforts should be made to develop improved models and data for use on a case by case basis to improve decisionmaking on emission control strategy alternatives.

Emissions Charges as an Instrument of Policy

Considerations of strategy with respect to sulfur compounds emission pertain not only to the application of alternative technologies to the problem but to the selection of policy instruments and administrative practices. In this connection an emissions charge appears to be a well suited policy instrument for inducing efficient sulfur emissions control. The application of an emission charge on SOx, perhaps at the level of the estimated incremental cost of the pollution consequences from the average power plant, would provide a strong, immediate, and across the board incentive to undertake emissions controls activities. At the same time, in view of the still existing disagreements about the applicability of particular technologies, the special circumstances of particular plants, supply constraints, and other complexities of the situation, it would permit flexibility of response. This flexible response would be achieved in a decentralized manner without the necessity of administrative agencies and the courts trying to decide every individual case in an adversary atmosphere. The latter approach invites delays and frequently arbitrary decisions, and establishes the incentive to hire lawyers rather than to proceed with emissions control. Emissions charges exert a persistent incentive to act whereas variances and delays in imposing requirements allow the emitter free use of environmental resources, with no incentive to act as long as these can be obtained. Moreover, a charges policy would have desirable efficiency characteristics. It would tend toward an application of controls first at those locations where costs per unit of SOx reduction are lowest. In the longer run it would provide a powerful spur for the development of more efficient technologies.

When first suggested the idea of emissions charges was greeted with some skepticism by many policy makers and environmentally concerned persons. For various reason industry was also opposed. In recent years this policy option has gained increasing acceptance among conservationists, environmentalists, policy makers here and abroad, and even industry as indicated by a recent Committee on Economic Development report. The sulfur emissions problem is a highly suitable one for the applications of emissions charges as a policy instrument.

PART TWO:

Section 1: Relationship of Emissions to Ambient Air Quality and Chemistry of Precipitation

CHAPTER 6

THE RELATIONSHIP OF SULFUR OXIDE EMISSIONS TO SULFUR DIOXIDE AND SULFATE AIR QUALITY

(Chapter 6 was written by John Trijonis under the general supervision of the committee, which reviewed the work at several stages and suggested modifications which have been incorporated. While every committee member has not necessarily read and agreed to every detailed statement contained within, the committee believes that the material is of sufficient merit and relevance to be included in this report.)

Sulfur oxide emissions from man-made sources consist primarily of sulfur dioxide. Both during emission and while in the atmosphere, gaseous sulfur dioxide can become oxidized to form sulfate (SO4)† particulate matter. The sulfate aerosol is mainly composed of sulfuric acid and corresponding salts such as ammonium sulfate. This chapter reviews the relationship between sulfur oxide emissions and ambient air quality levels for sulfur dioxide and SO4.

As illustrated in Figure 6-1, the relationship between sulfur oxide emissions and ambient air quality involves several complex processes. Atmospheric transport and diffusion control the dispersal of the emissions, while chemical oxidation processes lead to the formation of sulfate aerosol from gaseous sulfur dioxide. Removal processes of sulfur dioxide and particulate sulfate include deposition on plants, soil, and water bodies as well as washout by precipitation. The latter leads to further pollution problems associated with increased rainfall acidity.

† Parts of this report were prepared by a computer-assisted text editing process for which the high-speed line printer does not yet provide subscripts or certain special symbols. Two conventions have been adopted as necessary throughout Parts One and Two: (a) for µ read u; (b) numerical subscripts in formulas are indicated by underscoring (e.g., SO4).

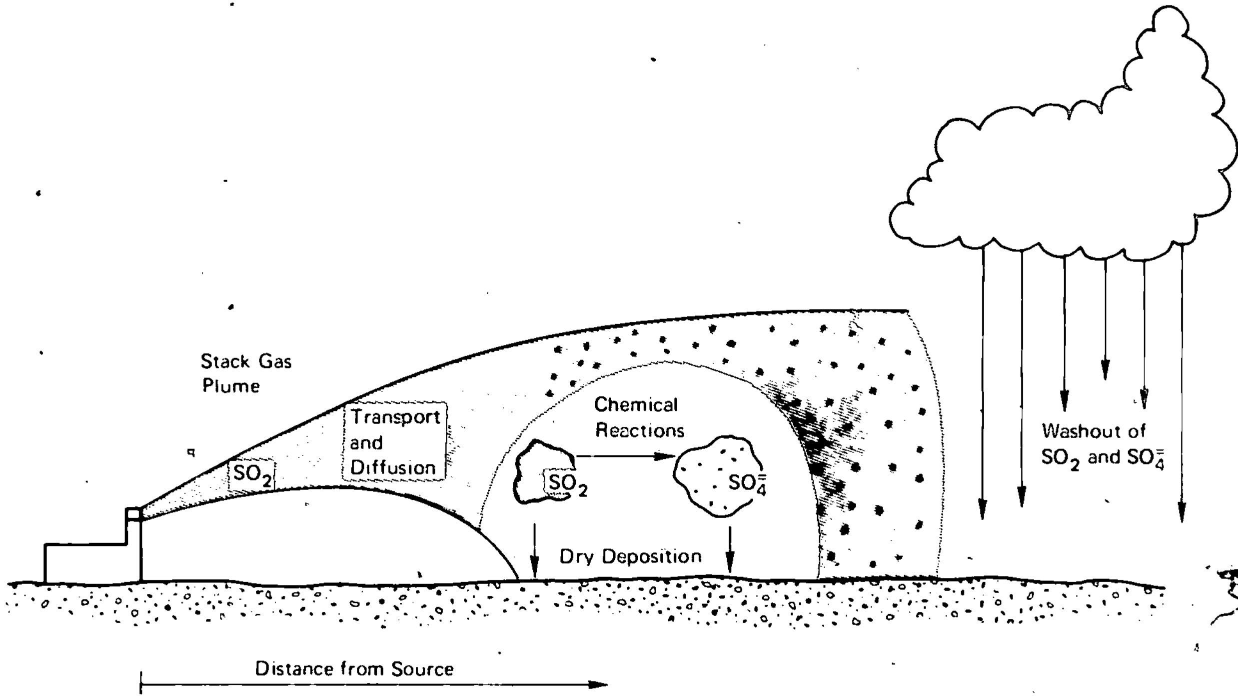

FIGURE 6-1: Processes Involved in the Relationship of Sulfur Oxide Emissions To Air Quality

The relationship between sulfur oxide emissions and air quality will be discussed below in five sections. Section 1 describes the nationwide SOx emission inventory; included are discussions of past trends and geographical features. Section 2 deals with sulfur dioxide air quality and its relationship to SOx emissions. Section 3 describes present ambient sulfate levels and examines the dependence of ambient sulfate formation on SOx emissions. In Section 4, recent trends in sulfur dioxide and SO_4 air quality are analyzed, and comparisons are made to corresponding SOx emission trends. Finally, Section 5 gives an approximate forecast of the air quality impact to be expected from the substantial increases in SOx emissions that have been projected for electric power plants.

SULFUR OXIDE EMISSIONS

The principal natural source of atmospheric sulfur oxides is the oxidation of hydrogen sulfide or dimethyl sulfide gas which results from decaying vegetation (Kellogg et al. 1972, Lovelock et al. 1972). Over the oceans, significant sulfate is also emitted as part of sea spray. Presently, on a global basis, these natural occurrences of atmospheric sulfur compounds are estimated to be about one and one half times the emissions from anthropogenic (man-made) sources (Cavender et al. 1973, Kellogg 1972). However, in industrialized regions, the concentrated emissions from technological processes, in particular fossil fuel combustion, are much greater than natural contributions.

Data on the composition of SOx from combustion and other man-made sources indicate that about 98 percent of emitted SOx is sulfur dioxide. The remaining fraction of typical SOx emissions, about 1 to 2 percent, is sulfur trioxide and its derivatives. The main sulfur trioxide derivative in emission gases is sulfuric acid; metallic sulfates appear to be directly emitted only in trace amounts.

Table 6-1 summarizes the 1972 inventory of man-made SOx emissions for the United States as well as for six selected air basins. On a national basis, more than 75 percent of SOx emissions result from coal and fuel oil combustion in stationary sources, and more than 50 percent of SOx emissions result from fuel combustion in electric power plants alone. Industrial process SOx emissions, mostly from the metallurgical, petroleum, chemical, and mineral industries, are also significant. On a national basis, other stationary sources and transportation sources are relatively minor contributors.

As indicated in Table 6-1, the relative importance of different SOx source types varies considerably from region to region. In the three eastern/midwestern air basins listed in Table 6-1, (Boston, Atlanta, and St. Louis), stationary fuel combustion is especially important, accounting for 83 to 97 percent of total SOx emissions in those areas. Industrial processes and transportation are relatively more significant in the three western air basins (Dallas-Ft. Worth, Denver, and Los Angeles), where they account for 50 to 67 percent of total SOx emissions.

Figure 6-2 presents the geographical distribution of estimated SOx emissions in the United States (EPA 1974). It is evident that these emissions are particularly concentrated in the northeast sector of the country, which accounts for about half the total SOx emissions in the United States.

The trends in the SOx emission inventory are summarized in Table 6-2. From 1960 to 1970, total SOx emissions in the United States increased by 45 percent; about seven eighths of this increase was due to an almost doubling of electric power plant emissions. The remaining portion of the increase basically resulted from growth in emissions from industrial process sources. Total emissions from all other sources have remained essentially constant.

This increase in total SOx emissions was accompanied by significant alterations in the spatial distribution of those emissions. The

TABLE 6-1

Sulfur Oxide Emission Inventories for the United States and for Selected Air Quality Control Regions, (NEDS Data for 1972).

	United States	Boston AQCR	Atlanta AQCR	St. Louis AQCR	Dallas/Ft.Worth AQCR	Denver AQCR	Los Angeles AQCR
Total Sulfur Oxide Emissions (Thousand tons/year)	32,000	332	94.7	1,234	17.3	33.5	238
Percentage of Sulfur Oxide Emissions by Source Category							
Stationary Source Fuel Combustion							
Electric Power Plants	54.3%	41.6%	70.8%	76.2%	3.0%	34.2%	16.8%
Industrial	15.3%	8.2%	5.6%	6.0%	5.0%	10.4%	14.6%
Commercial & Residential	7.1%	48.6%	5.7%	1.9%	19.8%	5.3%	18.8%
Industrial Processes	21.1%	0.5%	12.3%	15.3%	23.7%	40.7%	37.6%
Other Stationary Sources	0.2%	0.1%	0.5%	0.1%	5.3%	0.2%	1.6%
Transportation Sources	2.0%	1.0%	5.1%	0.5%	43.2%	9.2%	10.6%

Source: Reference ( )

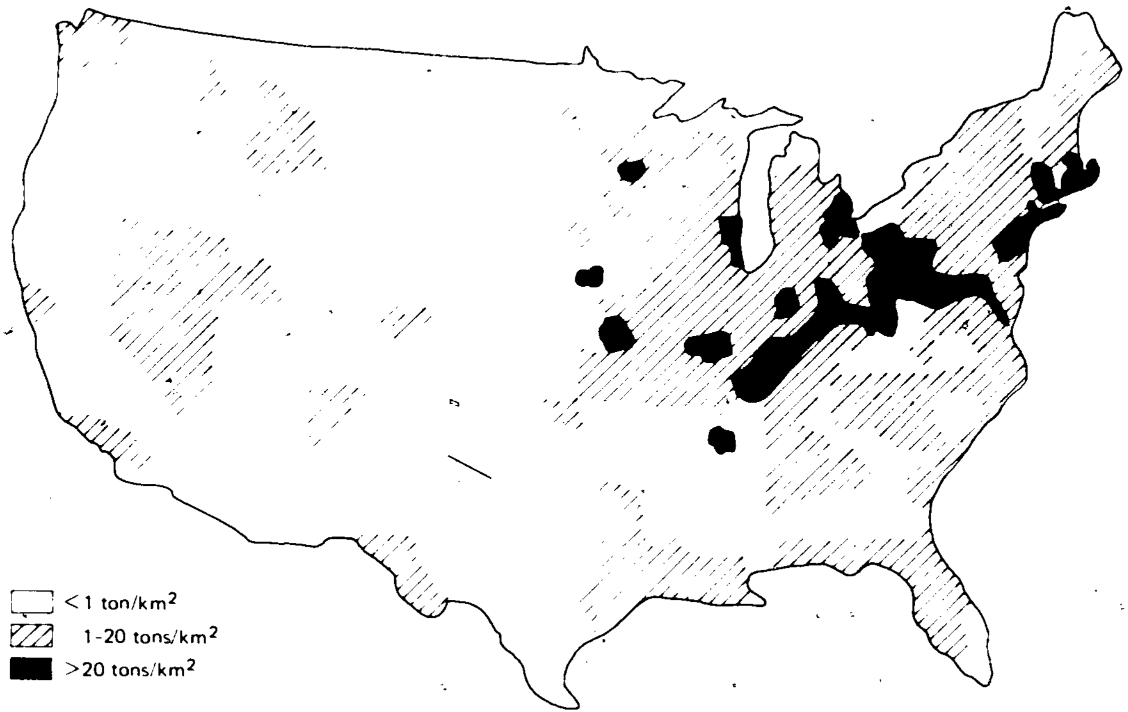

FIGURE 6-2: Nationwide Geographic Variation in SO_2 Emission Density

TABLE 6-2

Sulfur Oxide Emission Trends in the United States, 1960-1970

Source Category	United States Emissions, 1960 (1000 Tons/Year)	United States Emissions, 1970 (1000 Tons/Year)	% Change 1960 to 1970
Stationary Source Fuel Combustion			
Electrical Power Plants	10,100	19,400	+92%
Industrial	4,800	4,890	+ 2%
Commercial and Residential	2,590	2,160	-17%
Industrial Processes	4,720	6,030	+28%
Other Stationary Sources	360	380	+ 6%
Transportation	675	984	+46%
TOTAL	23,300	33,900	+45%

bulk of the increase occurred among sources which are located away from central-city areas and/or which emit pollutants through tall stacks. Further, the growth of ground-level sources tended to occur in a non-homogenous manner; the greatest growth occurred in suburban areas, leading to a spreading of emissions throughout metropolitan regions. Ground-level/central-city emissions appear to have actually been significantly reduced from 1960 to 1970. These changes in the spatial distribution of SOx emissions will be discussed in more detail in Section 4, which deals with ambient air quality trends for sulfur dioxide and sulfates.

With no further abatement programs, SOx emissions are expected to continue increasing in the future. The National Academy of Engineering has predicted a 66 percent national increase from 1970 to 1980 and a 135 percent national increase from 1970 to 1990 (National Academy of Engineering). These increases will again occur predominantly in the power plant category, where SOx emissions are expected to double from 1970 to 1980 and to triple from 1970 to 1990.

SULFUR DIOXIDE AIR QUALITY

Table 6-3 summarizes the present National Ambient Air Quality Standards for sulfur dioxide. Natural background sulfur dioxide levels are well below the air quality standards; measurements indicate that natural background sulfur dioxide concentrations are on the order of 0.5 to 4 $\mu g/m^3$ (Georgii 1970, Cadle et al. 1968, Lodge and Pate 1968). In urban areas, man-made sources lead to sulfur dioxide concentrations which are considerably greater than background levels. In the early 1970's, annual average sulfur dioxide concentrations in urban areas of the United States tended to range from 10 to 80 $\mu g/m^3$ (EPA 1972). In 1970 and 1971, the average level among all urban National Air Surveillance Network (NASN) monitoring sites was about 25 $\mu g/m^3$ (annual average), and only about 2 percent of measured annual averages at NASN urban sites exceeded the national primary

TABLE 6-3

The National Ambient Air Quality Standards for Sulfur Dioxide

Averaging Time	Primary Standard	Secondary Standard
1 year	80 $\mu g/m^3$	---
24 hours	365 $\mu g/m^3$	---
3 hours	---	1300 $\mu g/m^3$

standard. At nonurban NASN monitoring sites, annual mean sulfur dioxide concentrations tend to be around 5 to 15 ug/m³, slightly above estimated natural background levels (EPA 1972).

Figure 6-3 provides an approximate contour map for typical annual average sulfur dioxide levels in urban areas of the United States for 1970-1971. It can be seen that urban sulfur dioxide concentrations are generally higher east of the Mississippi. From 1964 to 1968, the average sulfur dioxide concentration at all eastern urban sites was three times the average at all western urban sites (Altshuller 1973). Particularly high urban sulfur dioxide concentrations are found in the industrialized northeast section of the country. As was illustrated in Figure 6-2, the northeast sector has the greatest SOx emission density.

The Relationship Between Sulfur Dioxide Air Quality and Emission Levels

The relationship between sulfur dioxide air quality and SOx emissions is simple in one sense but complex in another. It is simple in that the ambient sulfur dioxide contribution from a single source tends to vary in direct proportion with the emissions from that source. Thus, for an area dominated by a single source, the linear rollback formula is usually appropriate for relating sulfur dioxide air quality to SOx emission levels. The simple linear rollback formula is also valid for relating ambient sulfur dioxide levels to total emissions from a group of sources, provided that the emissions from all the sources are reduced or increased in proportion to one another. Another, more exact way of stating this proviso is that the temporal and spatial distribution of emissions remain fixed.

In reality, spatial distributions of emissions are altered by relocation of emission sources, by non-homogeneous growth patterns, and by non-proportional emission changes for different types of sources. Herein lies the complexity. When spatial emission patterns change there is no guarantee that the linear

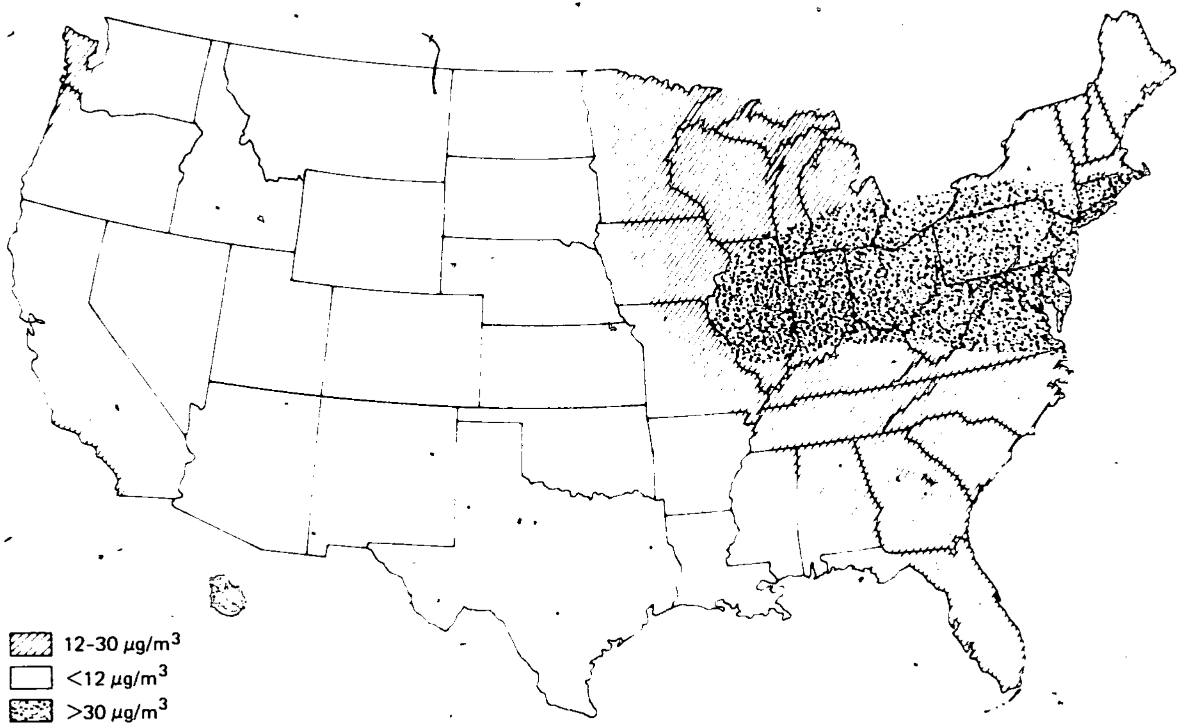

FIGURE 6-3: Geographical Distribution of Typical Annual Urban Ambient Sulfur Dioxide Concentrations

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rollback formula is valid; in fact, it can be grossly in error. For instance, consider an air basin which undergoes an increase in total SOx emissions. Assume that this increase is due to greater emissions from sources with tall stacks and from sources located away from the central-city area. Further assume that ground-level emissions in the central-city area remain constant. In this case, central-city/ground-level air quality will change little in comparison to the total emission increase for the region, i.e., the deterioration in central-city air quality will be considerably less than that which would be predicted by the linear rollback formula.* As will be discussed in Section 4, this hypothetical example has some resemblance to recent sulfur dioxide air quality history which involves an apparent paradox of increasing national SOx emission trends accompanied by decreasing urban sulfur dioxide concentrations.

For cases such as the example above, where significant changes in spatial patterns of emissions occur, mathematical-meteorological models are most appropriate for relating ambient

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*The linear rollback formula is as follows:

$$\frac{C_i(E_1) - C_{bkg}}{C_i(E_2) - C_{bkg}} = \frac{E_1}{E_2}$$

where C_i = the concentration of SO_2 at location "i",

C_{bkg} = the background SO_2 concentration,

and E_1 and E_2 refer to two different emission levels.

sulfur dioxide air quality to emission levels. Several of these models, which account explicitly for source location, sulfur dioxide oxidation rates, sulfur dioxide deposition rates, and regional meteorology, have been developed (Shir and Shigh 1974, Roberts et al., Randerson 1970). It is not necessary here to review these models but rather to point out the type of problem which requires their application.

SULFATE AIR QUALITY

There are two principal methods by which natural sources produce sulfate particulate matter: the formation of sulfate from the oxidation of naturally occurring sulfur dioxide and the emission of sulfate as part of the sea spray aerosol (Kellogg et al. 1972). In remote areas which are not downwind of concentrated man-made sources, sulfate levels tend to average in the range of 1 to 4 $\mu g/m^3$ (Georgii 1970, Junge et al. 1969, Junge 1963). (A major exception involves remote areas in oceanic or sea shore environments which are very near the influence of strong wave action. Close to intensive breaker activity, sulfate levels can average as high as 5 to 20 $\mu g/m^3$. However, this sea salt influence tends to drop off rapidly as distance inland increases. Recent measurements indicate that more normal background levels are reached within a few hundred yards of the breaker activity.)

This range appears to be a reasonable indication of natural background concentrations. However, it should be noted that even remote areas may be significantly influenced by long term transport from anthropogenic sources. Man-made SOx emissions, which are presently estimated to be about two-thirds as great as natural emissions on a global basis, are continually oxidized to form sulfate aerosol; these man-made contributions may constitute a significant fraction of sulfate measurements in remote areas.

Sulfate levels in urban areas are higher than background levels due to the influence of man-made emission sources. During the late 1960's, about 85 percent of measured annual average sulfate levels in urban areas east of the Mississippi ranged from 7 to 20 ug/m³ (Altshuller 1973, EPA 1972, 1971). The average of all eastern urban NASN monitoring sites was 12.5 ug/m³ from 1964 to 1968 (Altshuller 1973). Urban sulfate concentrations in the west are generally about half as great as in the east. During the late 1960's, west of the Mississippi, about 85 percent of annual average sulfate levels in urban areas fell in the range of 3 to 10 ug/m³ (Altshuller 1973, EPA 1972, 1971). The average among all western NASN urban monitoring sites was 6.4 ug/m³ from 1964 to 1968 (Altshuller 1973). One major anomaly in the western United States is the Los Angeles area, where annual average sulfate ranges from 10 to 15 ug/m³ (MacPhee and Wadley, EPA 1972, 1971). As will be discussed later, overall ambient sulfate levels have remained relatively constant in the past decade; therefore, the average concentrations measured in the late sixties are probably indicative of present conditions.

Figures 6-4 and 6-5 present the geographical distributions of typical sulfate concentrations for urban and nonurban locations, respectively (EPA 1974). A significant feature of each map is the particularly high sulfate level found in the industrialized northeast sector of the country which has the greatest SOx emission density (Figure 6-2.)

The Relationship Between Sulfate Air Quality and Sulfur Oxide Emissions

The relationship between emitted SOx and ambient SO_4 concentrations is very complex and is the subject of much current research. Considerable uncertainty exists concerning the present state of knowledge; models are not now available for accurately relating SOx emissions to atmospheric sulfate concentrations. However, the general form of the relationship can be

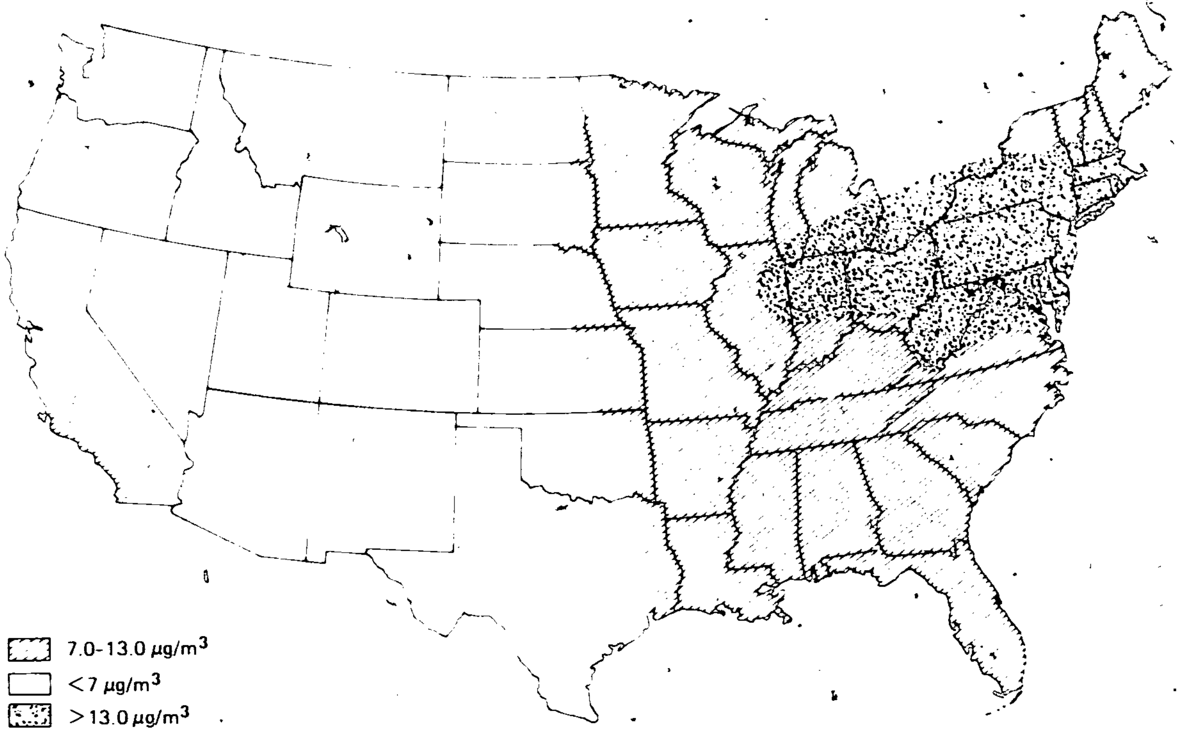

FIGURE 6-4: Geographical Distribution of Typical Urban Sulfate Levels in the United States (Annual Means)

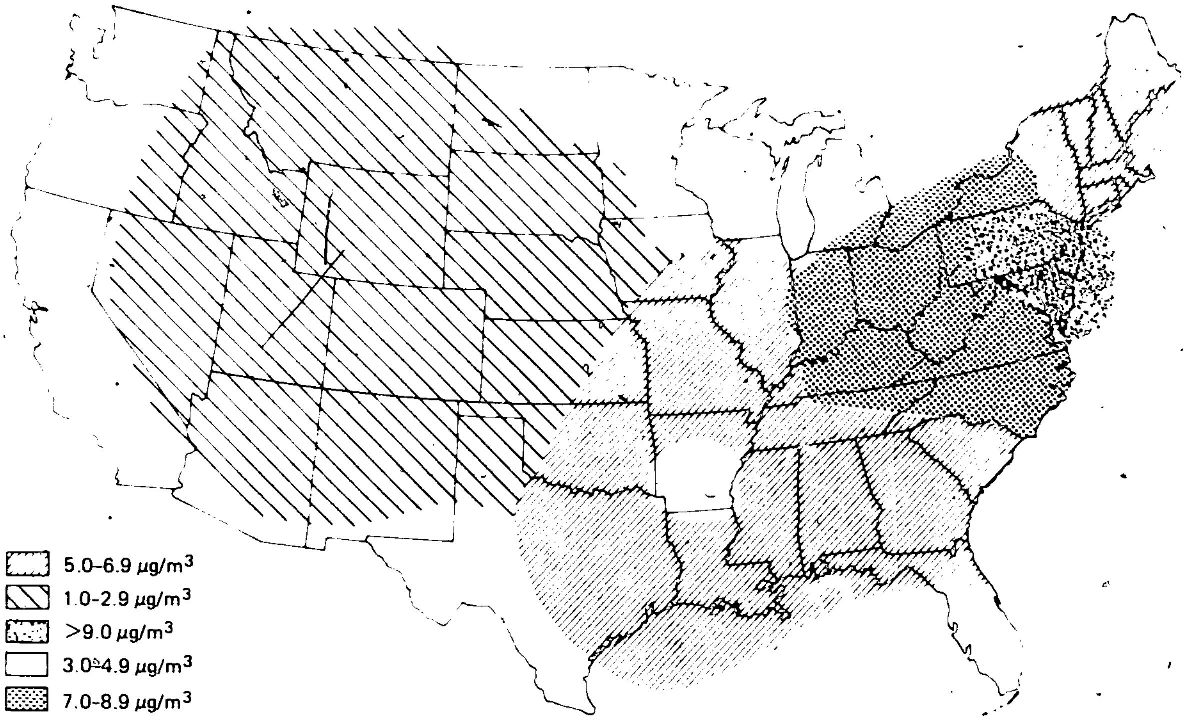

FIGURE 6-5: Geographical Distribution of Typical Nonurban Sulfate Levels in the United States (Annual Means)

analyzed by considering some known aspects of the chemical transformation processes and by examining certain atmospheric empirical evidence.

As noted previously, nearly all (>98 percent) SOx is emitted as sulfur dioxide. A variety of processes exist for oxidizing sulfur dioxide to yield sulfate aerosol. The relative importance of the various possible mechanisms is uncertain and depends heavily on local conditions. However, three general oxidation processes have been identified which appear to be significant for urban areas:

(1) Photochemically-induced oxidation of sulfur dioxide to sulfur trioxide with subsequent reactions yielding sulfuric acid or sulfates.
Irradiation of sulfur dioxide, water, and clean air mixtures yields oxidation rates which are too slow to be significant in urban areas, (Bufalini 1971, HEW 1970, Urone et al. 1968, Gerhard and Johnstone 1955). However, when hydrocarbons and nitrogen oxides are added to such mixtures, substantial oxidation rates can result (Bufalini 1971, HEW 1970, Wilson and Levy 1968, Renzetti and Doyle 1960). It has been postulated that sulfur dioxide is oxidized to sulfur trioxide by an intermediary product of the HC/NOx photochemical reaction system. Once sulfur dioxide is oxidized to sulfur trioxide, subsequent reactions of sulfur trioxide with water (and particulates) yield sulfuric acid and other sulfates.

(2) Absorption of sulfur dioxide into aqueous droplets with subsequent catalytic oxidation.
Numerous substances, particularly certain metallic salts (e.g., of iron, vanadium, or manganese), are effective in catalytically oxidizing sulfur dioxide (HEW 1970, Junge and Ryan 1958). The absorption of sulfur dioxide into water droplets, followed by catalytic oxidation from dissolved metallic salts, can be an important mechanism of sulfate production. In this chemical

mechanism, atmospheric ammonia may play a significant role by neutralizing some of the sulfuric acid built up in the droplet. (Sulfur dioxide becomes less soluble as acidity increases.) (Roberts 1974, HEW 1970).

(3) <u>Adsorption of sulfur dioxide on solid particles and subsequent reaction with adsorped oxygen</u>.

There is evidence that adsorption and subsequent oxidation of sulfur dioxide on particle surfaces may be an important mechanism of sulfate production in urban atmospheres. Carbonaceous (soot) particles as well as metal oxide particulates are potential sites for this process.

From the description of the above processes, it is apparent that several factors are important in determining the degree to which sulfur dioxide will be converted to sulfuric acid and other sulfate aerosols. Significant atmospheric factors include the:

- o presence of HC and NOx
- o presence of catalysts
- o presence of atmospheric ammonia
- o presence of adsorbent particles
- o solar radiation intensity
- o temperature and humidity

Actual measurements indicate that the rate of sulfate formation is very sensitive to these factors. Measured oxidation rates vary from approximately .01 to 50 percent per hour. Another important factor affecting the total amount of sulfate produced is residence time in the atmosphere -- the longer the time for reaction, the greater the conversion of sulfur dioxide to sulfate.

As to the dependence of sulfate on sulfur dioxide, it appears that at <u>low</u> sulfur dioxide concentrations all three of the chemical reaction mechanisms are such that the rate of sulfate production is directly proportional to sulfur dioxide concentrations. For given values of the other relevant parameters and for a given residence time, the amount of sulfate yield would be directly proportional to sulfur dioxide input. However, at <u>higher</u> sulfur dioxide concentrations, there are reasons to expect that sulfate yield would become less than proportional to total sulfur dioxide input. First of all, at higher sulfur dioxide levels, insufficient ammonia may be present in the ambient air to prevent aqeous droplets from becoming strongly acidic. Since sulfur dioxide is less soluble in acid solutions, the depletion of ammonia would reduce the rate of sulfate production from process (2) above (Roberts 1974, Junge and Ryan 1958). Second, theoretical analyses of the gaseous reactions in mechanism (1) have indicated that the relative rate of oxidation should decrease at higher sulfur dioxide levels (Roberts 1974, Cox and Penkett 1972). Third, process (3) may yield non-linear results at high sulfur dioxide levels due to saturation of adsorption sites (Roberts 1974).

The implications of the above discussion of the chemical reaction mechanisms are summarized in Figure 6-6a, which provides a qualitative illustration of the dependence of sulfate levels on sulfur dioxide input for fixed residence time and other parameters. At low sulfur dioxide levels, the dependence should be nearly linear. At higher sulfur dioxide levels, nonlinear saturation effects take hold and the relationship becomes less than proportional. The sulfur dioxide level at which nonlinearities set in is not known; this critical level will vary from case to case and will depend on the key parameters described previously (e.g., amounts of NH_3, HC, NOx present, adsorbent surface, etc.).

Actually, in a real air basin one would expect a slightly modified picture. A background sulfate level will exist due to natural sources as well as to man-made emissions external to the region in question. Thus, the effect of man-made sulfur dioxide emissions in a region on sulfate levels for that region should be as depicted in Figure 6-6b. Figure 6-6b is similar to Figure 6-6a except that sulfate levels reduce to a finite background level when sulfur dioxide emissions are eliminated.

Atmospheric data appear to be consistent with the above discussion of the dependence of sulfate on sulfur dioxide. Altshuller analyzed NASN sulfate and sulfur dioxide data from 18

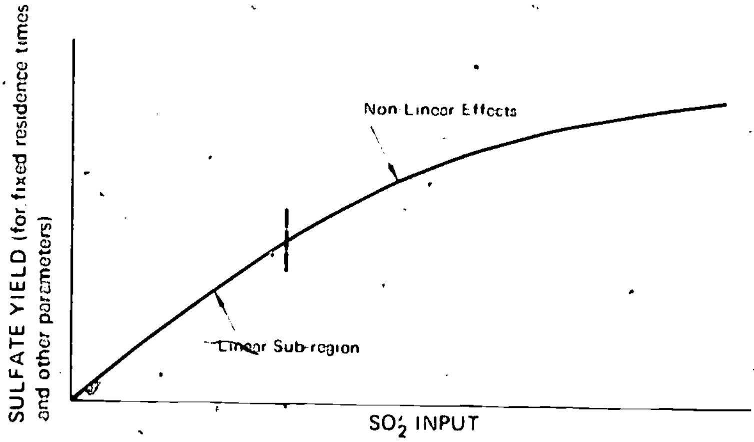

FIGURE 6-6a: Hypothetical Experiment

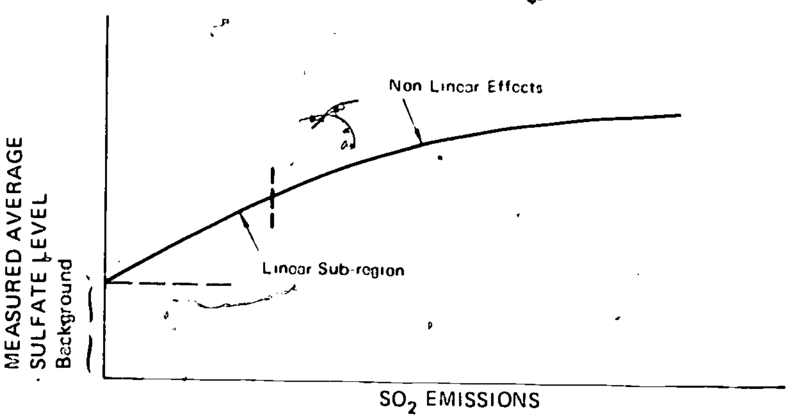

FIGURE 6-6b: Hypothetical Air Basin

urban monitoring sites for the period 1964 to 1968. He found that annual average sulfate levels initially increased linearly above mean background concentrations when plotted against annual average sulfur dioxide, but that sulfate formation levelled off at higher sulfur dioxide levels. A similar nonlinearity was observed in a recent study of Los Angeles data (Trijonis et al. 1974). However, these aerometric results do not necessarily confirm the theory that basic non-linearities exist in the relationship of sulfate yield to sulfur dioxide input, since alternative explanations have been suggested for the appearance of non-linearities in the atmospheric data (Frank 1974).

One further point should be mentioned concerning the relationship of sulfate air quality to SOx emissions. The above discussion of the dependence of sulfate on SOx emissions is appropriate to the case when all emission sources in a region are altered in the same proportion (i.e., when the relative spatial distribution of emissions remains fixed). Urban sulfate levels may not respond to total SOx emissions in a consistent manner if the spatial distribution of emissions is changed. It may be possible to increase total SOx emissions in a region but yet decrease urban sulfate concentrations (or alternatively to decrease total SOx emissions but increase sulfate), by changing the spatial pattern of emissions. Generally, for urban regions with significant photochemical smog activity or with high concentrations of atmospheric catalysts (e.g., from metallurgical industries), local SOx sources are much more important, on a per ton basis, than distant SOx sources to local sulfate air quality. This is because high local oxidation rates lead to considerable sulfate production from local SOx emissions. For regions with very low oxidation rates, e.g., regions with little photochemical smog and low levels of atmospheric catalysts, distant sources could be more important, on a per ton basis, than local sources to sulfate air quality.

One aspect of SOx emission distribution deserves special note. SOx emissions from stacks are probably more important than ground level SOx emissions to sulfate levels some distance downwind of the source area. SOx emissions from ground level sources encounter more initial interaction with plants and soil; thus, a good bit of the sulfur dioxide is lost by dry deposition before sulfate can be formed. Research has not yet delineated the general significance of this factor. In any case, before this factor can be used as an argument against tall stack control, it should be remembered that sources which are suitable for tall stacks usually will be fitted with some sort of stack; the dry deposition factor is most relevant in comparing ground level sources vs. stack sources and may not be very significant in comparing medium stacks vs. tall stacks (Gifford 1975; Bolin et al. 1974).

Mathematical-meteorological models which can accurately assess the impact of changes in spatial distribution of SOx emissions on sulfate air quality are only in the initial stage of development, (Altshuller 1974, Mills 1974). Reliable models of this type do not now exist. One of the main difficulties involved in formulating these models is the uncertainty that exists concerning sulfur dioxide oxidation rates under various atmospheric conditions. Uncertainty also exists concerning pollutant deposition to the ground and dispersal at the top of mixing layers.

AMBIENT AIR QUALITY TRENDS FOR SULFUR DIOXIDE AND SULFATE

Figure 6-7 summarizes recent trends (1964 to 1971) in ambient sulfur dioxide concentrations at urban monitoring sites in the United States (EPA 1973). Table 6-4 provides the trend in terms of the number of violations of the annual Federal primary air quality standard for sulfur dioxide (EPA 1973). It is evident that substantial improvement in sulfur dioxide air quality has occurred at urban monitoring sites from 1964 to 1971. The average sulfur dioxide level for urban NASN stations decreased by about 50 percent, and the number of stations violating

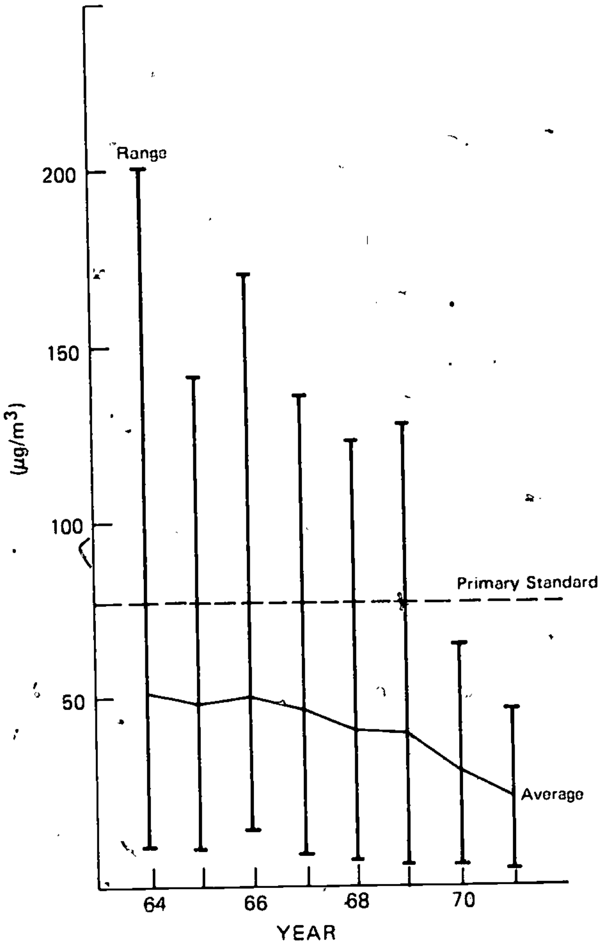

FIGURE 6-7: Average Sulfur Dioxide Concentrations at 32 Urban NASN Stations

TABLE 6-4

Number of NASN Stations Exceeding Primary and Secondary Annual Mean Standards for Sulfur Dioxide, 1964-1971

	1964	1965	1966	1967	1968	1969	1970	1971
Total Number of Stations	18	17	15	29	74	88	86	54
Number > Primary Standard (80 μg/m^3 Annual Mean)	6	6	5	6	10	6	3	0
Percent > Primary Standard	33	35	33	21	14	7	3	0

the national primary standard has dropped from about one-third of the total to zero.

Sulfate trends for urban NASN monitoring sites are summarized in Figure 6-8 (EPA 1973). This figure does not indicate any discernable long term trend in urban sulfate concentrations. A recent statistical study of NASN data (Frank 1974) has confirmed the conclusion that urban sulfate levels have remained essentially unchanged from 1964 to 1970.

The data on SOx emission trends (Table 6-2) and on sulfur dioxide and sulfate ambient air quality trends (Figures 6-7 and 6-8) present an apparent paradox. During the 1960's, total SOx emissions increased by 45 percent on a nationwide basis. In contrast, urban sulfur dioxide concentrations decreased by about 50 percent while average urban sulfate levels remained constant during this period.

A more in-depth analysis of changes in SOx emission during the 1960's provides a plausible explanation for these trends. Although total SOx emissions increased, there was also a significant shift in the spatial distribution of emissions. The SOx emission increase occurred primarily at large point sources, about seven-eighths of the increase being from electric power plants and about one-eighth from industrial process sources. Many of these facilities are located outside central-city areas and/or are equipped with tall stacks. Those large sources which were situated in central-city areas tended to be most affected by air pollution control policies; they either switched to cleaner fuels, relocated, or phased down their operating levels (Frank 1974, EPA 1973). Thus, the SOx emission increase tended to occur at facilities with large stacks and in locations away from central-city areas. In fact, emissions from point sources in urban areas might actually have decreased. Furthermore, the most significant ground-level sources of SOx emissions in urban areas, (residential and commercial sources) exhibited a significant decrease in SOx emissions.[3]

Central-city/ground-level emissions are substantially more important to central-city/ground-level ambient air quality than are

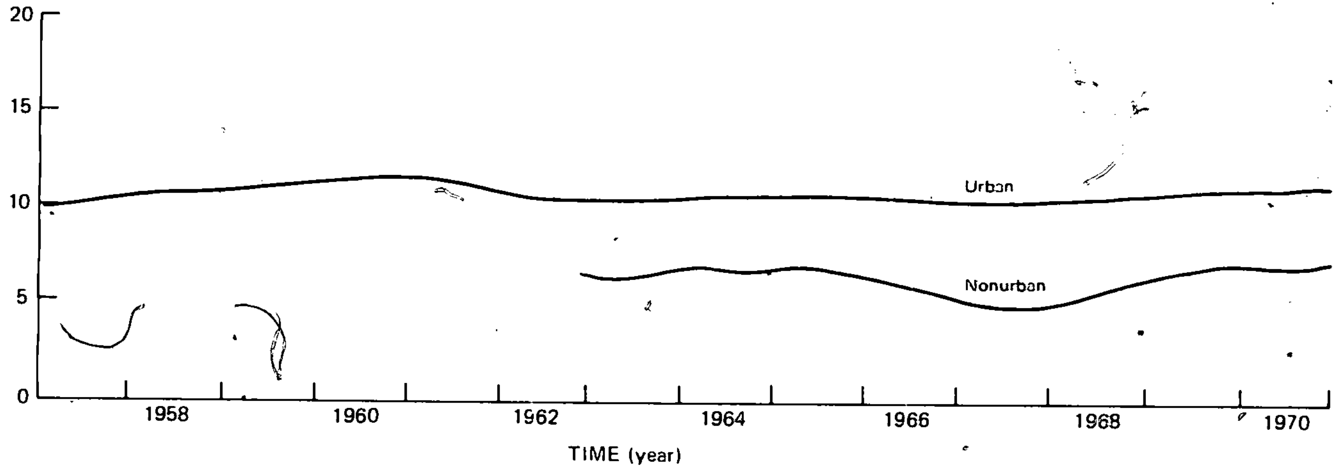

FIGURE 6-8: Long-term Sulfate Patterns in the United States

rural and/or tall stack emissions. Modeling studies, as well as some aeormetric data, indicate that (on a per ton basis) ground level emissions are typically about 5 to 10 times more important (EPA 1973, Wiedersum and Barr 1973, Woodburg 1972, Golden and Morgan 1971, National Air Pollution Control Adm. 1970). The relative importance (to central-city sulfur dioxide air quality) of central-city emissions compared to rural emissions is probably much greater. With these factors in mind, it seems reasonable to expect that reductions in urban SOx emissions could lead to significant improvement in central-city/ground-level sulfur dioxide air quality in spite of an increase in total, nationwide SOx emissions. Apparently this is what has happened.

The absence of significant urban sulfate trend can be accounted for in a manner consistent with the above discussion. Since sulfur dioxide emissions continually oxidize to form sulfate, distant sources are relatively more important for determining ambient sulfate levels than they are for determining ambient sulfur dioxide contributions.

An indication that long-term transport is more important for sulfate than for sulfur dioxide is that sulfate concentrations at non-urban sites are often greater than sulfur dioxide concentrations at those sites. This is particularly evident in the northeastern United States, where non-urban sulfate levels range from 6 - 12 ug/m³. The non-urban air quality picture is in contrast with the urban situation, where sulfur dioxide concentrations are typically 4 to 5 times greater than sulfate concentrations.

It has been estimated that distant sources are about one-half as important (on a per ton basis) as urban, ground-level sources to formation of urban, ground-level sulfate (EPA 1974). This estimated relative importance for urban sulfate air quality is significantly greater than the relative importance attached to the contribution of distant emissions to urban sulfur dioxide air quality. For urban sulfate formation, it is quite possible that the increase is SOx emissions from tall stacks and/or from rural areas was enough to negate the decrease in central-city/ground-level SOx emissions; thus urban sulfate levels remained relatively constant.

Another important factor in explaining the absence of an increase or decrease in ambient sulfate concentrations may be the nonlinearities involved in the sulfur dioxide/sulfate relationship. As mentioned in the previous section, sulfate should increase proportional to the increase in sulfur dioxide ambient concentrations at low sulfur dioxide levels. However, at higher sulfur dioxide concentrations, sulfate formation may level off due to saturation effects which tend to cause sulfate production to be insensitive to variations in sulfur dioxide levels.

Ambient Non-urban Trends

It was suggested above that trends in urban concentrations of sulfur dioxide and sulfate could be explained by the recent increase in rural, tall stack SOx emissions, accompanied by a concomitant decrease in central-city/ground-level SOx emissions. In order to further support this hypothesis, it would be useful to examine ambient non-urban sulfur dioxide and sulfate trends.

Unfortunately, sulfur dioxide data from non-urban locations are extremely sparse. Yearly sulfur dioxide summaries from NASN data are typically available only for about 5 to 7 non-urban locations, and the locations reporting information often differ from year to year (EPA 1972, 1972, 1971). A recent report on ambient sulfur dioxide trends concluded that sulfur dioxide data at non-urban stations are too scarce to justify formal analysis (EPA 1973).

Considerable data are available on non-urban sulfate concentrations. Appendix A presents the annual average sulfate levels measured at non-urban sites from 1960 to 1970. These data have been interpreted as indicating that non-urban sulfate levels have remained relatively constant during the 1960's. This interpretation is based on the non-urban sulfate line in Figure 6-8.

However, Figure 6-8 is a smoothed trend line fit to the averages of all non-urban NASN measurements. As indicated in Appendix A, a considerable number of new non-urban monitoring sites were added in the mid-1960's; thus, the non-urban trend line in Figure 6-8 represents different sets of locations during different years. In order to examine trends in a rigorous manner, the measurements from year to year should be examined for the same set of locations.

Figures 6-9a and 6-9b summarize average non-urban sulfate concentrations for fixed sets of locations during the 1960's. Figure 6-9a represents nationwide data, while figure 6-9b includes northeastern locations only. Data are sufficient to establish average non-urban trends from 1962 to 1970 at only 9 locations nationwide, and 7 locations in the northeast. Non-urban sulfate trends can be examined at 27 nationwide locations (including the aforementioned 9 sites) and 11 northeastern locations (including the aforementioned 7 sites) for the years 1965 to 1970.

Although the non-urban sulfate averages fluctuate from year to year, Figures 6-9a and 6-9b do indicate a general increase in sulfate concentrations, both nationwide and in the northeast. This result is in contrast with the non-urban sulfate data in Figure 6-8, which did not indicate a distinct trend. In Figure 6-8, the increasing non-urban sulfate trend was apparently disguised by the addition of data in the mid-60's from new monitoring sites which had lower average sulfate levels than the locations which provided data during the early 1960's.

The data in Figures 6-9a and 6-9b are not conclusive enough to support firm, quantitative analysis. However, if one accepts the validity of the upper (9 and 7 station) trend lines and assumes an average natural background level of 1 to 4 ug/m³, it appears that man-made sulfate has increased by about 40 to 100 percent in non-urban areas during the 1960's. This increase is qualitatively in agreement with the trends in SOx emissions which were discussed earlier.

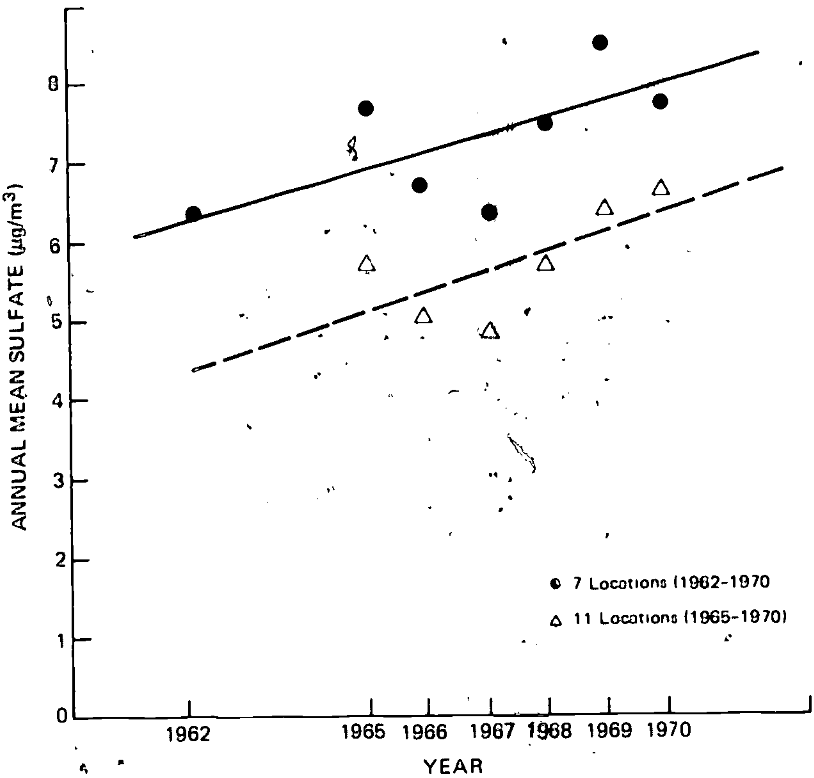

FIGURE 6-9a: Nonurban Sulfate Trends in the United States

(NASN Data, See Appendix A)

*Prior to 1965, data are sufficient to establish trend: only for 9 locations

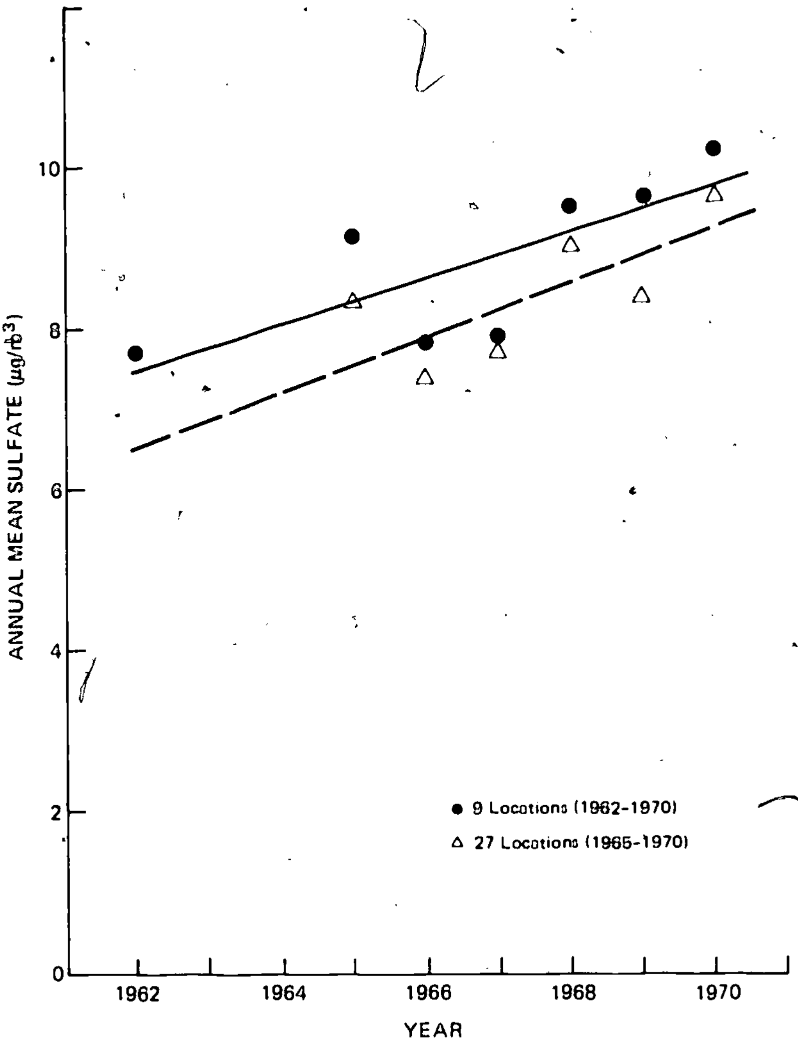

FIGURE 6-9b: Nonurban Sulfate Trends in the Northeastern United States

THE AIR QUALITY IMPACT OF PROJECTED INCREASES IN POWER PLANT EMISSIONS

As noted in Section 1 of this paper, recent forecasts indicate that power plant SOx emissions will double between 1970 and 1980, assuming that no new emission control programs are initiated. This section will examine the likely impact on sulfur dioxide and sulfate air quality to be expected if this increase in emissions is allowed to happen. The projected SOx emission increase will tend to occur at power plants which are located outside of central-city areas and/or which are equipped with tall stacks. Little deterioration in urban sulfur dioxide air quality would be expected form such an SOx emission increase. However, the increase in sulfate concentrations could be very significant.

A thorough and systematic investigation of the air quality impact associated with increased SOx emissions from power plants should be carried out on a <u>region by region</u> basis. Mathematical-meteorological models should be utilized to calculate changes in sulfur dioxide air quality due to increased total SOx emissions as well as to shifts in the spatial distribution of those emissions. Although sophisticated meteorological models are not available for explaining the effect on sulfate levels due to changes in SOx emissions, simplified quantitative analyses should be performed for sulfates which take into account local background levels and significant features of the spatial distribution of emissions. Such a detailed analysis is not possible within the time frame of this study; here, only a very approximate estimate will be made of the total <u>nationwide</u> impact on sulfur dioxide and sulfate levels.

For the purposes of this discussion, it will be assumed that power plant SOx emissions will double from 1970 to 1980 and that SOx emissions from other sources will remain constant. This hypothetical situation is summarized in Table 6-5. It will also be assumed that the increase in power plant SOx emissions will occur at plants which are equipped with tall stacks and/or which

TABLE 6-5

The Hypothetical National SOx Emission Increase to be Evaluated Here

	1970 Emissions (1000 tons/year)	1980 Emissions (1000 tons/year)	
Power Plants	19,000	38,000	(+100%)
Other Sources	14,000	14,000	
Total	33,000	52,000	(+58%)

TABLE 6-6

Urban SO_2 Air Quality Projections

	1970	1980	Change
A. Power Plant SOx Emissions (tons/year)	19×10^6	38×10^6	
B. Other SOx Emissions (tons/year)	14×10^6	14×10^6	
Factor: 0 Weighted Emissions (0 A + B)	14×10^6	14×10^6	(0%)
Factor: 1/5 Weighted Emissions (1/5 A + B)	17.8×10^6	21×10^6	(+21%)

Note that natural background SO_2 concentrations are neglected in these SO_2 air quality forecasts. This is reasonable since average natural background levels of SO_2 are much smaller than average urban SO_2 concentrations.

are located outside contral-city areas. As noted earlier, these "distant" emissions tend to be less important to urban/ground-level air quality than emissions from urban/ground-level sources. The impact on urban air quality will be calculated here by assigning relative importance factors to power plant emissions compared to emissions from other sources.

TABLE 6-5. THE HYPOTHETICAL NATIONAL SOx EMISSION INCREASE TO BE EVALUATED HERE

	1970 Emissions	1980 Emissions
Power Plants	19,000	38,000 (+100%)
Other Sources	14,000	14,000
Total	33,000	52,000 (+58%)

On a per ton basis, urban/tall stack SOx emissions are about 1/5 or 1/10 as important to urban/ground-level sulfur dioxide concentrations as are emissions from urban/ground-level sources, (see discussion in previous section). Also, on a per ton basis, rural SOx emissions are of essentially negligible importance to urban sulfur dioxide air quality as compared to urban SOx emissions. With these considerations in mind, it appears reasonable to assign a range of near 0 to 1/5 for the relative importance factor of power plant SOx emissions vs. other SOx emissions to urban/ground-level sulfur dioxide air quality. Using this range, the forecasted increase in national urban/ground-level sulfur dioxide concentrations from the projected increase in power plant SOx emissions would be expected to range from near 0% up to 20%. (See Table 6-6).

The relative importance of "distant" SOx emission sources as compared to urban/ground-level SOx sources to urban/ground-level sulfate depends a great deal on regional characteristics. For urban areas with substantial photochemical smog activity or with high concentrations of atmospheric catalysts (e.g., from metallurgical industries), local SOx sources are much more important to resulting local sulfate air quality on a per ton basis, than are distant SOx sources. In these urban areas, high local oxidation rates lead to considerable sulfate formation from local SOx emissions, and, the relative importance factor for distant emissions would be low (Finklea, personal conversation 1974). In the opposite situation, for urban areas with very low oxidation rates due to the absence of both photochemical activity and atmospheric catalysts, distant SOx sources can be of equal or greater importance to resulting sulfate air quality on a per ton basis than local SOx sources (Finklea, Personal Conversation 1974). In such cases, the relative importance factor for distant SOx emissions sources could reach (or exceed) one.

Recent EPA studies of trends in SOx emissions and sulfate air quality for different regions have shed some light on the issue of relative importance of distant vs. local sources to sulfate air quality (Finklea, personal conversation 1974). From the results of these studies, it would appear that the average nationwide relative importance factor for distant sources compared to local sources, on a per ton basis, lies between 1/4 and 1. It is assumed here that this range is appropriate in comparing power plant SOx emissions to SOx emissions from other sources. By accepting this range, by accounting for natural background sulfate concentrations, and by using a linear relationship of ambient sulfate concentrations, to SOx emissions, the forecasted increase in urban sulfate levels due to the projected doubling of power plant emissions would be +20 percent to +40 percent. (See Table 6-7). As noted in Section 3, the increase in sulfate may be less than this due to nonlinearities in the relationship of sulfate to sulfur dioxide. To allow for uncertainty concerning the non-linear effect, the lower bound of the estimated urban sulfate increase should be reduced. A predicted urban sulfate increase in the range of 10 percent to 40 percent appears reasonable.

To summarize, we have examined the impact on urban sulfur dioxide and sulfate air quality to be expected from a hypothetical doubling of

TABLE 6-7

Urban Sulfate Air Quality Projections

	1970	1980	Change
A. Power Plant SOx Emissions (tons/year)	19×10^6	38×10^6	
B. Other SOx Emissions (tons/year	14×10^6	14×10^6	
Factor: 1/4 Weighted Emissions (1/4A+B)	18.75×10^6	23.5×10^6	(+25%)
Factor: 1 Weighted Emissions (1 A+B)	33×10^6	52×10^6	(+58%)

Average 1970 urban sulfate concentrations were 10 $\mu g/m^3$, of which about 3 $\mu g/m^3$ was natural background. The weighted emission changes calculated above apply only to man-made contributions. Thus, accounting for the natural background levels, the total sulfate changes would be

$$\frac{(1.25\text{x}8+3) - 11}{11} = +18\% \text{ for the "1/4" factor}$$

and

$$\frac{(1.58\text{x}8+3) - 11}{11} + +42\% \text{ for the "1" factor}$$

power plant SOx emissions. Properly, such an analysis should be carried out on a region by region basis, using available models for sulfur dioxide and formulating simple quantitative models for sulfate. Because of time limitations, only a very approximate estimate of nationwide air quality changes is presented here. This estimate indicates that the expected increase in nationwide urban sulfur dioxide concentrations would be in the range of 0 percent to 20 percent, while the likely increase in nationwide urban sulfate concentrations would range from 10 percent to 40 percent.

TABLE 6-8

NASN Nonurban Annual Arithmetic Mean Sulfate Concentrations $\mu g/m^3$

	CITY	SITE	STATE	'60	'61	'62	'63	'64	'65	'66	'67	'68	'69	'70
	Wrangell-Petersburg Ed	001	Alaska			0.4								
ab	Grand Canyon Nat Park	001	Arizona			1.9			2.2	2.0	0.9	1.7	8.3	2.9
a	Montgomery Co	001	Arkansas							3.8	3.6	3.9	7.1	5.4
a	Humboldt Co	001	California						3.7	2.3	2.4	3.0	4.9	3.3
	Mesa Verde Nat Park	002	Colorado										1.8	3.2
abcd	Kent Co	001	Delaware					8.7	9.5	10.1	10.9	9.4		
	Hardee Co	001	Florida										3.9	5.6
	Monroe Co	001	Florida			3.0								
a	Butte Co	001	Idaho						2.4		0.7	1.1	2.0	2.1
ac	Monroe Co	001	Indiana							6.8	6.2	9.7	7.9	7.5
abcd	Parke Co	001	Indiana			9.2			8.0	7.9	5.5	6.9	8.6	13.5
	Porter Co	001	Indiana						6.5					
	Porter Co	004	Indiana						7.8					
	Delaware Co	001	Iowa			7.3			5.3					
abcd	Acadia Nat Park	001	Maine			6.6				5.4	6.2	5.8	4.9	7.0
abcd	Calvert Co	001	Maryland			7.9			11.6	8.0	8.0	14.1	9.8	
a	Jackson Co	001	Mississippi						5.7	5.2	5.8	5.6		
a	Shannon Co	001	Missouri						5.3	4.7	4.9	4.7		
	Shannon Co	002	Missouri										6.6	5.7
ab	Glacier Nat Park	001	Montana			0.9			3.4	1.4	1.3	1.4	1.7	
a	Thomas Co	001	Nebraska						3.5	2.1	1.5	1.7	4.9	2.2
a	White Pine Co	001	Nevada						3.8	1.7	0.5	1.2	2.3	2.3
ac	Coos Co	001	New Hampshire						6.1	5.9	5.9	7.3	3.4	6.5
	Rio Arriba Co	001	New Mexico						2.4	2.0	0.9			
abcd	Jefferson Co	001	New York			6.0			10.0	6.5	7.3	10.0	9.1	9.4
a	Cape Hatteras Nat Park	001	North Carolina						9.2	8.4	6.6	9.4	8.1	11.9
a	Cherokee Co	001	Oklahoma						4.3	4.8	4.4	3.4	5.6	9.9
a	Curry Co	001	Oregon						3.8	3.6	2.6	3.8	2.8	4.8
ac	Clarion Co	001	Pennsylvania						9.0	7.5	9.7	9.8	9.1	12.4
abcd	Washington Co	001	Rhode Island			7.1			12.6		10.1	10.5		
	Washington Co	002	Rhode Island										9.1	7.7
a	Richland Co	001	South Carolina							5.4	4.1	7.4	7.8	
a	Black Hills Nat Park	001	South Dakota						2.2	2.2	1.4	0.9	1.9	
	Cumberland Co	001	Tennessee										6.8	8.7
a	Matagorda Co	001	Texas						3.7	3.6	4.4	3.8	9.8	6.5
	Tom Green Co	001	Texas											
ac	Orange Co	001	Vermont						6.2	5.9	8.3	7.5	5.8	8.0
abcd	Shennandoah Nat Park	001	Virginia			8.3			6.1	5.7	6.2	8.1	13.3	9.2
	Wythe Co	001	Virginia										7.6	8.7
	King Co	002	Washington											2.6
	Door Co	001	Wisconsin										4.0	
a	Yellowstone Nat Park	001	Wyoming						1.7	1.8	1.0	1.5	2.6	

a. Included in 27 sites for nationwide trends for 1965-1970.

b. Included 9 sites for nationwide trends for 1962-1970.

c. Included in 11 sites for northeastern trends for 1965-1970.

d. Included in 7 sites for northeastern trends for 1962-1970.

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CHAPTER 7

SULFATES AND ACIDITY IN PRECIPITATION: THEIR RELATIONSHIP TO EMISSIONS AND REGIONAL TRANSPORT OF SULFUR OXIDES

(Chapter 7 was written by Ian Nisbet under the general supervision of the committee, which reviewed the work at several stages and suggested modifications which have been incorporated. While every committee member has not necessarily read and agreed to every detailed statement contained within, the committee believes that the material is of sufficient merit and relevance to be included in this report.)

INTRODUCTION

The principal route by which sulfates are returned to earth is in precipitation (Kellogg et al. 1972, Rodhe 1972, Robinson and Robbins 1968, Eriksson 1963, Bolin et al. 1971). Hence--with certain important limitations to be discussed below--the occurrence of sulfates in precipitation can provide useful information about the distribution and deposition of sulfates. Such information is needed to complement the somewhat limited data provided by direct measurements of ambient concentrations of suspended sulfates. This chapter of the report summarizes observations of sulfates in precipitation, and uses them to amplify the evidence given in Chapter 6 for regional transport of sulfur oxides and a secular increase in sulfate concentrations. This section also summarizes measurements of the acidity of precipitation, which has attracted much attention in recent years, both in North America (Likens et al. 1972, Likens and Bormann 1974, Cogbill and Likens 1974, Likens 1972) and in Europe (Bolin et al. 1971, Oden 1968, Brosset 1973).[1] The acidity of rainfall is not only of importance in itself, because of its known or suspected effects on biological systems (Chapter 5), but can give indirect information--otherwise lacking-about the acidity of the suspended sulfates.

THE SULFUR CYCLE AND SULFATE DEPOSITION

The occurrence of sulfates in rain and snow must be interpreted in the context of the sulfur cycle in nature (Kellogg et al. 1972, Rodhe 1973, Robinson and Robbins 1968, Eriksson 1963, Bolin et al. 1971). Sulfur is introduced into the atmosphere by three principal routes: in spray from ocean waves (primarily in the form of neutral sulfates); by decomposition of biological materials (primarily in the form of hydrogen sulfide); and by combustion of fossil fuels (primarily in the form of sulfur dioxide). After transport and oxidation, it is returned to earth by four principal routes:

(a) absorption of gaseous sulfur dioxide by the soil or vegetation;
(b) deposition of sulfur dioxide in rain or snow;
(c) deposition of sulfates (including sulfuric acid) in rain or snow;
(d) dry deposition of particles containing sulfates.

Studies of the sulfur cycle (Kellogg et al. 1972, Rodhe 1972, Robinson and Robbins 1968, Eriksson 1963, Bolin et al. 1971) suggest that (a) and (c)--direct absorption of sulfur dioxide and deposition of sulfates in precipitation--are the most important routes of deposition. Concentrations of sulfur dioxide in precipitation appear to be generally low in comparison with those of sulfates (Miller and de Pena 1972, Dana et al. 1973), and suspended sulfates are generally on small particles which are deposited very slowly (Garland 1974).

On a global basis, deposition of sulfates in precipitation is estimated to account for 40-80 percent of the sulfur deposited on land; direct absorption of sulfur dioxide accounts for much of the rest (Kellogg et al. 1972, Rodhe 1972, Robinson and Robbins 1968, Eriksson 1963, Bolin et al. 1971). According to the models of sulfur transport and deposition outlined in Chapter 6 and in Chapter 13 (Appendix A), direct absorption will be relatively important in circumstances where the oxidation rate of sulfur dioxide to sulfates is low (especially in dry, unpolluted air). Deposition of sulfates will be more important where the oxidation rate of sulfur dioxide is high, or far downwind from sources where ambient concentrations of sulfur dioxide are low.

Two principal processes lead to the occurrence of sulfates, including sulfuric acid, in precipitation: (a) the absorption of sulfur dioxide into cloud droplets, with subsequent oxidation (Miller and de Pena 1972, Dana et al. 1973, Beilke and Georgii 1968, Petrenchuk and Drozdova 1966)[2]; (b) the uptake of suspended particulate sulfates into raindrops (Kellogg et al. 1972, Miller and de Pena 1972). Where the ambient concentration of sulfure dioxide is very high, the former process may be more important (Kellogg et al. 1972, Beilke and Georgii 1968, Petrenchuk and Drozdova 1966). However, where the ambient sulfur dioxide level is moderate or low, scavenging of suspended particulates ("washout") is probably the dominant source of sulfates in precipitation (Likens 1972). Hence, close to major sources, the concentrations of sulfates and acidity in precipitation may be determined primarily by local oxidation of sulfur dioxide; far from major sources they are likely to reflect suspended sulfate levels. Thus, with due attention to the likely influence of local sources, regional patterns of deposition of sulfates can provide indirect information about the distribution of airborne sulfates.

SULFATES IN PRECIPITATION IN EASTERN NORTH AMERICA

Large-scale surveys of the chemical composition of precipitation in the United States were conducted in 1955-56 (Junge 1958, Junge and Werby 1958) and in 1965-66 (Pearson and Fisher 1971, Lodge et al. 1968, Gambell and Fisher 1966). By multiplying the average concentration of sulfates in precipitation by the observed rainfall for the year, it is possible to calculate the total amount of sulfur deposited in precipitation per unit area at each station. This has been done for the 1955-56 survey by Eriksson (1960), whose map of "excess"[3] sulfur deposition is reproduced here as Figure 7-1. It will be seen that the rates of sulfur deposition were relatively high throughout the northeastern United States; the highest rates [up to 13 kilograms of sulfur per hectare per year (kg/ha/yr)] were observed in western Pennsylvania and western New York. (Eriksson's map was based on data from the United States alone: in view of subsequent observations of high rates of deposition in southeast Ontario (Shiomi and Kuntz 1974)[4] it is likely that the area of maximum deposition should be extended somewhat further to the north).

The data from the 1965-66 survey (Pearson and Fisher 1971, Lodge et al. 1968, Gambell and Fisher 1966), when treated in the same way, show a very similar pattern, but with consistently higher rates of deposition. At nine stations that were in operation during both surveys, the average rate of sulfur deposition was higher by 68 percent in 1965-66 than in 1955-56. Within the area outlined by the contour line for 9 kg/ha/yr in 1955-56, all stations operated in 1965-66 reported deposition rates between 13 and 20 kg/ha/yr. Similarly the contour line for 6 kg/ha/yr in 1955-56 corresponds closely to that for 10 kg/ha/yr in 1965-66. Thus there appears to have been an increase in total deposition of sulfur by 60-65 percent during the decade, with little change in the geographical pattern of distribution. In addition, the 1965-66 survey indicated especially high rates of deposition at three urban stations: Chicago (32-62 kg/ha/yr),

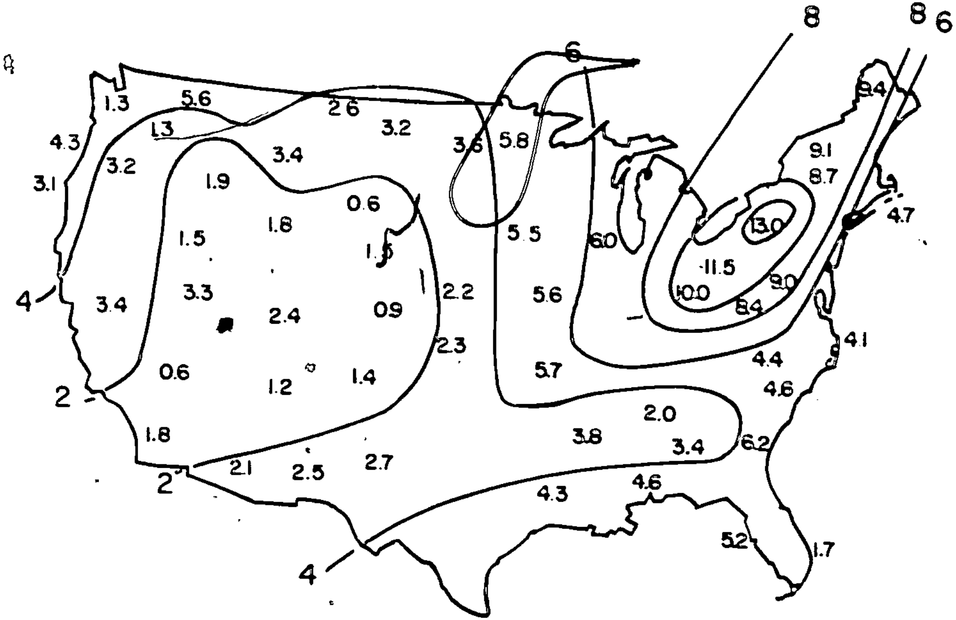

FIGURE 7-1: Excess Sulfur in Precipitation Over the U.S. in 1955-56 (After Eriksson 1960, Fig. 7.5: Original Data From Junge and Werby 1958). Units: KG Sulfur per Hectare per Year. Sulfates Associated With Sea Salt Have Been Subtracted From the Observed Sulfate Deposition Before Compiling the Map.

Albany (21 kg/ha/yr), and Philadelphia (19 kg/ha/yr).

No comprehensive survey has been conducted since 1965-66. Data obtained at Hubbard Brook, New Hampshire (Table 7-1) and in central New York (Likens 1972) suggest a levelling-off or even a decline in sulfur deposition rates between 1965-66 and 1970-71. However, relatively high deposition rates (average 17.2 kg/ha/yr) were recorded at seven stations in southern Ontario in 1970-71 (Shiomi and Kuntz 1974, see fn. 4); deposition at the Hubbard Brook station increased again to reach its highest level in 1972-73 (Table 7-1), and relatively high deposition rates were recorded at five other widely scattered stations operated by NOAA/EPA in 1972-73.[5] Thus the available data are consistent with a continued, if irregular, increase in sulfur deposition in eastern North America since 1965-66.

By integrating the observed deposition rates in the areas marked on the maps in Figure 7-1, it is possible to estimate the total amount of sulfur that fell out as sulfates in precipitation in eastern North America in 1955-56. This estimate, together with corresponding estimates for 1965-66 and 1972-73, is given in Table 7-2 and compared to estimates of emissions in the same area. According to these estimates, deposition of "excess" sulfates in precipitation accounted for about one-third of the sulfur emitted into the air in eastern North America. This estimate is a minimal figure for the fraction of sulfur dioxide converted to sulfates, because some airborne sulfates must be carried eastwards over the North Atlantic Ocean.

During the 17-year period under review, total deposition of sulfates in eastern North America has roughly doubled (Table 7-2). This is probably less rapid than the increase in emissions from power plants (about 200 percent), but more rapid than the increase in total emissions in the area (about 50 percent). This suggests that power plants are somewhat more important than other sources in leading to sulfate formation--perhaps because sulfur dioxide emitted from stacks is mixed better with the atmosphere than sulfur dioxide emitted from

TABLE 7-1. Sulfur Deposition in Rain and Snow at Hubbard Brook, New Hampshire, 1965-73 (Likens and Bormann 1974, Cogbill and Likens 1974, Likens et al. 1971, Cogbill, personal communication)

Year	1955-66	'66-'67	'67-'68	'68-'69	'69-'70	'70-'71	'71-'72	'72-'73
Mean sulfate concentration in bulk precipitation (mg/l)	3.3	3 1	3.3	2.4	2.4	2.8	2.7	2.9
Total precipitation (mm)	132	142	128	130	126	123	151	186
Calculated sulfur deposition (kg/ha/yr)	14.5	14.7	14.1	10.4	10.1	11.5	13.6	18.0

TABLE 7-2. Comparison of SO_2 Emissions in Eastern North America[1] with Total Deposition of Sulfates in Precipitation
(Units: Million tons sulfur)

	1955-56	1965-66	1972-73
Estimated emissions			
Canada[2]	1.3	1.8	1.3
U.S.: Electric power plants[3]	2.9	5.6	8.9
U.S.: Other emissions[3]	3.6	3.4	3.6
Natural sources[4]	1.8	1.8	1.8
Total	9.6	12.6	15.6
Estimated deposition in precipitation	2.9	4.7	5.7[5]
Deposition as % of emissions	30%	38%	37%

Notes: 1. The area considered is that from the Mississippi River east to the Atlantic Coast and Nova Scotia (60E), north of central Alabama (33N) and south of 50N (Winnipeg to the Gulf of St. Lawrence).

2. Canadian emissions for 1970 are given in Rennie and Halstead (1973): these were dominated by the emissions from the Sudbury smelters, which have probably been relatively constant during the period considered (Beamish and Harvey 1972, Balsillie, personal communication).

3. U.S. emissions were estimated by interpolating from the data for 1950, 1960, and 1970 in EPA(1973), assuming that 80 of power plant emissions and 50 of other emissions were in the area considered (cf. 13-10 and Figure 6-2).

4. Natural emissions were estimated by scaling the global estimate of ref. 1 in proportion to the area under consideration (about 2 percent of the earth's land surface) (cf. Rodhe 1972).

5. An increase of 3 percent per year is assumed for the period 1965-1972: this is consistent with Table 7-1 and other data quoted in the text, and also with 6-9b.

low-level sources, and therefore has a longer period in which it can be oxidized to sulfates before it can be absorbed by the ground. If emissions from tall stacks and low-level sources are weighted in the ratio 2:1, the observed increase in sulfate deposition would be roughly parallel to that in the weighted emissions.

Very few data are available on sulfates in precipitation prio to the 1955 survey.[6]

ACIDITY OF PRECIPITATION IN EASTERN NORTH AMERICA

Acid rain and snow have been reported in a number of localities in the eastern and northeastern United States (Likens et al. 1972, Likens and Bormann 1974, Cogbill and Likens 1974, Likens 1972, Johnson et al. 1972). The only large-scale synoptic survey is a student-conducted study (Anon 1974) carried out between 15-31 March 1973.[7] The results of this study, reproduced here as Figure 7-2, indicated that rain below the "normal" pH of 5.5[8] was falling throughout most of the eastern United States; the pH level was actually below 5.0 over extensive areas, especially in the Northeast. In addition there were small areas with precipitation pH below 4 near several cities, including Los Angeles, Chicago, Evansville, Louisville, Birmingham, Philadelphia, New York, Providence, and Boston.

No direct measurements of the pH of precipitation in the United States prior to 1959 appear to be available (Likens and Bormann 1974, Likens 1972). However, Granat (1972) and Cogbill and Likens (1974) have shown that is is possible to compute the pH of chemically analyzed rainwater samples with reasonable accuracy, by means of a stoichiometric balance between cations and anions. The principle of the method is shown in Figure 7-3: tests have shown that it can predict the pH of precipitation samples within ± 0.1 unit (Cogbill and Likens 1974; Figure 7-2).[9] Accordingly Cogbill and Likens (1974) have constructed maps of the average pH of precipitation,[10] based upon the precipitation chemistry data obtained in the surveys conducted in 1955-56 (Junge 1958, Junge and Werby 1958)

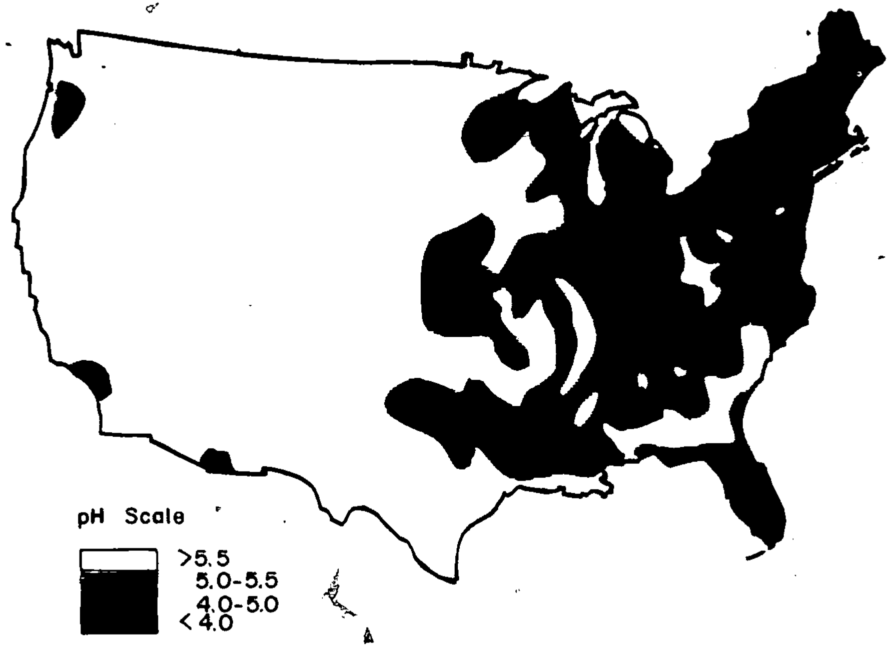

FIGURE 7-2: Acidity of Rainfall in the United States in the Period 17-31 March 1973 (ANON 1974). The map is based on a survey conducted by 16,000 High School Students at 1,100 Stations, Using a Method Accurate to ±0.5 Units or Better. See footnote 7 for Data on Rainfall in this Period.

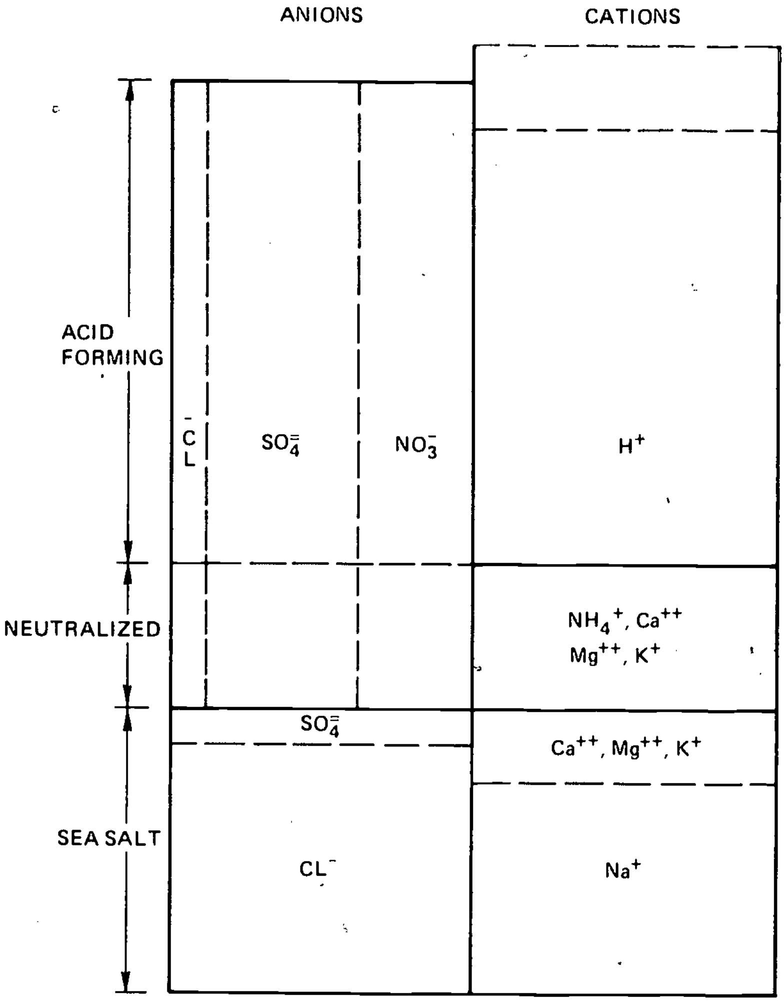

FIGURE 7-3: Chemical Composition of Rainfall at Hubbard Brook, New Hampshire (from Cogbill and Likens, in press, 1975)

and 1965-66 (Pearson and Fisher 1971, Lodge et al. 1968, Gambell and Fisher 1966). Both maps show that precipitation was significantly acidified (average pH below 5) throughout the northeastern United States; the area most affected (average pH below 4.5) was that between eastern Ohio and New Hampshire. The map for 1965-66 is reproduced here as Figure 7-4. Comparison of the two maps (Cogbill and Likens 1974) shows that a general increase in acidity had occurred in the intervening decade: the lowest computed pH's fell from 4.42-4.47 in 1955-56 to 4.20-4.35 in 1965-66.

Cogbill (personal communication) has compiled a similar map of precipitation pH for 1972-73, using data from nine stations in the United States (Coghill and Likens 1974, see fn. 5) and unpublished data from 12 stations in Ontario. This map indicates a marked extension of the area of acid precipitation, especially to the south and west: by 1972-73 the contour line for pH 5.0 included the entire United States east of the Mississippi River except for the Gulf Coast area; average pH's at several stations in New York and New Hampshire were as low as 4.05-4.09, and that at Gatlingburg, Tennessee was 4.19 (Cogbill and Likens 1974). Cogbill's 1972-73 map is generally consistent with the 1973 student survey (Figure 7-1), except that the latter shows more fine detail, including the pockets of very acid precipitation near industrial cities.

Other reported measurements of the pH of precipitation are generally consistent with the three maps prepared by Cogbill and Likens. A survey conducted between 1959 and 1964 showed consistently low pH's (below 5.5) at 13 stations in eastern North America[11]: the principal difference from Figure 7-4 is that the former survey showed lower pH values at Nantucket, Massachusetts, and in northern Maine, suggesting greater northeastward extension of the area affected by acid precipitation than indicated by Cogbill and Likens' maps. An earlier report had indicated significantly acid precipitation in Nova Scotia (Herman and Gorham 1957). The 1959-1964 survey also showed consistently high acidity (mean pH 4.0) at an urban station in

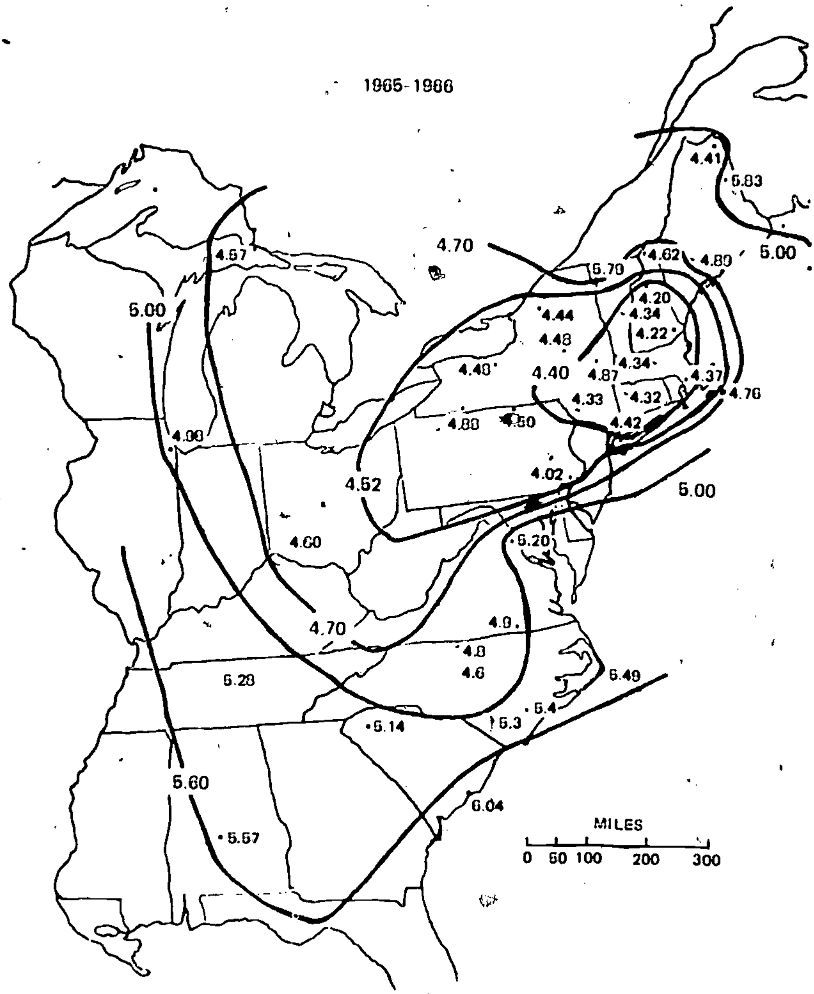

FIGURE 7-4: Acidity of Precipitation in the Eastern United States in 1965-66 (after Cogbill and Likens, 1974: Figure 4). The figures given are the annual average pH of precipitation (footnote 8) at each station (derived from data in Pearson and Fisher 1971, Lodge et al. 1968, Gambell and Fisher 1966).

Philadelphia (see fn. 11). Recent data from southern Ontario (Wiebe and Whelpdale 1974, Van Loon 1973), from ships in the Upper Great Lakes[12], from Massachusetts[13], Pennsylvania (Frohliger: personal communication), West Virginia and Maryland (Frohliger: personal communications, Gordon 1972) all show good agreement with Cogbill's unpublished map for 1972-73. Data from six stations in central New York state obtained in 1970-71 showed average pH of precipitation in the range 3.91 to 4.02 (Likens 1972).

Analyses of rain from individual storms in New York state, New Hampshire, and elsewhere frequently yield pH values between 3 and 4 (Likens and Bormann 1974, Cogbill and Likens 1974, Likens 1972), and there is one reported incident of rain with a measured pH of 2.1 (Likens and Bormann 1974). In New York and New Hampshire precipitation is significantly more acid in the summer months than in the winter (Likens and Bormann 1974, Likens 1972). The seasonal difference in pH at some stations is as great as 0.4-0.8 units, and at Ithaca, New York, the average pH of rain in June-July 1971 was as low as 3.53 (Likens 1972). A similar seasonal difference was recorded during the 1959-1964 survey at several stations in the northeastern United States, but not elsewhere (see fn. 11). The summer peak in acidity has been attributed tentatively to seasonal differences in storm tracks and to the greater humidity in summer, which leads to more rapid oxidation of sulfur dioxide to sulfuric acid (Likens and Bormann 1974).

There is no direct information about the acidity of rain in the eastern United States prior to 1955, but there is indirect evidence that the pH of precipitation was formerly above 5.7 over wide areas where it is now below 4.5.[14] Likens and Bormann have suggested that the acidification of rain began about 1950, as a consequence of the decrease in emissions of alkaline fly ash from coal-burning plants, in combination with increasing emissions of sulfur oxides and nitrogen oxides (Likens and Bormann 1974).

THE RELATIONSHIP OF ACID PRECIPITATION TO EMISSIONS OF SULFUR AND NITROGEN OXIDES

By multiplying the average hydrogen ion concentration observed at a meteorological station by the annual rainfall, an estimate can be derived for the total rate of deposition of hydrogen ions per unit area. Integrating these computed rates of deposition over the areas marked on the maps by Cogbill and Likens (1974), it is then possible to estimate the total quantity of hydrogen ions deposited in eastern North America in the course of a year. Such estimates for the two survey years 1955-56 and 1972-73 are tabulated on the first line of Table 7-3. These figures suggest tht the total amount of acid falling in precipitation increased by a factor of about 2.7 in the 17 years 1956-1973.

The principal acids involved are sulfuric and nitric acids, with small quantities of hydrochloric acid at some stations (see Figure 7-3). Data from all the surveys indicate that the ratio of sulfuric to nitric acids in precipitation has decreased substantially since 1955, because the concentrations of dissolved nitrates have increased more rapidly than those of sulfates. The best series of data is that from Hubbard Brook, New Hampshire, where the sulfate/nitrate ratio (expressed on an equivalent weight basis) has decreased from about 3.0 to 2.2 between 1965 and 1973 (Likens and Bormann 1974, Cogbill: personal communication), and from Ithaca and Geneva, New York, where this ratio has decreased from about 3.5 to 2.0 between 1955 and 1973 (Cogbill and Likens 1974, Likens 1972). Unfortunately the concentrations of nitrates in precipitation vary quite widely, and all three of these stations have atypically low sulfate/nitrate ratios [those at nearby stations range from 4 (Likens 1972: Table 13) up to 11 or higher (Shiomi and Kuntz 1974)]. Averaging all the data available for eastern North America, a sulfate/nitrate ratio of about 4 appears reasonable for 1955-56, and a ratio of about 3 for 1972-73. These figures have been used in Table 7-3 to calculate the changes in sulfuric acid and nitric acid deposition since 1955.

The last two lines in Table 7-3 compare the calculated deposition of sulfuric acid with the total deposition of sulfates, derived from Table 7-2. The fraction of sulfates deposited as sulfuric acid, averaged over the entire region, appears to have increased since 1955.[15] This result is consistent with the hypothesis of Granat (1972), that the supply of alkaline materials (such as ammonia or calcium) to the atmosphere is limited: once this supply is exhausted, emission of further acid-forming materials is expected to lead to a rapid increase in acidity. [16] Further support for Granat's hypothesis is provided by the regional distribution of sulfates and acidity: in areas where the sulfate loading is high, the acid fraction of the sulfates is disproportionately high. For example, at Ithaca, New York, about 80 percent of the excess sulfates are in the form of sulfuric acid (Cogbill and Likens 1974), in comparison with the regional average of 24 percent (Table 7-3).

Another noteworthy feature of Table 7-3 is that total nitrates in precipitation have increased more rapidly (roughly threefold since 1955) than sulfates (roughly twofold in the same period). This presumably reflects the fact that total nitrogen oxide emissions have increased more rapidly than sulfur oxide emissions (see Chapter 14 Figure 14-15). The increase in nitrate deposition appears to have been more rapid than the increase in total nitrogen oxide emissions (which roughly doubled between 1955-1972), but matches reasonably well the increase in emissions from stationary fuel combustion and industrial processes (which roughly tripled in the same period). This suggests that high level emissions are more important than ground level emissions in leading to formation and deposition of nitrates, [17] in just the way that was suggested earlier for sulfates. In fact, if attention is confined to high level emissions (power plants, smelters, and industrial processes), the geographical and temporal patterns of emissions of sulfates and nitrates appear to match well the patterns of emission of sulfur and nitrogen oxides.

TABLE 7-3. Deposition of Acid in Precipitation in Eastern North America, Assigned to Sulfuric Acid and Nitric Acid According to the Measured Sulfate/Nitrate Ratio
(Units: 10^4 ton-equivalents per year -- See Note 1)

	1955-56	1972-73	% change 1956-73
Total deposition of acid (as H+: unit 10^4 tons)	4.4	11.9	+170%
Estimated deposition as sulfuric acid (as 1/2 H_2SO_4: unit 49 10^4 tons)	3.5(80%)	8.7(73%)	+150%
Estimated deposition as nitric acid (as HNO_3:unit 63 10^4 tons)	0.84(19%)	2.86(24%)	+240%
Total deposition of sulfates (from Table 2) (as 1/2 SO_4:unit 48 10^4 tons)	18	35	+95%
Sulfuric acid as % of sulfates	19%	24%	+25%

Notes: 1. All deposition rates are expressed as multiples of the chemical equivalent weight, so that rates for different chemical species can be compared directly. 1 ton H^+ is equivalent to 49 tons sulfuric acid or to 63 tons nitric acid. For comparison with emissions, 1 ton H^+ is also equivalent to 32 tons SO_2, 16 tons sulfur, or 30 tons nitric oxide.

2. A small but increasing fraction of the acid in precipitation is attributable to hydrochloric acid (see Figure 7-3 and Likens and Bormann 1974, Cogbill and Likens 1974, Likens 1972).

EVIDENCE FOR LONG RANGE TRANSPORT AND DEPOSITION OF SULFUR OXIDES

The pattern of sulfate deposition indicated in Figure 7-1, with its peak in the northeastern United States, agrees closely with the distribution of suspended sulfates plotted in Figures 6-4 and 6-5. A slight difference is that Eriksson's map (Figure 7-1) suggests relatively low deposition rates near the Atlantic coast; however, this was based on data from very few stations and high deposition rates were recorded in the 1965-66 survey (Pearson and Fisher 1971).

The distribution of acidity in precipitation is very similar to that of sulfates (Figures 7-2, 7-4). Pearson and Fisher found a good correlation between acidity and sulfate deposition in the northeastern United States (Pearson and Fisher 1971: Figure 5). Likens found that this correlation was less strong in New York state (Likens 1972: Figure 12), but an exact correlation would not be expected because of the importance of nitric acid in that area (see discussion above).

Cogbill and Likens (1974: Figure 5) plotted the trajectories of air parcels associated with precipitation in central New York state. They found a strong association between rainfall with low pH values and air which had passed through the East-Central and Mid-Western states 1-2 days earlier. The area defined by these trajectories (broadly, the Mississippi and Ohio valleys north to the Great Lakes) coincides with the area of greatest emission density for both sulfur oxides (Figure 13-10) and nitrogen oxides (Table 14-8 in the nitrogen oxides report). Air parcels arriving in New York state from areas further north (i.e., Canada) or further southeast (i.e., the Appalachian mountains and beyond) were associated with rain that was much less acidic (Cogbill and Likens 1974: Figure 5). This finding is fully consistent with the hypothesis that acid precipitation results from regional transport of sulfur oxides (and perhaps nitrogen oxides) emitted 300-1500 km upwind (Cogbill and Likens 1974). It thus provides indirect support for the models of regional transport of sulfur oxides discussed in Chapter 6 and Chapter 13.

According to the nitrogen oxides report (see Chapter 15), rates of conversion of nitrogen oxides to nitric acid are typically 3-4 times faster than those for conversion of sulfur dioxide to sulfates. Accordingly it might be expected that nitric acid would be more concentrated near major sources than sulfuric acid (see Chapter 15). However, there is no direct evidence for this in the precipitation chemistry data. The only stations where the reported nitrate/sulfate ratios in precipitation were greater than 0.3 are St. Albans Bay, Vermont (Pearson and Fisher 1971), Hubbard Brook, New Hampshire (Likens and Bormann 1974, Cogbill and Likens 1974), Ithaca and vicinity, New York (Likens 1972), Mntgomery, Alabama (Junge 1958, Junge and Werby 1958, Lodge et al. 1968), Greenville, South Carolina, and Chicago, Illinois (Lodge et al. 1968). With the exception of Chicago, where the concentrations of both nitrates and sulfates were exceptionally high, these are predominantly rural stations away from major sources. At other urban stations, including those such as Albany and Philadelphia where highly acidic precipitation has been reported, nitrate/sulfate ratios are consistently lower than 0.3 (Junge 1958, Junge and Werby 1958, Pearson and Fisher 1971, Lodge et al. 1968, Gambell and Fisher 1966, Van Loon 1973). Thus the concentration of acid precipitation around cities (Anon 1974) cannot be explained as being due to local fallout of nitric acid; it probably reflects primarily the rapid oxidation of sulfur dioxide in polluted urban atmospheres, as discussed in Chapter 6 and in Chapter 13, Appendix A.

SUMMARY OF COMPARABLE OBSERVATIONS IN EUROPE

Since the above survey of sulfates and acidity in precipitation in North America is based on somewhat limited data, it is at least reassuring that similar conclusions have been derived independently in Europe, where the phenomena of long-range sulfur transport and acid precipitation have been studies for a longer period (Rodhe 1972, Bolin et al. 1971, Oden 1968, Brosset 1973, Granat 1972, Barrett and Brodin 1955, Munn and Rodhe 1971, Garland 1974, Nord 1974). Deposition of sulfur has been increasing at 2-3 percent per year, roughly in parallel with the increase in emissions of sulfur dioxide in western Europe (Bolin et al. 1971, Munn and Rodhe 1971). The total amount of sulfur deposited as sulfate in precipitation in western Europe has been estimated as between 27 and 45 percent of the total amount emitted (Rodhe 1972, Garland 1974). The acidity of precipitation has increased more rapidly than the sulfate content, locally more than 200-fold over a 10-year period (Bolin et al. 1971, Oden 1968). The area subject to acid precipitation has expanded to include most of western Europe (Bolin et al. 1971, Brosset 1973). The area most affected (average pH less than 4.0, sulfur deposition greater than 15 kg/ha/yr in 1965) is in the eastern Netherlands and N.W. Germany, a few hundred km downwind from the major sources (Rodhe 1972, Bolin et al. 1971, Oden 1968, Brosset 1973). By plotting atmospheric trajectories it has been possible to relate acid precipitation and sulfate deposition in northern Europe to sources in the industrialized regions to the south and west (Rodhe 1972, Bolin et al. 1971, Brosset 1973, Nord 1974): the data are consistent with a lifetime of 2-4 days for sulfur in the atmosphere and a corresponding range of 1000-2500 km (Rodhe 1972). By summing the rates of deposition mapped in Bolin et al. (1971: Figure 3:5), I have estimated the total rate of deposition of hydrogen ions in western Europe to have been about 4.5 10^4 tons/yr in 1965. If all this were attributable to sulfuric acid, it would account for 15-25 percent of the sulfates deposited in the same area (Rodhe 1972, Garland 1974). However, as in North America, there are substantial contributions of nitric acid to total acidity (Granat 1972). All these findings are similar both qualitatively and quantitavely to those summarized above for eastern North America.

PROJECTED CONSEQUENCES OF INCREASED EMISSIONS IN 1980

Following Trijonis (Chapter 6), I will consider the effects of a projected doubling in sulfur oxide production in power plants between 1970 and 1980, with other emissions projected to remain constant. Two extreme cases will be considered:

No SOx control: power plant emissions will increase to 38 million tons sulfur dioxide in 1980, mainly through tall stacks;

SOx control: power plant emissions in 1980 will remain unchanged at 19 million tons sufure dioxide.

For nitrogen oxide emissions in 1980, I will use the high projection in the nitrogen oxides report,[18] assuming no control of emissions.

Canadian emissions of both SOx and NOx will be assumed to change in the same proportion as United States emissions.

To calculate the likely impact of these projected emissions on the chemistry and acidity of precipitation, the following assumptions will be made.

(a) the fraction of SOx emissions that takes place in the eastern United States will be unchanged (assumed to be 80 percent of power plant emissions and 50 percent of others);

(b) similarly, the fraction of NOx emissions that takes place in the eastern United States will remain constant at about 70 percent (Table 14-8 in the nitrogen oxides report).

(c) the use of tall stacks and intermittent control systems will not affect the fractions of emitted sulfur dioxide and NOx that are ultimately converted to sulfates and nitrates (in fact, some increase is likely);

(d) the total amount of sulfates deposited in precipitation in eastern North America will increase in proportion to weighted emissions of sulfur dioxide, where stack and ground-level emissions are weighted in the ratio 2:1 (see Table 7-2 and accompanying discussion in the text);

(e) the total amount of nitrates deposited in precipitation in eastern North America will increase in proportion to stack emissions of NOx (stationary source fuel combustion and industrial processes) (see text discussion above);

(f) to estimate the fraction of nitrates and sulfates deposited as acids, two alternative assumptions will be made:

(i) constant percent acid: the fraction deposited as acid will be the same in 1980 as in 1972-73;

(ii) all acid: all the nitrates/and sulfates produced in 1980 in excess of those deposited in 1972-73 will be deposited as acids (following the hypothesis of Granat (1972) that the neutralizing capacity of the atmosphere has been exhausted).

Table 7-4 presents the results of calculations of the rates of deposition in 1980 of sulfates, nitrates, and the corresponding acids, under the four sets of assumptions. These results suggest two major conclusions:

(i) even if sulfur oxide emissions are not permitted to increase, there is likely to be a small increase in the acidity of precipitation, as a consequence of increased nitrogen oxide emissions;

(ii) if sulfur oxide emissions from power plants increase to twice the 1980 levels, the acidity of precipitation will increase by a larger factor. The exact magnitude of the increase depends on the fraction of the additional sulfates and nitrates that will be deposited as acids. Granat's hypothesis (1972) that this fraction is likely to be high, because the neutralizing capacity of the atmosphere is nearly exhausted, is at least plausible and is supported by the observed rapid increase in acidity of precipitation both in North America and in Europe. Accordingly the "all acid" assumption is probably more likely to be correct than the "constant percent acid" assumption, which requres an additional source of alkaline materials into the atmosphere. Thus, if sulfur oxide emissions are uncontrolled, the total acidity of precipitation in eastern North America is likely to increase by a factor of 2-3 between 1972 and 1980. Projecting the trends of the period 1965-1973, it is likely that this increase will be accompanied by an extension of the area affected. In particular, if emissions

TABLE 7-4. Projected Deposition of Sulfates, Nitrates, and Acids in Eastern North America in 1980, under 4 Sets of Assumptions about Emission Controls and Acid Formation.

	1972 estimates (from Tables 7-2 & 7-3 and footnote 17)	1980 projections: SO_x control, constant % acid[5]	1980 projections: SO_x control, all acid[6]	1980 projections: No SO_x control, constant % acid[5]	1980 projections: No SO_x control, all acid[6]
Sulfur emissions (10^6 tons S)					
Tall stacks	12.5	12.5	12.5	18.9	18.9
Low level	3.1	3.1	3.1	3.2	3.2
Weighted total (1:0.5)	14.1	14.1	14.1	20.5	20.5
NO_x emissions (10^6 tons NO)					
Tall stacks[2]	10.8	14.9	14.9	14.9	14.9
Sulfate deposition (10^6 tons S equivalent)					
Total[3]	5.7	5.7	5.7	8.2	8.2
As sulfuric acid	1.4	1.4	1.4	2.0	3.9
% acid	24%	24%	24%	24%	46%
Nitrate deposition (10^6 tons NO equivalent)					
Total[4]	3.5	4.9	4.9	4.9	4.9
As nitric acid	0.86	1.17	2.21	1.17	2.21
% acid	24%	24%	45%	24%	45%

TABLE 7-4 (Cont.)

Total acid deposition (10^4 tons H+)	11.6	12.7	16.2	16.4	31.8
% increase in acid,1972-80	--	+10%	+40%	+40%	+175%

Notes: 1. Power plants, smelters and industrial emissions. It is assumed that 80 of power plant emissions and 50 of other emissions will be in the eastern United States.

2. Stationary fuel combustion and industrial emissions. It is assumed that 70 of these emissions will be in the eastern United States, as in 1972 (Table 14-8) in nitrogen oxides report).

3. Assumed to remain constant at 40 of weighted emissions.
4. Assumed to remain constant at 32 of stack emissions.
5. Sulfuric and nitric acids assumed to remain constant at 24 of deposited sulfates and nitrates respectively.
6. All sulfates and nitrates incremental between 1972 and 1980 assumed to be deposited as the corresponding acids.

increase in the Midwest, it is likely that significantly acid precipitation will extend over a wider area of prime agricultural land from Ohio and Tennessee westwards.

NEUTRALIZATION AND RUN-OFF OF ACIDIFIED PRECIPITATION

When acid rain falls on vegetation, part of the acid is neutralized in the leaves. In a study carried out in New Hampshire, for example, rain falling in summer on a deciduous forest at an average pH of 4.06 was 90 percent neutralized in the foliage and reached the ground at an average pH of 5.01 (Eaton et al. 1973). Neutralization of acid results in losses (leaching) of cations such as potassium, magnesium, and calcium from the leaves (Eaton et al. 1973, Tukey et al. 1965, Wood and Bormann 1975).

In winter, and on agricultural land, acid rain falls directly on the soil and is partly neutralized there. Again the process of neutralization results in the removal of cations such as calcium and magnesium from the soil in the form of soluble sulfates (Overrein 1972). Unelss the soil is well buffered, continued exposure to acid precipitation can lead to progressive depletion of nutrients and acidification of the soil (Bolin et al. 1971, Overrein 1972, Malmer 1973). Direct absorption of sulfur dioxide contributes to these effects, because it is oxidized to sulfuric acid in the soil. Unless soils are naturally alkaline or well buffered, neutralization of acid is usually incomplete, so that run-off water is often significantly acidic (Bolin et al. 1971). In the New Hampshire study cited above, for example, the average pH of run-off water was 5.15 (Likens and Bormann 1974, Johnson et al. 1972). Experimental studies with a poorly buffered (podzol) soil showed marked acidification of run-off water after only 80 days when treated with simulated rainfall at pH 4.0 (Overrein 1972).

In Scandinavia acidification of a number of rivers and lakes has been reported in recent years (Bolin et al. 1971), Almer et al. 1974, Hagen and Langeland 1973); typical changes in pH between 1965 and 1970 were of the order of 0.1 to 0.4 (Bolin et al. 1971). Similar phenomena have been recorded locally in North America (Beamish and Harvey 1972, Johnson et al. 1972, fn. 13). The rate at which streams and lakes become acidified appears to be highly variable and to depend upon the nature of the underlying rocks and of the soils in the watershed, and the buffering capacity of the lakes themselves (Bolin et al. 1971, Beamish and Harvey 1972, Malmer 1973). Where the underlying rocks are acidic and the soils poorly buffered, lakes may become significantly acidified after a few years of exposure to precipitation at pH 4.5 or below (Beamish and Harvey 1972, Almer et al. 1974).

Studies in Scandinavia (Bolin et al. 1971) and in the northeastern United States (Likens: personal communication, fn. 13) have shown that there is often a marked peak in the acidity of streams and lakes at the time of snow-melt in the spring. Hagen and Langeland (1973) found that acid contained in winter snowfall is concentrated in the surface layers of lakes immediately after the ice melts. These seasonal peaks in the acidity of surface waters are of special importance because they coincide with the spawning of fish; moreover in some lakes the deeper layers are depleted of oxygen during the winter, so that fish are forced to use the surface layers which are acidified in early spring (Hagen and Langeland 1973).

The full extent of the acidification of soils and surface waters in eastern North America has not been studied. However, as a broad generalization, much of the area of the United States and Canada most affected by acid precipitation is also characterized by acidic rocks and relatively thin soils. Accordingly, if the Scandinavian experience is any guide, continued exposure to highly acidic precipitation can be expected to lead to widespread acidification of soils and surface waters over a period of years or decades.

SUMMARY AND CONCLUSIONS

1. Deposition of sulfates in rain and snow results both from oxidation of sulfur dioxide absorbed into water droplets, and from washout of suspended sulfate particulates. With due regard for the local importance of the former process near major sources of sulfur dioxide, the occurrence of sulfates in precipitation gives information about the regional distribution of sulfates.

2. Concentrations of sulfates in precipitation are highest in the northeastern United States: their distribution parallels that of suspended sulfate particulates.

3. About one-third of the sulfur dioxide emitted to the atmosphere in the eastern United States is subsequently deposited as sulfates in precipitation.

4. The rate of deposition of sulfates in precipitation has increased in the past 20 years. The increase has probably been more rapid than the increase in total emissions of sulfur dioxide; it agrees better with the increase in high level emissions (from power plants and industrial processes).

5. Concentrations of nitrates in precipitation have increased even more rapidly than those of sulfates. The increase in the rate of deposition of nitrates is similar to the increase in high level emissions of nitrogen oxides.

6. The acidity of precipitation in the northeastern United States has increased rapidly in recent years. The areas affected by acid precipitation now covers most of eastern North America, including parts of southeast Canada. In some small areas in the Northeast the average pH of precipitation is near or below 4.0.

7. The distribution of acidity in precipitation is generally similar to that of dissolved sulfates. However, the fraction of the acidity attributable to nitric acid has increased and now amounts to roughly 24 percent.

8. Acid rain in the northeastern United States is associated with air parcels that have traveled through the major emitting areas in the mid-western and east-central states one or two

days earlier. This supports the hypothesis of regional transport of sulfur oxides and nitrogen oxides.

9. Closely similar phenomena have been reported from western Europe.

10. Even if sulfur oxide emissions are held constant, a small increase in the acidity of precipitation is likely by 1980, as a consequence of increased nitrogen oxide emissions.

11. If sulfur oxide emissions are allowed to double between 1970 and 1980, the average acidity of precipitation in the northeastern United States and in southeastern Canada is likely to increase by a factor of 2-3. The area affected may be enlarged also.

12. Acid precipitation is partly neutralized when it falls on vegetation and further neutralized in soil. The process of neutralization removes cations, including important nutrients.

13. The processes of neutralization in foliage and soil are usually incomplete, so that run-off water is often significantly acidic.

14. If the acidity of precipitation is maintained or increased, soils and surface waters are likely to become progressively acidified. The rate of acidification depends on the buffering capacity of the soil or water and is difficult to predict in individual cases.

FOOTNOTES

1 A cooperative study is in progress under the auspices of the OECD, and some preliminary results are now available (Garland 1974, Nord 1974).

2 The mechanisms of absorption and oxidation of sulfur dioxide in water droplets are complex and cannot be reviewed fully here (see Miller and de Pena 1972, Dana et al. 1973 for discussion). Uptake of sulfur dioxide is strongly limited by the acidity of the raindrops (Dana et al. 1973). The rate of uptake and oxidation is increased in the presence of ammonia (Scott and Hobbs 1967) or other catalysts. Observations of

precipitation around point sources of sulfur dioxide indicate marked variations in the rate of oxidation. In a plume from a heating plant at Uppsala, Sweden, much of the sulfur dioxide emitted during precipitation was deposited as sulfate within 50-100 km (i.e., within 2-3 hours) (Hogstrom 1973). Studies around a sulfur extraction gas plant in Alberta Canada, showed similarly rapid oxidation of sulfur dioxide during rain (32-46 percent deposited as sulfate within 40 km), but much slower oxidation in snow (Summers and Hitchon 1973, Summers 1974). However, studies around a smelter at Sudbury, Ontario, showed very slow oxidation of sulfur dioxide (less than 1 percent deposited as sulfate within 50-100 km) even when emitted during rain (Muller and Kramer 1974, Wiebe and Whelpdale 1974). The low rate of oxidation in the Sudbury area may be related to the peculiar chemistry of the plume, which has high concentrations of iron, nickel, and copper, but low concentrations of vanadium and manganese which are effective catalysts for oxidation of sulfur dioxide to sulfates (A. Wiebe: personal communication).

3 Dissolved sulfates in precipitation include a neutral fraction derived from ocean spray: this can be estimated from concentrations of sodium and chloride and is customarily subtracted out to derive an "excess" sulfate concentration (Junge and Werby 1958, Eriksson 1960). For details see notes to Figure 7-3.

4 The deposition rates recorded in Shiomi and Kuntz (1974) were probably slightly low because the collectors did not sample snow efficiently. An earlier study of precipitation in southeastern Ontario in 1965-66 (Rutherford, Can. J. Earth Sci. 4: 1151-1160, 1967) had shown anomalously high rates of sulfur deposition (average 39 kg/ha/yr), but the presence of silicates and aluminum in these samples suggests the possibility of contamination by wind-blown dust. For discussion of other technical problems see Likens (1972) and Pearson and

Fisher (1971). In particular, most of the studies cited in this section were of bulk precipitation (which includes dry fallout), whereas those reported in Junge and Werby (1958) and Lodge et al. (1968) were designed to exclude dry fallout. However, there is indirect evidence that dry fallout of sulfates is only a small component of deposition (Gambell and Fisher 1966). Where the surveys of Lodge et al. (1968) and Gambell and Fisher (1966) overlap there is no discernible discrepancy in calculated deposition rates.

5 Sulfate concentrations supplied by J. Miller per C.V. Cogbill; precipitation data from NOAA Environmental Data Service.

6 At two stations in central New York, sulfate levels in precipitation were much higher between 1915 and 1950 than after 1952 (Likens and Bormann 1974, Likens 1972). However, both stations were close to sources burning fossil fuels; the occurrence of higher concentrations in winter is consistent with a local origin (Likens 1972).

7 The period 15-31 March, 1973 was unusually wet in much of the eastern United States. The total rainfall during this 17-day period at selected stations was as follows: Boston 1.08"; New York City 2.17"; Philadelphia 2.49"; Chicago 2.78"; Louisville 3.88"; Birmingham 6.84" (from Daily Weather Maps, NOAA Environmental Data Service). Hence, except perhaps for Boston, the high acidity recorded near these stations cannot be explained by low dilution factors.

8 pH is the symbol for the negative logarithm of the concentration of hydrogen ion [H^+ solution in grams per liter. The more acid a solution, the lower the pH. Doubling the [H^+] concentration reduces the pH by 0.301 units. Unpolluted rain water is slightly acid because it contains dissolved carbon dioxide, which dissociates to form [H^+] and bicarbonate ions. If in equilibrium with atmospheric carbon dioxide, rain water would have a pH of 5.5-5.7. As strong acids are added, carbon dioxide is displaced and the pH falls below 5.5

9 Several reviewers have questioned the use of bulk precipitation samples or monthly samples for determining pH, because of the possibility of chemical changes prior to analysis. However, pH estimates obtained by the methods of Cogbill and Likens (1974) appear to agree well with those obtained from weekly or monthly samples (Cogbill and Likens 1974, Likens 1972). In this survey both single-storm and monthly samples are used indiscriminately, except where there was evidence of contamination by airborne dust or insects (for discussion see Likens 1972).

10 "Average" pH's quoted in this section are derived from weighted averages of hydrogen ion concentrations throughout a year. They thus represent the pH that corresponds to the average chemical composition of precipitation.

11 Unpublished data from the National Precipitation Network, supplied by R.A. McCormick (EPA).

12 Unpublished data of Canada Center for Inland Waters: personal communication from F.C. Elder.

13 Unpublished data of Massachusetts Division of Fisheries and Game.

14 Published measurements of bicarbonates in rainwater in the eastern United States prior to 1932 indicate that the pH of precipitation was uniformly above 5.7 at that period (Likens and Bormann 1974, Likens 1972).

15 A similar conclusion holds <u>a fortiori</u> for nitrates, because the stoichiometric analysis summarized in Figure 7-3 assigns hydrogen ions to sulfates and nitrates in proportion to their ratio in the precipitation sample.

16 This hypothesis does not necessarily conflict with that of Trijonis (Chapter 6), that oxidation rates of sulfur dioxide to sulfates are limited by the acidity of particles or water droplets when neutralizing materials are depleted. This mechanism would limit the <u>local</u> rate of oxidation in places where ambient levels are

high. It would not prevent oxidation of the sulfur dioxide after further dispersal and dilution.

17 The figures in Table 7-3 can be used to estimate the total amount of nitrates deposited in precipitation in eastern North America. For 1972-73 this turns out to be about 7 million tons expressed as nitrate, equivalent to 3.5 million tons NO. Although this estimate is indirect and subject to some error, it is clearly much smaller than the total NOx emissions in the area (about 17 million tons) or even those derived from stationary fuel combustion (9 million tons). Evidently deposition in precipitation is a substantial, but not the major sink for nitrogen oxides emitted into the atmosphere.

18 Use of the high projection is based on the consensus of opinion in the Committee that a massive increase in nuclear generation of electric power is unlikely to take place by 1980. However, even the low projection involves an increase in NOx emissions about three-quarters as large as that considered here.

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PART TWO

Section 2: Techniques for Reducing Emissions from Power Plants

CHAPTER 8

PRICING POLICY AND DEMAND FOR ELECTRICITY

(Chapter 8 was written by Alfred Kahn under the general supervision of the committee, which reviewed the work at several stages and suggested modifications that have been incorporated. While every committee member has not necessarily read and agreed to every detailed statement contained within, the committee believes that the material is of sufficient merit and relevance to be included in this report.)

EFFICIENT PRICING AND CONSERVATION

One important means of achieving the broad goals of our national energy policy (i.e., reduced dependence on foreign crude oil and minimization of damage to the environment consistent with economic welfare) is to promote conservation or, put another way, avoid waste in our consumption of energy. Referring to our mandate in this chapter, one important way of minimizing damages to health and the environment from the use of coal to produce electric power is to limit the consumption of electricity or reduce its rate of growth.

There are various definitions of "conservation" and "waste." But whatever the disagreements over the choice of definitions, there can be no disagreement that we want to eliminate inefficient uses in the economic sense, which means, most broadly, uses whose benefits are less than the costs they impose on society. There is wide social consensus that our principal mechanism for assessing these benefits and costs should be the competitive market system in which ultimate choices are made by the "sovereign" consumer.

If this system is to work efficiently, the prices of all the competing costs and services that buyers confront, and on the basis of which they make their choices, must accurately reflect the respective costs to society of providing them. More precisely, the price of each product must equal its marginal cost, the cost of producing a little bit more, or the cost that would be avoided if buyers consumed a little bit less. The reason for this is that the demand for all goods and services is, in some degree, elastic--if price goes up, buyers will typically take a little bit less, if price goes down, they will typically, take a little more. If then, buyers are to make their individual choices in such a way that they will, as a group, get the maximum total satisfaction from our limited total productive capacity, the prices by which they are guided must reflect the cost to society of producing a little bit more, or its savings from producing a little bit less. Only then will consumers know what sacrifices they are imposing on society (and therefore on themselves as a body) by behaving correspondingly, and (according to this idealized conception) consume just the right amount of each good and service--neither too much (carrying their consumption beyond the point where incremental costs exceed incremental benefits) nor too little (refraining from consumption whose incremental benefits exceed the incremental costs to society).

By this reasoning, one prerequisite for achieving the proper degree of conservation, or economizing, in the use of energy, is that its price be equated to its marginal social cost of production. In fact, our pricing of electricity falls far short of this requirement in many ways, of which we can mention only the most prominent.

Electric utilities are in most jurisdictions regulated on an original, or historic, cost basis. This means that their capital costs (depreciation, return on investment, income taxes), which bulk extraordinarily large in this industry compared with most others, are measured by, and as a return on, the cost historically incurred. But the only measure of marginal cost that has any economic significance is current costs, or, as one sets rates for the future, the cost that will be incurred or saved during the period when those rates are in effect. In times of rapid inflation, those marginal costs tend, naturally, to rise relative to average company revenue requirements, when the latter are based heavily on historic costs. For example, the current cost of a kilowatt of additional base load generating capacity may range between $300 and $800; the average book cost, on the basis of which we fix the depreciation and return component of electric rates, is on the order of $150 to $200. This means that the prices that purchasers confront (to which they will, if they are rational, equate the benefits of additional consumption) understate the additional sacrifices imposed on society by their consumption or the savings that society would realize if they consumed somewhat less. In this way, such prices encouage wasteful consumption.

Operating in the same direction is the failure of electricity rates typically to reflect peak responsibility principles. The demand for electricity varies widely from one hour of the day and one season of the year to another. Since the current itself cannot be stored, the only way demand can be satisfied when it is at its highest levels of the day or year is by having the required capacity available at those times. The required level of investment in generating and transmitting capacity thus depends specifically on the level of demand at the times of system peak consumption.

If the economically proper amount of capacity is to be constructed, therefore, it is that particular consumption, i.e., consumption at the time of peak utilization, that must be charged the full marginal costs of making that capacity available: only purchases at the peak are marginally responsible for the industry's incurring the costs of providing that capacity; every kilowatt hour by which consumption might be reduced on peak would save society the cost of providing that much additional capacity. In contrast, consumption truly and inalterably off peak should not pay any capacity costs: the inustry would have to be adding to capacity to meet peak demand regardless of whether consumption off peak continued or ceased.

There is not time here to discuss all the problems that would be involved in administering such a pricing system. It is clear, generally, that the failure of most electric utility pricing to reflect these peak responsibility principles, as well as to measure capacity costs in current rather than historical terms, gives rise to a/greater demand for electricity and a consequent greater construction of plant than would otherwise occur.

The embodiment of these principles in electric rates would be expected to both restrict total consumption and shift some of it from peak to off-peak times. The latter shift by itself, incidentally, would in the first instance conserve mainly construction of capacity, rather than the consumption of energy. And yet both energy and environmental conservation would nevertheless be served by such a shift in consumption, even in the absence of a decline in the total kilowatt hours, for several reasons.

First, the construction of capacity itself uses energy, in producing the materials that go into the generating and transmission plant, and in the construction process itself, energy that would be saved to the extent that the same total of consumption could be supplied by fuller utilization of a smaller plant.

Second, the plant used for peaking purposes is typically highly energy-intensive. Consumption for relatively short peaks is most economically served by plant, such as gas turbines, involving relatively low capital and high fuel costs: it does not pay to incur the much higher capital costs of, say, nuclear capacity for plant that will operate for only a small fraction of the year. In terms of energy utilization, peaking capacity is therefore extremely inefficient. To the extent that consumption is, therefore, shifted from peak to off-peak, kilowatt hours will be supplied at energy costs of only a few mills instead of several cents per kilowatt hour.

Third, in important measure impairment of the environment is a function of the amount of capacity constructed. The construction of economically unnecessary generating and transmission plant involves unncessary additional injury to the environment, even though the total amount of electricity generated is unchanged.

Fourth, to some extent the injury to the environment from the generation of electricity is an increasing function of the amount generated at any particular time. Air and water can, within limits, handle a given infusion of pollutants with little injury; but it seems likely that increasing that total load on the environment at any given time involves rapidly mounting external costs. Anything therefore that levels out the generation of energy, reducing its amount at times of peak utilization, even though increasing it correspondingly at off-peak times, is likely to result in a net diminution in environmental costs.

For all these reasons, more efficient pricing that has the effect only of leveling out the consumption of electricity, without changing its total, will in itself be promotive of conservation.

In addition, however, it may confidently be expected that efficient pricing will diminish total consumption, as well, and thus contribute further to the same end.

DEMAND PROJECTIONS AND ELASTICITY

One important part of our national energy policy must be a strenuous effort to encourage conservation to the extent that this is consistent with economic welfare. There are many justifications for such a policy; one benefit that is directly pertinent to our inquiry is that one way to limit sulfur dioxide emissions from electricity generation is to decrease the need for so much electricity in the first place. Conservation thus becomes part of a national policy to control these emissions. We suggest elsewhere in Part Two several ways of encouraging this result, including that of pricing electricity, in so far as practicable, at its long-run marginal cost and on the basis of peak responsibility (see the discussion of pricing below.)

It is not part of our task here to predict the effects of such efforts, or to supply estimates of the rate at which the demand for electricity is likely to grow in the next several years or the level it is likely to reach in 1980 or 1985. These estimates and the methods of making them vary widely, largely because of the enormous uncertainties that have been introduced into our energy equations by the dramatic events of 1974, and partly because of the difficulty of predicting the consequences of national energy policies that are still in process of formulation.

The electric companies themselves have tended to use an essential eclectic method of projecting demand as the basis for their long-range planning. There is a popular assumption that most of them have, until recently, relied essentially on naive extrapolations of past trends, and many of them may have done so. Others have relied on projections of such factors as demographic trends, market saturation rates for various appliances, predictions of general economic conditions, the prospects for certain key industries in their market areas, and the market shares of electricity as compared to other sources of energy in their various submarkets. These techniques have often produced the same kind of results as simple extrapolations on the assumption of constant growth rates, and have proved reasonably reliable during the last several decades because most of the determining economic factors have in fact grown at reasonably stable rates from the late 1940s to the early 1970s.

The validity of these methods and their projections of essentially unchanged growth rates have been subjected to intense criticism during the last few years, under the gradual impact of environmentalist and conservationist objections to perpetuation of the 7 percent growth rate of the last two decades, and especially in the wake of the energy problems that have emerged since the 1973-74 boycott by the oil exporting countries. The critics have argued that the industry clings unrealistically to the basic assumption that, apart from the loss of a year's growth because of the shortages and unusual conservation efforts of 1974, growth may be expected to resume in 1975 at something like its former rate. The critics contend, instead, that the altered consumption habits manifested in 1974, and particularly the drastic increases that have occurred in the price of energy in the recent past and may be expected to occur in the future, are likely to have a permanent dampening effect on the rate of demand growth.

Using the 1973 demand of 1.85 trillion kilowatt-hours as a base, the 1980 demand at various growth rates would be as follows:

rate (%/yr)	1980 demand (10^{12} kwh)
0	1.85
2	2.12
4	2.43
6	2.78
8	3.17

In calling into question the need for constructing additional electric generating plants, these observers have tended to insist on the application of more sophisticated and formal statistical techniques for estimating future demand than the electric companies have traditionally employed. These techniques involve a formal effort to identify the major external determinants of electricity consumption--such economic factors as income, the price of electricity, the prices of substitute fuels, as well as demographic factors--and, on the basis of an analysis of past experience, to specify mathematically the relationship between each of these external determining (or exogenous) factors and the variable to be estimated--electricity consumption in this case.

These various competing methods of making electricity demand projections have produced a wide range of differences in result. The most recent forecast of total electricity (net) generation in 1980 made by the National Electric Reliability Council (NERC) for the current National Power Survey is 3.07 trillion kWh, as compared with an actual level of 1.85 trillion in 1973. In contrast, Chapman et al. have produced an econometric model that emerged with a forecast of 2.21 kWh in 1980, obviously projecting a growth rate only a fraction of that implied by the NERC estimate. The major reason for the difference between these two results is that Chapman et al. estimate an elasticity of demand for electricity in the range of -1.00 to -1.22 for the three classes of customers (residential, commercial, and industrial). This implies that a 10 percent increase in price (measured in constant dollars) would eventually lead to sales of 10 to 12.2 percent below the levels which would result if price had not increased. This comparatively high estimated elasticity of demand has an important effect on the ultimate level of consumption projected, because the authors assume an increase of 5 percent per year in the real price of electricity from 1972 to 1980.

Other statistical analyses have produced results implying a considerably lesser elasticity of demand for electricity, and, in consequence, estimate a considerably more rapid rate of growth in the demand for electricity than the rate forecast by Chapman. The Data Resources Inc. model, for example, projects the consumption of electricity will grow by more than 5 percent per year by 1985, which correspons to a generation level of 2.59 trillion kWh in 1980, as compared with the 3.07 the NERC forecast and the 2.21 by Chapman. The estimate by the Project Independence Blueprint of the Federal Energy Administration is roughly consistent with the NERC forecast for 1980.

Moreover, whatever the elasticity of the demand for electricity, it will be greater in the long than in the short run. It takes time for consumption patterns to change in response to alterations in price, not only because habits are always slow to change, but because, even more important, the kinds of energy employed and the efficiency with which they are employed is determined preponderantly by the characteristics of the existing stock of appliances, buildings, industrial and transportation equipment, which can change only as that population is replaced; and finally, because altered price relations can induce adaptations in the technology of utilization only over time. For all these reasons, the full response of demand to recent price changes is unlikely to be achieved in the 1975-1980 period.

For our purposes, it suffices to observe that the demand for electricity must certainly have some price elasticity, and that the prospective increase in the price of electricity may confidently be expected to exert some dampening effect on the growth of consumption; but that, on the other hand, the sharply increasing price of alternative sources of energy, the drying up of supplies of natural gas, and the uncertainties on the part of consumers about the continued availability of oil imports, after the experience of the Arab boycott, will all tend to work in the other direction--that is to say, inducing a shift from those alternative sources of energy to electricity. We conclude that electricity demand will continue to grow, that it will have to be supplied in increasing proportions from coal as well as nuclear power, and that the problem of reducing sulfur dioxide emissions cannot therefore be exorcised by the comfortable assumption that additional generating capacity will be unnecessary.

CHAPTER 9

EFFECTS OF IMPROVED FUEL UTILIZATION ON DEMAND FOR FUELS FOR ELECTRICITY

(Chapter 9 was written by Elias Gyftopoulos and Thomas Widmer under the general supervision of the committee, which reviewed the work at several stages and suggested modifications that have been incorporated. While every committee member has not necessarily read and agreed to every detailed statement contained within, the committee believes that the material is of sufficient merit and relevance to be included in this report.)

INTRODUCTION

This chapter estimates the potential reduction of fuel for electricity in 1985 resulting from improved utilization of fuel in industrial, residential, and commercial end-uses. Because a fraction of the fuel used for electricity is coal, improved fuel utilization is a method complementary to other available methods for reducing sulfuric oxides and sulfates discharged into the atomosphere by electric powerplants. The chapter also evaluates the potential effect on fuel-demand for electricity resulting from some alternate methods of space heating.

PATTERNS OF FUEL SUPPLY AND DEMAND

This section presents statistical data for the U.S. patterns of fuel supply and demand (SRI 1972) in 1968 and projections for these patterns for 1985 (Dupree and West 1972). Although many projections have been made for 1985, for the purposes of this report, we will consider only the projections of the U.S. Department of the Interior (Dupree and West 1972).

In 1968, the amount of fuel consumed in the U.S. was about 57 quads (1 quad = 10^{15} Btu) exclusive of about 3 quads used as feedstock materials. It was distributed among the fuel sources approximately as follows:

Petroleum products	43.5%
Coal	23.0%
Natural Gas	32.0%
Nuclear and hydrostatic head	1.5%
	100 %

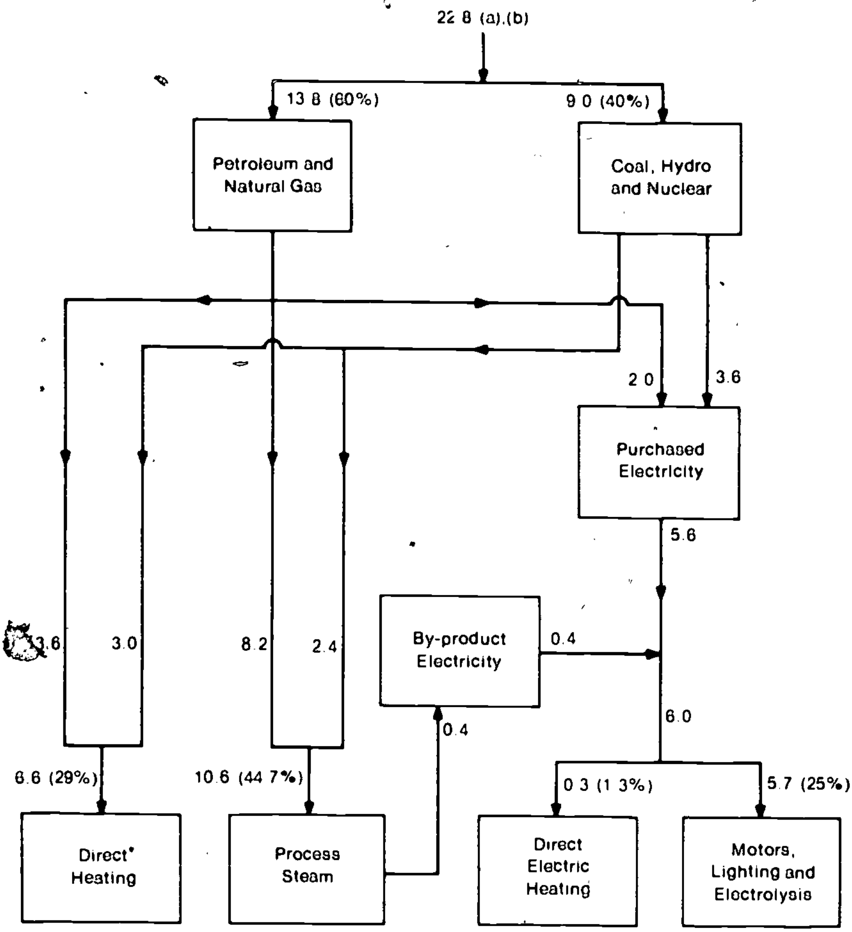
It was consumed in the major sectors of the economy in the amounts shown in the first and second columns of Table 9-1, namely 41 percent in the industrial sector, 34 percent in the residential and commercial sector, and 25 percent in the transportation sector. Some of the fuel was consumed in the form of electricity (columns 3 and 4, Table 9-1) which was primarily (92 percent) from utilities and to a lesser degree (8 percent) generated as by-product of industrial processes. The fuels used in electricity generation were 53.5 percent coal and 46.5 percent others.

The principal end-uses of fuels in industry in 1968 can be classified in the four major catagories shown at the bottom of Figure 9-1 among which the fuels are distributed as follows:

TABLE 9-1

Fuel Consumption by Sector in 1968

Sector	All fuels 10^{15} Btu	All fuels Percentage %	Electricity 10^{12} kwh Generated by Utilities	Electricity 10^{12} kwh Total
Industrial	23.0	41	0.6	0.72
Residential and commercial	19.5	34	0.73	0.73
Transportation	14.5	25	NIL	NIL
Total	57.0	100	1.33	1.45



(a) All fuel values in 10^{15} Btu/year

(b) Does not include 2.1 x 10^{15} Btu equivalent fuel value for gas and petroleum materials used in 1968 as feedstock for chemical products.

FIGURE 9-1: Sources and End-uses of Fuel by U.S. Industry in 1968.

Direct combustion heating	29.0%
Process steam	44.7%
Direct electric heating	1.3%
Motors, lighting and electrolysis	25.0%
	100 %

The principal end-uses of fuels in the residential and commercial sector are shown in Table 9-2. We see from the data in Figure 9-1 and Table 9-2 that process steam raising, space and process heating, and refrigeration and air conditioning in 1968 represented the major end-uses of fuels in sectors other than transportation. These processes consumed over 50 percent of the coal in 1968 as illustrated by the data in Table 9-3.

Several projections have been made about the fuel demand in 1985. For the purposes of this report, the projections of the U.S. Department of the Interior have been used (Dupree and West 1972). The projected demand in the major sectors is shown in the first column of Table 9-4 exclusive of fuels for feedstock materials. It will be distributed among the principal fuel sources approximately as follows:

Petroleum product	42%
Coal	28%
Natural Gas	25%
Others	5%
	100%

Some of the anticipated fuel demand will be supplied by utilities in the form of electricity as whown in the third column of Table 9-4. Comparing the data in Tables 9-4 and 9-1 we see that in 1985 the demand for all fuels is projected to be about two times as large, and for electricity about three times as large as those in 1968. The demand for fuels for electricity generation is projected to be as shown in Table 9-5. We see from this table that 37 percent of electricity will be generated from coal in

TABLE 9-2

Major End-Uses of Residential and Commercial Fuel in 1968 (excluding feedstock)

End-use		Percentage of sector fuel	FUEL			
			Electricity	Petroleum and gas for direct firing	Coal for direct firing	Total
		%	10^{15} Btu[1]	10^{15} Btu	10^{15} Btu	10^{15} Btu
Heating	Space	56	0.48	9.84	0.57	10.89
	Water	13	0.9	1.55	NIL	2.45
Refrigeration and air conditioning		16	3.0	0.1	NIL	3.1
Total		85	4.38	11.49	0.57	16.44

(1) 1 kw-hr of electricity = 10,000 Btu fuel in power plant.

TABLE 9-3

Selected End-Uses of Coal in 1968[1]

End-use		10^6 tons	Percentage of coal consumption %
Industrial	Process steam	87	18.5
	Heating	51	11
Residential and Commercial	Heating	49[2]	10.5
	Refrigeration and air-conditioning	56[3]	12
Total		243	52

(1) Total consumption 13.1 quads or 470 x 10^6 tons at 28 x 10^6 Btu/ton.

(2) Weighted average of direct coal usage and electricity produced by using 53.5% of fuels in the form of coal.

(3) Based on 53.5% of electricity produced from coal.

TABLE 9-4

USDI Projected Fuel Demand by Sector in 1985

Sector	All fuels		Electricity generated by utilities	
	10^{15} Btu	Percentage	10^{12} kw-hr	Percentage
Industrial	41.9	38.5	1.86	45
Residential and commercial	39.7	36.5	2.23	54
Transportation	27.2	25.0	0.04	1
Total	108.8	100	4.13	100

TABLE 9-5

USDI Projection of Demand of Fuels for Electricity Generation in 1985[2]

Fuel	Electricity 10^{12} kw-hr	Percentage of total electricity %
Coal	1.53	37
Hydrostatic head and geothermal	0.25	6
Petroleum and gas	1.07	26
Nuclear	1.28	31
Total	4.13	100

1985 whereas 53.5 percent of electricity was generated from coal in 1968.

POTENTIAL FOR IMPROVED EFFECTIVENESS

Industrial Sector

As discussed in a report to the Energy Policy Project of the Ford Foundation (Gyftopoulos et al. 1974) many opportunities exist for the application of existing technology to the enormous fuel flow in industrial heating processes so as to yield large fuel savings. For example, the bulk of industrial fuel (about 45 percent in 1968) is consumed in raising process steam. Wherever process steam is required in reasonable amounts, an opportunity exists to produce electricity at small cost in fuel consumed. For example, if process steam at 200 psi or 382 F is generated by burning a hydrocarbon fuel, $(CH_2)_n$, over 60 percent of the available useful work of the fuel is lost. Much of this loss may be prevented by burning fuel in a gas turbine and using the turbine exhaust to generate steam (Figure 9-2a), by generating steam at a pressure higher than 200 psi and expanding the steam in a steam turbine to 200 psi at which pressure it is exhausted to process (Figure 9-2b), or by a combination of these two (Figure 9-2c). Figure 9-3 compares a combined system (Figure with the more widely used present practice of spearate generation of steam and electricity.)

Typical results of the electricity generated by the various topping systems are summarized in Table 9-6. The electricity produced, if considered as a by-product of the process heat, should be charged with the fuel consumption over and above that required when process steam is produced directly without the intervening

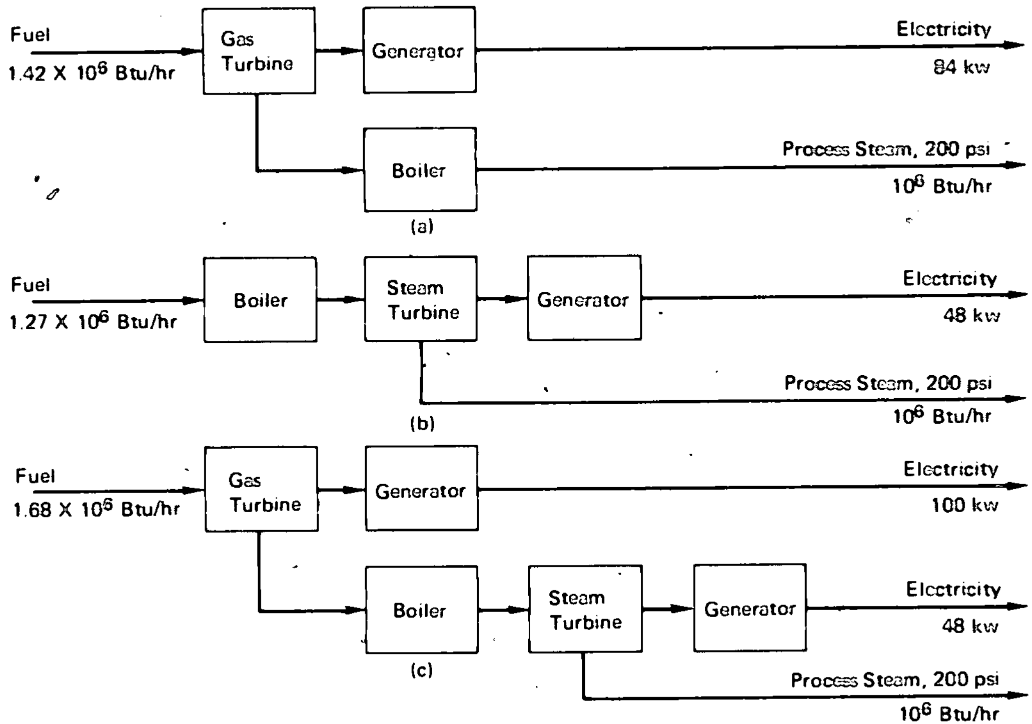


FIGURE 9-2: Combined Process Steam Raising and Electricity Generation Options for Process Steam at 200 psi and 10^6 BTU/hr.

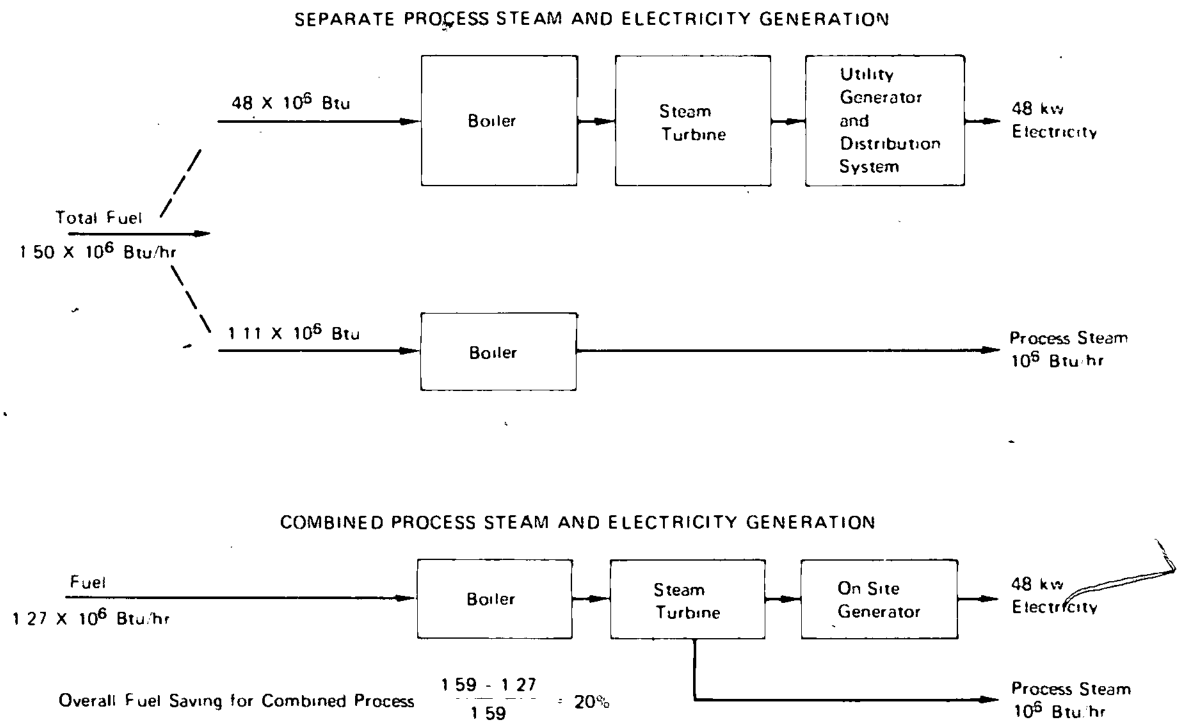


FIGURE 9-3: Comparison of Overall Fuel Requirements for Steam and Electricity Generation With Separate Versus Combined Processes

TABLE 9-6

Kilowatts of By-Product Electric Power for 10^6 Btu/hr of Steam Supplied to Industrial Process

Process Steam Pressure, psi	Steam-Turbine Power, kw		Gas-Turbine Power, kw		Total Combined Gas and Steam-Turbine System Power. kw.
	Alone	Fed From Exhaust of Gas Turbine	Alone	Followed by a Steam Turbine	
50	77		84		
200	49	48	84	100[a]	148
400	34		84		

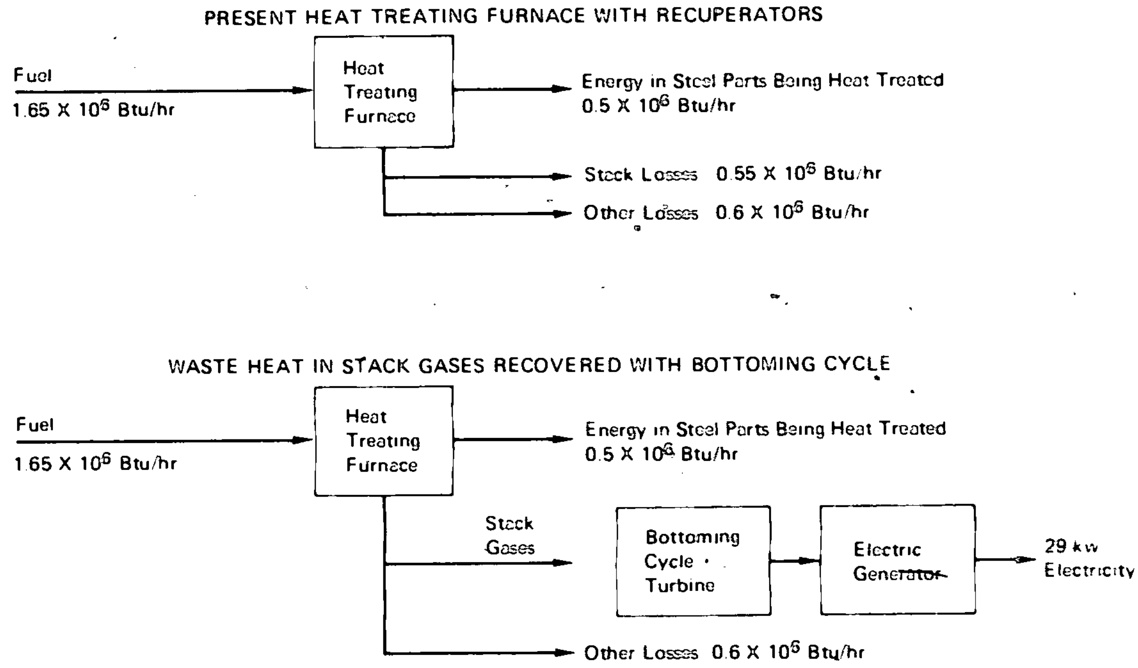
[a] The power of the gas turbine is increased from 84 to 100 kw because some of the available useful work of the fuel necessary for the steam turbine is consumed in the gas turbine.

topping system. On this basis, the fuel consumption for each of the cases shown in Table 9-6 is about 4230 Btu of available useful work for each kw-hr of electricity. These figures translate into an effectiveness of electricity generation of 0.8. The corresponding figure for the best central station powerplant is less than 0.4; that is, electricity is produced at less than half the fuel consumption of the best central-station powerplant.

Other fuel savings can be achieved through use of organic Rankine bottoming systems for recovery of availability from waste heat of industrial combustion processes. Wherever heat is rejected at temperatures 700 F or higher, an opportunity exists to produce electricity at no fuel consumption. A typical arrangement of a bottoming system combined with a radiant tube furnace is shown in Figure 9-4.

In 1968, industrial by-product electricity was 0.12 x 10^{12} kw-hr reducing the amount of fuel consumed by utilities by about 1.2 quads (assuming that, on the average, 1 kw-hr generated by a utility consumes 10,000 Btu of fuel in the powerplant). It is estimated that the amount of incremental fuel consumed by industry for the generation of this by-product electricity is about 0.5 quads and, therefore, that a net fuel saving of 0.7 quads was achieved.

If all process steam could be raised in combination with electricity generation, then the upper limit for industrial by-product electricity generation in 1968 was 0.7 x 10^{12} kw-hr and could be achieved for an incremental fuel consumption of 2.9 quads (Table 9-7). The fuel that would have been saved by the utilities would have been 7 quads and, therefore, the net fuel saving would have been 4.1 quads. In addition, it is estimated that bottoming systems could have generated 0.1 x 10^{12} kw-hr at no fuel consumption in 1968 (Table 9-



Note Diagrams are based upon 1 ton/hour of steel parts processed. Electrical output amounts to 29 kw-hrs per ton of parts and fuel saved by electric utility powerplants averages 300,000 Btu per ton of parts.

FIGURE 9-4: Bottoming Cycle Applied to Radiant Tube Heat Treating Furnace for On-site Generation of By-product Electric Power

TABLE 9-7

Upper Limit for Industrial By-Product Electricity Generation in 1968

Industrial process	Fuel saving method	Total fuel consumption 10^{12} Btu	By-product electricity 10^{12} kw-hr	Incremental fuel required 10^{15} Btu
Process-steam raising	Topping cycles	13.2	0.7	2.9
Direct combustion heating	Bottoming cycles	6.5	0.1	NIL
Total		19.7	0.8	2.9

7). The corresponding fuel that could have been saved by the utilities was 1 quad. In summary, in 1968 total fuel consumed by industry was 23 quads, and fuel saved by means of by-product electricity generation was 0.7 quads out of a maximum possible 5.1 quads.

In 1985, total fuel demand by industry is projected to be 41.9 quads. If by-product electricity generation continues at the 1968 rate, the fuel saving will be:

$(0.7x10^{15})(41.9x10^{15}\ 23x10^{15}) = 127.x10^{15}$Btu
out of a possible maximum of
$(5.1x10^{15})(41.9x10^{15}\ 23x10^{15}) = 9.3x10^{15}$Btu
It follows that maximum by-product electriity generation could result in additional fuel saving of 8 quads in 1985.

Decrease of fuel for electricity can also be brought about through improved effectiveness of industrial processes. An illustration of this decrease is provided by the Hall process for reduction of A1$\underline{2}$O$\underline{3}$ to aluminum metal. In this process, electrolysis is carried out in carbonlined boxes into which carbon rods project. An electrical potential is applied so that the box serves as the cathode and the rods as the anode. Upon electrolysis the alumina is decomposed; the aluminum metal is deposited at the cathode in a molten condition and the oxygen is deposited at the anode.

Considerable variations in the electricity requirements for primary aluminum production exist from plant to plant; typical numbers of production cells range from 13,600 to 16,400 kw-hr per ton of aluminum. Primary aluminum production in the U.S. was 3.25 x 10^6 tons in 1968. Assuming an average electrical demand of 15,000 kw-hr per ton, the electrical consumption by the aluminum industry amounted to 4.9 x 10^{10} kw-hr in 1968, or about 3.7 percent of total U.S. electricity needs.

Analysis of the Hall cell voltage shows that only 1.6 to 1.8 volts, out of almost 5 volts drop across the cell, is required for the basic electrolysis process. The remainder is necessary as a result of voltage drops (resistive losses) across various electrical resistances in the cell circuit. Because the electrolysis voltage is relatively independent of current through the cell, aluminum production is approximately proportional to the current. The parasitic resistive losses, on the other hand, are proportional to the square of the current. It follows that the fraction of electricity effectively utilized for the electrolytic reduction of Al_2O_3 increases as the cell current is decreased. For example, decreasing the current of a typical cell from 105,000 amps to 82,000 amps would decrease electricity consumption per ton of aluminum by 16 percent. Although such current decrease would decrease production per cell by 22 percent total production can be maintained at the desired level by installing more cells, namely at the expense of higher capital costs. In general, the optimum cell current density decreases as power costs increase. At the lower current electrical consumption is only about 12,500 kw-hr per ton of aluminum (Gyftopoulos et al. 1974). Assuming that primary aluminum production will be 10 x 10^6 tons in 1985, the electricity saving would be 2.5 x 10^{10} kw-hr and, therefore, the fuel saving 0.25 quads.

Residential and Commercial Sector

An opportunity exists for reduced electricity demand for refrigeration and air conditioning equipment in the residential and commercial sector. In 1968, refrigeration and air conditioning consumed 20.7 percent of all U.S. electricity and 5.4 percent of all fuels. If the same percentage is valid for 1985, the fuel demand for this end-use would be 5.9 quads. The opportunity exists for application of existing technology to improve the effectiveness of refrigeration equipment. For example, present central air-conditioning systems for homes have a performance index of 8.5 Btu per watt-hr. This can be readily increased to 12 Btu per watt-hr by means of well-known heat-transfer methods. It is estimated that a 30 percent average improvement in performance of all refrigeration equipment is reasonable for 1985. This improvement would result in a fuel saving of 1.8 quads.

Another large factor in electricity consumption is lighting for commercial and public buildings. Recent FEA guidelines for lighting and thermal operations indicate a potential saving of 43 percent in this end-use (FEA). This would represent 133 x 10^9 kw-hr or 1.3 quad fuel saving by electric powerplants in 1985.

POTENTIAL FOR SHIFTING TO ALTERNATE SOURCES FOR SPACE HEATING

This section presents the limiting effects of using alternate methods of space heating.

In 1968, space heating consumed 10.89 quads or 19 percent of all fuels of which 0.57 quads was in the form of coal. If the same percentage is valid for 1985, the fuel demand for space heating will be 20.8 quads (0.19 x 10^{15} Btu). In addition, it is estimated that electrical and coal space heating will gradually increase from 0.48 and 0.57 quads in 1968 to 1.1 and 1 quads in 1985, respectively.

In direct-firing space heating, only a fraction of the heating value of the fuels is used in raising the space temperature, the remaining being lost up the stacks of the burners. The fraction that is used varies widely depending on the type and maintenance of the burner. For our purposes, we will assume that 70 percent of the heating value of the fuels contributes to space heating.

If all the space heating needs in 1985 were to be switched either to pure resistance electric heating, electric heat pump heating, or coal gas heating then the fuel demand would be modified as follows:

Pure resistance heating

Increase in electrical load = $0.7(20.8-1.1)10^{15}$
= 13.8 quads
= 4.04×10^{12} kw-hr
Increase in fuel demand = 40.4-19.7=20.7quads

In the extreme, this increase might result in the following distribution between fuels:

Net increase in coal demand, = 39.4 quads/year = 1640×10^6 tons/year
(1 ton of coal = 24×10^6 BTU)

Net decrease in oil and gas demand, -18.7 quads/year =-8.5×10^6 barrels/day
(1 barrel of oil = 6×10^6 BTU)

Heat pump heating

Assuming a national average coefficient of performance (COP) for heat pumps of 1.8, then

Increase in electrical load $=4.04 \times 10^{12}/1.8$
$=2.25 \times 10^{12}$ kw-hr
Increase in fuel demand =22.5 - 19.7
= 2.8 quads

For the maximum shift to coal, this increase would result in the following distribution between fuels:

Net increase in coal demand = 21.5 quads = 896×10^6 tons/year
Net decrease in oil and gas demand =-18.7 quads =-8.5×10^6 barrels/day

Another possible means of shifting home heating load from oil and natural gas to coal-based energy is the alternative of gas from coal gasifications; assuming a gasification and distribution efficiency of 0.62, demand would be modified as follows:

Net increase in coal demand	$= (20.8 - 1)/0.62 = 31.9$ quads/year $= 1330 \times 10^6$ tons/year
Net decrease in oil and gas demand	$= -19.8$ quads/year $= -9 \times 10^6$ barrels/day

EVALUATION OF CAPITAL COST FACTORS

Costs Related to Improved Effectiveness

The evaluation of the relative benefits of various fuel saving methods necessitates consideration of both capital requirements and fuel pricing practices.

From an aggregate capital availability point of view, it is important to compare the capital for supplying additional fuel with that for saving an equal amount of fuel through improved effectiveness measures.

Some estimates for capital required to supply various forms of energy are listed in Table 9-8. All figures are normalized to the equivalent of one barrel of oil per day.

For an industrial installation needing 1 megawatt of electricity, if this electricity were to be provided by a coal-fired powerplant, with a load factor of 0.7, the capital required would be:

Coal-fired powerplant (AGA 1974)	\$ 456,000
Distribution	180,000[a]
Coal supply (28 barrels of oil per day equivalent annual average)	48,000
Total	\$ 684,000

(a Note: Capital investment in distribution system for industrial customers assumed to be \$13,000 per 1760 kw-hr of electricity per day which is energetically equivalent to 1

TABLE 9-8

Approximate Capital Costs for Supplying the Fuel Equivalent of One Barrel of Oil Per Day (6×10^6 Btu/day) in Various Forms

1. GAS FROM COAL GASIFICATION

Coal production (Eastern deep mine)	\$ 2,800 (a)
Gasification plant (AGA 1974)	10,000
Transmission and distribution system	7,400 (b)
	\$20,200

(a) At 0.62 plant and distribution efficiency

(b) Assumes \$1.35 per million Btu (\$8.10 per barrel of oil equivalent) as the average cost of transmission and distribution to residential customers in the Eastern United States. Fifty percent of this figure is assumed to derive from capital charges computed at 20 percent annually, namely

$$\text{Capital cost} = 8.10 \times \frac{365}{0.2} \times 0.5 = 7400.$$

2. OIL FROM NEW DOMESTIC SOURCE

Production (off-shore) (7)	\$5,000-\$8,000 (c)(d)
Refining	1,000
Transportation and distribution	3,000 (e)
	\$9,000-\$12,000

(c) Includes bonuses paid on leases

(d) Estimates for shale oil, synthetic crude from coal, or tertiary recovered oil vary from \$10,000 to \$20,000 per barrel per day.

(e) For Alaskan oil, the pipe line alone costs \$5,000 per barrel per day.

3. ELECTRICITY FROM COAL-FIRED POWERPLANT

Coal production (Eastern deep mine)	\$ 5,500 (f)
Electric plant (AGA 1974)	36,500 (g)
Transmission and distribution system	20,500 (h)
	\$62,500

(f) Electricity generated at 0.34 plant and distribution efficiency.

(g) Estimate based on average capital cost \$456/Kw for new coal-fired generating plants greater than 1,300 Mw capacity that could be on-stream by 1981, and load factor 1.0.

(h) Assumes 1.28¢ per kw-hr as average cost of distribution to residential customers, with 50 percent of this figure attributed to capital costs, as in note (b) above.

barrel of oil per day; comparable figure used for residential customers in Table 9-8 is $20,500.)

On the other hand, suppose that the industrial installation has a potential application for a 1.0 megawatt bottoming-cycle engine generator to recover waste heat from a large continuous metal processing furnace with an annual utilization factor of 0.7. At $400 per kw, the capital required would be $400,000 and the fuel consumed would be zero. To this may be added $62,000 for 50 percent emergency supply diesel generators and, therefore, the capital investment would be $462,000.

From these results we see that the investment for incremental electricity from a coal-fired powerplant would require about 48 percent more than that for the on-site bottoming-cycle system. For an oil-fired powerplant, the advantage of the bottoming cycle is even greater.

On the other hand, whether the advantage of the fuel-saving over the increased fuel supply method will be evident to the industrial firm depends on fuel pricing policies. If the price of fuel reflects the true cost of new fuel supplies then the bottoming cycle is advantageous. If the price of fuel is based on averages over old and new sources then the bottoming cycle and, therefore, the advantage of the fuel-saving method may not be as decisive as the preceding capital requirement estimates indicate.

To illustrate this point, we shall assume 2.5 cents per kw-hr as being representative of the price paid by an industrial customer for electricity. By assuming a ten-year sum-of-year- digit depreciated life time for the bottoming cycle generator, a 0.3 cent per kw-hr operating and maintenance cost and a 70 percent duty cycle, we obtain the following break-even capital costs for on-site power generation with bottoming cycle system:

Required after tax (52%) return on investment	Break-even capital cost for bottoming cycle system
12%	546 $/kw
15%	469 $/kw

We see that the bottoming cycle capital requirement of 462$/kw is comparable with the break-even cost determined from the price of electricity of about 2.5 cents per kw-hr. It follows that for the assumed price of electricity, the user most likely will decide to buy electricity rather than install a bottoming-cycle system. The reason for such a decision is, of course, that the assumed price of electricity does not reflect the true cost of new supplies.

Costs of Fuel Shifting for Space Heating

The demand for fuel for residential and commercial space heating could be shifted from oil and natural gas to either electricity generated from coal or to alternate sources such gas produced from coal.

Table 9-9 lists estimates of capital requirements for three alternate methods of space heating, electric resistance, electric heat pump, and gas from coal, all of which use coal as the primary fuel. The calulations are based on residential heating units requiring 150 x 10^6 Btu per year, or 0.07 equivalent barrels of oil per day.

We see from this table that electric heat pumps offer the lowest total fuel consumption of the three cases. Gas from coal gasification on the other hand, affords a significant saving in capital investment over either form of electrical space heating. It should be noted that the investment advantage for the gas from coal

TABLE 9-9

Capital Investment of Alternate Home Heating Methods Using Coal as Primary Fuel

	Home heating method		
	Electric resistance	Electric heat pump	Gas from coal gasification
Plant efficiency:			
$\frac{\text{energy to home}}{\text{energy from coal}}$	0.34	0.34	0.62
Home furnace yield:			
$\frac{\text{heat to home}}{\text{energy to home}}$	1.0	1.8	0.7
Barrel per day equivalent coal consumed per equivalent barrel of oil per day of heat supplied to home	2.94	1.63	2.30
Capital investment per equivalent barrel of oil per day of heat supplied to home			
Supply plant[a]	$ 96,100	$ 53,400	$ 44,400
Home heating plant[b]	7,200	28,600	14,300
Total	$103,300	$ 81,000	$ 58,700

[a] Load factor for all plants 0.65.
[b] Based on home heating unit costs of $500, $2,000 add $1,000 per home from baseboard resistance, heat pump, and gas-combustion furnaces, respectively.

gasification approach will be increased even further when adjustment is made for the high percentage of existing gas home-furnaces which would have to be replaced if either electric heating concept were adopted.

SUMMARY OF DEMAND MODIFICATION ALTERNATIVES

The limiting incremental values of effects of demand modifications established in the preceding sections can be allocated to coal. The results are summarized in Tables 9-10 and 9-11 in million tons of coal per year on the basis of 1 ton of coal = 24×10^6 Btu.

Only a fraction of these effects can be achieved, however, by 1985 partly because some industrial plants may be too small in size to justify a modification, partly because of fuel-pricing policies that do not make changes attractive, and partly because of institutional constraints. For example, a plant may need process steam in amounts which do not justify economically the installation of a topping system, or the price of electricity may be low enough so that the investment for an on-site system cannot be recovered in sufficiently short time. Finally, there may be state or local utility regulations which prohibit the sale of surplus electricity by an industrial plant to a utility.

EFFECTIVENESS OF FUEL UTILIZATION IN A PROCESS

In attempting to evaluate the opportunity for fuel saving in a particular process, we need to know the minimum fuel requirement for the process so that we can compare it with the fuel consumed under current practice and obtain a measure of the effectiveness of that practice. The minimum fuel requirement can be evaluated by means of the thermodynamic concept of

TABLE 9-10

Maximum Potential Shift in Coal Requirements Resulting from Selected Improvements in Electrical Effectiveness at Point of Use. (1 quad = 24×10^6 tons of coal)

Demand modification	Maximum incremental coal consumption (tons/year from 1985 baseline USDI forecast)
On-site generation of by-product electricity in industrial processes	-333×10^6
Re-optimization of Aluminum electrolysis process to lower current density (1)	-10×10^6
Improved performance residential & commercial refrigeration air conditioning equipment	-75×10^6
Relamping of commercial & public buildings to FEA lighting standard	-54×10^6

(1) Aluminum electrolysis, which accounts for about 7.5% of industrial electricity, is shown as an example of improved industrial process effectiveness. In order to determine potential savings for improvements in other electrical-intensive processes, it will be necessary to perform a detailed study of each individual industry.

TABLE 9-11

Maximum Potential Shift in Coal Requirements Resulting from Shifting All Residential and Commercial Space to Methods Based on Coal as Primary Source

Demand modification	Maximum incremental coal consumption[a] (tons/year from 1985 baseline USDI forecast)
Shift all space heating to electric resistance	$+1640 \times 10^6$
Shift all space heating to electric heat pumps	$+396 \times 10^6$
Shift all space heating to gas from coal gasification	$+ \quad \times 10^6$

[a] The corresponding reduction in oil and natural gas consumption is about 9 million barrels of oil equivalent.

available useful work. Readers unfamiliar with the foundations of thermodynamics and the concept of available useful work might consult the article on "Principles of Thermodynamics" in the 1974 Edition of the Encyclopedia Britannica

In a report prepared for the Energy Policy Project of the Ford Foundation (Gyftopoulos et al. 1974) the concept of available useful work was used to evaluate the effectiveness of fuel utilization in five energy-intensive industries. Table 9-12 lists the industries, outputs, specific fuel consumptions, and total fuel consumed in 1968. In addition, the table lists the minimum specific fuel requirements, and minimum total fuel requirements for these industries. It is seen from these data that the average fuel effectiveness for the five industries under consideration is 1.17 x 10^{15}/9.2 x 10^{15} =13 percent. The average fuel effectiveness of 13 percent should not be confused with the efficiency value of 70 percent or higher reported in the literature. The latter figure represents the average fraction of the heating value of the fuels that are used in industrial processes.

The large margins that exist between current practives and minimum theoretical requirements indicate the potential which is available for major long-term reductions in fuel consumption through basic process modifications.

TABLE 9-12

1968 Product Output and Fuel Consumption for Selected U.S. Industries

Industry	Industry Output (tons/yr)	Specific Fuel Consumption (Btu/ton)	Total Fuel Consumption (Btu/yr)	Percentage of Industrial Sector Fuel	Theoretical Minimum Specific Fuel Consumption Based Upon Thermodynamic Availability Analysis (Btu/ton)	Minimum Total Fuel Requirement (Btu yr.)
Iron and Steel	131×10^6	26.5×10^6	3.47×10^{15}	15.2	6.0×10^6	0.79×10^{15}
Petroleum Refining	590×10^6	4.4×10^6	2.6×10^{15}	11.4	0.4×10^6	0.24×10^{15}
Paper and Paperboard	50×10^6	39×10^6	1.95×10^{15}	5.4	[b]Greater than -0.2×10^6 smaller than $+0.1 \times 10^6$	0.00
Primary Aluminum	3.25×10^6	190×10^6	0.62×10^{15}	2.8	25.2×10^6	0.08×10^{15}
Cement	72×10^6	7.9×10^6	0.57×10^{15}	2.5	0.8×10^6	0.06×10^{15}
		TOTAL	9.2×10^{15} [a]	388	-	1.17×10^{15}

[a] Includes heating value of waste products (bark and spent pulp liquor).

[b] Negative value means that no fuel is required.

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CHAPTER 10

SOME METHODS OF REDUCING SULFUR OXIDES FROM POWER PLANTS

(Chapter 10 was written by Harry Perry under the general supervision of the committee, which reviewed the work at several stages and suggested modifications that have been incorporated. While every committee member has not necessarily read and agreed to every detailed statement contained within, the committee believes that the material is of sufficient merit and relevance to be included in this report.)

In this chapter a number of possible techniques for reducing emissions of sulfur oxides from powerplants are discussed and evaluated. They include:

(1) improved efficiency of conversion of fuel to electricity (this would reduce pollutant emissions per unit of electricity generated);
(2) shift to nuclear generation as rapidly as possible since no sulfur oxide (or particulate) is emitted from nuclear plants;
(3) shift fossil fuel plants to lower sulfur fuels;
(4) removal of sulfur from coal before combustion, or of the sulfur oxide after combustion, but before it enters the stack: the techniques to be considered are

(a) coal preparation (conventional and advanced technology);
(b) solvent refined coal;
(c) low sulfur, low BTU gas from coal;
(d) fluidized bed combustion;

(5) shift fuel consumption from electricity to pipeline grade gas made from coal or to solar energy.

Other possible techniques are discussed in Chapters 11 and 12.

ASSESSMENT OF THE POTENTIALS FOR IMPROVED EFFICIENCY IN THE CONVERSION OF FUEL TO ELECTRICITY

Summary

Methods of generating electricity at increased efficiencies (in order to reduce the pollution load per unit of electricity generated) are not expected to come into widespread use until 1985 or later. As important as it is to continue R&D on these advanced power cycles, they offer no sollution for reducing sulfur oxide or particulate emissions in the period between 1975 and 1985.

The average heat rates (i.e. the number of heat units required to generate one net kw-hr. of electricity) for utility power plants in the U. S. declined steadily from 25,175 BTU/kw-hr. in 1925 to 10,479 BTU/kw-hr. in 1972. This increase in conversion efficiency was the result of a number of technologic improvements, the most important of which were those that permitted steam turbine plants to operate at higher temperatures and pressures. Through experience it has been found that the best current practical efficiency of operation is 38 to 39 percent. (Heat rate approximately 9000 BTU/kw-hr.).

The average heat rate was lowest in 1968 when it was 10,371 BTU/kw-hr., and it has increased steadily since that time for a number of reasons. The leveling off in the efficiency of new plants, the more widespread use of nuclear

plants with their higher heat rates and the need for additional electricity for pollution control are among the most important causes for the increase.

With the efficiency limitation that exists when using the steam turbine cycle, the electric utilities have started to examine the potential for using advanced power cycles which have the potential for using less fuel per kw-hr. and thus reducing the amount of pollution generated. Among those systems receiving the greatest attention are:

(a) combined steam turbine-gas turbine systems (open cycle);
(b) magnetohydrodynamics (open cycle); and
(c) the use of binary cycles or working fluids other than steam.

Steam Turbine-Gas Turbine Combined Cycles (open cycle)

In this cycle a fuel is burned in a gas turbine and part of the energy released generates electricity in the gas turbine. The hot gases leaving the turbine are then used to generate steam for use in a conventional steam turbine. Steam turbine-gas turbine combined cycles are currently being used commercially on medium sized boilers using natural gas as a fuel. However, plants using these cycles would be unable to use coal as a fuel (unless it were first gasified and the particulates removed); they do not operate at sufficiently high temperatures to get the high conversion efficiencies desired, and when the temperatures are increased, do not have the service life required in a base load utility plant.

A number of new developments will be required if these shortcomings are to be overcome. Efficient and low cost processes will have to be devised to convert coal into a clean gaseous fuel, turbine designs will have to be greatly improved to obtain the turbine life needed at central stations, and materials of construction will have to be developed that are able to operate at the high temperatures needed for obtaining high conversion efficiencies.

Estimates vary as to when commercial plants using this type of advanced technology with coal as a fuel will be in operation, but no large number of such plants could be operational before 1985.

Magnetohydrodynamics (open cycle)

Magnetohydrodynamics (MHD) is a method for converting the energy in a hot moving gas stream into electricity by passing a conducting gas through a magnetic field. The gas must be heated to very high temperatures to make it conducting, and even the, "seed" materials may have to be added to obtain the levles of gas conductivity required to operate satisfactorily. In open cycle MHD, the hot gases leaving the MHD duct would be used to raise steam for generating electricity in a conventional steam generator.

Numerous studies have been conducted both in the U. S. and abroad aimed at solving the multitude of problems that still must be overcome before MHD is commercially available. A demonstration size unit, using natural gas as a fuel, is now being tested in the USSR. Even when using clean fuel, MHD ducts still need to be developed that will have a useful life in the extreme temperature conditions expected in ducts; methods for recovering the seed material and removing nitrogen oxides must be found and materials must be developed for the electrodes that will have the needed electrical properties and the ability to withstand high temperatures for extended periods.

If coal were first converted into a clean gas (free of particulates) before it was burned in an MHD unit, the same problems would have to be overcome as when natural gas is the fuel. If coal were to be used directly as the fuel for MHD generation, all of these already difficult problems would be compounded. The presence of coal ash in the gases in the form of a liquid slag, could cause serious erosion and corrosion problems that could shorten insulation, duct and electrode life. In addition, seed recovery will be much more complex in the presence of coal ash.

If MHD could be used commercially, conversion efficiencies of from 50 to 60 percent should be attainable. While there is considerable controversy over whether a greatly expanded research effort can be justified, there is general agreement that widescale commerical application will not occur until 1985 or later.

The Use of Binary Cycles or Working Fluids other than Steam

The efficiency limitations of modern steam electric plants arise from the high temperatures and pressures required when using water (steam) as the working fluid. In order to extend these currently used steam conditions, special steels would be required and the capital costs would exceed the costs of the fuel savings. If a working fluid which possessed a higher temperature than steam at a given pressure were used, it would be possible to increase the quantities needed of the more expensive second fluid, a binary cycle would be used with the second fluid being used for the high temperature part of the cylce and a conventional steam cycle being used at the lower temperatures.

A few such binary plants were built and operated in the mid-1930s using mercury as the second fluid. Because of health hazards and a variety of maintenance and operating problems, these plants were abandoned. The binary cycles now under consideration would probably use potassium (or a mixture of sodium and potassium) as the high temperature fluid because of the experience gained with using these fluids in turbines in Atomic Energy Commission and NASA programs. Other working fluids (carbon dioxide, helium) might cause fewer operating problems and are also being considered for possible use.

Efficiencies as high as 50-55 percent might be obtained in binary plants but, as with the other advanced power cycles, extensive commercial utilization will not occur until the period beyond 1985.

SHIFT TO NUCLEAR GENERATION AS RAPIDLY AS POSSIBLE

Summary

If the necessary Federal and State actions are taken to restore the originally planned nuclear capacity (with associated reduction in particulate and sulfur oxide emissions) coal consumption could be reduced by about 75 million tons in 1980.

Commercial nuclear power plants use light water reactor technology (except for one demonstration and one full scale gas-colled reactor) but are only now beginning to produce significant quantities of electricity. There are currently 32,500 MW of capacity with operating licenses, 56,800 MW of capacity are under construction and 147,000 MW that have limited work authorizations are being planned. Because of the difficulty that utilities are now having in raising capital for construction of new plants and the higher capital costs of nuclear plants compared to fossil fuel plants, a number of utilities have announced that they will delay or cancel construction of nuclear plants that had been ordered. Moreover, since many utilities are able to "pass through" to the consumer (without waiting for regulatory approval) the increased costs of fuels, an economic incentive is provided to continue to operate existing fossil fuel plants and to build new ones. Under other conditions new nuclear plants which do not emit particulates or suflur oxide, but which have higher capital costs and do not benefit from the automatic fuel clause would be constructed. As a result, construction schedules for nuclear power plants are being delayed. The AEC's 1972 projection of 1980 nuclear capacity was 132,000 installed megawatts. By last year, (1973) the forecast had been reduced to approximately 100,000 megawatts. The current projection (late 1974) is for nuclear capacity in the range of 60 - 70,000 megawatts by 1980.

FEA estimates[1] that nuclear plant construction schedules could be accelerated so as to permit recovery of about 30,000 megawatts between now and 1980 yielding a total installed capacity in that year approaching 100,000 megawatts. To accomplish this, it would be necessary for state regulatory commissions to take steps to alleviate the financial strains on utilities. Some action on the federal level to assure priorities for critical materials and components will also probably be needed. Without this assistance, it is unlikely that the industry will be able or willing to finance capacity in excess of 60 - 70,000 megawatts by 1980.

If these actions are taken and 30,000 MW of nuclear capacity, which would otherwise not be operational, are on stream by 1980 it would reduce coal consumption in that year by about 75 million tons.

However, using nuclear fuels for generating electricity creastes other types of environmental problems. These include possible radiation releases during routine plant operation and in the event of an accident, unresolved problems with respect to the long term disposal of high level radioactive wastes, and prevention of the theft of highly radioactive material from which nuclear weapons could be produced.

SHIFT TO LOWER SULFUR FUELS

Summary

Existing and projected shortages of natural gas will further reduce the amount of this clean fuel (low in sulfur, low in particulates) available for electric utility use. Domestic petroleum production has not been able to supply domestic demand for a number of years and new national policies are aimed at further reducing the imports that have made up the deficiency. As a result the electric utility industry will have even less petroleum to use in the future than it has in the past. Eastern low-sulfur coal reserves are large but much ~~of the reserves~~ are not available to the electric industry. Existing production capacity for these low-sulfur coals is inadequate and is expected to remain so. By shifting available low-sulfur coal to plants not meeting primary standards from plants which could burn higher sulfur coal and still meet the primary standards, some improvement in ambient air quality could be achieved. It is estimated that a shift of as much as 36 million tons could be made to reduce the tonnage in violation by about 15 percent. Low sulfur western coals will be usable in new coal fired plants designed to burn them if transportation capacity is increased but their use in retro-fitted plants will be limited.

Natural Gas

Since 1968, except for the Alaskan natural gas discoveries, the amount of gas that has been consumed each year has been greater than that which has been discovered. As a result there has been a steady deterioration in the absolute quantity of gas in the proven reserve category. Natural gas production on the other hand continued its rapid growth until about 1973; since then production has about leveled off. This is the result of inadequate gas supplies since unfilled demand for gas, the cleanest of the fossil fuels, remains at an all time high.

As a result of the apparent peaking of production, gas curtailments have been made by the pipeline transmission companies to a large number of gas distribution systems with the greatest curtailments made for those that serve geographic areas far from the gas producing regions. During the winter period of 1973/1974 curtailments to East Coast gas distributors were approximately 25 percent. Some transmission companies have announced that curtailments on their system during the 1974/1975 heating season will be even greater. Transco, the gas transmission line serving many East Coast distributors, has indicated that curtailments could reach 30-35 percent this winter.

Priorities have been assigned to various types of gas consumers. The highest priority customers are mainly the residential and small commerical users of gas who would have the greatest difficulty in shifting to other fuels and who, even from a pollution abatement standpoint, should receive the highest priority since, in nearly all instances these customers are equipped with short stacks for discharging the combustion products into the atmosphere. If they used a polluting fuel they would make a much greater contribution to deteriorating ambient air quality than large consumers with tall stacks the use of which dilutes air pollutant concentrations by providing for mixing of the pollutants with large volumes of cleaner air before reaching ground levels.

As a result of all of these factors <u>less</u> natural gas will be available to the electric utility industry and to large industrial customers than there has been in the past. In the President's Message to Congress on October 8, 1974, he urged that all existing plants burning oil and gas that could be converted back to coal should do so, and that all plants due to come on stream that were designed for these other fuels should be modified to burn coal.

Petroleum and its products

A small quantity of petroleum products is indispensable to the operations of the utility industry. Coal fired plants need petroleum to start up new furnaces and to stabilize coal fired flames when the boilers are operated at low loads. In addition, small quantities of petroleum products are needed for diesel electric generator sets and to fuel gas turbines used largely for peaking operations.

The bulk of the petroleum used by the utility industry, however, is residual fuel oil burned to supply the heat for base load steam electric generating plants. Some of the plants using residual oil were converted from coal to oil to meet the sulfur oxide air pollution regulations.

National policy is now aimed at reducing the use of oil in utility boilers in order to reduce oil imports; shifting to low sulfur oil to reduce sulfur oxide and particulate emissions would frustrate this national goal. Even the discovery of large domestic resources of either oil or gas on the Outer Continental Shelf (particularly the OCS Atlantic which is close to the oil burning plants and to regions with high ambient concentrations of sulfur oxides) would probalby not result in any significant change in this situation. If the OCS Atlantic leasing were to start as early as 1975 and if important discoveries were made, it would be 1979 or 1980 before large volumes of these fuels would be available for use from this source. Moreover, it is abvious that use of these fuels, which are in short supply now and are certain to be in the future, by the utility industry would not represent the best use of these fuls to obtain short term environmental relief.

There are approximately 64,300 MW of oil burning steam electric capacity in operation in 1974 with 24,000 MW of new oil burning capacity being constructed. Of the existing capacity, 23,600 MW at 73 plants, or about 30 percent, could be converted to coal. These plants are concentrated mainly along the East Coast--New England and the Mid-Atlantic states--with a few in mid-western areas where residual fuel can be delivered at low cost by water. Because of the geographic locations of these plants with respect to coal suppliers and the high concentration of plants in a relatively few areas, supplying coal that they require, even coal with a high sulfur content, may be more difficult than it first appears because of a shortage of coal production capacity and of railroad cars.

Coal

The coal resource base of the U.S. is large anough that, no matter what projections are made for its use as a boiler fuel or for its conversion into synthetics, there will be adequate supplies available to last until beyond the end of the century. Unfortunately, most of the coal currently being mined in the U.S. has a higher sulfur content than the new SOx source performance will permit (in the absence of stack gas desulfurization). In 1972, the electric utility industry consumed 352.9 million tons of coal of which only somewhat over 1 percent had a sulfur content 0.5 percent or less, about 12 percent had a sulfur content of 0.6 to 1.0 percent, 56 percent had sulfur content between 1.1 and 3.0 percent, while 31 percent had a sulfur content over 3 percent. The average sulfur content sulfur content of coal provided to electric utilities was approximately 2.5 percent. From FPC records (which have been kept since February 1973) the average sulfur content of coals delivered each month to electric utilities was 2.3 percent until May 1974 when it rose to 2.4 percent. In commenting on this increase in sulfur content the FPC noted "In the past few months as coal purchases have continued to register year-to-year gains, the bulk of the increase has been in the higher sulfur categories."

The increase in coal production in the first ten months of 1974 of about 5.0 percent over the comparable period of 1973 appears to have come mainly from increasing the capaicty of old mines (working additional time, and opening up new sections) rather than from the opening of new mines. Immediately following the passage of the Coal Mine Health and Safety Act of 1969, 21.5 million tons of deep mine capacity was closed in 1970, 19.7 million tons in 1971 and 21.0 million tons in 1972, for a total of 62.2 million tons. This represented a total of 1585 mines of which 1341, or about 85 percent, were mines producing less than 50,000 tons per year. With the disappearance of the small underground mines that were unable to meet the new health and safety regulations and with the large demand for coal and the high price it commands, the number of closings in 1973 and 1974 was probably smaller but is still probably in the range of 15 million tons per year.

Deep mine openings in these same resulted in added capacities of 33.2 million tons, 23.5 million tons and 17.6 million tons respectively, for a total of 45.6 million for the three year period. Deep mining capacity, therefore, decreased 16.6 million tons in three years.

The new strip mine productive capacity that was installed was 26.5 million tons in 1970, 56.1 million tons in 1971 and 54.6 million tons in 1972 for a total for the three years of 137.2 million tons. No data are available on strip mine closings but the total capacity that was involved can be estimated to be very small. The major new strip mining capacity added in 1970 was mainly in Arizona, Kentucky (mostly Eastern) and West Virginia. By 1972, although development of Western coal deposits had started in order to exploit their low sulfur content, the largest capacities added were still in Kentucky (17 million tons), Ohio (9 million tons), Alabama (7.6 million tons) and West Virginia (3.9 million tons). Five new mines were opened in Wyoming with a total capacity of 5.4 million tons.

Total net new mine capacity added between 1970 and 1972 was about 120 million tons. With the uncertain future of sulfur oxide regulations as well as the deep division of opinion about whether sulfur oxide control technology had been "proven" to be commercial, it appears that construction of new Eastern underground mines has been slowed down appreciably. This uncertainty has probably also had an impact on the rate of development in the low sulfur coals that can be strip mined at low cost in Wyoming, Montana and North Dakota. If sulfur oxide control technology is commercially available, the new markets for the low sulfur Western coals that had been anticipated in order to meet sulfur oxide regulations would be depressed and the very large investments in these new mines would have been premature. On the other hand, if stack scrubbing technology is still unproven and sulfur oxide regulations are to be waived by regulators for only limited periods, then investments in new and expensive underground mining capacity for high sulfur coals cannot be justified. It is for these reasons that it is important that the coal and utility industry know what the state of technology really is and what national policies will be followed with respect to sulfur oxide control. Until these questions are resolved, investments in the new mines and generating plants (that it is now clear will be badly needed) will not be made.

In the meantime some of the available low sulfur coal is being used at utility plants which would be able to meet the national emission standards using coals with a higher sulfur content than they are now using. In January 1974, the Bureau of Mines[2] estimated a shortage of low sulfur coal of 243 million tons in 1975 and 275 million tons in 1980. The EPA issued a report[3] using updated Bureau of Mines data that indicated that the demand for low sulfur fuels would be 273, 295 and 329 million tons in 1975, 1977 and 1980 respectively. These estimates were based on restrictive assumptions transferring coal from where it is unacceptable to where it is acceptable. Estimates of the shortages of clean coal for 1975 under three alternative transfer assumptions were:

	1975 Deficit (millions of tons)
No transfers	273
Intrastate Transfer	253
Intersector transfer	237
Intraregional transfer	191

The most reasonable assumption is that intersector transfer could be arranged so that this practice, if implemented, would reduce the deficit in 1975 by 36 million tons.

With proven stack scrubbing technology, any U.S. coal could be used regardless of its sulfur content. The rate at which stack scrubbers could be installed is discussed below, but the time required is in the same order as the time to open up new mines[4]. However, if stack scrubbing (or other control devices) are not "proven" there are still adequate reserves of low sulfur coal to meet the air quality standards set for some areas. The recoverable reserves of low sulfur Eastern coal with a sulfur content of 1.0 percent or less are estimated at about 19 billion tons[5,6]. Much of this is high quality coking coals. Some of the coal is owned by steel companies and are dedicated to use in the manufacture of coke needed to produce pig iron. Moreover, there is insufficient mining capacity to produce this coal and there is no rush to construct new capacity until the sulfur oxide issues are more clearly resolved.

Estimate of coal production (in thousands of tons) by sulfur content in the Appalachian and Midwestern regions are shown below for 1975, 1977 and 1980:

	1975			1977			1980		
	Sulfur Content, Percent			Sulfur Content, Percent			Sulfur Content, Percent		
	<1.0	1.1-2.0	>2.0	<1.0	1.1-2.0	>2.0	<1.0	1.1-2.0	>2.0
Appalachian	202,100	94,050	135,250	218,150	100,800	144,325	251,850	113,900	160,350
Mid-Western	5,700	8,150	152,300	6,200	8,700	163,000	6,775	9,450	179,875
	207,800	102,200	287,550	224,350	109,500	307,325	257,925	123,350	340,225
Total	597,550			641,175			721,500		

Production is expected to increase by 123,950,000 tons (20 percent) in this period (1980) but the share of coal with a sulfur content that is 1 percent or less is expected to remain about the same, 35 percent of the total, while the share with a sulfur content between 1.1 and 2.0 percent will also remain constant at 17 percent of the total. However, and additional 50 million tons of less than 1 percent sulfur coal will be produced and could be used where it is needed most.

The very large reserves of low sulfur coal are found in the West at great distances from the markets and in areas where the construction of large new mines would have a major impact on socio-economic conditions in the region. If this coal is to be used, it will require three or four years to bring on the first of the capacity and up to 10 years or more to construct all the new capacity that might be required in order to meet SOx regulations. Many of the existing coal-fired plants would be unable to use these Western coals, even if the mining capacity was available since these coals require different types of mills, feeders and combustors. Even if they were physically able to be used, in nearly all instances it would be at the expense of boiler derating and this large cost would have to be included in making tradeoffs between stack scrubbing and using Western coals.

There already exists a serious shortage of coal cars for transporting coal from existing mines to markets for what is usually a relatively short distance. If the much longer haul to bring Western coals to Appalachian markets were used, the transportation shortage would be severely aggravated for at least 2 to 3 years even with Government intervention and longer without.

REMOVE SULFUR FROM COAL BEFORE AND DURING COMBUSTION

Summary

Proven conventional coal cleaning methods can reduce the sulfur content of coal significantly. However, unless the original sulfur content is already low enough to nearly meet the sulfur oxide emission standards, conventional coal cleaning methods will not bring most coals into compliance with source performance standards for SOx. For existing plants in rural areas, coal cleaning can reduce sulfur content of the coals used by about 40 percent. Advanced coal cleaning methods may be able to increase the amount of sulfur and ash removed but all of the processes are in their early stages of development and many may be high cost for the extra sulfur removal that is accomplished.

A low sulfur - low ash product can be produced from coal and commercial plants may be in operation by 1982-1983. The products made from a coal with costs of 80 cents per million BTU will cost in the range of $1.75 to $2.00 per million BTU. Alternatively, a low BTU - low sulfur gas can be made from coal (80 cents per million BTU) with costs in approximately the same range. Commercial plants making gas may be operational in 1981 or 1982.

High sulfur coals may be burned directly in an environmentally acceptable way in a fluidized bed operated either at atmospheric or elevated

pressure. A full scale unit designed for atmospheric pressure could be in operation by 1980 or 1981 and a pressurized unit approximately two years later. If fluidized bed combustion can be successfully developed, it should produce a clean fuel for boiler use that is less costly than either a low sulfur - low ash coal or low BTU - low sulfur gas produced from coal.

Conventional Coal Cleaning Technology

Besides substituting a low sulfur for a high sulfur fuel to reduce the SOx formed, the sulfur can be removed from the fuel before combustion. Methods for removing sulfur from coal before it is burned have been widely used for many years. The basic purpose of coal treatment for most uses to which coal was put, was to reduce the ash content of the coal but during the treatment process some of the sulfur was also removed. When the coal is to be used to make coke, special efforts are also made to remove as much sulfur as possible while reducing the ash content, since the presence of sulfur in the coke significantly increases the costs of making pig iron.

Sulfur does not occur as an element in coal but it is found in chemical combination in three forms: combined with the organic coal substance, with iron which is found as discrete particles of pyrite in the coal substance and with calcium and iron in the form of sulfates. Sulfate sulfur is always present in small quantities.

The percentage of sulfur that occurs in coal, as pyrite, varies from about 20 to 80 percent of the total sulfur. For U. S. coals with a low sulfur content most of the sulfur is in the organic form while for high sulfur content coals most of the sulfur occurs in one or more of the pyrite forms. For example, an Iowa coal with a sulfur content of 6.21 percent has 4.80 percent has 4.80 percent of the sulfur in the form of pyrites while a West Virginia coal with 0.66 percent sulfur has 0.61 percent of the sulfur in the form of organic sulfur.

The methods that have been used to remove ash and sulfur depend on physical separation of the coal from the impurities (ash and pyrite) and rely on the difference between the desities of coal and its impurities. A variety of processes have been used in attempts to make the separation more efficient, to increase and simplify the process in order to reduce costs and to increase the amount of impurities removed.

The organic sulfur does not respond to physical separation based on differences in density since it is an integral part of the coal substance. The amount of pyritic sulfur that is removed depends on the fineness of the pyrite particles. If most of the pyrite occurs in larger pieces, then more pyrite will be removed than if the pyrite is fine. Crushing the pyritic-containing coals to smaller sizes frees more of the pyrite from the coal and a higher degree of pyrite (and sulfur) removal is possible. However, these finer sized coals are more difficult to handle and coal cleaning costs are higher.

The amount of sulfur removed by coal cleaning is a function of a large number of variables and each coal seam must be evaluated separately in determining its amenability to sulfur reduction. In a series of tests conducted on a number of different U. S. coals by the Bureau of Mines the average content of the raw coal was 3.23 percent. When crushed to a top size of 3/8" these coals, after cleaning, averaged 1.95 percent sulfur with a 90 percent yield. The average pyritic sulfur content of these raw coals was 2.05 percent (out of the total sulfur content of 3.23 percent) and this was reduced at 90 percent yield to 0.75 percent. The average organic sulfur content was 1.2 percent and was not reduced during cleaning. At a 90 percent yield only about 15 percent of the coals could be cleaned to 1 percent sulfur or less.

The Illinois Geological Survey conducted tests on 67 Illinois coals, with most of the raw coals having a sulfur content of 3 to 5 percent. Only 6 could be reduced to a 1.0 percent sulfur level, with an 80 percent yield.

In general, the sulfur content of the higher sulfur coals can be reduced more than those with lower sulfur content. At a 1 percent sulfur level in the raw coal sulfur content is reduced by about 0.2 percent while at a 3 percent sulfur level it is reduced to about 1.5 percent. Conventional physical cleaning will reduce the total sulfur emitted by power plants by significant amounts (in the order of 40 percent overall) but it will not, in general, result in coals with a sulfur content meeting the new source performance standards for the SOx. However, coal preparation can reduce the sulfur content to levels that are permitted by the State Implementation Plans for existing plants in some rural areas.

Under normal circumstances a coal preparation plant using any of the various commercial technology can be installed in from 1 1/2 - 2 1/2 years.

Advanced Coal Cleaning Technology

All of the coal preparation techniques now used commercially use physical processes for the separation of impurities and these methods are unable to remove all of either the ash or sulfur. It is possible to leach out ash and part of the sulfur through the use of aqueous media or by other means, and a number of different processes are now being tested to accomplish this although none is being used commercially.

TRW has investigated two different sulfur removal processes--one for removal of pyritic sulfur and one for organic sulfur but the latter has not been studied extensively. The process for pyritic sulfur removal has been tested using a ferric salt [$Fe_2(SO_4)_3$ or $FeCl_3$] in solutions in which the coal was treated at 50 to 130 C and at pressures of 1 to 10 atm, with residence times of 1 to 16 hours and with the top coal size ranging from 1/4" to 100 mesh. Results on 4 coals showed removal of the pyritic sulfur with the extracted sulfur appearing as soluble iron sulfate and elemental sulfur.

A large pilot plant is being constructed using the TRW pyritic sulfur removal process and it will probably require at least one year to construct and another year to test the process on this scale. At that time a decision will have to be made as to whether the greater costs (estimated at $7 - $10 per ton)[7] associated with this process (compared to the $2.00 to $2.50 per ton for the physical separation methods that are used conventionally) can be justified, in view of relatively small amount of additional pyritic sulfur that is removed. If this process appears to be commercially viable, a few plants might be constructed and be operational by mid 1978 and a large number by 1980. However, this process does not remove organic sulfur and reasonably good pyritic sulfur reduction is obtained by conventional methods at much lower costs, it is improbable that any of the advanced sulfur removal processes will be widely used by 1980 or even 1985.

Low-Sulfur, Low-Ash Coal

It has been known for many years that pulverized coal can be dissolved in a suitable solvent both in the presence and absence of hydrogen gas at elevated pressure. Several process variants have been investigated which would use this property of coal as part of an overall methods for producing oil from coal. By a suitable choice of the severity fo the processing conditions, a low-sulfur and low-ash solid fuel can be produced from coal without converting the coal to a liquid product. The fuel produced is a satisfactory clean fuel for use under boilers (for one coal tested 0.5 percent sulfur and 0.1 percent ash) and should not be as expensive as a liquid produced from coal that could be used as a refinery feedstock.

One variant of this process, solvent refined coal (SRC), has been under study for a number of years and construction of a 50 ton/day pilot plant was completed in the fall of 1974. It was designed to make a product containing about 0.8 percent sulfur and have a heating value of about 16,000 BTU per pound. The pilot plant will be operated to (1) test scale-up factors from the bench scale studies, (2) demonstrate the solid/liquid separation step and (3) assess the effects that higher hydrogen pressures may have.

A much smaller pilot plant (5 tons/day) using the same process was constructed by Southern Services Inc. at Wilsonville, Alabama. Although the pilot plant has been in operation for some time, no data have yet been released about its operation.

Sohio has announced that it is attempting to arrange for private financing of a 900 ton per day prototype plant using this type of process. It would be constructed near Toledo, Ohio adjacent to an existing power plant. The project is designed to demonstrate production of lcean fuel from coal on a scale sufficiently large so there would be little or no risk in scaling up to a full size plant and to demonstrate that the fuel produced can be used in a full size boiler.

Even if the Sohio project is consummated, it will probably be late 1978 before testing can begin. With a one year test period, the first commercial plants could not be operational before 1982 or 1983 and it is unlikely that many plants would be built simultaneously until at least one plant has actually been used on a commercial scale.

The Electric Power Research Institute has engaged Babcock and Wilcox to study the combustion of SRC. A combustion test on 30 tons of SRC is planned for February 1975.

Costs of producing a low-sulfur, low-ash coal are still difficult to estimate even roughly. A large number of different estimates have been published but the rapidly increasing costs of both the coal and heavy construction make most of the earlier estimates low. With coal at $20 per ton (80 cents per million BTU) the low-sulfur, low-ash coal would cost (in 1974 dollars) in the range of $1.75 - $2.00 per million BTU.

A number of other coal liquefaction processes are under study but they are either being tested on a smaller scale, so that commercialization will be at an even later date, or they are designed to produce a refinery feedstock rather than a clean boiler fuel.

Low-BTU Low-Sulfur Gas from Coal

Gasification of coal was a commerically used process both in the U.S. and abroad for many years, but was abandoned in favor of natural gas when large supplies of natural gas became available. Town gas produced for distribution by the local gas companies was made from the distillation of bituminous coals and by producing "water gas", a mixture of carbon monoxide and hydrogen, made from coal. The water gas was enriched with light gaseous hydrocarbons made by thermally cracking petroleum to make a gas with a 550 BTU per cubic foot heating value. The process was cyclic, inefficient and expensive.

When a clean dust-free gaseous fuel was needed by industry (e.g. at glass works), it was made by gasifying coal with a mixture of steam and air in a continuous process. The hot gas produced (producer gas) had a low BTU content (130 BTU per cubic foot) because of the dilution with the nitrogen in the air from which it was made. As a result it could not be transported economically very far. On the other hand the gas could be made at lower cost than the higher heating value "water gas." Most of the sulfur that was in the coal appeared in the producer gas as hydrogen sulfide. Since air pollution from sulfur oxides was not considered a problem at the time, the hot gas was burned with the hydrogen sulfide still in it. In a few installations the hydrogen sulfide was removed.

When interest in coal gasification was revived in this country it was because the pipeline transmission companies and the gas distribution companies became concerned that they would be unable to continue their growth as natural gas supplies were depleted. Thus, all the early research was directed at making a high BTU gas as a substitute for natural gas. A large number of new processes to make town gas had been tried in Europe after World War II since at that time coal was still their main energy source. Almost all the processes were designed to be continuous in order to avoid the high costs of cyclic operation and the air that was used in making producer gas was replaced by oxigen. The large scale use of oxygen became possible as the result of development of such plants for use in other types of commercial industrial processes. In addition, if the process was able to operate under pressure, there were economic advantages over the older atmospheric pressure operations. With the continuous processes made possible by the use of oxygen in place of air, the new processes were also designed to operate at pressure.

Three processes for making town gas from coal had been used in a significant number of installations so that they can be considered commercial. There are the fixed bed, high pressure Lurgi process, the atmospheric pressure entrained Kippers Totzek process and teh Winkler fluid bed atmospheric process of Davey Power Gas, Inc. Commercial scale plants using any of these processes could be built with a high degree of confidence that satisfactory operation would be achieved. However each of these processes has disadvantages so that more advanced processes are being studied in an effort to overcome these shortcomings.

A large number of new processes are under study and two have been operated intermittently on a large pilot plant scale for several years (IGT-Hygas and CO_2 Acceptor). A third large pilot plant, the Synthane process, is due to start operation in late 1974 or early 1975. Construction on a fourth plant using the Bigas process was started recently. Unless there are unexpected breakthroughs the first commerical plant using any of these technologies will not be operational until about 1981 to 1983.

In the last several years, interest has turned to making a low-BTU low-sulfur content gas for use under industrial and utility boilers. This type of gas should be able to be produced at lower costs than high BTU gas since oxygen is replaced with air and a number of downstream process steps are eliminated. Moreover, the overall efficiency of conversion is expected to be as much as 85 percent compared to the 65-70 percent for high BTU gas.

Although it is believed that most of the processes that were under investigation for making high BTU could also be used for making low BTU gas there has been no testing of any of the commercial processes (Lurgi, Kioppers Totzek or Davey Power Gas, Incorporated) in the U.S. A Lurgi generator, using air instead of oxygen, has been under test in Germany for several years. The Lurgi generator is operated under pressure, then to a pressurized steam boiler to produce steam to generate electricity and to a gas turbine to produce additional electricity. The exist gases from the turbine are used to preheat steam. Total output of the power plant is 170 MW of which 74 MW is produced by the gas turbine. The test results on this plant have not been reported in detail so that it is not known how successful it has been, but no new installations using the process have been announced.

Plans are being made to test a Lurgi unit in the U.S. for making low-BTU low-sulfur gas. Commonwealth Edison COmpany and the Electric Power REsearch Institute have announced that they intend to construct a Lurgi unit which will produce a clean gas to operate one of Commonwealth Edison's small coal-fired electric-generating units. As of January 1975 no announcements have been made that contracts have been awarded for this plant.

Full scale, long term tests are required before even the Lurgi gasifier for making low-BTU low-sulfur gas can be considered to be available for commercial use. Illinois coals that are used extensively by Commonwealth Edison possess some mild coking properties. Tests in a specially designed Lurgi gasifier were conducted in the U.K. and appear to indicate that coking coals can be used successfully, but this still must be confirmed by additional testing. The gas cleaning system that has been developed creates potential environmental problems and improved and lower cost methods for gas clean-up would be desirable.

According to Commonwealth Edison's timetable, their first commercial plant using this process (should the tests be successful) would be operational in about 1981 or 1982. As with solvent refined coal it is difficult to estimate the costs accurately in the absence of any large scale plants. With coal at $20 per ton (80 cents per million BTU) low-sulfur, low-BTU gas (in 1974 dollars) would cost in the range of $1.75 to $2.00 per million BTU.

In order to operate the Kippers Totzek process with air instead of oxygen, changes in design would be required. No experimental data have been reported although Koppers is said to be planning to run a test with air on one of the commerical gasifiers that is already in operation abroad. No recent data has been reported on air operation of Davey Power Gas, Incorporated's gasifier so that commerical plants using either of these processes can probably be expected to be operational only some time beyond 1982. The timetable for commerical operation of the advanced processes to make a low-BTU low-sulfur gas that are still in the prototype or pilot plant stage to make a high-BTU gas is even less favorable. Combustion of low-BTU gas produces very low emissions of NOx.

Low Sulfur Oil from Coal

Liquid fuel from coal was produced during World War II in Germany using two different processes and the product was used as a refinery feedstock. Because of the existence of these processes and the expectation that when oil resources were depleted, coal liquefaction to a refinery feedstock would again be needed; most of the research on coal liquefaction was directed toward making this type of product.

The need to make a clean fuel from coal for boiler use has changed the emphasis on coal liquefaction R&D, just as it did for coal gasification research. Unfortunately, coal liquefaction research was not pursued as intensively as coal gasification during the 1950s and 1960s. The oil industry expected that our limited domestic oil resources would be first supplemented by imported oil since oil transport costs by sea are low and large reserves are found in other parts of the world.[8]

One of the two German processes (The Fischer Tropsch) would not be a useful method to pursue if a low-cost low-sulfur boiler fuel is all htat is needed. The Fischer Tropsch process first completely gasifies the coal and then recombines the carbon monoxide and hydrogen over a catalyst in a fluidized bed to produce relatively low molecular wight products. If a fixed catalyst bed is used, higher molecular weight products are formed.

The other German process used (Bergius) dissolved the coal in a suitable hydrogen donor solvent. However, the process would not have to be modified extensively to take advantage of the development of new types of hydrbogeneration catalysts and of advances in chemical engineering. As a result, ~~if~~ a low sulfur oil is now the desired commercial product, one of the several coal hydrogenation processes that have been tested only on a bench scale would have to be used. In addition to operating the low-sulfur low-ash process at more severe pressures and temperatures to make a liquid product, (see the discussion above of low-sulfur, low-ash coal) the Synthoil process of teh Bureau of Mines and the H-coal process of Hydrocarbon Research Incorporated could be considered as likely process candidates. If a successful modification of any of these processes can be accomplished, it might be possible to have a first commercial plant in operation in the priod 1982-1983. If any of the processes still at an early state of investigation must be developed for producing a low-sulfur oil for boiler fuel, commercial plants will probably not be in operation until 1985 to 1990.

Fluidized Bed Combustion

Because of the shift in interest away from producing synthetic high BTU pipeline gas and refinery feedstock from coal to producing a clean boiler fuel, methods for using coal directly have also been receiving increasing attention. Pulverized coal combustion (used almost universally in large power plants) has reached a high degree of perfection although there remain areas in which improvements are possible. These include optimization of the configuration of the heat transfer surfaces, the use of alloys capable of handling higher steam temperatures and pressures and the development of methods to reduce the fouling and corrosive effects of the ash of certain coals. Other important drawbacks to using pulverized coal boilers include the fact that nearly all of the sulfur in the coal is converted into sulfur oxides which appear in the flue gases and that emissions of nitrogen oxides are high.

Fluidized bed combustion, another method of burning coal directly in boilers, offers the potential for overcoming most of the difficulties that arise when pulverized coal is used. With fluidized bed combustion sulfur oxide and nitrogen emissions can be reduced, the efficiency and reliability of the units are expected to be increased and the size, weight and cost of the boiler may be reduced.

In fluidized bed combustion, crushed coal is burned in a bed of limestone or dolomite particles (1/16" to 1/8") which absorb the sulfur that is released from the coal on combustion to form CaSOx. In some pilot scale tests, it was possible to operate so that only about 1 to 4 percent of the sulfur in the coal appeared in the flue gas with pressurized fluidized bed combustion and about 10 percent with atmospheric combustion. The heat transfer tubes are embedded in the fluid bed so that combustion tempertures are much lower than in pulverized fuel furnace while still giving much greater heat release rates per unit of boiler volume.

Fluidized bed combustion studies have been supported by several government agencies for a number of years. In the early work of EPA it was contemplated that fluidized beds would be used both in smaller units such as those represented by the industrial water tube boiler market and in the large base load coal fired utility plants. This research drew heavily on the fluidized combustion experience of the oil industry. Limited tests on American coals were run for EPA in the U.K. on a small fluidized pressure boiler operating at 5 to 6 atmospheres pressure.

The research supported by OCR was done at atmospheric pressure in a small fluidized bed unit having a capacity of 7000 pounds of steam per hour. Furnace temperatures of 1500°F were used and it was demonstrated that all types of coals could be burned in the unit in an environmentally acceptable manner.

A 30 MW atmospheric unit, which is a scale-up of the successful pilot plant design, is being constructed and installed in a power plant in West Virginia. The plant is scheduled to be operational in June of 1975. If no unexpected difficulties are encountered during testing of this unit, a 200 MW plant would be constructed. The design of this larger unit has already started and it could be in operation by late 1977 or early 1978. If successful, an 800 MW unit would be designed and constructed and could be operational by 1980 or 1981.

Research on pressurized fluid bed combustion is not as far advanced as atmospheric operations. However, pressurized operation should further reduce the size and capital cost of the combustion equipment and would be particularly attractive for use in combined gas-turbine steam-turbine plants (see the dicussion above of nuclear generation). A pressurized fluidized bed 1.8 MW pilot development unit is to be used to provide design, materials and environmental information for the construction of larger pilot plant unit. A 20 to 60 MW, combined cycle plant is now being designed, as is a component test program for larger scale equipment. The pressurized fluid bed research is thought to be about 2 years behind that of the atmospheric pressure research. This would indicate that the first full scale commerical pressurized fluid bed combustion units might be achieved in the 1982 to 1983 period.

SHIFT FUEL CONSUMPTION FROM ELECTRICITY TO PIPELINE GRADE GAS MADE FROM COAL

Summary

The average cost of heating structures with natural gas made from coal is about one-half that of the average costs of heating the structure with electricity made from coal when using resistance heating. When using a heat pump with electricity, the average costs are about the same as for gas made from coal. Solar heating (including hot water) might also be competitive in some geographic areas with either gas or electricity made from coal.

A national policy of energy self-sufficiency will require an increase in the use of nuclear fuels and coal and a decrease in the use of natural gas and oil. The rate at which new nuclear electric generating capacity would be installed in the next 10 years was estimated above. The shift away from oil and gas can be accomplished most easily in the industrial markets, less readily in the commerical/residential sector and with greatest difficulty in the transportation sector.

For many uses, particularly in new installations, either electricity or a pipeline gas made from coal could be used to supply energy requirements. The choice of which route to select should be based on supplying energy at the lowest marginal social cost to the user.

Pipeline gas from coal can be produced at the gasification plant for approximately $2.50 to $3.00 per million BTU. Electricity produced at a new coal-fired base load plant (equipped with air and water pollution controls) would cost about 2.6 cents per kw hour or $8.10 per million BTU. With resistance heating (100 percent efficiency conversion of electricity to useful heat) electric costs would be approximately twice that of natural gas (excluding transmission and distribution costs for both fuels). Under the same assumptions, with a heat pump with a seasonal performance factor of 2, electricity would cost 30 to 60 percent more than gas.

Average transmission distances for synthetic gas made from coal would be about 500 miles so that transmission costs would be between 25 and 30 cents per million BTU. Distribution costs vary widely from city to city but the average costs for all types of consumers in the U. S. were 40 cents per million BTU in 1971. All the synthetic gas would not require new distribution facilities since part of that gas would serve as a replacement for the decline in natural gas production. However, assuming that new facilities would be needed for <u>all</u> of the gas and that new distribution costs would be 50 percent more than the 1971 costs, then total costs per usable BTU to the consumer would be \$5.70 to \$6.50 per million BTU. This assumes 60 percent efficiency in use.

Average transmission costs for electricity in 1968 were 1.98 mills per kw hour or about 60 cents per million BTU. As with gas, distribution costs for electricity vary widely among cities but the average distribution cost for electricity was 5.69 mills per kw hour or \$2.76 million BTU in 1968. Total costs to the consumer for electricity (after adjusting transmission and distribution costs upward by 50 percent over the 1968 costs) using resistance heating (at 100 percent efficiency) would be about \$12.80 per million BTU or from 100 percent to 120 percent more than synthetic natural gas made from coal. If a heat pump were used (with a seasonal performance factor of 2) the cost would be about \$6.40 per million BTU, or about the same (or perhaps 10 percent higher) than the same usable BTUs from gas made from coal.

Capital investments in the gas conversion plants are estimated at \$7.25/million BTU/year. After correcting for a 60 percent efficiency in use, this becomes \$12/million BTU/year. For the electricity generating plant, the cost is \$22/million BTU/year. If a heat pump with a seasonal performance factor of 2 is used, the investment becomes \$11/million BTU/year.

The incremental investment cost over gas to the home-owner of using a heat pump, however, is approximately \$1500/home for heating services above and about \$500/home higher than gas for heating and air conditioning service (a separate electric air conditioner for the gas heated home is assumed). A 1000 MW electric power plant would be able to serve 500,000 homes using a heat pump so that the investment cost (by the consumer) for using electricity is then $750 million or more than the generating plant.

Another alternative for heating structures is the use of solar energy. Capital investment by the homw-owner for a solar heating (including hot water) system would be in the range of $1500 to $2500 (collector costs of $2 to $4 per square foot). Depending on the amount of backup electrical installation that must be available for reliable service, solar energy for heating might be less costly than electric heating even when using a heat pump in some geographic areas.

FOOTNOTES

1 Derived from preliminary information supplied by Office of Energy Conservation, FEA.

2 Assessment of the Impact of Air Quality Requirements on Coal 1975, 1977, 1980. U.S. Bureau of Mines, January 1974.

3 The Clean Fuels Deficit - A Clean Air Act Problem Federal Energy Administration, August 1974.

4 Under normal circumstances a large new strip mine can be installed in 2-3 years and an underground mine in 3-4 years. Because of shortages of certain types of equipment and other factors, the time to open a new mine (either underground or strip) is now somewhat longer than this.

5 Bureau of Mines, Demonstrated Coal Reserve Base of U.S. on January 1, 1974, June 1974.

6 Bureau of Mines Circular 8655 - Reserve Base of Bituminous Coal and Anthracite for Underground Mining in Eastern U.S., 1974.

7 EPA Contract No. 68 - 02 - 1302 - Proj. No. 30, Nov. 1974.

8 Supplemental natural gas imports were not thought to be a useful method to supplement domestic supplies until the late 1960s because of the high cost of liquifying and

transporting the natural gas in the form of a very low temperature liquid.

CHAPTER 11

FLUE GAS DESULFURIZATION

1.0 INTRODUCTION

Are flue gas desulfurization (FGD) systems reliable and operable for scrubbing stack gas effluents from the combustion of high sulfur coal of the eastern United States?

It is important to consider this question both in light of the recent large increase in knowledge of FGD technologies and also with sober regard to the disappointments anad failures that have contributed to the new knowledge.

In 1970, a panel of the National Academy of Engineering (NRC 1970) advised that ". . . there is an urgent need for commercial demonstration of the more promising processes, to make reliable engineering and economic data available to engineers who are designing full-scale facilities to meet specific local and regional conditions. [Emphasis in orginal.] The panel's definition of proven industrial-scale reliablility is satisfactory operation on a 100-Mw or larger unit for more than 1 year. Also, technical and economic data developed must be adequate for confident projection to full commercial scale. Pilot scale refers to investigation using flue gas in the capacity range of 10 to 25 Mw. Smaller sizes and studies using synthetic gas mixtures are considered to be bench scale."

Spokesmen who affirm that industrial-scale reliability is now available, as well as spokesmen who deny it, often quote the NAE panel's requirement of 1 year of operation at the 100-Mw scale. They argue whether or not this has been acheived in a particular unit, and whether or not the experience in this unit is generally applicable.

Not much attention has been paid to the other important ingredient by the panel as necessary to insure industrial process availability: the requirement that technical and economic data must be available to permit design of full-scale units to meet specific local and regional conditions.

The NAE panel did its work at a time when the chemistry of sulfur oxides scrubbing appeared far simpler than it does today. The panel considered 16 stack gas control procedures. A reflection of the subsequent advance in knowledge is the fact that 10 of the 16 were not represented by presentations at a meeting that EPA held in Atlanta in early November of 1974 to review the status of control technology. The Atlanta meeting considered 13 processes, of which 7 were not on the list of the 1970 NAE panel.

It should also be remembered that there have been expensive large-scale development failures in sulfur oxide emission control (see Table 11-1). One process, limestone injection into a boiler followed by a scrubber, that EPA urged upon utilities as late as early 1972 (Walsh 1972), is no longer being offered for sale.

The record would stand as an indictment of the engineering profession were it not for the fact, now evidient, that the engineer was compelled to press forward into design and construction of scrubbing equipment of unprecedented size in absence of adequate chemical knowledge. Never before had the chemical engineer been asked to treat such a large flow of gas even for a chemistry that was well understood. It is not surprising, therefore, that many of the early disappointments involved failure of large

TABLE 11-1

Large-Scale Development Failures in Sulfur Oxides Emission Control

	Estimated cost
Dry limestone injection processes	
175-MW E.P.A. test at Shawnee Station of T.V.A.	$4 - 5 million
80-MW test at Dairyland Power Co-op's Alma Station	?
Limestone injection followed by lime scrubbing	
125-MW test at Meramec Station of Union Electric	$10 - 15 million
125- and 400-MW units at Lawrence Station of Kansas Power & Light (the former is badly corroded, operating poorly, and will be replaced by a new scrubber of a different process; the latter will be converted to a limestone scrubbing unit)	?
100-MW unit at Kansas City Power & Light's Hawthorn Station (has been converted to limestone scrubbing)	?
[This system is no longer being offered for sale.]	
Potassium solution scrubbing	
25-MW test at Baltimore Gas & Electric's Crane Station	$3.5 million
Sulfoxyl process	
22-MW test at Commonwealth Edison's State Line Station	$8.5 million
Molten carbonate process	
10-MW test (oil fired) at Consolidated Edison's Arthur Kill Station	$4 million
Rheinluft process	
10- and 15-MW tests in Germany	?
Manganese oxide process (DAP-Mn process)	
110-MW test in Japan	?

scrubbing equipment to perform properly either because poor materials of construction had been chosen or because the design failed to provide adequate contacting of gas and scrubbing medium. For some FGD processes under development (notably limestone injection followed by a scrubber), it was only after these mechanical problems began to come under control that the chemical problems began to be appreciated.

Much progress can now be acknowledged. Table 11-2 provides a list of commercial scrubbing units believed to be successful. It should be noted that little time has been available to make the critical judgments needed for compiling Table 11-2, and there may well be important omissions.

A feature of Table 11-2 is that many of the successful scrubbers operate on oil-fired boilers. Another feature is that many operate with an "open water loop", a manner of operation to be explained shortly.

Two broad changes in the outlook for scrubber technology have occurred since 1970 NAE panel's study:

(1) It is now appreciated that a process successful for oil firing cannot in general be transferred wholesale to coal firing without process refinement and a new commercial demonstration on a coal-fired boiler. The trend of thinking for wet scrubbing processes has been toward providing an electrostatic precipitator for removal of most of the fly ash ahead of the scrubber, instead of relying upon the scrubber for particulate control. This is because fly ash can interfere with both scrubber chemistry (see section 2.05) and mechanical operation. The 1970 NAE panel did not distinguish between scrubbing and the products of combustion of oil and coal, nor did most of the literature on scrubbing of that time. (It should be assumed that scrubbing flue gas from oil firing is necessarily always the easier task. See Appendix 11-A for a discussion of the difficulty of removing fume particles that are produced in an oil-fired boiler.)

(2) It is also now better appreciated that new difficulties arise in the chemistry of a wet scrubbing process if it must be operated

TABLE 11-2

Partial List of Commercial Scrubbers Handling Boiler Flue Gas and Believed To Be Successful

Note: The criterion for listing a unit here has been a belief that it has been continuously available for commercial service for a period of at least several months. This is not necessarily a complete list.

Carbide Lime	Fuel	Equivalent Electricity Capacity	Inlet SO_2, ppm	Water Loop
Paddy's Run (see 2.01)	Coal	100-MW	1800-2000	Closed
Mitsui Miike (see 2.02)	Coal	160-MW	2000-2350	Open
Lime				
Mohave (see 2.12)	Coal	170-MW	200	Closed*
Kansai Electric, Amagasaki	Oil	120-MW	1100	Open
Kansai Electric, Kainan	Oil	120-MW	500	Open
Tohoku Electric, Hachinohe	Oil	115-MW	820	Open
Limestone				
Cholla (see 3.10)	Coal	115-MW	420	Closed*
Will County (see 3.02)	Coal	84-MW	1200**	Open
LaCygne (see 3.03)	Coal	700-MW	4500***	Open
Tokyo Electric, Hokosuka	Oil	130-MW	250	Open
Chugoku Electric, Mizuchima	Oil	104-MW	300	Open
Ishihara Chemical, Yokkaichi	Oil	77-MW	2000	Open
Sodium-Lime Double Alkali				
Showa Denko, Chiba	Oil	150-MW	1200-1500	Open
Tohoku Electric, Shinsendai	Oil	150-MW	600-800	Open

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*Mohave and Cholla experience little rainfall, and water losses due to evaporation from their sludge ponds are significant.

**Earlier operation at higher inlet SO_2 levels was plagued by formation of deposits.

***Cleanout of deposits is necessary about every 5 days, but the operator deems the installation to be successful.

TABLE 11-2 (cont.)

Chiyoda (sulfuric acid-limestone double alkali)	Fuel	Equivalent Electricity Capacity	Inlet SO_2 ppm	Water Loop
Fuji Kosan Co., Kainan	Oil	50-MW	?	Open
Daicel Co., Aboshi	Oil	30-MW	1500	Open

[Three units larger than 100-MW were scheduled to begin operating on oil-fired boil rs in Japan during 1974.]

Wellman-Lord (sodium salts)				
Japan Synthetic Rubber, Chiba	Oil	70-MW	1800	Open
Chubu Electric, Nishi Nagoya	Oil	220-MW	1500	Open
Sumitomo Chiba Chemical Co., Chiba	Oil	120-MW	1300	Open

[Seven units larger than 100-MW were scheduled to begin operating on oil-fired boilders in Japan during 1974.]

subatantially without discharge of salty water to the environment. That is to say, it is more difficult to operate if all water leaving the scrubber must be returned to the scrubber except the water that is discharged along the wet solid waste or sludge. Operation without discharge of salty water is termed "closed loop", and an operation that discards salty water is said to have an "open water loop". Although operation with á closed water loop is not a recent concept, the 1970 NAE panel did not mention its special problems.

These developments reinforce the 1970 NAE panel's judgment that an adequate technical data base must be available on which to rest a commercial design for each given specific situation.

An ideal base in support of a new commercial design for a sulfur oxides scrubbing process would include:

(a) complete and detailed knowledge of the scrubber chemistry selected,

(b) understanding of the mechanical and process performance of the scrubbing hardware selected as well as the proper materials of construction,

(c) adequate correlations between performance of bench scale, pilot scale, and commercial scale scrubbers of the selected hardware and chemistry, and

(d) adequate numbers of chemists who share and agree upon the relevant chemical knowledge, as well as adequate numbers of chemical engineers who understand the scrubbing hardware, in the employ of engineering firms that supply scrubbing systems.

As Table 11-2 shows, only lime and limestone scrubbers have yet operated successfully on coal at the commercial scale for extended periods of time. The question of scrubber reliability and operability must be addressed here in detail only for these alternatives. The status of other FGD processes is discussed briefly in Section 4.0.

Lime and limestone scrubber experience will be discribed in Sections 2.0 and 3.0 with emphasis upon the question, is a design support basis available to allow engineering firms to build large-scale scrubbers for medium and high sulfur coal with confidence? It may be noted that "medium sulfur coal", containing between 1 and 3 percent sulfur, accounted for 33 percent of all deliveries of coal in the United States in 1973. "High sulfur coal", with more than 3 percent sulfur, constituted 29 percent. The remaining "low sulfur coal" delivered, containing less than 1 percent sulfur, were mostly taken by the steel industry.

The discussion in Sections 2.0 and 3.0 is written on the assumption that most locations in the eastern United States are such as to require operation of a scrubber in the closed loop mode. The discussion also emphasizes scrubbers of the vertical design characteristic of the great majority of scrubbing installations now undergoing commercial trials or under construction (however, see Sections 2.12, 2.15, and 3.12).

In reference to the foregoing ideal base, item (b) need be considered only briefly. Although there is room for improvement and especially need for wider dissemination of the available knowledge, there have been major advances during the past five years in knowledge of scrubber performance and of materials of construction. The chemical engineer judges a scrubber's performance in terms of its efficiency in the contacting of gas and liquor. In a large scrubber, the engineer can expect to see some local variation in performance, since it is a practicable impossibility to effect an absolutely uniform distribution of gas and liquor moving through the scrubber, so that each small quantity of liquid would come into contact with exactly the same small quantity of gas. Often, one of the points to be settled by a large trial is a determination whether or not the efficiency of contacting that is afforded by the practicable scrubber is adequate for the inherent requirements of the chemistry of the process under trial. As a result of recent advances, a failure in a large-scale test will probably not result from a design failure that causes the test scrubber to fall far short of the best for the scrubber's type.

Item (d) of the ideal technical base will be treated in Section 8.0.

Sections 2.0 and 3.0 will concentrate upon (a), availability of chemical knowledge, and (c), the adequacy of performance comparisons among bench, pilot, and commerical scrubbers.

A technical data base falling somewhat below the ideal may be adequate, to the extent that a good empirical knowledge in respect to (c) may be used to offset some ignorance in respect to (a). However, (c) is a sine qua non, and the comparisons of commercial experience with bench and pilot units should cover the range of variables important for meeting the desired range of specific local and regional conditions.

The discussion to follow might seem to imply criticism of some industrial operators who may not have sufficiently appreciated the experimental nature of their scrubbers. The discussion might also seem to imply criticism of some designers who may not have appreciated problems that now seem obvious. Further, the discussion might sometimes seem to imply criticism of experimentalists, whom only the naive critic might expect to have mounted an earlier attack on the unobvious problems that are only now coming clearly into view. No criticism is intended here. Hindsight is easy, and the questioning of motives, cheap.

Progress in a complex technological art is often crabwise. The need today is for a keener appreciation of the difficulties and the fastest possible dissemination of information, bad as well as good. Fortunately, the power industry is geared for rapid exchange of information. It is accustomed to attacking its problems through industrial committees. Historically, it found need to hire relatively few chemical engineers, and so it has been far better prepared for exchange of information concerning electrical or mechanical arts than chemical. The industry has recently begun to hire more chemical engineers, and this fact along with the advent of the Electric Power Research Institute should greatly improve the transfer of scrubbing experience.

The large number of scrubbers now on order (see Appendix B) is often cited as proof of commerical availability. It is of course no such thing. Their owners must regard the units as experimental, needing to be staffed in expectation of discoveries and of need for revision.

In view of the record, it will be remarkable if at least a few of the installations do not experience serious difficulties.

The record also justifies an optimistic view of the future of scrubbing technologies. The issue of scrubber availability on power plants burning high sulfur coals can be resolved in the near future by a program of experimentation that can now be specified with reasonable confidence. There is a reasonable expectation that scrubbers will become available for routine purchase for a wide range of specific conditions, if an analysis of cost versus benefit shows a purchase to be justified.

It may also be noted that both lime and limestone scrubbers appear to be reliable for application on power plants burning low-sulfur western coals (See Sections 2.12 and 3.10).

2.0 LIME SCRUBBING FOR MEDIUM AND HIGH SULFUR COAL

The chemistry of lime scrubbing is too complex (Hollinden 1974, Borgwardt 1974) to summarize briefly. It will be sufficient here to understand that the alkalinity needed to scrub sulfur dioxide from the flue gas stream is supplied by the dissolving of calcium sulfite particles in the scrubbing liquor as it passes through the scrubber. Although the solubility of calcium sulfite in water is relatively small, the liquor is unsaturated in respect to this species. As the calcium sulfite enters solution, sulfite ions react with sulfur dioxide to form bisulfite ions:

$$CaSO_3 \cdot 0.5\ H_2O = Ca^{+2} + SO_3^{-2} + 0.5\ H_2O \qquad (1)$$

$$SO_2 + SO_3^{-2} + H_2O = 2\ HSO_3^{-1} \qquad (2)$$

Removal of sulfite ions by reaction (2) tends to cause additional sulfite ions to enter the solution by reation (1), and the ions further react with Sulfur dioxide. Typically, about 3 percent of the entering calcium sulfite particles might be expected to dissolve as the liquor flows through the scrubber.

The spent liquor that leaves the scrubber is rich in bisulfite ion, and is conducted to a tank where it is mixed with a slurry of lime. In a rapid reaction, bisulfite ions in the liquor are neutralized by the lime to form calcium sulfite:

$$Ca(OH)_2 = Ca^{+2} + 2\ OH^{-1} \qquad (3)$$

$$Ca^{+2} + OH^{-1} + HSO_3^{-1} = CaSO_3 \cdot 0.5\ H_2O + 0.5\ H_2O \qquad (4)$$

The calcium sulfite precipitates to form small crystals of $CaSo_3 \cdot 0.5\ H_2O$. The greater part of the scrubbing liquor, carrying a burden of these crystals, is returned to the scrubber. A small part is sent to a step for clarifying the liquor to provide a concentrated sludge of calcium sulfite particles for discard. The clarifier returnes a clear stream to the scrubber.

Early problems of lime scrubbing relating to corrosion of materials of construction are now largely solved, provided the process is controlled to maintain the pH of the scrubbing liquor within the proper range.

Problems of formation of scale and deposits remain a major concern in scrubbers of designs typical of most existing and pending installations. These problems are related to the degree of oxidation of sulfite to sulfate in the scrubber, a subject that will be treated more fully below.

Critical points are the passages in spray nozzles for introducing the liquor into the scrubber and the passages in the mist eliminator that must be provided beyond the active scrubbing zone in order to prevent droplets of scrubbing liquor from leaving the system. The danger at these critical points is that tney will become plugged by either soft mud-like deposits, or blocked by a scale consisting of dense crystalline deposits of gypsum.

Essential to continous operation is that the passages of the mist eliminator be washed to keep them clear. Water is lost from the scrubbing system both in the sludge waste and in the form of water vapor in the stack gas that did not arise from combustion of hydrogen in the coal. Relatively small amounts of fresh water must be added to the scrubber system to make good these losses, and this water may be used to wash the mist eliminator. Alternatively, or in addition, the passages of the mist eliminator may be washed with some of the clear stream returned to the scrubber from the clarifier. If these procedures succeed, it is possible to operate the system as a closed loop. If the washing procedures do not succeed, the mist eliminator must either be cleaned out periodically during a shutdown or washed with additional fresh water to keep its passages open. Additional fresh water would force the operation to discard at least some of the clear liquor from the clarifier, opening the water loop.

Trouble may arise from deposits of both calcium sulfite and calcium sulfate:

(1) If the pH of the scrubbing liquor is too high, because lime is present in excess, lime will react directly with sulfur dioxide and carbon dioxide in the flue gas undergoing treatment to form a hard scale consisting of calcium sulfite and calcium carbonate.

(2) Some oxidation of sulfite to sulfate, both within the scrubber and in the reaction tank, appears to be inevitable. This leads to danger of precipitation of gypsum crystals, $CaSO_4 \cdot 2\ H_2$, from the solution.

The tendency for deposits to prove troublesome is greater for a coal of higher sulfur content, that leads to a flue gas from which more sulfur dioxide must be removed.

Until a little more than 1-1/2 years ago, it was believed that a lime scrubber in the closed loop mode inevitably operated with a liquor that was supersaturated in respect to gypsum, and that successful operation depended upon keeping the degree of supersaturation below a critical level beyond which scaling by deposits of gypsum became intolerable.

It is now recognized that a lime scrubber can preferably operate with a liquor that is unsaturated in respect to gypsum, thereby obviating difficulties from deposits of this species (Borgwardt 1974). Indeed, the only recent successful operation of commercial-scale scrubbers on medium sulfur coal has been in the unsaturated mode (see Sections 2.01 and 2.02 below). Accordingly, the discussion which follows concentrates upon operation in the unsaturated mode. Operation in the saturated mode will be considered in Section 2.14.

Whether or not a given scrubber can operate in the unsaturated mode depends upon the degree of oxidation of sulfite to sulfate in the scrubber. At low levels of oxidation, the calcium sulfate in the solution, although unsaturated in respect to gypsum, nevertheless coprecipitates with the calcium sulfite crystals that form in the reaction tank. The crystals that are produced have the geometry of crystals of pure $CaSO_3 \cdot 0.5\ H_2O$, but contain some calcium sulfate as if it were in a "solid solution" (Borgwardt 1974). There is an upper limit to the concentration of the coprecipitated calcium sulfate (to be discussed below). If the degree of oxidation exceeds this upper limit, the solution becomes supersaturated in respect to gypsum, and danger from deposits of this material arises.

Of all aspects of lime scrubbing chemistry, least well understood are the factors that cause or prevent oxidation of sulfite to sulfate. Lowering of the pH of the scrubbing liquor promotes oxidation by increasing the concentration of bisulfite ions, making more of these ions available for reaction with oxygen in the flue gas. A greater amount of excess air, leading to higher oxygen level, is believed to promote oxidation. Presence of fly ash is known sometimes to promote oxidation (see Section 2.05), and it is reasonable to expect that the effects may be greater for some ash compositions than for others. A lengthening of residence time of liquor in the reaction tank is believed to promote oxidation (Borgwardt 1974). Some researchers suspect that nitrogen dioxide in the flue gas may tend to oxidize sulfite to sulfate, and that a flue gas with an unusually high concentration of nitrogen dioxide may cause difficulties in respect to oxidation level (Rochelle 1975).

An aspect of scrubber operation that emerges from the foregoing discussion is that control of pH is important for success: to prevent scaling by coprecipitated calcium sulfite and calcium carbonate when pH is too high, and to prevent oxidation of sulfite to sulfate when pH is too low.

Control of pH depends, among other things, upon providing a scrubber that effectively promotes excellent and uniform contacting of all of the flue gas with all of the scrubbing liquor. If a part of the liquor, for example, sees too much flue gas, its pH will drop too low. In a scrubber of poor design, with maldistributed gas and liquor flows, the operator will tend to run at higher liquor rate in an effort to assure that all of the flue gas sees an adequate amount of liquor.

In equipment of the general types exemplified by the scrubber and reaction tank, chemical engineers are not surprised to see poorer contacting in equipment of larger size. Often, much of the development task involves learning how to live with a deterioration in contacting efficiency that has accompanied scale-up. It will be important to bear this in mind in considering differences between bench, pilot, and commercial scrubbers.

Presence of magnesium ions can help the performance of a lime scrubber (Borgwardt 1974). The amount of calcium sulfate that can be purged in the solids without supersaturation is increased. The solubility of calcium sulfate is enhanced, reducing risk of scaling. The solubility of calcium sulfite is also increased, tending to promote greater efficiency of sulfur dioxide removal, but also creating a higher level of sulfite ions in the liquor, a factor tending to promote oxidation.

It is probable that sodium ions would affect performance in similar ways (Weir 1975).

Some workers have appreciated these effects for a number of years: for example, the Tennessee Valley Authority experimented with additions of magnesium several years ago, and both M.W. Kellogg and Dravo have promoted use of magnesium. It appears, however, that an appreciation of the potential os such additions has become general only recently [2]. There may have been some resistance to the idea because presence of magnesium puts a greater premium upon maintaining a strict closure of the water loop, in order to prevent all possibility that substantial amounts of water containing magnesium sulfate in solution will be discharged.

Also, it has become generally appreciated only recently that presence of chloride ions can make it more difficult to operate with a liquor unsaturated in respect to gypsum (Borgwardt 1974).

(1) The solubility of calcium sulfate is less at high chloride ion levels, increasing danger from deposits of gypsum.

(2) In presence of chloride ions, less calcium sulfate can coprecipitate with $CaSO_3 \cdot 0.5\ H_2O$. This lowers the degree of oxidation of sulfite to sulfate that is allowable before the liquor becomes supersaturated toward gypsum.

(3) Chloride ions cause a greater drop to occur in pH of liquor flowing through the scrubber. This is apparently the result of a lowered solubility of calcium sulfite, whose dissolution is important to the capability of the liquor to absorb sulfur dioxide. As a result of this effect, it is advisable to operate with a greater rate of flow of scrubbing liquor if chloride ions are present (Borgwardt 1974).

The harmful effect of chloride ions can be offset by addition of magnesium, which suppresses calcium ion, so that the chloride ions are in effect tied up with magnesium rather than calcium. In addition, as noted above, magnesium ions enhance the amount of calcium sulfate that may coprecipitate with calcium sulfite.

For a solution that is low in both chloride and magnesium ions, the limiting amount of calcium sulfate that can coprecipitate with calcium sulfite is about 18 percent. Laboratory data (Borqwardt 1974) suggest that the limiting amount is primarily a function of the activity of sulfate ion in the solution. This decreases with increase of chloride ion, and increases with increase of magnesium ion. A coprecipitate containing as much as 30 percent sulfate has been observed for a laboratory lime scrubber (Borgwardt 1974), although at such a high level of magnesium ion as to make the sustaining of the operation in the closed loop mode almost imperative.

It follows from effect (3) above that a scrubber design of marginal contacting efficiency, that might be adequate for a liquor low in chloride ion content, may lead to trouble at a high chloride ion level, because maldistribution of gas and liquor flows could lead to a region of excessively low pH, where the local flow of flue gas exceeds the capability of the local flow of liquor to remove all of the sulfur dioxide in the gas.

The Tennessee Valley Authority has measured hydrogen chloride content of flue gas from a wide variety of the coals that TVA burns, to find that most of its coals produce a flue gas containing in the vicinity of 50 to 75 ppm of HC1 (Hollinden 1975). This concentration of HC1 would correspond, very roughly, to about 0.6 to 0.9 percent chlorine in the coal. Such a chlorine content would not ordinarily be considered to be particularly troublesome from standpoint of boiler fouling and corrosion, and generally speaking, the fuels technologist would term the TVA coals to be low in chlorine. Some "high chlorine" coals, in the usual meaning of this expression, occur in Illinois, where chlorine contents as high as 0.65 percent are known (Simon 1975). Analysis of 82 coals of the Illinois Basin (Illinois, Indiana, and Western Kentucky) gave values distributed as follows (Ruch et al. 1974):

19 coals between 0.00 and 0.2 percent chlorine

13	0.03 - 0.05%
8	0.06 - 0.11%
22	0.12 - 0.23%
11	0.24 - 0.35%

9 coals greater than 0.36 percent chlorine

Analysis of 9 coals from Ohio, Pennsylvania, and West Virginia gave 2, 3, 3, 0, 1, and 0 coals in the respective categories listed above.

All HC1 present in flue gas entering a lime scrubber is absorbed (an advantage of the process), in general with formation of calcium chloride. This salt is highly soluble in water. Although it is believed that up to about one-half of the chloride present in a lime scrubbing liquor can coprecipitate with calcium sulfite (Borgwardt 1975), much of the chloride ion can leave the system only in water solution, either in water that accompanies the waste sludge, or in an undesirable water discard to the environment. The level to which the chloride ion concentration builds in the scrubbing liquor, before input of HC1 balances the discharge of chloride salts, is greatest for closed loop operation. Since the quantity of water discharged in the sludge depends directly upon the quantity of sludge, the chloride ion level is inversely proportional to the sulfur content of the coal. If the scrubber handles fly ash, the quantity of sludge will be greater and the chloride levle of the liquor will be lower.

Before considering experience gained in the several lime scrubbers now in operation, it is important to recall that the chemistry of the operation is more complex than their brief review might make it appear. It is also sobering to remember that much of the chemistry became generally recognized only in November of 1974, and even now this knowledge does not appear to be widely disseminated, even among operators of lime scrubbing equipment.

2.01 Paddys Run

A lime scrubbing system (Louisville Gas & Electric's Paddys Run Station) has been demonstrated to be reliable in intermittent service on a 65-megawatt peaking unit fired with a coal of 3 to 3.5 percent sulfur. The level of Sulfur dioxide in the flue gas is about 1800 to 2000 ppm, reflecting a greater than usual degree of excess air, viz., about 80 percent excess air in contrast to the usual 20 to 30 percent. Accordingly, the scrubber operation is best regarded as simulating the cleaning of gas from combustion of coal of roughly 2 to 2.5 percent sulfur in a 100-MW unit.

The operation is unusual in two respects:

(1) The coal is low in chlorine content, about 0.03 to 0.04 percent. Chloride ion concentration in the scrubbing liquor runs between about 300 and 500 ppm, with a highest measured value of 600 ppm.

(2) The lime is a waste carbide lime sludge accumulated from manufacture of acetylene.

The operators believe the unit to function without discharges of salty water to the environment (i.e., closed loop). Residence time of liquor in the reaction tank is about 25 minutes.

The system receives flue gas from an electrostatic precipitator of about 95 percent efficiency, fly ash enters the system at a loading between about 0.2 and 0.4 grains per standard cubic foot.

Turndown of the unit is accomplished by reducing both liquor flow and gas speed, although when the load drops below about 80 percent of capacity, flue gas is recirculated to maintain gas velocity. During a period of 45 days of service, the operation was typically at 55 to 70-MW in the day, about 30 to 35 at night, and about 3-MW over the weekend.

The degree of oxidation is only about 1.5 to 3 percent, the lowest value reported for any lime scrubber, in spite of the large amount of excess air. The system operates with liquor less than 50 percent of saturation in respect to calcium sulfate, this species being coprecipitated with calcium sulfite.

A first set of bench-scale tests by Combustion Engineering with use of the carbide lime in a small scrubber suggested that the lime contained an oxidation inhibitor, but later tests failed to confirm this finding (Martin et al. 1974).

The operator believes that the low oxidation experienced in this unit may be a result of a relatively short time during which the flue gas remains in contact with scrubbing liquor.

2.02 Mitsui Miike Industrial Boiler

A second lime scrubbing system (a Mitsui Miike Industrial boiler in Japan, discharging combustion products roughly equivalent to a 160-MW utility boiler) has also operated successfully, and in continuous service.

By an unfortunate coincidence, this system, too, is unusual in exactly the same two respects noted for the Paddys Run system above.

The unit operates steadily, and its operators have not had the problem of following changes in load.

Water management is such as to guarantee discharges of water to the environment from time to time, thereby rendering the operation open loop. Many visitors to the unit have now seen water overflowing from the sludge pond during rainfall.

The oxidation level is 10 percent, the sludge containing 10 percent calcium sulfate coprecipitated with calcium sulfite. The liquor is probably unsaturated in respect to gypsum.

2.03 Phillips Station

A lime scrubbing system (Duquesne Light's Phillips Station) operating on a part of a 387-MW station's stack gas is still in trouble. Mechanical problems appear to be coming under control, but process control of the pH of the scrubbing liquor is poor. The pH varies with load on the station, tending to fall at high flue gas throughout. The capability to supply lime is not adequate, and pH control is poor lacking automatic features and timely response The operators believe that the chloride ion content of the liquor would be beyond 1100 ppm

588-587 O - 75 - 29

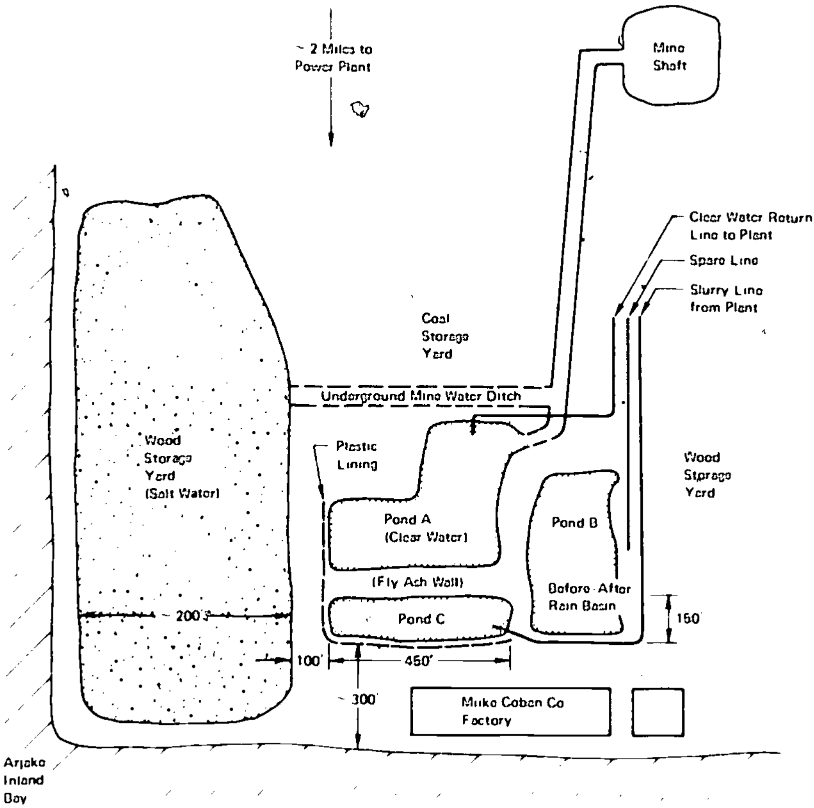
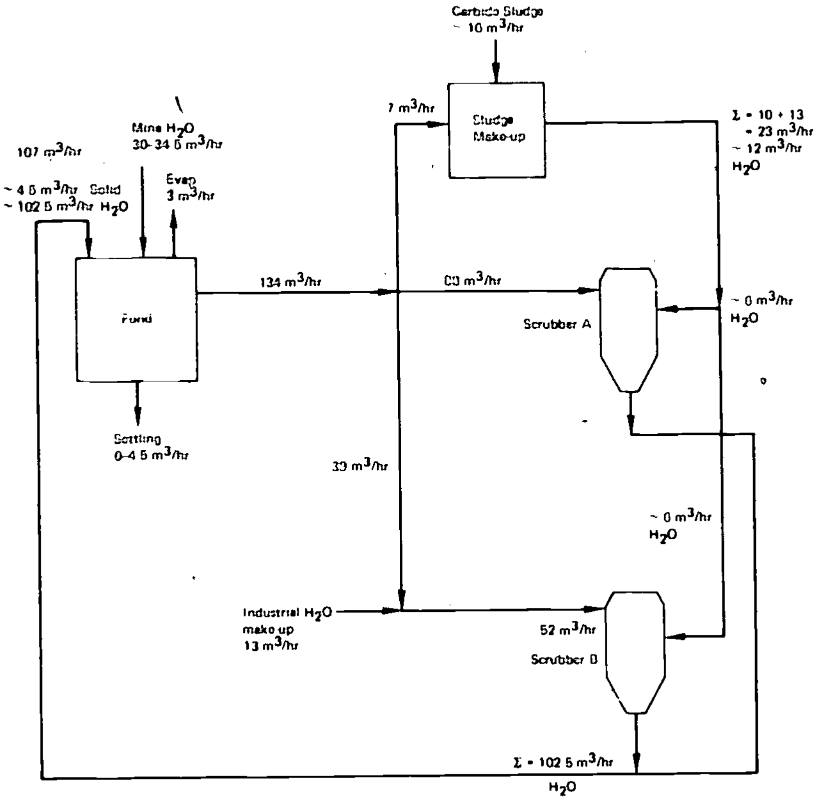


FIGURE 11-1: Mitsui Aluminum Co. Slurry Ponds as of July 5, 1973



H_2O "Balance" Around Scrubbers

in	out
88	102.5
52	34.5 Evaporation
6	~2.5 Entrainment
6	139.5 m^3/hr
152 m^3/hr	

FIGURE 11-2: Material Balance on H_2O System as of July, 1973 Mitsui SO_2 Scrubber

if the operation were closed loop, but recent operation has been at around 400 to 600 ppm, reflecting the extent to which the operation is taking in and discharging extra water. The need for the extra fresh water reflects in large part the need to use fresh water to wash mist eliminators and induced-draft fans, as well as to reduce deposits in other parts of the equipment. The operators in the past have expected to shut down and enter the equipment for cleaning about once or twice a month, and there have also been forced shutdowns due to plugging of the demisters and bleed lines, as well as to accumulation of deposits at dampers. It is possible that performance of the unit could be greatly improved by better pH control. Tests with a lime high in magnesium showed better sulfur dioxide removal.

2.04 EPA's 10-MW Pilot Lime Scrubber

Operation in the unsaturated mode has been observed in EPA's 10-MW pilot scale time scrubbing system (at TVA's Shawnee Station) for 392 hours with a closed water loop (Princiotta 1975). Chloride ion was 2000 to 3000 ppm during the latter two-thirds of the run, while magnesium ion was about 3000 ppm. Oxidations ranged from 15 to 28 percent, and the liquor was 45 percent saturated. The liquid-to-gas ratio, L/G, was 90. (L/G is expressed in terms of gallons per minute of liquor supplied to the scrubber per one thousand actual cubic feet per minute of stack gas treated by the scrubber.) This is a much higher liquor rate than that used in the Paddys Run scrubber, where L/G is between 32 and 40. The EPA pilot scrubber takes gas upstream of the Shawnee Station precipitator, the gas containing about 3 grains of fly ash per cubic foot.

The 10-MW scrubber has always operated steadily, and has not been subjected to an experiment simulating the following of load variations that might typically be expected in operation of a commercial utility boiler (Moore 1975).

The run in the unsaturated mode was shut down voluntarily, but scale had formed on the mist eliminator early in the run. This has chronically given trouble in the 10-MW operation, scaling having appeared even in trials of fresh water for washing the mist eliminator. The original design of the mist eliminator appears to have provided a sort of "mini-scrubber", where residual sulfur dioxide in the flue gas can react with calcium ions in the wash liquor to produce dense gypsum scale. After the shutdown of the run in the unsaturated mode, emphasis of the work returned to attempts to solve the problem of the mist eliminator in tests conducted in the supersaturated mode. A new mist eliminator of design similar to that used at Paddys Run was installed, but this also scaled, and has now been removed.

It should be noted that even a base-loaded commercial operation might be willing to shut down to clean out mist eliminators every few months. However, a non-scaling design is greatly to be desired.

2.05 TVA's 1-MW Bench Lime Scrubber

The Tennessee Valley Authority has a 1-MW bench scale scrubber at TVA's Colbert Station that is normally used for test operations looking toward design of the limestone scrubber for TVA's Widows Creek Station. The 1-MW scrubber was operated continuously for a month as a lime scrubber well in the unsaturated mode (Hollinden 1975). Flue gas was supplied to the scrubber from a point downstream from the electrostatic precipitator of the coal-fired Colbert Station. The flue gas averaged 0.022 grains per standard cubic foot and 2430 ppm sulfur dioxide. The operation was effectively closed loop in respect to build-up of chloride and magnesium ion levels, which ran about 5000 ppm and 2300 ppm respectively. (The mist eliminator of this scrubber is arranged so that it can be washed with fresh water that does not combine with the primary flow of scrubbing liquor. During the first two of three months of lime operation, the wash water was discarded;

during the final month, the wash system operated with a closed water loop.) The test was intended to simulate opertion of the Paddys Run system, insofar as possible. However, in addition to the higher levels of chloride and magnesium ions, L/G was 60 versus 32 to 40 at Paddys Run. Also, the Colbert Station operates at far less excess air. Oxidation level was 5 to 7 percent, appreciably higher than at Paddys Run. Since both chloride and magnesium ions tend to promote oxidation, not much can be said concerning the comparison of the oxidation levels in the 1-MW bench scale scrubber and at Paddys Run. Approach to saturation was 20 to 30 percent.

The lime scrubbing operation was continued in the 1-MW unit for a second and third month with flue gas supplied from a point upstream of the precipitator. Fly ash in gas entering the scrubber averaged 4 grains per standard cubic foot. The oxidation level rose to 10 to 12 percent, with two peaks as high as 17 percent. Approach to saturation rose to 60 to 90 percent. Because of the greater amount of sludge, chloride ion and magnesium ion levels fell to about 4000 and 1500 ppm respectively. Reaction tank residence time was 18 minutes early in the run, and was reduced to 5 minutes with no deterioration in performance.

The unit was not subjected to a test simulating load variation during the lime run.

2.06. EPA's 0.1-MW Bench Lime Scrubber

EPA's 0.1-MW oil-fired bench scale lime scrubbing system (300 ft^3/min) at Research Triangle Park has recently provided valuable new chemical information, that has cast light upon the operation in the unsaturated mode that characterizes the successful scrubbing system cited in 2.01 above, and probably also the system cited in 2.02 (Borgwardt 1974). Broadly, results from the bench scale scrubber may be summarized as follows:

(1) A test simulating the operation conditions in the Paddys Run system provided an oxidation level of 4

percent, only a little higher than that experienced in the larger unit. The liquor was at 30 percent of saturation in respect to gypsum. The test differed from Paddys Run in several respects:

(a) Ordinary lime was used instead of carbide lime.

(b) The liquor-to-gas ratio, L/G, was 50 versus 32 to 40 at Paddys Run. The higher L/G would be expected to hold down oxidation.

(c) The residence time of liquor in the reaction tank was 2 minutes versus 25 minutes at Paddys Run. The shorter residence time would be expected to tend to hold down oxidation.

(d) The excess air was appreciably less, so that the gas to be scrubbed contained about 4.5 percent oxygen versus typically 9 percent at Paddys Run.

(2) Most of the remaining test data, usually obtained in 5-day runs, were at an L/G of 77, roughly double that at Paddys Run.

(3) Tests at low level of chloride ion (between 600 ppm) were in the unsaturated mode (between 50 and 70 percent of saturation). Runs with high magnesium ion (about 1000 ppm) tended to be higher in oxidation but no higher in degree of saturation.

(4) Most tests at low levels of magnesium ion and at 3000 to 8300 ppm of chloride ion were substantially at the borderline between the unsaturated mode and supersaturation. Most oxidation levels were markedly higher, between 7 and 18 percent.

(5) Tests at chloride ion levels between 2400 and 5100 ppm and at magnesium ion

levels of about 1100 ppm were in the unsaturated mode (67 to 85 percent).

Oxidation levels tended to scatter, and replication of oxidation levels in similar runs appeared to be difficult. The oxidation levels could not be readily interpreted in terms of the relevant variables thought to affect oxidation. No general correlation of oxidation with these variables has yet emerged from the work.

Some tests were made with carbide lime instead of ordinary lime, and some tests were made with addition of coal fly ash to the reaction tank along with the lime. (Most workers would regard adding fly ash to the reaction tank to provide a less satisfactory test of the effect of fly ash than taking flue gas from a point upstream from an electrostatic precipitator of a coal-fired boiler, because of the possibility that "young" fly ash has properties not preserved during storage.) The operators believed (Borgwardt 1974) that these factors did not affect oxidation level, but the unexplained scatter in oxidation values from all of the runs renders this judgment tentative.

Although the work has cast new light upon lime scrubber chemistry, it is hard to escape the impression that much work remains before the chemistry is well understood. This is not to depreciate the value of the new information, especially in allowing a better understanding of the troubles that scrubbers have experienced.

2.07 Conclusion for Medium Sulfur Coal of Low Chlorine Content

In light of the above facts, the probability would appear to be greater than 90 percent that a lime scrubbing unit could be ordered with a reasonable prospect for reliable performance in a closed loop unsaturated mode for use in a power plant burning a medium sulfur coal (1 to 3 percent sulfur) where the chlorine content is below about 0.04 percent. The design should provide an electrostatic precipitator for removal of fly ash at high efficiency, preferably beyond 99 percent. A prudent design would provide the possibility of operating at an L/G least as high as 77.

The probability for this prospect would be enhanced (to around 99 percent, say) if the Paddys Run unit were operated successfully with ordinary lime instead of carbide lime to provide assurance that Paddys Run's success has not depended upon some unknown factor peculiar to carbide lime.

2.08 Conclusion for Scrubbing without Electrostatic Precipitator

The chemical knowledge base is not yet adequate to permit confident design of a commercial lime scrubber for a coal of medium (1 to 3 percent) or high (beyond 3 percent) sulfur content without provision of an electrostatic precipitator. Fly ash has been convincingly demonstrated to promote oxidation in the 1-MW Colbert Station scrubber. Some workers suspect that the fly ash of some coals may contain metal species that are catalytic toward oxidation of sulfite to sulfate, and this is a reasonable suspicion. It is plausible that the catalytic activity might be a function of the "freshness" of the fly ash. Much more research in respect to these possibilities would seem to be required.

2.09 Conclusion in Respect to Chemical Knowledge Base

The chemical knowledge base is not yet adequate to permit confident design of a commercial lime scrubber for a coal of medium or high sulfur content at a chlorine level beyond about 0.04 percent. No generalized correlation of oxidation is yet available in respect to the relevant scrubber variables:

Inlet sulfur dioxide level
Degree of excess air
Overall efficiency of the scrubber
Dispersion of efficiencies for local regions of the scrubber, created by less than

perfect distribution of gas and liquor flows
Ratio of liquor to gas rate (L/G)
Chloride ion concentration in liquor
Magnesium ion concentration
Time of contact of scrubbing liquor with flue gas
Reaction tank residence time
Presence or absence of fly ash
Kind of fly ash

One cannot even be certain that this list of variables is complete. Is nitrogen oxide level in the flue gas important? Is the level of iron ions in the scrubbing liquor important? The effects of the variables known to be important are understood only qualitatively.

The known or suspected interactions among the variables are such that the chemical knowledge base would have to be very good indeed in absence of strong assurance, provided by a relatively large number of comparisons of performance among bench, pilot, and commercial scale scrubbers, that performance of the latter can be expected to duplicate performance of the former with high probability.

2.10 Conclusion in Respect to Adequacy of Performance Comparisons

The comparisons between bench scale scrubbers at 0.1-MW and 1-MW, the pilot scale scrubber at 10-MW, and Paddys Run are insufficiently exact to provide a basis for confident design, even if the chemical knowledge base were adequate.

Table 11-3 summarizes performance comparisons for scrubbers treating flue gas from coal combustion. The higher L/G that was apparently required for the spray tower of the 10-MW EPA scrubber may reflect this unit's poorer distribution of gas and liquor flows through its contacting region.

Although the oil-fired 0.1-MW bench unit at Research Triangle Park observed an oxidation level of only 4 percent in one five-day test simulating Paddys Run conditions, the scatter in

TABLE 11-3

Comparisons among Bench, Pilot, and Commercial Lime Scrubbers Operating in Closed Loop, Unsaturated Mode on Flue Gas from Coal-Fired Boilers

	Paddy's Run	EPA Scrubber at Shawnee	TVA Scrubber at Colbert Station	
Scrubber type	Marble bed	Spray tower	T.C.A.*	
Kind of lime	Carbide	ordinary		
Size	100-MW equivalent	10-MW	1-MW	
Fly ash present or not	No**	Yes	No***	Yes
Length of run	45 days	17 days	30 days	60 days
Load following	Yes	No	No	No
L/G (gpm/Macfm)	32 to 40	90	60	60
Chloride, ppm	300 to 500	2000 to 3000	5000	4000
Magnesium, ppm	low	3000	2300	1500
Oxygen in fl[illegible]	9%	about 4.5%		
Residence time of liquor in reaction tank, minutes	25	10	18	5 to 18
Oxidation of sulfite to sulfate	1.5 to 3%	15 to 28%	3 to 7%	10 to 12%
Degree of saturation in respect to gypsum	50%	45%	20 to 30%	60 to 90%

*"Turbulent Contact Absorber", in which mobile, hollow polyethylene spheres are retained by horizontal grids.

**Precipitator at 95% efficiency.

***Precipitator at 99% efficiency.

oxidation results on the 0.1-MW unit greatly reduces the significance of this comparison.

The low oxidation level at Paddys Run remains, in the eyes of most workers in the field, an unresolved mystery.

2.11 Conclusion in Respect to Confidence in Early Resolution of Question of Availability of Lime Scrubbers

In spite of the reservations stated in 2.09 and 2.10, the comparisons of Table 11-3 as well as the recent advance in chemical understanding warrant considerable confidence that a well-planned program, put into effect promptly, could provide a sufficient number of performance comparisons as well as a marked improvement in chemical knowledge.

It is possible to be reasonably confident that the program can resolve the question of availability of lime scrubbers for wide application in the affirmative.

Without an intent to specify a program, and purely by way of illustration, it might be suggested that a program could include:

(1) Statistically planned experiments in bench scale scrubbers to obtain a better understanding of the effects of variables listed in 2.09 on oxidation and upon approach to saturation. Such experiments could include studies of effects of carbide lime and addition of fly ash (preferably by taking gas from a coal-fired power station ahead of a precipitator).

(2) Similar experiments in 10-MW pilot scale scrubbers, that might reasonably be less extensive in covering the range of variables, if work under (1) provides adequate direction. The experiments should include simulations of load following. The present APA 10-MW lime scrubber at Shawnee, a venturi followed by a spray tower, does not provide much built-in positive control

over distribution of liquor and gas flows to assure that the rates of both of these flows are well matched in each small zone of the scrubbing equipment. It is tempting to associate the relatively high rate of oxidation in the recent experiement in the unsaturated mode (15 to 28 percent) with the scrubber design, and one might wish to see data for lime scrubbing at the 10-MW scale in a design that provides trays or grids, with or without packing. Resolution of the problem of the mist eliminator scaling is an urgent matter.

(3) As insurance against the possibility that factors affecting oxidation may remain obscure, it might be advisable to establish a pilot scale scrubber (larger than 10-MW) with its own captive coal-burning equipment. Such an installation could be used for pilot scale tests on a variety of coals, and especially on a specific coal projected for use with a specific proposed commercial scrubber.

(4) In interests of speed, an alternative would be to supply flue gas to a pilot scale scrubber from a boiler fitted with a precipitator working at high efficiency, and then to spike the flue gas with HC1 and/or fly ash from coal intended for a specific proposed commercial scrubber.

(5) Consideration might be given to trails at Paddys Run with spiking of flue gas with HC1 and/or fly ash from other power stations. Such tests should bear in mind that the L/G at Paddys Run are markedly lower than the L/G's of the bench scale scrubbers, and it might be found necessary to increase the L/G at Paddys Run, if this should be possible. The trials should include load following. If magnesium is added to

offset the adverse effects of chloride ion, the operation should continue long enough to provide a realistic assessment of the problems of operating with a strictly closed water loop at high levels of chloride and magnesium ions.

(6) Since the Phillips Station scrubber (see 2.03) will operate in any event, consideration might be given to a study looking toward its revision for an attempt to operate in the closed loop, unsaturated or borderline mode. The revision ought to bring pH under firm control, to separate the main scrubber loop from loops serving venturi scrubbers whose job is to remove fly ash, and no doubt to effect other improvements as well.

(7) The Phillips Station scrubber is a retrofit, and a more advantageous alternative might be found among the 7 lime scrubbing installations (totalling 3,312-M) now under construction or on firm order for high-sulfur coals.

In connection with (6) and (7), it should be remembered that research on lime scrubbing chemistry on the 100+-MW scale must be regarded, under normal circumstances, as imprudent. Certainly, (6) would be costly, and Duquesne Light might well feel that it would need financial help if the task is to be done quickly, and perhaps a loan of personnel as well.

2.12 Horizontal Scrubber at Mohave Station

A novel horizontal scrubber has been in successful use of the 170-M scale at Southern California Edison's coal-fired Mohave Station. Most of the operation of this unit has been with an inlet sulfur dioxide concentration of only about 200 ppm. Cloride ion levels are extremely high. Susbtantially all of the absorbed sulfur is oxidized to the sulfate form. The effective L/G is 80, but liquor arrives to the scrubber at an L/G at 20: there are four stages of scrubbing with repeated use of the same liquor in countercurrent flow in respect to the gas. The pH may be adjusted separately in each stage, although this has not been done to date. The unit is fitted with a mist eliminator arranged so that it can be washed with fresh water that does not combine with the primary flow of scrubbing liquor.

The Mohave installation experiences loss of water by evaporation from the sludge pond, and there is ordinarily no return of water from the pond to the scrubber.

The horizontal design has also been tested at the 10-M scale in intermittent service (10 hours a day, 4 days a week) where chloride ion levels are negligibly small. Tests at the 10-M scale have extended to inlet sulfur dioxide concentrations of 3000 ppm.

The horizontal scrubber may prove to be a valuable "what then?" if vertical scrubbers of the more common designs are proved not to provide sufficiently uniform distribution of gas and liquor flows for operation at high chloride ion levels or at other conditions promoting oxidation.

Advocates of the horizontal scrubber also feel that it could operate well into the supersaturated mode without trouble from deposits.

2.13 Open versus Closed Water Loop

As noted earlier, the point of view here has been that substantially all locations in the eastern United States where a lime scrubber might be situated are such as to require operation of the scrubber in the closed loop mode. In a few locations, operation of an open loop system may be allowable, and this will be easier. No commercial system, however, can yet be said to be operting with complete success on a medium or high sulfur coal in the open loop mode, except for the unusual system cited in 2.02 above.

2.14 Lime Scrubbing in the Supersaturated Mode

The path of development emphasized here has been one toward an operation in the unsaturated or borderline mode. In view of the success of the Paddys Run scrubber, this path seems closest to achieving commercial results for medium or high sulfur coals.

The Fulham Station scrubber in England (see 3.01 below) is reported to have operated with lime in a supersaturated mode in the late 1930s.

Operators of EPA's 10-M lime scrubber believe their work to have demonstrated a viable practice in a supersaturated mode, although the operation was marred by trouble at the mist eliminator and has not yet simulated load variations. One might also worry about the sensitivity of the degree of supersaturation in some of the runs to the sulfur oxide level in the flue gas, small variations in the latter producing large changes in the former (Epstein et al. 1974). It is not necessary here to examine this option in depth, for no recent commercial practice is available for a performance comparison. It is possible that some of the 7 lime scrubbing installations now under construction may intend to operate in the supersaturated mode, and if so, the necessary performance comparisons may be forthcoming.

2.15 Operation at a High Degree of Oxidation

Some workers take the view that operation far into the supersaturated mode, and at a high degree of oxidation of sulfite to sulfate, will be possible if the scrubber is designed physically to operate without trouble from gypsum deposits, even if gypsum forms freely within the scrubber.

Such an operation would enjoy the advantage that gypsum settles from a water suspension more quickly than crystals of calcium sulfite, and the settled material occupies less volume. These properties of gypsum would ease the preparation of a concentrated sludge of waste solid for disposal, as well as reduce the volume of the sludge deposit.

These points of view seem valid, but there is little experience to back them up. They require a new path of development that might be said to have only barely been begun.

3.0 LIMESTONE SCRUBBING FOR MEDIUM AND HIGH SULFUR COAL

The chemistry of limestone scrubbing is also complex (Hollinden 1974, Borgwardt 1974), and a brief summary can touch only highlights. The alkalinity needed to scrub sulfur dioxide from the flue gas stream is supplied by the dissolving of calcium carbonate particles in the scrubbing liquor as it passes through the scrubber. Although the solubility of calcium carbonate in water is small -- indeed, far smaller than the solubility of calcium sulfite, which provides alkalinity to the lime scrubber -- nevertheless, the liquor is unsaturated in respect to this species. As the calcium carbonate enters solution, carbonate ions react with sulfur dioxide to form bisulfite ions and to release carbon dioxide gas:

$$CaCO_3 = Ca^{+2} + CO_3^{-2} \qquad (1)$$

$$CO_3^{-2} + 2\ SO_2 + H_2O = 2\ HSO_3^{-1} + CO_2 \qquad (2)$$

Typically, about 1.5 percent of the entering limestone might be expected to disolve as the liquor flows through the scrubber.

The spent liquor that leaves the scrubber is rich in bisulfite ion, and is mixed with a slurry of limestone in a reaction tank. The reaction of the bisulfite ions is relatively slow, to produce calcium sulfite:

$$CaCO_3 = Ca^{+2} + CO_3^{-2} \qquad (1)$$

$$CO_3^{-2} + 2\ HSO_3^{-1} = 2\ SO_3^{-2} + CO_2 + H_2O \qquad (3)$$

$$Ca^{+2} + SO_3^{-2} + 0.5\ H_2O = CaSO_3 \cdot 0.5\ H_2O \qquad (4)$$

Calcium sulfite precipitates to form crystalline $CaSO_3 \cdot 0.5\ H_2O$. Liquor leaving the reaction tank contains unreacted limestone, as well as calcium sulfite crystals. The greater part of the scrubbing liquor is returned to the scrubber. A small part is sent to a step for clarifying the liquor to provide a concentrated sludge for discard. It will be appreciated that inherently the sludge must contain some limestone. Generally, the supply of stone must amount to about 150 percent of the stoichiometric requirement for conversion of sulfur dioxide in the flue gas. In contrast, a lime scrubber requires but 100 percent of stoichiometric. Lime, however, is much more expensive than limestone.

Early problems of limestone scrubbing relating to materials of construction are now largely solved, provided large pH excursions are avoided.

Oxidation of sulfite to sulfate is inherently a more serious problem for a closed loop limestone scrubber than for the lime system. This is because the pH in the limestone system is buffered by presence of limestone at a pH in the vicinity of 5.5. Thus, the liquor might enter a limestone scrubber typically at a pH of about 5.8 and leave the scrubber at 5.4. In contrast, liquor entering a lime scrubber might be at about 8 to 8.5 pH, and the liquor would typically leave at about 5.6, since crystals of $CaSO_3$ in this liquor provide no buffering action. As a consequence, the bisulfite ion level is generally higher throughout the limestone scrubber, providing more opportunity for oxidation.

All commercial limestone scrubbers, whether open or closed loop, appear to have operated with a liquor that was supersaturated in respect to gypsum. In scrubbers of the general type employed in most tests underway or projected in the United States, successful operation depends upon keeping the degree of supersaturation below a critical level, generally believed to be about 130 percent, beyond which scaling by deposits of gypsum become intolerable. [However, see Section 3.12.]

Operation of a limestone scrubbing system has been observed in a closed loop, unsaturated mode in one tiny bench scale scrubber. There is general agreement that operation in such a mode will be considerably more difficult in a large limestone system than in a large lime system.

Accordingly, the emphasis here will be upon operation in the supersaturated mode.

Although the limestone system is buffered in respect to pH, nevertheless, excursions of pH in the downward direction can occur and are to be feared. This is because the dissolution of limestone is so slow that a sudden downward excursion of pH of the total scrubbing liquor flow can be corrected only relatively slowly by adding more limestone to the reaction tank. This characteristic of the system may make load following more difficult for the limestone system.

Local downward excursions of pH are possible in the scrubber itself in a zone oversupplied with gas and undersupplied with liquor.

If a pH excursion dips below about 4.8, experience at both a bench scale scrubber (TVA's 1-M unit at Colbert Station) and a large unit (Will County) has shown that extreme difficulty may arise from the blinding of the limestone particles by a "candy coating" of calcium sulfite when stone is added in an atempt to restore the pH. When this happens, experience has shown that the operator's only resort is to dump the liquor system and begin again with a new inventory of limestone. Blinding is difficult to duplicate, and hence difficult to study, and so the kinetics are unknown, and it cannot be said whether this is a problem only for a mishap that affects the overall liquor system, or is also a problem for a scrubber with bad internal distribution of gas and liquor.

3.01 Fulham Station

Full scale limestone scrubbers began to be used in England in 1933, and experience was acquired there for both open and closed loop operation. The coal-fired Fulham Station was fitted with a closed loop system in about 1938, and this system operated until 1940, when it was shut down because it was believed that the plume attracted enemy aircraft. The system employed four scrubbers each 45-M in capacity. As a result of pilot tests in the closed loop mode, developers of the Fulham Station system provided higher rate of flow of scrubbing liquor relative to flow of flue gas (higher L/G) and a larger burden of circulating limestone than the levels of these variables that have been used in commercial limestone scrubbers built recently in the United States. The designers also provided a scrubbing tower of a larger size, so that the speed of the gas was appreciably lower, and a longer residence time was provided in the reaction tank. Reports on the operation at Fulham Station contain remarks suggesting presence of all of the problems that scrubbers have experienced here in respect to materials of construction, scaling, fouling by deposits, and corrosion, although operations were deemed successful. It is understood, however, that the Fulham Station scrubber was so badly corroded when it was shut down that it would have had to be rebuilt if further operation had been required after the War.

3.02 Will County Station

A limestone scrubber for 84-M capacity at Commonwealth Edison's Will County Station has operated in the open loop mode. Much truble was experienced with fouling by deposits, among other problems, as long as attempts were made to operate on coal containing 3 percent sulfur. Since April of 1974, availability of the scrubber has been good (still in open loop mode), but the station has fired mixtures of low sulfur Western coal (about 1 percent suflur) and the Illinois coal. The sulfur level has averaged about 1.5 percent, and it might be suspected that this represents an approximate upper limit for operation of a system of the Will County design in the open loop mode without difficulty from gypsum deposits. It should be noted, however, that the design uses an appreciably lower liquor rate, higher gas velocity, lower solids burden, and shorter reaction tank residence time than the Fulham Station design that was reported to be successful in closed loop operation (see 3.01). It is doubtful that the Will County unit could be modified to approximate the Fulham Station experience in view of space limitations. In any event, such modification would be expensive.

3.03 LaCygne Station

A limestone scrubbing installation for 820-MW at Kansas City Power & Light's LaCygne Station is mechanically similar to the installation just described at Will County. The LaCygne Station burns coal containing 5.5 percent sulfur. The coal also has a high ash content. Experience in operating the scrubbers at LaCygne has been generally similar to the experience at Will County for 3 percent sulfur coal.

The operation is open loop, although it should be pointed out that the system does not discharge water beyond the station's property line having a salt content in violation of Missouri or Kansas regulations in respect to quality of industrial waste water. This is because the property includes a 2600 acre lake to receive scrubber effluent and hold it for dilution by rainfall before it eventually crosses the property line.

The original design was recognized to be experimental. Because of factors not fully appreciated when the design was executed, the LaCygne Station has had to be derated to 700 megawatts. The derating can be overcome by installing one scrubber module in addition to the seven modules already present, and by installing another forced draft fan.

The operator expects to shut down one or two scrubber modules each night on the graveyard shift, and to enter them to clean out deposits. Each scrubber is entered for cleaning about once every five days. Although the operator is not yet completely satisfied with the system availability, he regards the installation as a viable one from the standpoint of his particular needs, since he is content to reduce the production of electricity during the early morning hours. It should be remarked that such a reduction might not be congenial to most utility operators of new baseload power units, whose good fuel efficiency depends upon operating them steadily around the clock for long periods at a time. It may also be noted that the State of Kansas Public Service Commission at first took the position that the operation was not satisfactory, and delayed until recently its approval of the entering of the investment in the scrubbing system into its owner's rate base.

Alternation of the system to approximate the Fulham Station experience would be costly, if not infeasible.

3.04 EPA's 10-M Pilot Limestone Scrubber

The operators of EPA's 10-M pilot limestone scrubbing system (at TVA's Shawnee Station) have developed a mode of operation on flue gas high in sulfur dioxide and HC1 in the closed loop mode. The liquor is supersaturated in respect to calcium sulfate and high in chloride ion content. The success appears to depend upon keeping the degree of supersaturation below about 120 percent, and this has been done by

- (a) maintaining a high liquor-to-gas ratio in the scrubber (L/G = about 80 gallons per minute per thousand actual cubic feet per minute of gas entering the scrubber),
- (b) maintaining a high solids content in the liquor (about 15 percent by weight),
- (c) providing for a residence time of liquor in the reaction tank of about 20 minutes, and
- (d) reducing the gas velocity from 12.8 to 9 feet per second in the scrubber, thereby derating the unit to about 7 megawatts.

These conditions are closely comparable to the reportedly successful Fulham Station design.

A recent run lasted for 2000 hours under steady load. The unit has not been tested with simulated variations of load.

Attempts to operate the EPA pilot unit in the unsaturated, closed loop mode have failed to date, but the failure may be explained by the long liquor residence time in the reaction tank of the pilot scrubber.

3.05 EPA's 0.1-M Bench Limestone Scrubber

EPA's 0.1-M oil-fired bench scale limestone scrubbing system (300 ft^3/min) at Research Triangle Park has recently demonstrated operation in a closed loop unsaturated mode at 7000 ppm chloride ion and high magnesium ion levels (2400 ppm and beyond). Work with this unit has only very recently revealed that operation in an unsaturated mode is a possibility. The factors promoting or hindering such operation are much the same as those discussed in Section 2.0 for lime scrubbing, except the effect of chloride ion is greater for limestone scrubbing, and the ameliorating effect of magnesium ion is less. Operation in an unsaturated mode without presence of a substantial quantity of magnesium does not appear possible. Operation in the unsaturated mode requires that oxidation of sulfite to sulfate remain below some critical level, and no generalized correlation of oxidation is yet available.

3.06 TVA's 1-MW Bench Limestone Scrubber

The Tennessee Valley Authority's 1-MW bench scale scrubber at TVA's Colbert Station has been used to study design variables looking toward construction of a large limestone scrubber (550-MW) at TVA's Widows Creek Station.

In general, it appears that the planners of the large scrubber have not been willing to incur the extra expense entailed by the high liquor rate, low gas rate, and long reaction tank residence time suggested by the Fulham Station practice. The residence time in the bench scale scrubber is 5 minutes.

A major innovation in respect to the mist eliminator, howeve, gives the planners hope that a design somewhat closer to that used at Will County and LaCygne, can be made to function satisfactorily, although with a somewhat higher liquor rate than these existing commercial scrubbers provided. The mist eliminator is arranged so that it can be washed with fresh water that does not combine with the primary flow of scrubbing liquor. At first, it was expected that the water discard from the mist eliminator could be discharged from the system, and it may still turn out that this is the case. However, in absence of definitive regulations in respect to the quality of the water that may be discharged, strenuous efforts have been made over the past three years to close the mist eliminator wash water loop, by recirculating this water and bleeding water make-up for the scrubbing system proper from the mist eliminator loop. These efforts had been unsuccessful until recent encouraging experiments in which a relatively small quantity of sodium carbonate has been added to the wash water loop to precipitate calcium carbonate and raise the alkalinity of the water.

The 1-M limestone scrubber has been subjected successfully to tests simulating variations in load.

3.07 Paddys Run Operated as a Limestone Scrubber

A six-week test of the scrubber at Lousville Gas & Electric's Paddys Run Station (see Section 2.01) with limestone was deemed not successful by the operators. The scrubbing liquor was supersaturated in respect to calcium sulfate and low in chloride ion. The plant's operators are aware of the possibility of operating in an unsaturated mode, and the unit normally operates successfully in this mode as a lime scrubber. It should be remarked that the residence time of the liquor in the reaction tank when in use for lime scrubbing is 25 minutes, and the experience with the 0.1-M bench scale scrubber suggests that shortening this time appreciably might be conducive, if not indeed necessary, to achieving operation in the unsaturated mode. It is not known whether or not the operators reduced the residence time for the limestone test.

3.08 Conclusion in Resepct to Commercial Availability of Three Modes of Operation

It is not possible at this time to select a certain winner among three possible modes of closed loop operation revealed by the foregoing experience:

(1) The Fulham Station mode, tested at 10-Mw
(2) The Widows Creek mode, projected for TVA's Widows Creek Station and demonstrated at 1-Mw
(3) The unsaturated mode with high magnesium ion level, observed at 0.1-Mw

It would appear that the earliest comparison of bench scale and commercial performance will be provided by TVA's Widows Creek scrubber, which TVA rightly regards as an experimental unit.

The nature of the designs provided by Peabody Engineering for Detroit Edison's St. Clair Station (180-M), by Riley Stoker/Environeering for Central Illinois Light Co.'s Duck Creek Station (100-M), and by U.O.P. for Springfield Utility Board's Southwest Station (200-M) is not known. Perhaps further experiments might be arranged in these units on order or under construction to provide trials for the first or third options.

The third option, however, could hardly be taken seriously until it has been observed in something larger than the 0.1-M bench scale unit at Research Triangle Park.

3.09 Conclusion in Respect to Technical Base for Design

The chemical knowledge base is not yet adequate to permit confident design of a commercial limestone scrubber for a medium or high sulfur coal to operate in a closed loop mode.

Performance comparisons among bench, pilot, and commercial scale scrubbers operating in such a mode are not available.

3.10 Cholla Station

Information acquired from the successful operation of the 115-M limestone scrubber at the Cholla Station of Arizona Public Service has limited value for judging the operability and reliability of limestone scrubbing in the Northeastern United States, because of the low sulfur content of the coal burned at this station. The inlet sulfur dioxide concentration is typically 420 ppm.

The Cholla scrubber system loses water by evaporation from its sludge pond, and there is ordinarily no return of water from the pond to the scrubber.

3.11 Open versus Closed Water Loop

As noted in Section 2.13, the point of view here has been that substantially all locations in the Eastern United States would require operation of a limestone scrubber in the closed loop mode. Open loop systems have come onstream in Japan recently, or are about to come onstream, for coal-fired boilers, the objective being to produce gypsum for wallboard manufacture. With this end in view, the systems are tailored to promote oxidation of sulfite ion to sulfate. One of the systems in the Mitsui Miike scrubber described in Section 2.02, which has recently begun to operate as a limestone scrubber.

3.12 Operation with Free Formation of Gypsum in Scrubber

The path of development emphasized here has been one toward operation in scrubbers of the general vertical design that appears in substantially all of the commercial systems now under test, under construction, or on order.

Some workers take the point of view that scrubbers of significantly different design would be immune from troubles caused by scaling and deposits, even if gypsum were to form freely within the scrubber. (See Sections 2.12 and 2.15.) This would require a new path of development that at present is at an early stage.

4.0 ENVIRONMENTAL CONSIDERATIONS

4.01 Sludge Disposal

One of the most troublesome problems associated with lime/limestone processes is the difficulty of sludge disposal. The EPA has estimated the following rates of waste production rates for a typical 1000-Mw coal fired power plant (3 percent sulfur, 12 percent ash, 6400 hours/year operation, limestone scrubbing).

Production Rate, 10^3 Tons/Year

	Dry	Wet, Separate Collection and Disposal	Wet, Common Collection and Disposal
Scrubber Sludge	381	763 - 50% Solids	763 (50% Solids)
Coal Ash	338	422 - 80% Solids	676 (50% Solids)
	719	1185	1439

The EPA has also estimated the land requirements at 377 acres for sludge and fly ash disposal and 108 acres for fly ash disposal only. (EPA estimates that 90,000 MW of FGD systems will be installed in the eastern U.S. Not all of these will necessarily be lime/limestone.) If 90,000

NW of lime/limestone scrubbing capacity were installed, 130,000,000 tons per year of wet sludge including ash would be produced (50 percent solids basis).

EPA has summarized the following qualitative comparison between scrubber sludge and fly ash.

(a) The sludges typically contain calcium sulfite, calcium sulfate, calcium carbonate and fly ash in varying compositions. Fly ashes typically contain silica, alumina and hematite. The compounds in the scrubber sludge are more soluble than those in the fly ash. Table 11-3 shows the characteristics of sludges obtained from operating sulfur dioxide scrubbers.

(b) Both sludge solids and ash will contain trace elements and other species originating in the coal, lime/limestone or water. The primary source of trace metals is the coal (ash).

(c) Sludge and ash liquors will both contain dissolved species. Untreated sludge liquors normally have a lower pH than fly ash liquors, hence trace metal solubility is generally greater.

(d) Sludge liquors may contain significant quantities of chlorides.

(e) Untreated sludge settles to about 50 percent solids, and will require more storage volume per unit weight than ash which settles to about 80 percent.

At this time disposal of scrubbers sludges by ponding and land fill appears to be the only important near term alternative. Table 11-4 presents the current sludge disposal practices of several utilities. Nationwide ponding represents about 60 percent of the disposal methods and landfill 40 percent.

If ponding is used, water pollution problems can be prevented by proper pond design, installation of a pond liner and operating in a closed loop mode (do not discharge water from the pond). Costs for pond liners vary from $1 per square yard for clay and thin plastic to $9 per square yard for 30 mil rubber coated fabrics. A cost of $1 per square yard is

TABLE 11-4

Current Sludge Disposal Practices in the Utility Industry in the United States

Facility	Location	Efficient Management	Pretreatment Method: Oxidation	Clarifier	Vacuum Filter	Centrifuge	Pond	Chemical Fixation	Ultimate Disposal Method: Ponding	Landfill	Utilization
Lawrence 4	Lawrence, Kansas	Closed loop					X		X		
Lawrence 5	Lawrence, Kansas	Closed loop					X		X		
Hawthorn 3	Kansas City, Missouri	Open loop[a]		X			X		X		
Hawthorn 4	Kansas City, Missouri	Open loop[a]		X			X		X		
Will County 1	Lockport, Illinois	Closed loop		X				X[d]		X	
Stock Island	Key West, Florida	Open loop					X		X		
La Cygne	La Cygne, Kansas	Closed loop					X		X		
Cholla	Joseph City, Arizona	Open loop[b]					X		X		
Paddy's Run	Louisville, Kentucky	Closed loop		X	X					X	
Phillips	South Heights, Pa.	Closed loop		X			X	X[e]		X	
Mohave 2	South Point, Nevada	Closed loop[c]		X			X	X[c]	X	X	
Parma	Parma, Ohio	Closed loop		X	X					X	

(a) Closed loop with respect to clarifier and open loop with respect to pond.
(b) Solar evaporation.
(c) Aided by solar evaporation.
(d) Chicago Fly Ash method.
(e) Dravo method.

roughly equivalent to $5/kw capital and 0.15 mills/kwh annualized operation.

If landfilling is used, chemical fixation will most likely eliminate land deterioration problems. Fixation if successfully applied will avoid water pollution problems in unlined landfill sites, but this has not been demonstrated. However, Dravo has received a permit in Pennsylvania for disposal of fixed sludge at the Bruce Mansfield plant. As is shown on Table 11-5, at least three utilities are using chemical fixation methods but there do not seem to be any reliable published measurements of leaching, ground water runoff etc., that will determine the commercial effectiveness of fixation. Dravo and Will County are both currently conducting leachate tests.

Fixation methods have been developed by three companies (Chenfix, Dravo, and IUCS). In general, they are a means of increasing the sludge strength and shear properties. Selmeczi and Elnaggar have published the screen and sub-sieve analysis for sludges from eastern coals:

Diameter, microns	50	40	30	20	15	10	7	5	2
Cumulative weight %	1	2	5	12	21	39	51	68	68

This indicates that the particle sizes fall in the range of silt with minor percentages in the fine sand and clay size particles. The water content of settled sludge from the thickener may vary from 250 percent to 150 percent of the dry weight (160 percent is most typical) or 35 - 40 percent solids. The specific gravity is 2.5.

The settled sludge has a very soft consistency and its structure is unstable in that it liquefies when disturbed.

Fixation (stabilizing) is the process of adding chemicals to the sludge which will strengthen and harden the sludge. Each of the three companies have different additives but they are basically intended for the same purpose. Costs of chemical fixation are reported to be $15/ton on a dry solids basis (50 percent solids stream to be treated).

TABLE 11-5

Sludge Treatment/Disposal Techniques for Four Selected Utility FGD Systems Nearing Completion.

		Dewatering	Final Disposition
Detroit Edison	Limestone Eastern Coal	Pond	Landfill - no fixation
TVA Widows Creek	Limestone Eastern Coal	Pond	Ponding - lined
Ohio Edison Bruce Mansfield	Limestone Eastern Coal	Clarifier	Landfill - fixation
Northern States Power Sherburne	Limestone - flyash Eastern coal	Clarifier	Pond - clay lined

Because of the uncertainties regarding sludge stabilization (performance and economics), further study will be done in an advanced program at the Shawnee test facility under the direction of the Aerospace Corporation. The three different proprietary processes will be studied: Davo, IU Conversion Systems and Chemfix. There will be two large ponds for untreated sludge (one lime, one limestone) and three small ponds for treated sludge (one for each process). Each pond will have a leachate well and a ground water well to monitor any ground water contamination.

Utilization of scrubber sludges as a building material is technically feasible, and the process of gypsum production for wall board is practiced in Japan. The first large scale application of sludge utilization in the U.S. will be at the limestone scrubber installation at Mohave Unit No. 1, where IU Conversion Systems will convert the sludge into a building material.

One of the difficulties in sludge disposal is that a potentially useful product (sulfur) is being thrown away. EPA estimates have shown lime/limestone FGD systems are probably not competitive with regenerable FGD systems (where the sulfur is recovered) if the sludge disposal costs exceed about $4-6 per wet ton. (However sulfur recovery (not H_2SO_4) in an integrated FGD unit on a utility boiler will not be demonstrated until 1975-76 at the NIPSCO plant, Wellman-Lord process.) The magnesium oxide process, to be installed in two locations, will also recover a useful byproduct.

Table 11-5 summarizes the sludge disposal chosen by four utilities who have FGD systems that are fairly close to completion.

5.0 WATER POLLUTION

As pointed out above there are potentially serious water pollution problems from leaching and/or runoff from unlined ponds. There appears to be no problem for a lined and well engineered pond. Experimental work is now underway to improve the technology and further define the costs of sludge disposal.

The Wellman-Lord Process, currently under installation at NIPSCO does not produce a sludge. This process does require a bleed stream from the scrubber circulating liquid. Roughly 5 to 10 percent of the incoming sulfur (sulfur dioxide) is oxidized to sulfate and may be discharged as sodium sulfate (dissolved in water), or as dry sodium sulfate. There is well known technology available to treat the problem, but its application will be relatively expensive. Davy Powergas (licensor for the Wellman-Lord Process) has an active research program underway to find methods of minimizing or eliminating the sulfur oxidation. Until actual operating data becomes available, the magnitude and cost of this problem in a high sulfur eastern coal utility is difficult to assess with reliability.

6.0 PARTICULATE REMOVAL EFFICIENCIES

Almost all of the FGD systems report fly ash removal efficiencies in terms of grains/SCF. These data indicate that outlet particulates concentrations of 0.01 to 0.03 grains/SCF are very typical for a wide range of inlet loadings. These outlet concentrations are adequate to meet air quality criteria.

Two important parameters have not, in general, been measured. These are the removal efficiencies as a function of size range, and analysis of the outlet particulates for fly ash and sulfates, if any.

There are two sets of published data on particulates removal efficiencies as a function of size range.

Weir et al. (1974) have recently presented the results of particulate removal analysis at the Southern California Edison plant. (This is a horizontal scrubber designed by Weir, so interpretation of data for vertical scrubbing may be difficult). Figure 11-3 shows the total outlet particulate loading (grains/SCF) as a function of total inlet particulate loading (grains/SCF) for the 170 MW scrubber. The

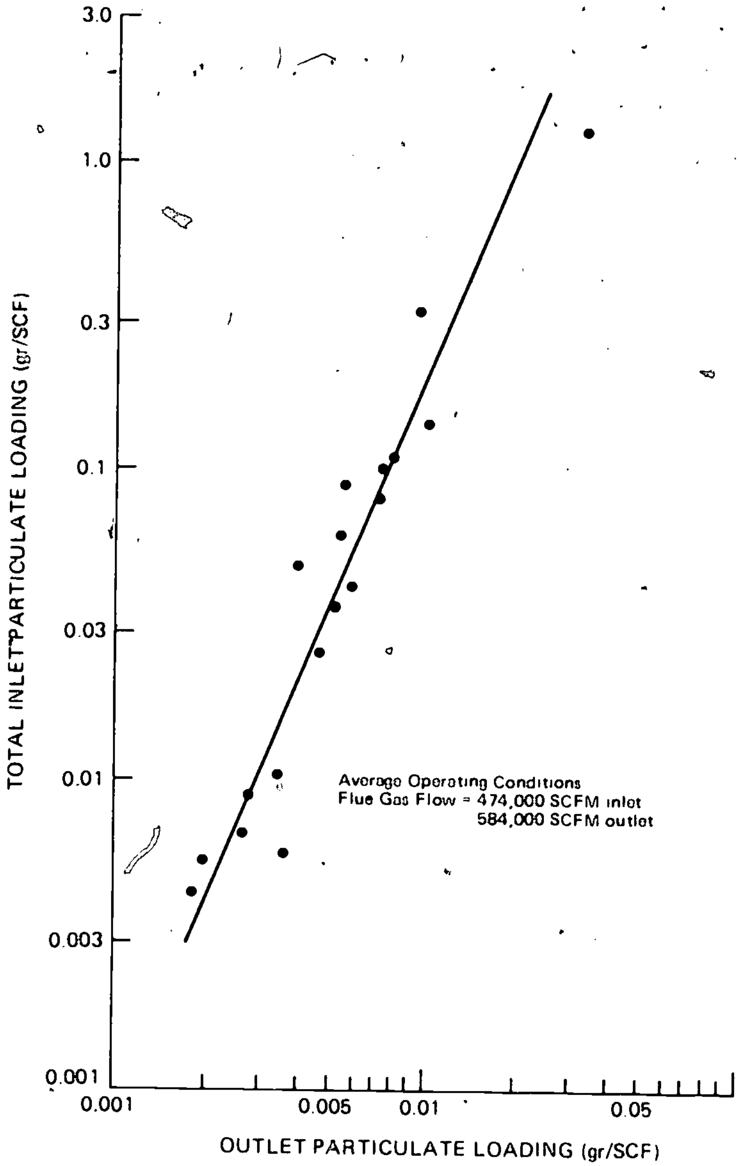


FIGURE 11-3: Inlet vs. Outlet Grain Loading 170 MW Horizontal Module Southern California Edison Plant

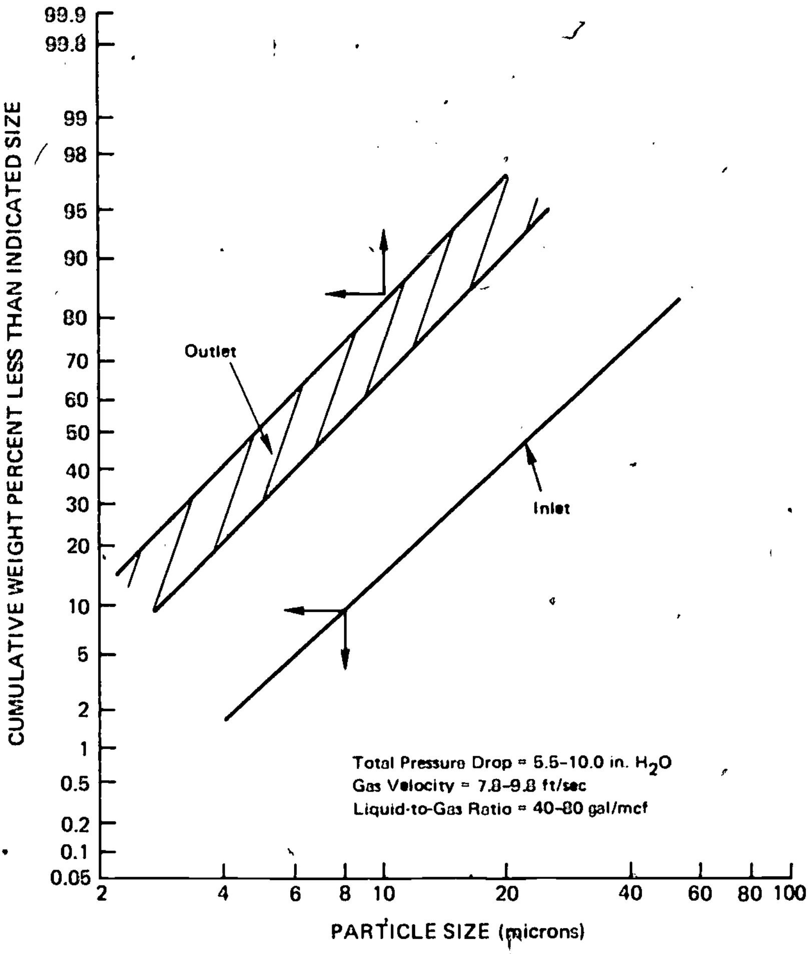


FIGURE 11-4: Particle Size Distributions at TCA Scrubber Inlet and Outlet - TVA Shawnee Plant

particulate removal varies from 70 percent at 0.01 grains/SCF inlet to 98 percent at 1.00 grains/SCF inlet.

The particle size in the flue gas at Mohave is usually small - 90 cumulative wt. percent less than 4 microns, 70 wt. percent less than 2 microns, 40 wt. percent less than 1 micron and 15 wt. percent less than 0.5 microns. The plant does have an electrostatic precipitator, and these measurements are downstream from it.

Fractional size collection efficiencies for both the pilot plant and the commercial plant are shown below:

Particle Size Microns	% Pilot Plant (.02 GR/SCF inlet)	% Commercial Unit (.08 GR/SCF inlet)
>1.5	97	92
1.0-1.5	96	91
0.5-1.0	87	85
0.3-0.5	75	76

Epstein et al. (1974) and Bechtel (for EPA) have published particulate removal data for TVA Shawnee operations, for limestone and lime scrubbing. The Bechtel report contains data on overall particulate removal efficiencies for several different scrubbers--venturi and spray scrubbers, TCA scrubber with five grids and a marbel bed scrubber. All removal efficiencies appear to be in the 98-99+ percent range, but the inlet gas particulate loadings are also high.

Figure 11-4 shows inlet and outlet particle size distributions for the TCA scrubber. Figure 11-5 shows the TCA particulate removal efficiency as a function of particle size. (The data are given as percent penetration, which is 100 minus percent removal). As would be expected, the removal efficiency drops rapidly with decreasing particle size, especially at low pressure drops. Because of the limited number of tests (shown on these figures), firm conclusions regarding collection efficiency

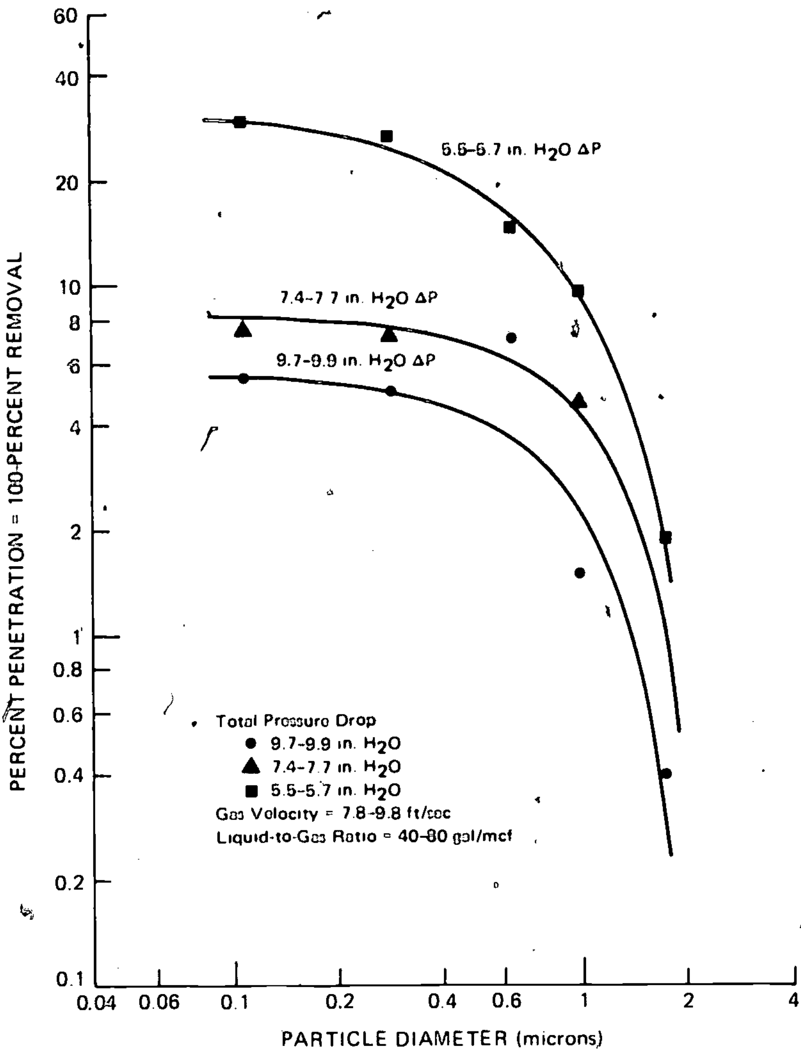


FIGURE 11-5: TCA Scrubber Particulate Removal Efficiency as a Function of Particle Size - TVA Shawnee Plant.

should await additional testing to be carried out.

Very few data are available on the fly ash content of the particulates leaving the FGD system. What data are available suggest that the fly ash represents about 60 percent of the total and the remainder (40 percent) comes from entrainment from the slurry liquor. This is apparently calcium and magnesium salts but very little definitive work has been done in this area.

7.0 COSTS OF FLUE GAS DESULFURIZATION SYSTEMS

McGlamery and Torstrick (1974) have presented results of an EPA sponsored cost appraisal of the most advanced FGD systems, using as a base case a new 500 MW unit burning a 3.5 percent sulfur coal. The unit costs were estimated at $45/kw for lime slurry and ranged up to about $80-90/kw for catalytic oxidation. Most utility companies believe that these costs are in error and much too low. In addition these are for new units; retrofit installations will cost more. The results of studies on retrofit plants are discussed later. Also, the costs from McGlamery's study are based on early 1974 data, and do not include the rapid construction cost escalation experienced in recent months. The major cost factors used are summarized below:

1. Project schedule and location. Project assumed to start in mid-1972 with 3-year construction period ending mid-1975. Midpoint of construction costs mid-1974; *Chemical Engineering* Cost Index - 160.2. Startup - mid-1975. A midwestern plant location is assumed.
2. Power unit size. Costs for three unit sizes--200, 500, and 1000 MW--are projected.
3. Fuel type. Systems for both coal- and oil-fired units are costed--coal 12,000 Btu/lb, 12 percent ash, oil - 18,500 Btu/lb, 0.1 percent ash.
4. Sulfur content of fuel. Costs for three sulfur levels are evaluated for each fuel--

2.0, 3.5, and 5.0 percent for coal; 1.0, 2.5, and 4.0 percent for oil.

5. Plant status. Although systems for both new and existing power units are evaluated, only a simple, moderately difficult (scrubbing system installed on vacant space beyond the stack) retrofit is estimated since such systems can vary over such a wide range of configurations and restrictions. New units are designed for a 30-year life, 127,500 hours of operation. Costs for new and existing systems are not directly comparable.

Clearly the costs of a new plant ordered today will be higher. However the relative costs, i.e. differences between cases, is probably valid.

Table 11-6 is a summary of the total capital investments as presented by McGlamery. The effects of power plant size and sulfur content of the coal on unit investments are shown in Figures 11-6 and 11-7 respectively. Table 11-7 is McGlamery's summary of the total operating costs for the same cases as shown on Table 11-6 for capital investments. Again these numbers 2.0-2.5 mills/kwh. are probably too low, because the capital charges (resulting from the low investment costs) will be too low. Also note that investment and operating costs for disposal of fly ash and byproduct credit have been excluded. The effect of excluding product credits from annual operating cost estimates is shown below.

Process	Total Average Annual operating cost, $	Annual credit for byproducts, $[a]	Net annual operating cost, $
Limestone	7,702,700	-	7,702,700
Lime	8,101,900	-	8,101,900
Magnesia	9,210,800	883,200	8,327,600
Sodium	11,601,500	1,077,500	10,524,000
Cat-Ox	8,873,900	659,400	8,214,500

[a]Corresponds to credit fo \$8/ton 100% H_2SO_4 as 98% H_2SO_4 for magnesia slurry process; \$25/short ton sulfur, \$20/ton Na_2SO_4 for the sodium solution process; and \$6/ton 100% H_2SO_4 as 80% H_2SO_4 for the catalytic oxidation process.

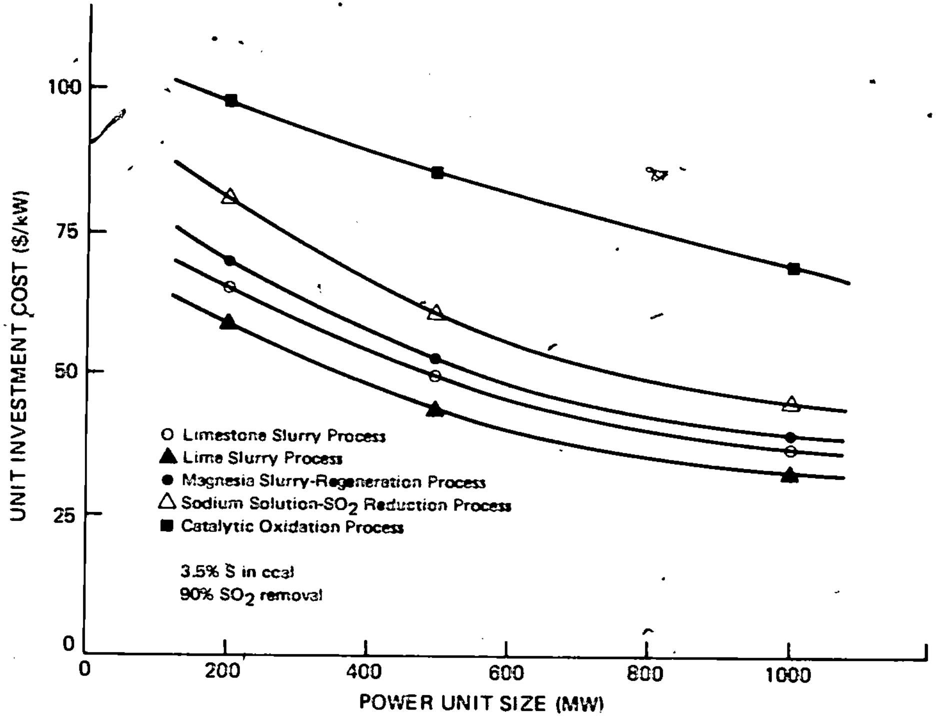


FIGURE 11-6: New Coal-fired Units - the Effect of Power Unit Size on Unit Investment Cost, Dollars per Kilowatt (McGlamery and Torstrick 1974)

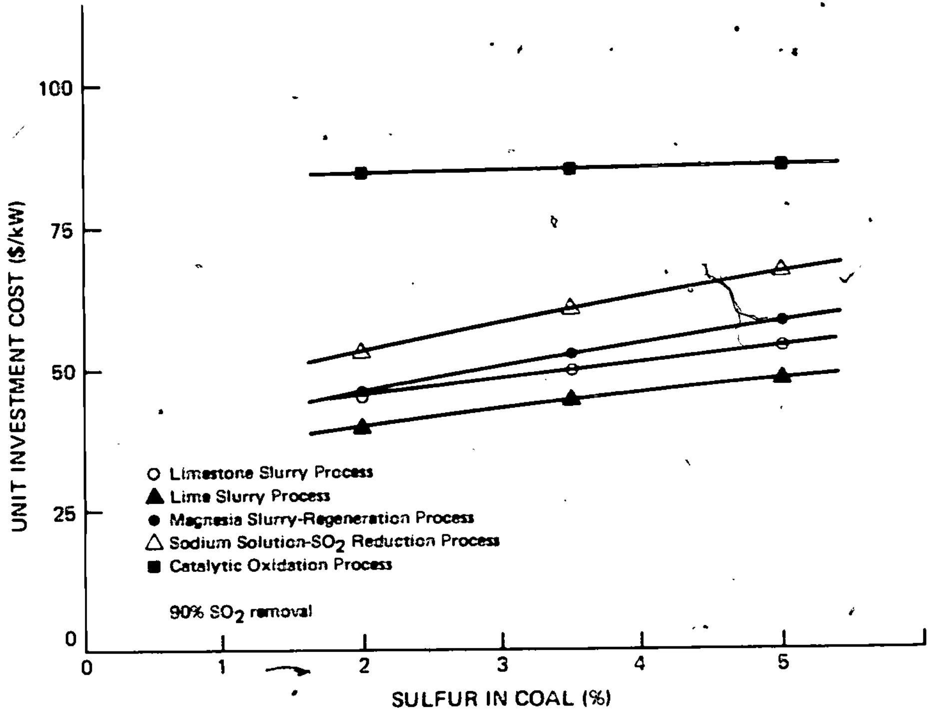


FIGURE 11-7: 500-MW New Coal-fired units - Effect of Sulfur Content of Coal on Unit Investment Cost (McGlamery and Torstrick 1974)

TABLE 11-6. Summary[a] - Total Capital
For Flue Gas

Case	Years life	Limestone process $	Limestone process $/kw	Lime process $	Lime process $/kw
Coal-fired power units					
90% SO_2 removal; onsite solids disposal					
200 MW new, 3.5% sulfur	30	13,031,000	65.2	11,749,000	58.7
200 MW existing, 3.5% sulfur	20	11,344,000	56.7	13,036,000	65.2
500 MW existing, 3.5% sulfur	25	23,088,000	46.2	26,027,000	52.1
500 MW new, 2.0% sulfur	30	22,600,000	45.2	20,232,000	40.5
500 MW new, 3.5% sulfur	30	25,163,000	50.3	22,422,000	44.8
500 MW new, 5.0% sulfur	50	27,345,000	54.7	24,272,000	48.9
1000 MW existing, 3.5% sulfur	25	35,133,000	35.1	38,133,000	38.1
1000 MW new, 3.5% sulfur	30	37,725,000	37.7	32 765,000	32.8
30% SO_2 removal; onsite solids disposal					
500 MW new, 3.5% sulfur	30	24,267,000	48.5	21,586,000	43.2
90% SO_2 removal; offsite solids disposal					
500 MW n-w, 3.5% sulfur	30	20,532,000	41.1	18,323,000	36.6
90% SO_2 removal; onsite solids disposal (existing particulate collection facilities)					
500 MW existing, 3,5% sulfur	25	29,996,000	60.0	26,090,000	52.2
Oil-fired power unit					
90% SO_2 removal; onsite solids disposal					
200 MW new, 2.5% sulfur	30	8,263,000	41.3	9,482,000	47.4
500 MW new, 1.0% sulfur	30	12,935,000	25.9	15,961,000	31.9
500 MW new, 2.5% sulfur	30	15,473,000	30.9	18,148,000	36.3
500 MW new, 4.0% sulfur	30	17,481,000	35.0	19,861,000	39.7
500 MW existing, 2.5% sulfur	25	18,657,000	37.3	21,817,000	43.6
1000 MW new, 2.5% sulfur	30	23,384,000	23.4	26,341,000	26.3

[a] Midwest plant location represents project beginning mid-1972, ending mid-1975. Average cost basis for scaling, mid-1974. Minimum in process storage; only pumps are spared. Investment requirements for disposal of fly ash excluded. Construction labor shortages with accompanying overtime pay incentive not considered.

Investment Requirements
Desulfurization Processes (McGlamery and Torstrick 1974)

Magnesia process $	Magnesia process $/kw	Sodium process $	Sodium process $/kw	Cat-Ox process[b] $	Cat-Ox process[b] $/kw
14,139,000	70.7	16,198,000	81.0	19,537,000	97.7
14,372,000	71.9	17,149,000	85.7	17,735,000	88.7
26,026,000	52.1	31,208,000	62.4	37,907,000	75.8
22,958,000	45.9	26,706,000	53.4	42,520,000	85.0
26,406,000	52.8	30,491,000	61.0	42,736,000	85.5
29,355,000	58.7	33,709,000	67.4	42,928,000	85.9
38,717,000	38.7	47,721,000	47.7	62,913,000	62.9
38,865,000	38.9	45,832,000	45.8	69,889,000	69.9
25,568,000	51.1	29,127,000	58.3	-	-
-	-	-	-	-	-
32,213,000	64.4	37,957,000	75.9	43,816,000	87.6
8,861,000	44.3	10,324,000	51.6	13,069,000	65.3
12,695,000	25.4	15,198,000	30.4	28,067,000	56.1
16,080,000	32.2	18,945,000	37.9	28,277,000	56.6
18,765,000	37.5	21,893,000	43.8	28,449,000	56.9
20,376,000	40.8	24,445,000	48.9	32,824,000	65.6
23,656,000	23.7	28,765,000	28.8	46,356,000	46.4

[b] All Cat-Ox installations require particulate removal to 0.005 gr/scf prior to entering converter. Because existing units are assumed to already meet EPA standards (0.1 lb particulate/MM BTU of heat input). Only incremental additional precipitator is required.

Most utilities and vendors today will estimate FGD costs of $60-100/kw, with the majority believing that the costs will be closer to $100/kw.

The latest investment (December 1974) figure for the FGD system being installed at the Ohio Edison Bruce Mansfield plant is $133/kw. These are two-800 MW coal fired boilers. The breakdown of the costs is given in Table 11-8. Ash removal and disposal costs are included in the $133/kw.

McGlamery has also published cost estimates of "limestone slurry process investment with modified scope," which is shown on Table 11-9. These costs are intended to reflect possible additional capital charges associated with FGD systems, and include such items as overtime to accelerate the project, replacement of energy consumed by the FGD system, retrofit difficulty etc. The total of these potential charges is about $60/kw. Obviously not all charges are applicable to any given case, but the point is that some of them may be, and the capital charges cited above of $60-100/kw may not include all necessary charges. For example, provisions for redundancy (item D of Table 11-9) may be needed to provide adequate assurance of reliable operation.

A major component (about one-half of the operating costs) is the charge to reheat the stack gases before discharge to the atmosphere. Experimental data now becoming available indicates that addition of magnesium to the scrubber liquor can improve sulfur dioxide removal efficiency substantially (e.g. from 90 to 95 percent). This would allow bypassing a small amount of hot gas to provide at least some of the reheat needed for the scrubbed gas and therefore reduce operating costs.

Further the data available indicates that 5 to 7 percent of a utility plant's power output will be required by an FGD system. Aside from energy costs, this can have important implications in estimating reserve capacity.

As pointed out in Section 5.01 ponding increases operating costs from 0.15 mills/kwh to over one mill/kwh. Sludge fixation, instead of ponding, will further increase the operting costs

TABLE 11-7. Summary[a/] – Total Average Flue Gas

Excluding Credit for Byproducts

Case	Years life	Limestone process: Total annual operating cost, $	Limestone process: Mills/kWh	Lime process: Total annual operating cost, $	Lime process: Mills/kWh
Coal-fired power units					
50% SO_2 removal; onsite solids disposal					
200 MW new, 3.5% sulfur	30	3,921,500	2.80	4.163,900	2.97
200 MW existing, 3.5% sulfur	20	3,867,100	2.76	4,822,000	3.44
500 MW existing, 3.5% sulfur	25	7,892,600	2.26	9,612,400	2.75
500 MW new, 2.0% sulfur	30	6,774,700	1.94	6,915,100	1.99
500 MW new, 3.5% sulfur	30	7,702,700	2.20	8,101,900	2.31
500 MW new, 5.0% sulfur	30	8,522,200	2.43	9,170,100	2.62
1000 MW existing, 3.5% sulfur	25	12,752,900	1.82	15,301,400	2.19
1000 MW new, 3.5% sulfur	30	11,874,100	1.70	12,553,100	1.79
30% SO_2 removal; onsite solids disposal					
500 MW new, 3.5% sulfur	30	7,378,000	2.11	7,806,900	2.23
90% SO_2 removal; offsite solids disposal					
500 MW new, 3.5% sulfur	30	8,376,500	2.39	8,641,000	2.47
90% SO_2 removal (existing unit without existing particulate collection facilities)					
500 MW existing, 3.5% sulfur	25	9,573,400	2.74	9,728,300	2.78
Oil-fired power unit					
90% SO_2 removal; onsite solids disposal					
200 MW new, 2.5% sulfur	30	2,842,000	2.03	3,413,500	2.44
500 MW new, 1.0% sulfur	30	4,732,500	1.35	5,748,600	1.64
500 MW new, 2.5% sulfur	30	5,564,400	1.59	6,852,800	1.96
500 MW new, 4.0% sulfur	30	6,281,800	1.79	7,742,300	2.21
500 MW existing, 2.5% sulfur	25	6,587,300	1.88	8,001,500	2.29
1000 MW new, 2.5% sulfur	30	8,987,400	1.28	10,795,200	1.54

a/ Power unit on-stream time, 7000 hr/yr. Midwest plant location, 1975 operating costs.

Annual Operating Costs
Desulfurization Processes (McGlamery and Torstrick 1974)

Magnesia process Total annual operating cost, $	Magnesia process Mills/ kWh	Sodium process Total annual operating cost, $	Sodium process Mills/ kWh	Cat-Ox process Total annual operating cost, $	Cat-Ox process Mills/ kWh
4,776,800	3.41	5,971,700	4.27	4,232,700	3.02
5,091,200	3.64	7,377,700	5.27	5,849,400	4.18
9,607,900	2.75	14,658,000	4.19	12,399,600	3.54
7,523,400	2.15	9,101,700	2.60	8,801,200	2.51
9,210,800	2.63	11,601,500	3.31	8,873,900	2.54
10,768,500	3.08	13,983,300	4.00	8,940,500	2.55
15,481,900	2.21	25,118,500	3.59	21,460,800	3.07
14,347,000	2.05	18,391,300	2.63	13,957,600	1.99
8,789,700	2.51	10,834,300	3.10	-	-
-	-	-	-	-	-
11,227,300	3.21	16,389,200	4.68	13,598,300	3.89
3,204,400	2.29	4,269,200	3.05	2,750,100	1.96
4,633,100	1.32	5,854,700	1.67	5,743,600	1.64
6,092,700	1.74	8,305,100	2.37	5,677,500	1.62
7,393,500	2.11	10,640,500	3.04	5,565,100	1.59
7,308,700	2.09	10,261,600	2.93	11,126,100	3.18
9,715,900	1.59	13,686,200	1.96	8,911,900	1.27

Investment and operating cost for disposal of fly ash excluded.

TABLE 11-8

Installation Cost, Bruce Mansfield Plant, Flue Gas Desulfurization System

Scrubber System	
Chemico Contract	$32,000,000
Foundations, Electrical, I&C, Linings, Oil Storage, Control Room, Erection	32,200,000
Sludge Ponds with Lining	2,500,000
Lime Dock, Handling and Storage Facilities	11,700,000
Chimney with LIning	7,600,000
Subtotal	$86,000,000
Distributables	18,300,000
Contingency	3,000,000
Escalation	5,000,000
Engineering	1,500,000
Owner's Cost Including Allevi-ance for Funds During Construc-tion	15,100,000
Total Scrubber System	$128,500,000
Off-site Waste Disposal	
Dravo Contract	$59,500,000
Land, Right-of-way, Electrical, Calçilox Harbor, Grits Conveyor, Pipe Rack	2,800,000
Subtotal	$62,300,000
Contingency	6,000,000
Escalation	5,300,000
Dravo Engineering	4,000,000
Owner's Costs Including Allowances for Funds During Construction	6,700,000
Total Off-site Waste Disposal	$84,300,000

TABLE 11-9

Limestone Slurry Process Investment with Modified Project Scope

	Investment, $/kw
BASE INVESTMENT - LIMESTONE SLURRY PROCESS (Including Fly Ash Removal But Not Disposal)	
500-MW new coal-fired unit burning coal with 3.5% S, 12% ash, 90% SO_2 removal, 30-year life 127,500 hours operation, onsite solids disposal, proven system, only pumps spared, no bypass ducts, experienced design and construction team, no overtime, 3-year program, 5% per year escalation, mid-1974 cost basis for scaling	50.30
A. Overtime to accelerate project or cover local demand requirements (50% of construction labor requirements)	3.20
B. Research and development costs for first of a kind process technology (as allowed by FPC accounting practice)	5.00
C. Power generation capital for lost capacity (normally covered by appropriate operating costs for power used in process)	4.50
D. Reliability provisions with added redundancy of scrubbers, other equipment, ducts and dampers, instrumentation for change over (assumes no permission to run power plant without meeting SO_2 removal emission standards at all times)	7.00
E. Additional bypass ducts and dampers	1.00
F. Retrofit difficulty--moderate, space available beyond stack, less than three shutdowns required for tie-ins, field fabrication feasible	10.00
G. Fly ash pond including closed-loop provisions	5.50
H. 500-ft stack added to project cost	6.00
I. Air quality monitoring system, 2-15 mile radius, 10 stations	0.70
J. Cost escalation of 10% per year instead of 5%	4.80
K. Possible delay of up to 2 years in equipment and material deliveries (1977 completion instead of 1975)	15.00
Total	113.00

(of a non-regenerable FGD system.) For example, a cost of $4.50 per ton (wet basis) is roughly equal to 0.6 mills/kwh. As a rough guide EPA believes that if the cost of sludge disposal exceeds $4 to $6 per ton (wet basis), a regenerable process which recovers a useful byproduct, will be more economic than a non-regenerable process. (This assumes byproduct credit.)

7.1 Retrofit Flue Gas Desulfurization Systems

Cost estimation for retrofit FGD systems is extremely difficult because of the wide variation in specific details for each power plant. The most recent studies have been completed by Radian Corporation, which extended and complemented a previous study by M. W. Kellogg, and a second study by Kellogg. Radian's study was completed in December 1973 and was primarily addressed to the utility industry in Ohio. However, the conclusions reached are probably applicable to the eastern U.S. Radian studied retrofitting of lime/limestone processes, the magnesium oxide process and the Wellman-Lord process. They concluded that the space requirements and ease of retrofit should not vary significantly with the process selected (of those considered).

Space required for the scrubbing section of the process is the major concern in retrofit since this equipment must be placed adjacent to the stack. Process equipment outside of the scrubber area is of less concern to the retrofit problem since it can usually be located on the peripheral areas of the plant. In some cases vertical space limitations may cause problems, but this should not occur in most plants where retrofit is possible.

Radian estimated that if both hold tanks and scrubbers are placed adjacent to the stack, a plot area of 45 square feet/MW will be required. If the hold tanks can be placed on the periphery of the powerhouse (or placed directly under the scrubbers), Radian estimates a requirement of 23.8 square feet/MW. This estimate was made for a 550 MW power plant, but the unit space requirements (sq. ft./MW) are probably fairly independent of power plant size.

Radian then studied in some detail the power plants in the state of Ohio, using as a criterion that any unit with an available area less than 20 square feet/MW can be retrofitted only with great difficulty (if at all). The data on utility plant size and plot availability was taken from the first Kellogg study. A high percentage of large, new boilers have sufficient space for flue gas cleaning systems. More than 87 percent of the surveyed units ten years old or less may be retrofitted. About 85 percent of capacity in existing units larger than 500 MW have sufficient space.

In terms of total capacity, about 70 percent can be retrofitted by including only boilers less than 20 years old or greater than 100 MW capacity. About 74 percent of the total capacity surveyed had available area equal to or greater than 20 square feet/MW. The results of the study are shown graphically in Figure 11-8.

Two fairly detailed cost estimates were made in 1973 comparing costs for new and retrofit FGD. One, done for EPA, by Catalytic Incorporated (Jain 1972) estimated a 30 percent greater price for a retrofit. Radian compared two designs for FGD systems done by the same vendor (Babcock and Wilcox)--Commonwealth Edison's Will County and Kansas City Power and Light La Cygne Station. After adjusting for size difference (using curves developed by Radian), they concluded that the actual cost increase due to retrofit was 23 percent. McGlamery's and Torstrick's (1974) data and estimates suggest that a 25 - 30 percent cost increase would seem to be typical.

Radian also presented estimates of the incremental cost trends for flue gas cleaning in the State of Ohio. The combination of higher unit investment costs, low load factor and short operating life results in a dramatic increase in "incremental control costs" associated with the degree of retrofit. Incremental costs are those associated with moving from larger new units to smaller old units. Since most of the capacity is less than 20 years old or greater than 100 MW in size, the incremental present cost of flue

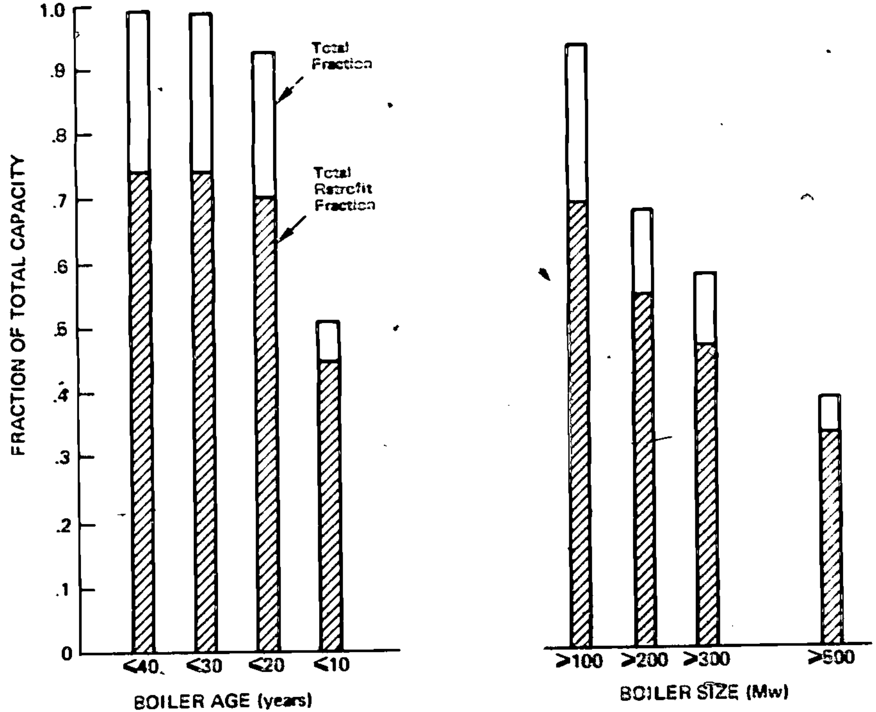


FIGURE 11-8: Fraction of Total Retrofittable Capacity in U.S. Versus Age and Size

gas cleaning remains constant for most units. As smaller, older units are included in further retrofit capacity, incremental control costs escalate rapidly.

Figure 11-9 illustrates this concept. The data was developed for the State of Ohio, but is probably in line with the Eastern U.S., since Ohio boilers account for about 14 percent of the total U.S. sulfur emissions from coal-fired generating capacity.

The second Kellogg study investigated retrofitting a limestone process at eight specific plants. They estimated installed costs of $40 to $100/kw, excluding the cost of the pond ($7 to $12/kw) and the price of the land. The report also states that the overall accuracy of the estimates is 30-35 percent with very little probability of underrun. Further no space scrubbing capacity was installed. Consideration of escalation factors, and the comments above would suggest that the Kellogg estimates would provide a comparable retrofit cost as suggested by McGlamery and Radian, i.e. a 30 percent increase from new plant costs which, in turn are $60 - $100/kw with $100/kw being most likely.

8.0 INSTITUTIONAL BARRIERS TO THE APPLICATION OF SULFUR OXIDE CONTROL SYSTEMS

There have been two recent studies discussing institutional barriers to the installation of FGD systems. These have been done by EPA (SOCTAP 1973) and by Radian Corporation. The SOCTAP report is much more detailed than the Radian report but both reach essentially the same conclusions, which are summarized below.

Typically utilities required about a 20 percent reserve capacity within a power region. In 1972 the actual reserves were between 7.8 and 24.3 percent, and about 15 percent on the east coast. Installation of FGD systems requires about 5 to 7 percent of the utility plant output and this will, of course, eventually decrease the reserve capacity.

In existing plants, scrubbers would have to be installed during a shutdown and take into

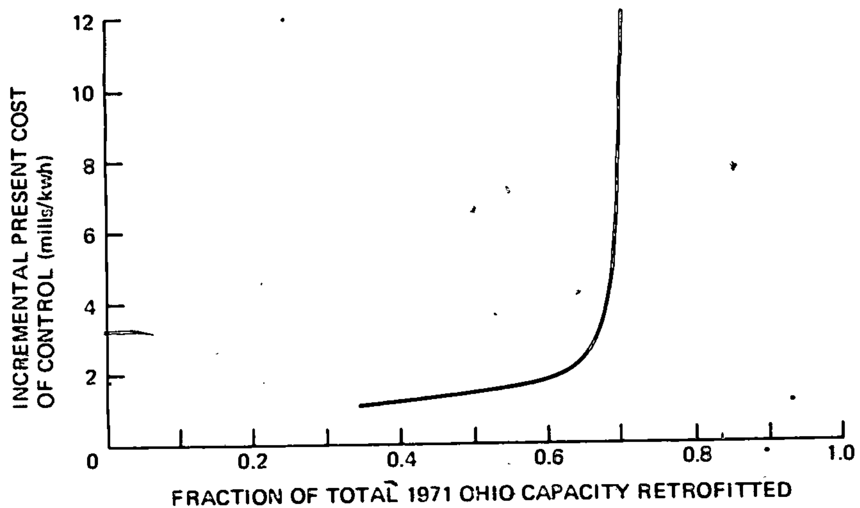


FIGURE 11-9: Incremental Cost of Control Versus Capacity Retrofitted

account the reserves available. It has been estimated that (because of these scheduling difficulties) no more than 10 to 20 percent of the capacity can be retrofitted each year.

Most utilities do not yet feel that they will be directly involved in chemical processing, (either SOx control technology or chemical cleaning of fuels), and their engineering staffs have remained relatively unchanged. There is a general feeling in the utilities that they can ultimately rely on the vendors; yet there is general skepticism of the vendors' claims at this time. If a decision is made at the management level that the utilities must turn to stack gas scrubbing as an abatement strategy, there will be heavy demand for in-house engineering talent to prepare specifications, review bids, provide liaison during the construction and shakedown phases, and assume responsibility for reliable operation of the scrubbers. That type of manpower will probably be in a very short supply.

Similarly, most utility companies do not have available on their staffs a supply of chemical process engineers. This manpower must be available to provide technical service capabilities and maintenance capabilities which are absolutely essential to keeping FGD systems operating once they are installed.

Currently there are about 15 vendors that have some established expertise in flue gas desulfurization and of these three or four have substantial experience and the capability to expand their services rapidly. This would suggest that the data given in Appendix 11, showing new scrubber installations, provides a reasonable estimate of the possible rate of installation--some 20 or so installations per year. Radian has estimated that vendors can produce 3 - 5 systems per year each. Radian's report also states that three specific vendors felt they could only have a combined total of 30 systems on line within three years of the contract award.

ACKNOWLEDGMENTS

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FOOTNOTES

1 Trouble from formation of this type of scale was one of the problems that led to abandonment of the process using a scrubber that followed a boiler into which limestone was injected. The procedure introduced free lime directly into the scrubber.

2 It is noteworthy that the beneficial effect of magnesium ions was not mentioned during hearings held in March through May of 1974 by Hearing Examiners of the Ohio Environmental Protection Agency. Indeed, testimony was heard concerning the question whether or not adequate supplies of limestone of low magnesium content were available.

APPENDIX 11-A

1.0 CONTROL OF EMMISSIONS OF SULFURIC ACID VAPOR AND MIST

Why have sulfate particulates in New York City air remained substantially constant over the past decade, while sulfur oxide emissions have decreased more than 85 percent? Any examination of this question ought to consider the hypothesis (Heller 1975) that emissions of sulfuric acid vapor and mist in New York City have declined nowhere nearly in proportion to the decline in sulfur content of the fuel.

The data that it was possible to gather in the time available tend to confirm the hypothesis, and point to need for experimental studies to determine sulfur dioxide emissions from representative space heating equipment in New York City while it burns oil containing 0.3 per cent sulfur, the level presently in use. Measurements would have to extend over a period of time, to recognize the rather wide variations in Sulfur dioxide production that might be expected at different firing rates and with upsets in the firing conditions.

Additional measurements at the higher sulfur levels typical of oils fired 10 years ago would be needed for comparison.

It would appear that emissions of sulfuric acid vapor and mist are typically much less for coal firing than for oil firing, even when coal at high sulfur level is compared with oil at much lower sulfur level. This raises the question whether or not the incidence of acid rains in Sweden late in the 1950's (Eriksson 1971) might have not have been associated with the massive conversion from coal to oil that took place in Sweden at about that time. A similar question would be raised concerning the acid rains of the northeastern United States in recent years.

1.01 Production of Sulfur Trioxide during Combustion

The production of sulfur trioxide in a furnace or boiler can vary widely, as data for oil and coal firing, to be presented in Section 4.03 and 4.05, will illustrate. This section will introduce the subject briefly.

Sulfur trioxide can be a marked function of the age of a combustion device, its design, and the method of firing. For a given device, it is a function of sulfur level in the fuel when a fuel of a given type is fired at varying sulfur content. In comparing different combustion devices, the sulfur content of the fuel can be far outweighed in importance as a factor affecting sulfur trioxide production by such things as the degree of excess oxygen and by the method of mixing of fuel with the oxygen (Jackson et al. 1969, Krause 1959).

One mechanism for producing su ur trioxide operates at the temperature of the flame (Krause 1959, Widersum 1967, Brown 1966, Widell 1953, Williams 1964, Barrett et al. 1966). Sulfur dioxide reacts with atomic oxygen to yield concentrations of sulfur trioxide that paradoxically exceed the thermodynamic equilibrium yield for reaction of sulfur dioxide with elemental oxygen at the temperature of the flame.

The equilibrium conversion of Sulfur dioxide to Sulfur trioxide increases as the temperature in the flue gas drops. For the amounts of excess combustion air that are normally supplied, the equilibrium conversion closely approaches 100 percent at normal stack temperatures. Catalytic conversion becomes important at lower temperatures, especially, it would appear in the general temperature range between about 1200 degrees and 700 degrees F (Krause 1959, Widersum 1967, Barrett et al. 1966).

1.02 Conversion of Sulfur Trioxide to Sulfuric Acid Vapor

Below about 750 degrees F, sulfur trioxide combines with water vapor in the flue gas to form sulfuric acid vapor (Gundry et al. 1964).

If the flue gas were to be cooled indefinitely, a temperature would be reached at which a mist of sulfuric acid particles would begin to appear in the gas. This temperature is termed the dew-point of the flue gas, and is a function of both the concentration of sulfuric acid vapor in the gas and also the concentration of water vapor. Muller's relation for dew-point versus sulfur trioxide of flue gas appears to have been confirmed by the best available data and to have general support among workers on the subject (Lisle and Sensenbaugh 1965, Dismukes 1975). Another view (Gimitro and Vermeulen 1964) reflects the complexity of the thermodynamic situation, and there is room for further research.

Small combustion devices, such as equipment for space heating and small industrial boilers, generally operate at stack temperatures far above the dew-point, so that substantially all emissions of sulfur trioxide from small devices are in form of sulfuric acid vapor. This would be true for coal or oil firing.

Large utility boilers fired with oil frequently operate at a stack temperature just above the dew-point. If the temperature were allowed to fall below the dew-point, serious corrosion of heat transfer surface in the air heater would occur. The dew-point is lower if the combustion uses less excess air (see Section 1.04).

Large utility boilers fired with coal are able to operate at lower stack temperatures, without serious air heater corrosion, than are generally possible for oil-fired units, except for units that use low excess air. The lower stack temperatures possible in coal-fired units at usual amounts of excess air may in part reflect a smaller production of sulfur trioxide, but also, they seem to reflect an uptake of sulfuric acid vapor by adsorption upon fly ash or reaction with alkaline material in fly ash. Emissions of sulfuric acid vapor or mist are generally lower for coal-fired boilers, unless the comparison is with an oil-fired unit at low excess air. Total sulfate emissions, of course, are significantly lower for a coal-fired unit only if an efficient electrostatic precipitator or an efficient scrubber is provided to remove fly ash.

1.03 Levels of Sulfur Trioxide in Flue Gas from Oil Firing

The wide variation in production of sulfur trioxide in oil-fired furnaces can best be appreciated by discussing in turn the several experiments for which data are summarized in Figures App. 11-A 1 and 2.

The hour glass symbol I in Figure App. 11-A1 indicates the range of data for firing natural gas spiked with H_2S, to simulate fuel at 1, 2.5, and 6 percent sulfur, in a small laboratory combustor (Barrett et al. 1966). The unit had stainless steel walls cooled to 500 F, and the stainless steel surface at this temperature was demonstrated to lack any catalytic virtue for conversion of sulfur dioxide and sulfur trioxide. The tests were conducted at 15 percent excess air, and the data represent the production of sulfur trioxide in the flame itself, unaided by catalytic effects, for the specified experimental conditions.

The open and closed triangles, △ and , ▲ Figure App. 11-A1 are data for combustion of oil in a small laboratory furnace with a refractory lining (Crumbley and Fletcher) which undoubtedly contributed some catalytic effect (Barrett et al. 1966). The open triangles are for operation at a furnace-wall temperature of 2300 F. The closed triangles are for 2010 F, illustrating the pronounced effect of firing procedure upon

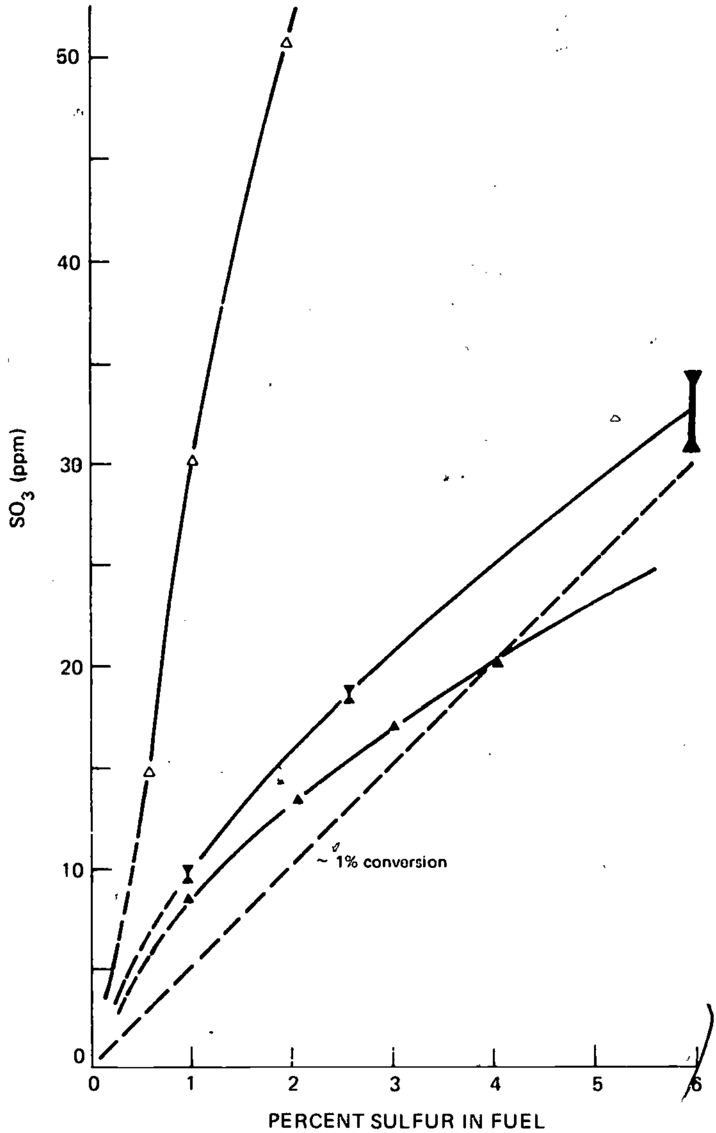

FIGURE App. 11-A1: Sulfur Trioxide Levels in Flue Gas from Careful Laboratory Studies or Combustion of Oils (the Triangles) and of Natural Gas Spiked with Hydrogen Sulfide (the Hour Glass Points).

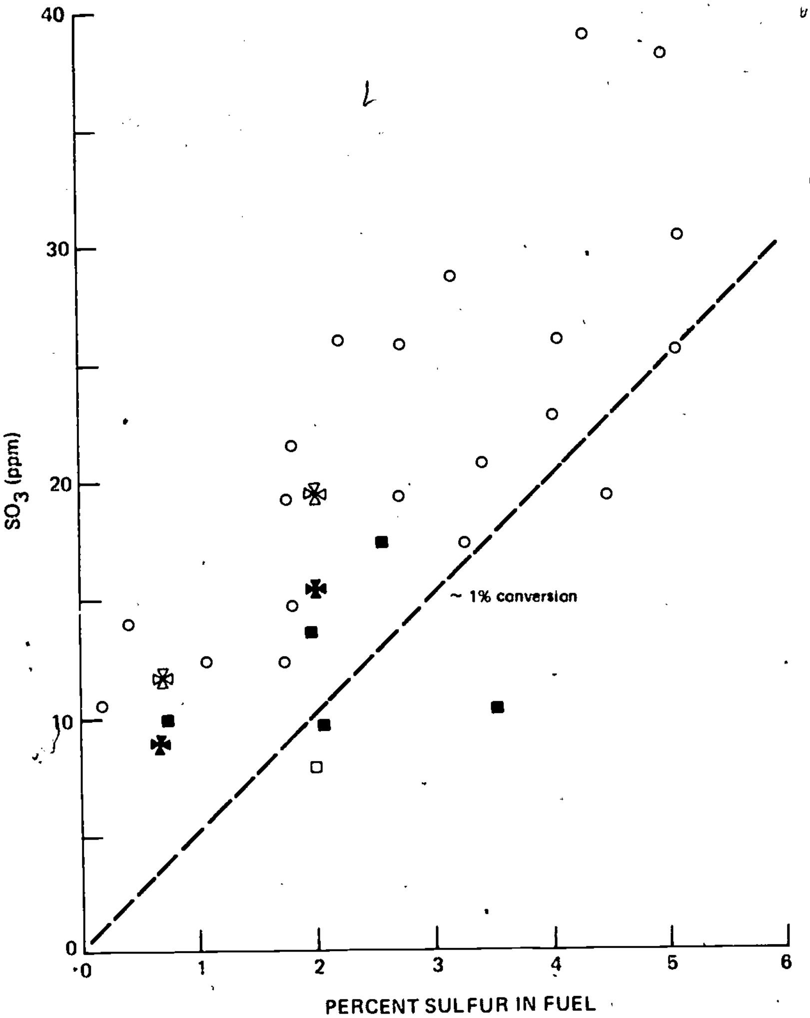

FIGURE App. 11-A2: Sulfur Trioxide Levels in Flue Gas from Oil Combustion in

(a) Laboratory Furnace Tests (the Circles) More Nearly Representative of Practical Small-Scale Combustion Devices, and

(b) Small, Industrial Boilers (the Remaining Points), between 40 and 175 Thousand Pounds per Hour of Steam.

(See text for explanation of test arrangements).

sulfur trioxide production in the same furnace. The series of experiments at 2300 F gave 67 ppm Sulfur trioxide at 3 percent sulfur in oil.

If each of the three carefully done laboratory tests for which data are presented in Figures is considered in isolation, a smooth curve can be drawn through each set of data that includes the origin.

The open circle ○ in Figure App. 11-A2 give data for a small laboratory furnace with a refractory lining (Rendle and Wildsdon 1956). The combustion chamber was stated to be at 1830 F. Each point plotted for this furnace in Figure App. 11-A2 is the average of Sulfur trioxide readings for a sample of gas taken from the combustion chamber and a sample taken from a "cool" exit chamber at about 650 F. The tests used 25 percent excess air. Fuels were a variety of oils, both oils inherently containing sulfur, and oils to which sulfur was added in form of carbon disulfide. The data shown by the circles in Figure App. 11-A2 are probably more nearly representative of the performance of practical small-scale oil-fired combustion devices than the data of the more carefully done laboratory tests given in Figure App. 11-A1.

The closed squares ■ in Figure App. 11-A2 are data for an industrial boiler able to supply about 50,000 lbs/hr of steam (Corbett 1953). Each of the four points at the higher sulfur levels in the oil is an average of 15 to 20 readings for samples taken at several temperature levels in the boiler and at several steaming rates. The point at 0.75 percent sulfur is deduced from an average of dew-point readings for several steaming rates, judging from the average dew-points for the operations at the higher sulfur levels. The excess air ranged upward from about 12 percent.

The open square □ is for a 40,000 lbs/hr industrial boiler burning 2 percent sulfur oil at about 25 percent excess air (personal communications from confidential source 1975).

The open and closed Maltese crosses, ✠ and ✠ are for two larger industrial boilers, for 175,000 and 130,000 lbs/hr of steam respectively, at about 10 percent excess air

(Dichl and Luksch 1964). (These are boilers B and C respectively in Figure App. 11-A3).

A feature of the data is that the production of sulfur trioxide remains above about 8 ppm of sulfur trioxide in the flue gas from the lowest sulfur level in oil that was reported.

Although the careful laboratory tests yield data consistent with gentle curves that included the origin, the rougher tests illustrated in Figure 2 App. 11-A2 suggest that a high percentage of the sulfur in a low-sulfur oil, even an oil at 0.3 percent sulfur, can sometimes be converted to sulfur trioxide. Notice that the dashed lines in Figures App. 11-A1 and 2 represent very approximately 1 percent conversion of sulfur dioxide in flue gas to sulfur trioxide.

It is possible to suppose that the situation in respect to space heating equipment and other small furnaces in New York City may be worse, possibly even much worse, than the data of Figure App. 11-A2 might suggest.

An old oil-fired boiler is reported to convert typically 5 percent of the sulfur to sulfur trioxide when using oil at 2 percent sulfur (Weir 1975). After a thorough cleaning, this boiler characteristically does not make much sulfur trioxide for a week or so, but conversion to sulfur trioxide is renewed thereafter. The boiler burned oil containing vanadium. Similar effects of cleaning and time of operation upon sulfur trioxide levels have been reported by others (Crumley and Fletcher), as well as excursions upward in sulfur trioxide production with changes in load (Jackson et al. 1969).

Iron rust is catalytic toward the conversion of sulfur dioxide to sulfur trioxide (Krause 1959, Barrett et al. 1966). Vanadium fly ash has also been shown to be catalytic. Small-scale tests have shown conversions as high as 20 percent of the sulfur in a Venezuelan residual oil when residence time of about 4 seconds was provided in the 600 to 1000 F temperature range (Kapo et al. 1973), and conversions beyond 90 even for oils containing very little vanadium percent were observed at a residence time of about 1 minute (Kapo 1975).

Old combustion equipment in New York City may well be characterized by vanadium fly ash deposits and rust. Space heating equipment generally operates at efficiencies no better than about 70 to 75 percent, so that stack temperature can be expected often to be in a range where catalytic effects can operate. Tall buildings mean tall flues, and, often, a relatively long residence time for conversion to occur.

1.04 Control of Sulfur Trioxide Emissions from Oil Firing

1.041 Low Excess Air

A control measure accessible to the operator of a large utility boiler, and not to the user of small combustion equipment, is the limitation of excess air. Figure App. 11-A3 illustrates the sharp decline in Sulfur trioxide when the excess air is reduced below about 10 percent.

Two curves are shown in Figure App. 11-A3 for boiler A (Jackson et al. 1969). The lower Sulfur trioxide values were for a fairly usual firing arrangement. The upper values were observed when special means were introduced to obtain intimate mixing of oil and air, in order to acheive "better" combustion conditions. A sister boiler produced over 100 ppm Sulfur trioxide at 1.5 percent oxygen in flue gas (Jackson et al. 1969).

Two sets of curves are given in Figure App. 11-A3 for boilers B and C (Diehl and Luksch 1964). The decrease in sulfur trioxide when 0.72 percent sulfur oil was substitutes for oil at 2.05 percent sulfur was not in proportion to the sulfur level in the two oils. As before, in connection with Figure App. 11-A2, one is led to the conclusion that sulfur trioxide emissions fron the firing of oil at 0.3 percent sulfur will not be negligibly small at the levels of excess air usual in small combustion equipment.

Boiler D had a "standard" firing arrangement (Jackson et al. 1969). The low emissions of sulfur trioxide from this boiler, as well as the

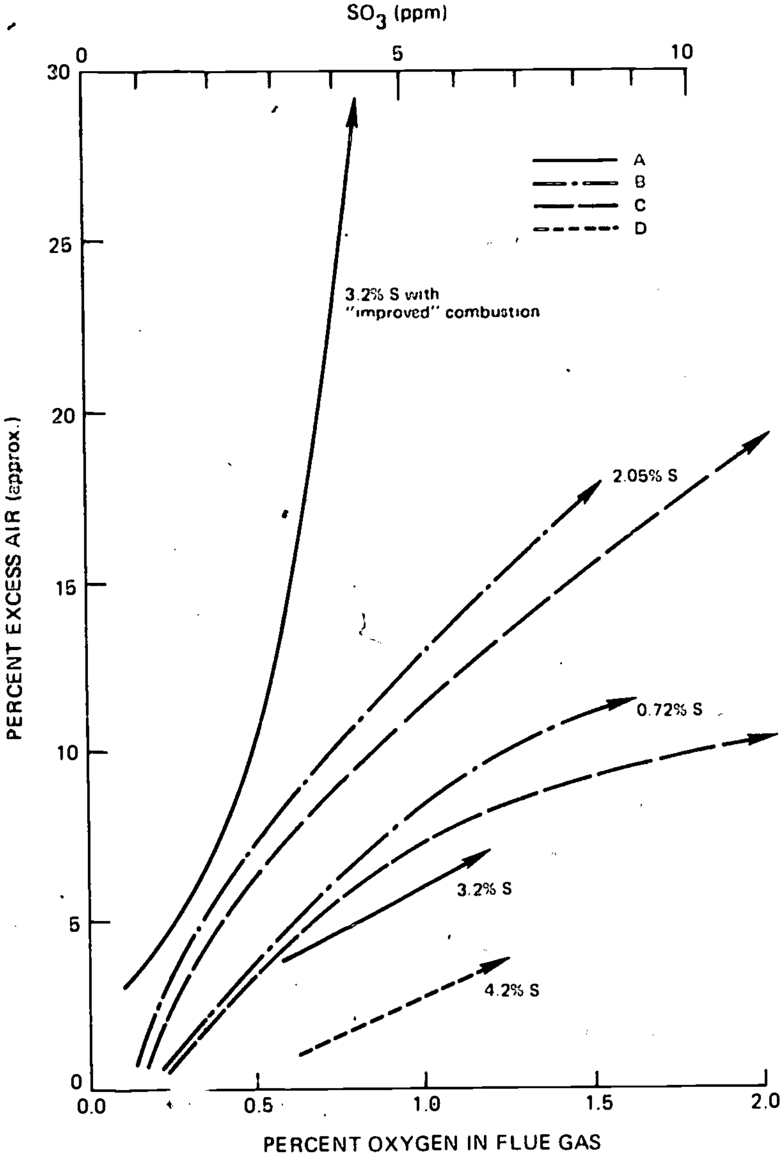

FIGURE App. 11-A3: Sulfur Trioxide in Flue Gas from Boilers at Low Excess Air. (Note: Data for boilers A and D are from Jackson et al. [1969]. Data for boilers B and C are from Diehl and Luksoh [1964]).

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comparison of emissions from boiler A with "standard" and "improved" combustion, lead to the impression that sulfuric acid vapor emissions might be reduced by appropriate adjustments, as yet not well understood, in the firing practice.

That emissions from large utility boilers can be kept small be proper firing at low levels of excess air is illustrated by the following data (personal communication from a confidential source 1975):

Boiler Size	% S in Oil	% O2 in Flue Gas	ppm SO3
100-MW	2.5%	1.6%	2
		2.5%	3
	1.5%	1.7%	0.8
		2.3%	1.7
200-MW	0.8%	1.5%	2.6
400-MW	0.9%	2.5%	3.5

The first four sets of data were from tests on the same 100-MW boiler.

It should be appreciated that operation at low excess air requires extreme care and good control procedures. There is risk that failure of the air rate to track changes in fuel rate will lead to a fuel-rich mixture and development of an explosive gas mixture in the boiler. The difficulties become serious below about 5 percent excess air, and routine operation at 3 percent excess air would demand the utmost caution.

1.042 Scrubbing

A 10-MW horizonal scrubber (see Chapter 11, Section 2.12) on an old oil-fired boiler sometimes receives sulfuric acid mist at 50 ppm when oil at 2 percent sulfur is used. The efficiency of removal is typically 50 percent (Weir 1975), although the efficiency has been varied by changes in scrubber operating parameters. Presumably higher removal efficiencies can be acheived by operating the scrubber at greater drop in pressure in the flue gas being treated.

If the primary object of a scrubber on an oil-fired boiler is to achieve a plume of low opacity, rather than to remove sulfur dioxide, then scrubbing may be more difficult for oil combustion than for coal. The particle size of ash resulting from oil firing is typically 0.1 tp 0.3 micron, much smaller than the fly ash from coal combustion. As the gas is cooled for scrubbing, the oil ash serves as condensation nuclei for sulfuric acid vapor (Weir 1975). A removal efficiency for the oil ash beyond about 50 percent may be difficult to acheive.

1.043 Electrostatic Precipitation

Tests by Consolidated Edison Company of New York (Ramsdell 1975) have shown that electrostatic precipitators working at about 300 F on flue gas from firing of oil at 1 percent sulfur can provide a collection efficiency of about 50 percent. The collected material comprised oil ash particles, typically 0.1 to 0.3 micron in size, on which some sulfuric acid vapor had condensed. The amount of sulfuric acid removed in this way is not known.

Operation of the precipitator at this temperature level is difficult, and Con Edison does not normally keep the precipitators in service. The difficulty arises from the hygroscopic character of the ash particles collected and their tendency to form a gummy mass.

It may be noted that Con Edison's precipitators are unusually large, having been increased in size during the latter 1960's to afford 99+percent collection efficiency for fly ash from combustion of low-sulfur coal.

One of Con Edison's precipitators, on its Ravenswood No. 3 boiler, was installed to work at about 700 F. It afforded test efficiencies between 99.2 and 99.6 percent when working some years ago on coal containing 1 percent sulfur, the results averaging at 99.4 percent.

A test of this precipitator on ash from oil of 1 percent sulfur gave an efficiency of 75 percent. A fire in an ash hopper has temporarily put this precipitator out of service, and illustrates another difficulty in the handling of oil ash.

A precipitator operating at 700 F does not prevent sulfuric acid vapor from reaching the environment, for either coal or oil firing.

1.044 Additives

For many years, Long Island Lighting Co. has injected finely divided magnesium oxide into boilers fired with high-sulfur Venezuelan residual oil. The effect is to suppress sulfur trioxide emissions and to capture vanadium in form of magnesium vanadate. The Company enjoys a considerable revenue from sale of the vanadium-rich fly ash, and indeed, supplies a significant fraction of the nation's requirement for vanadium.

1.045 Control of Small Combustion Equipment

Control of sulfuric acid vapor emissions from small oil-fired combustion devices may not be easy. In any case, measurements are needed before too much thought is given to control procedures and appropriate levels of control.

It is possible that a good measure of control may be obtained simply by subjecting such devices and their flues to a thorough cleaning.

It might be added that space heating equipment accounted for about 57 percent of sulfur oxide emissions in 1973 in New York City. Electricity generation accounted for about 37 percent.

1.05 Control of Sulfate Particulate Emissions from Coal Firing

1.051 Precipitation

A summary of data for a large number of boilers (Rosohl 1956) suggests that dew-points for flue gases from coal firing are generally far below those of oil firing at usual levels of excess air.

The differences may arise not so much from a lower production of Sulfur diovide in a coal flame as from a tendency for the vapor pressure of sulfuric acid to be considerably lower in the presence of fly ash. Data suggesting such a tendency have been reported for a boiler fitted with an electrostatic precipitator working at about 95 percent efficiency (Archbold 1961). The level of sulfur trioxide at 750 to 700 F, just ahead of the air heater, was 22.9 ppm. The temperature at the outlet of the air heater was about 280 to 285 F, and the sulfur trioxide level was 7.1 ppm, far below the equilibrium content of sulfuric acid vapor in coal combustion gas at 280 to 285 F. The gas entered the precipitator at 7.1 ppm of Sulfor trioxide. The outlet of the precipitator showed 2.0 ppm, suggesting that the uptake of Sulfur trioxide by fly ash, whether by adsorption or by chemical reaction with alkalinity in the ash, had continued during the approximately 1.6 seconds of gas residence in the precipitator. It does not seem probable that the sulfur trioxide level would have been reduced much further if the gas residence had been longer, as is characteristic of modern precipitators working at 99+percent efficiency.

Tests on at least one coal-fired power station showed fly ash collected by a precipitator at 95 percent efficiency had a pH of 7 or even a bit above, while the fines passing through this precipitator has a pH of 2.5 to 3 (Grob 1961). This observation is consistent, it might be noted, with a finding that the greatest accumulation of sulfur in California urban aerosels is on particles in the 0.1 to 0.65 micron range of size (Flocchini et al. 1974).

These facts suggest that fly ash particles act as condensation nuclei for sulfuric acid vapor in flue gas from coal combustion. Inherently, it would appear that the content of sulfuric acid vapor in coal flue gas from a utility boiler operating at high efficiency is usually small. It is plausible to suppose that the vapor condenses more or less uniformly over all of the particulate surface that is available, so that smaller particles will take up more of the acid vapor, unless alkalinity in the ash directs acid to the larger particles.

This view is consistent with the fact that fly ash from a coal of low sulfur content is difficult to collect by electrostatic precipitation of around 300 F, but can be made more collectible by addition of sulfur trioxide to the flue gas. If done properly, the conditioning of fly ash by addition of sulfur trioxide does not lead to significant quantities of sulfuric acid vapor in the stack effluent (personal communication from two confidential sources 1975).

It would appear that an electrostatic precipitator working at high efficiency should prevent most sulfate particulate matter, as well as sulfuric acid vapor, from reaching the atmosphere. Of course, there seems to be general agreement that larger particles of such matter are not injurious to health, and a chemical examination of the ultra-fine particles that escape the precipitator would seem to be in order.

1.052 Scrubbing

The level of sulfuric acid mist typically reaching the 10-MW limestone scrubber at Shawnee Station (see Chapter Section 3.04) is about 8 to 10 ppm (Moore 1975). This is a representative level for T.V.A.'s coal-fired stations, that generally produce a flue gas containing between about 5 and 10 ppm of sulfur trioxide. Although the 10-MW limestone scrubber displays excellent efficiency for removing fly ash, the efficiency for removing sulfuric acid mist is poor (Princiotta 1975), the effluent usually containing about 5 ppm of sulfuric acid mist (Moore 1975).

1.06 Conclusion in Respect to Emissions of Sulfuric Acid Vapor

The hypothesis (Heller 1975) that sulfate particulate levels in the air of New York City and other urban communities have remained high because of significant emissions of sulfuric acid vapor even from the firing of oil at 0.3 percent sulfur is sufficiently credible to warrant an experimental test.

Measurements are needed to compare emissions today and ten years ago from typical oil-fired devices, especially small furnaces and space heating equipment. The tests should reflect typical operating practices.

The evidence that it has been possible to find in the time available supports a hypothesis that emissions of sulfuric acid vapor have not declined in New York City in proportion to decline in sulfur content of fuel. It might be further hypothesized that the limiting factor that has determined the level of sulfate particulate matter in New York City has been the surface area of the total particulate matter that is available for adsorption of sulfuric acid vapor. Under the latter hypothesis, contours of sulfate particulate matter and of the acidity of rainfall around New York City might be expected to reveal the City as a source of sulfuric acid vapor that was nor adsorbed by particulates within the city. Such contours would also, of course, shed a sharp light upon the hypothesis that sulfate particulate matter in New York City has originated from sulfur dioxide emitted by power stations at a distance, this sulfur dioxide having been converted to sulfate as it moved toward the city. In other words, is New York City a "hot spot", or does its atmosphere represent a region extending westward, say, to Ohio?

APPENDIX 11-B

This appendix contains tables presenting a summary of electric utility flue gas desulfurization facilities in the United States, and a breakdown of the installed FGD systems and those under construction.

TABLE App. 11-1

Summary of Electric Utility Flue Gas Desulfurization Facilities in the United States

	MW Capacity (No. of Plants)			
PROCESS	CURRENTLY INSTALLED(1)	UNDER CONSTRUCTION	PLANNED	TOTALS
Limestone Scrubbing	1,904(8)	2,950(7)	8,897(20)	13,351(35)
Lime Scrubbing	715(4)	2,944(6)	4,651(17)	8,310(27)
Limestone or Lime	30(1)	650(1)	6,100(10)	6,780(12)
Magnesium Oxide Scrubbing	370(3)	---	576(2)	946(5)
Catalytic Oxidation	110(1)	---	---	110(1)
Wellman-Lord	---	115(1)	715(2)	830(3)
Aqueous Sodium Base Scrubbing	250(2)	125(1)	125(1)	500(4)
Double Alkali	32(1)	20(1)	---	52(2)
Process Not Selected	---	---	6,140(11)	6,140(11)
TOTAL	3,411(20)	6,404(17)	27,204(63)	31,019(100)

(1) Not necesaarily in operation. Some are plants which have recently been shut down.

TABLE App. 11-2

Summary of Installed Electric Utility Flue Gas Desulfurization Systems in the United States

FGD PROCESS/ POWER STATION	NEW OR RETROFIT	SIZE MW	FUEL TYPE	% ASH	% S
LIMESTONE SCRUBBING					
Arizona Public Service Cholla No. 1	R	115	Coal	5-15	0.4-1.0
City of Key West	N	37	Oil	-	2.75
Commonwealth Edison Will County No. 1	R	167	Coal	10	2.0 (1)
Kansas City Power & Light					
La Cygne No. 1	N	820	Coal	10-20	4.0
Hawthorne 3 & 4	R	240	Coal	-	0.6 - 3.0
Lawrence 4 & 5	R/N	525	Coal	-	3.5
LIME SCRUBBING					
DuQuesne Light Phillips	R	410	Coal	10	2.3
Louisville Gas & Electric Paddy's Run No. 6	R	65	Coal	14	3.7
Southern California Edison Mohave No. 2	R	160	Coal		0.5-0.8
LIME/LIMESTONE SCRUBBING					
TVA Shawnee No. 10 (2)	R	30	Coal	12-15	3.0
MAGNESIUM OXIDE SCRUBBING					
Boston Edison Mystic No. 6	R	150	Oil	-	2.8
Potomac Electric, Dickerson 3	R	95	Coal	10-20	2.0

TABLE App. 11-2 (continued)

FGD PROCESS/ POWER STATION	NEW OR RETROFIT	SIZE MW	TYPE	FUEL % ASH	% S
SODIUM CARBONATE SCRUBBING					
Nevada Power					
Reid Gardner No. 2	R	125	Coal	9	0.6
Reid Gardner No. 1	R	125	Coal	9	0.6
DOUBLE ALKALI					
General Motors Chevrolet Parma 1,2,3,4 1	R	32	Coal	10-12	20.25
CATALYTIC OXIDATION					
Illinois Power Wood River No. 4	R	110	Coal	10	3.3

Notes:

1. Will County is now (1974) burning low sulfur Montana coal at least part of time.
2. EPA sponsored facility operated by Bechtel. Used as experimental facility.
3. Not all systems are now in operation.

TABLE App. 11-3

Flue Gas Desulfurization Systems Under Construction in the United States

Size, MW	New or Retrofit	Fuel %S	Fuel Type	Process	Contractor	Location
250	N	0.44	Coal	Limestone	Research Cottrell	Arizona Public Service Cholla No. 2
100	N	2.5-3	Coal	Limestone	Riley Stoker	Central Illinois Light Co., Duck Creek No. 1
375	N		Coal	Lime	UOP	Columbus and Southern Ohio, Conesville 5
375	N		Coal	Lime	UOP	Conesville 6
180	R	3.7	Coal	Limestone	Peabody	Detroit Edison St. Clair No. 6
510	R		Coal	Lime	Chemico	Duquesne Light Elrama
64	R	3.8	Coal	Lime	American Air Filter	Kentucky Utilities Green River 1,2,3
178	R	3.5-4.0	Coal	Lime	American Air Filter	Louisville Gas & Elect. Cane Run 4
425	R	3.5-4.0	Coal	Lime	American Air Filter	Mill Creek 3
360	N	0.8	Coal	Lime	C.E.A.	Montana Power Colstrip 1
360	N	0.8	Coal	Lime	C.E.A.	Colstrip 2
125	R	0.5-1.0	Coal	Sodium Carbonate	C.E.A.	Nevada Power Reid Gardner 3
115	R	3.2-3.5	Coal	Wellman Lord	Davy Powergas	NIPSCO Mitchell 11

TABLE App. 11-3 (cont.)

FGD Systems Under Construction

Size, MW	New or Retrofit	Fuel %S	Fuel Type	Process	Contractor	Location
680	N	1.0	Coal	Limestone	Combustion Engineering	Northern States Power Sherburne 1
680	N	1.0	Coal	Limestone	Combustion Engineering	Sherburne 2
880	N	4.3	Coal	Lime	Chemico	Pennsylvania Power Co. Bruce Mansfield 1
800	N	4.3	Coal	Lime	Chemico	Bruce Mansfield 2
120	R	2.5	Coal	Magnesium Oxide	United Engineering	Philadelphia Electric Eddystone 1 (now in startup phase)
160	R	0.5-0.8	Coal	Limestone	UOP	Southern Calif. Edison Mohave 1
345	N	0.5	Coal	Lime/ Limestone	Combustion Engineering	Southwest Public Serv. Harrington 1
200	N		Coal	Limestone	UOP	Springfield Utility Bd. Southwest 1
550	R	3.7	Coal	Limestone	TVA	TVA Widows Creek 8
793	N	0.4	Coal	Limestone	Research Cottrell	Texas Utilities Martin Lake 1
793	N	0.4	Coal	Limestone	Research Cottrell	Martin Lake 2
650	N	1.5	Coal	Lime/ Limestone	Combustion Engineering	Pub. Serv. Co. of Ind. (Gibson #2)

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CHAPTER 12

CONTROL OF AMBIENT SULFUR DIOXIDE CONCENTRATIONS WITH TALL STACKS AND/OR INTERMITTENT CONTROL SYSTEMS

(Chapter 12 was written by Robert W. Dunlap under the general supervision of the committee, which reviewed the work at several stages and suggested modifications which have been incorporated. While every committee member has not necessarily read and agreed to every detailed statement contained within this report, the committee believes that the material is of sufficient merit and relevance to be included in this report.)

INTRODUCTION

Air quality implementation plans were enacted by the states in 1972 to bring about local compliance with national ambient air quality standards (AAQS). Not surprisingly, sulfur dioxide emission limitations for electric utility power plants varied widely in these plans. Much of the variation can be ascribed to regional fuel availability considerations, to state-imposed air quality standards or timetables, and to differing methodologies for relating emissions and air quality. Some of the variation, however, represents moot recognition of the different capacities of regional atmospheres to safely assimilate specific emission levels. For example, Pennsylvania's regulations (Table 12-1) allow about an eightfold difference in emission rates, dependent upon source location. These allowed spatial differences in emission rates, with all rates designed to meet the same sulfur dioxide

TABLE 12-1

Pennsylvania Sulfur Dioxide Regulations for Steam Power Plants (Existing Sources)

Area	Allowed Lbs $SO_2/10^6$ BTU	Allowed Coal % S*
Philadelphia	-	0.3
Allegheny County (Pittsburgh)	0.6 - 1.0	0.4 - 0.6
Beaver Valley, Monongahela, and SE Pennsylvania Air Basins	0.6 - 1.0	0.4 - 0.6
Allentown-Easton, Erie, Harrisburg, Johnstown, Lancaster, Reading, Scranton-Wilkes Barre, York Air Basins	1.8 - 3.0	1.1 - 1.9
All Other Regions	4.0	2.5

* Assumes 12,500 BTU/Lb Coal.

AAQS, are understandable in terms of different numbers of sources, background sulfur dioxide levels, meteorological characteristics, and topography for Pennsylvania's air basins.

Temporal variations in emission limitations were not part of most states' implementation plans, except as part of emergency episode criteria. However, the situation here is roughly analogous to the spatial variation situation. At any given emission rate, it can be generally stated that carefully chosen temporal fluctuations about that rate will provide air quality benefits, since variability in meteorological conditions causes significant changes in the air quality impact of any given level of emissions. With temporal controls, when dispersive characteristics of the atmosphere are unfavorable, minimum emission rates are demanded; when optimal atmospheric dispersion conditions exist, higher emission rates than the base rate are permitted.

This air quality management concept, i.e., that emissions can be spatially and temporally varied to achieve compliance with air quality standards, has only been partially accepted by regulatory agencies over the past several years. In particular, temporal emission controls for sulfur dioxide, here termed intermittent control systems (ICS), have been recognized in this country only in specific situations, and have been a focal point of a lively debate between regulatory agencies and some parts of the electric utility industry.[1] This position paper details some of the aspects of this debate in order to assess current and future public policy concerning ICS programs.

TALL STACKS AND ICS PROGRAMS

An intermittent control system can be defined as "...system whereby the rate of emissions from a source is curtailed when meteorological conditions conducive to high ground-level pollutant concentrations exist or are anticipated" (FR 1973). For power plants, two intermittent control strategies are potentially available: (a) fuel switching, i.e., using a temporary supply of low sulfur fuel; (b) load shifting, i.e., shifting a portion of the electrical load to an interconnected generating station with capacity in excess of demand.

Tall stacks are closely associated with ICS programs, since increased stack height can yield decreased needs for intermittent emission reductions, assuming that control of ambient sulfur dioxide concentrations is the sole objective. For example, TVA's comparison of estimated maximum ground level concentrations of sulfur dioxide associated with different meteorological dispersion conditions and stack heights between 60-360 meters is shown in Figure 12-1 (Carpenter 1971). Three critical meteorological conditions are depicted, each corresponding to a different plume transport condition: coning, inversion breakup, and trapping (mixing depths between 760-1065 meters).[2] For coning and inversion breakup conditions, an inverse relationship between stack height and concentration applies. For trapping conditions, concentrations are primarily determined by the elevation and magnitude of the subsidence inversion or stable layer aloft; stack height does play a role if the plume emitted from the stack penetrates the layer of stable air and is not trapped below it (concentrations for plume penetration cases are approximately zero and are not shown in Figure 12-1). Based on dispersion model results such as these, as well as ambient air quality data, TVA suggests the anticipated requirements for ICS measures at its Kingston steam plant will drop from 55 days per year (7-hour average duration) for the plant's current stack configuration (four 250-foot stacks and five 300-

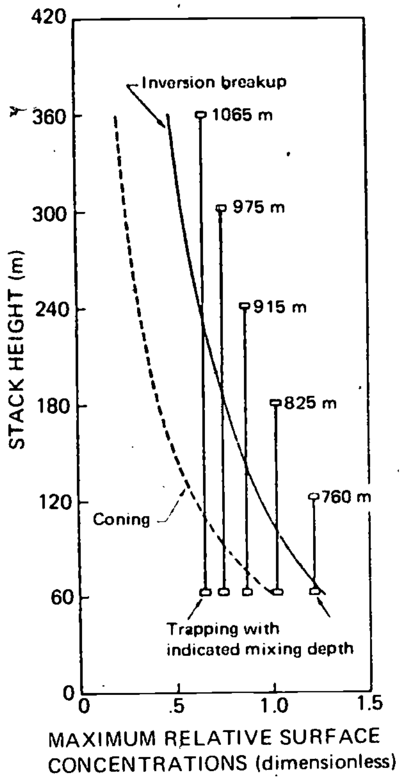

FIGURE 12-1: Maximum Relative Concentration of SO_2 as a Function of Meteorological Conditions and Stack Height (Carpenter et al. 1971)

foot stacks) to zero days per year for two 1000-foot stacks (TVA 1973).

Two basic types of tall stack-ICS programs are thus apparent. In the first, stacks of sufficient height are built so that ground-level concentrations do not abrogate standards, even without use of ICS measures. This approach is advocated in Great Britain (Stone and Clark 1967, Lucas 1974), and also by some utilities in this country for plants located in relatively flat terrain where no limitations on stack height (e.g., limitations imposed by flight safety requirements near airports) exist (Frankenberg 1970, Smith and Frankenberg 1974). In the second approach, applicable where ambient sulfur dioxide standards cannot be met simply by increasing stack height, tall stacks are coupled with ICS measures to control ground-level sulfur dioxide concentrations. Controversy has existed over whether tall stacks alone can adequately control ground-level sulfur dioxide concentrations. It seems clear, however, that as stack height increases, the required frequency of use of ICS measures decreases for most, if not all, given control situations. The frequency of ICS use is also dependent on emission rate, other stack parameters (stack gas velocity, temperature), meteorological conditions, and topography.

Regulations proposed by the Environmental Protection Agency (EPA) do not accept increases in stack height beyond levels of "good engineering practice" as acceptable air quality control measures, unless this is accomplished as part of an approved ICS program (FR 1973). These same proposed regulations impose stringent limitations on the types of sources and situations for which ICS programs are acceptable, and include the following constraints:

•The ICS program must be a supplement to constant emission controls; the source must undertake research and development programs to accelerate development of applicable constant emission reduction technology.

•The system must exhibit a high degree of reliability (ability to protect against violation of AAQS) and must be legally enforceable. Elements of such an ICS are shown in Figure 12-2 (PEDCO 1974) and include:

•A sulfur dioxide monitoring network sufficient to allow calibration of a dispersion model, as well as interpolation between samplers.

•An operating model which relates meteorological inputs, emission rates, source data, terrain and location factors to current and future air quality.

•Meteorological inputs suitable for use in air quality forecasting.

•Objective rules for emission control, relating air quality predictions to controlled emission schedules.

•A requirement for continuous evaluation and systematic improvement of ICS reliability (upgrade system).

Figure 12-2 indicates the two different modes in which an ICS may be operated: (1) open-loop emission control, in which the decision to reduce emissions is made based on the output of the predictive model; and (2) closed-loop emission control, an emergency operating mode, in which real-time measurements from the air quality monitoring network are used to override the operating model in making the control decision. The closed-loop mode occurs when the open-loop mode of operation has failed, i.e., when ambient air quality measurements exceed specified threshold values and/or AAQS.

From the system diagram presented in Figure 12-2, ICS reliability and performance depend on a number of factors:

(1) The adequacy and accuracy of monitoring the pertinent emissions, air quality, and meteorological parameters;

(2) The ability to forecast meteorological inputs to the operating model, including transition periods between different types of weather and potential interactions between meteorology and terrain;

(3) The adequacy of the air quality forecast model or models, which must represent all possible meteorological conditions, account for terrain and location factors, and estimate anticipated emission rates;

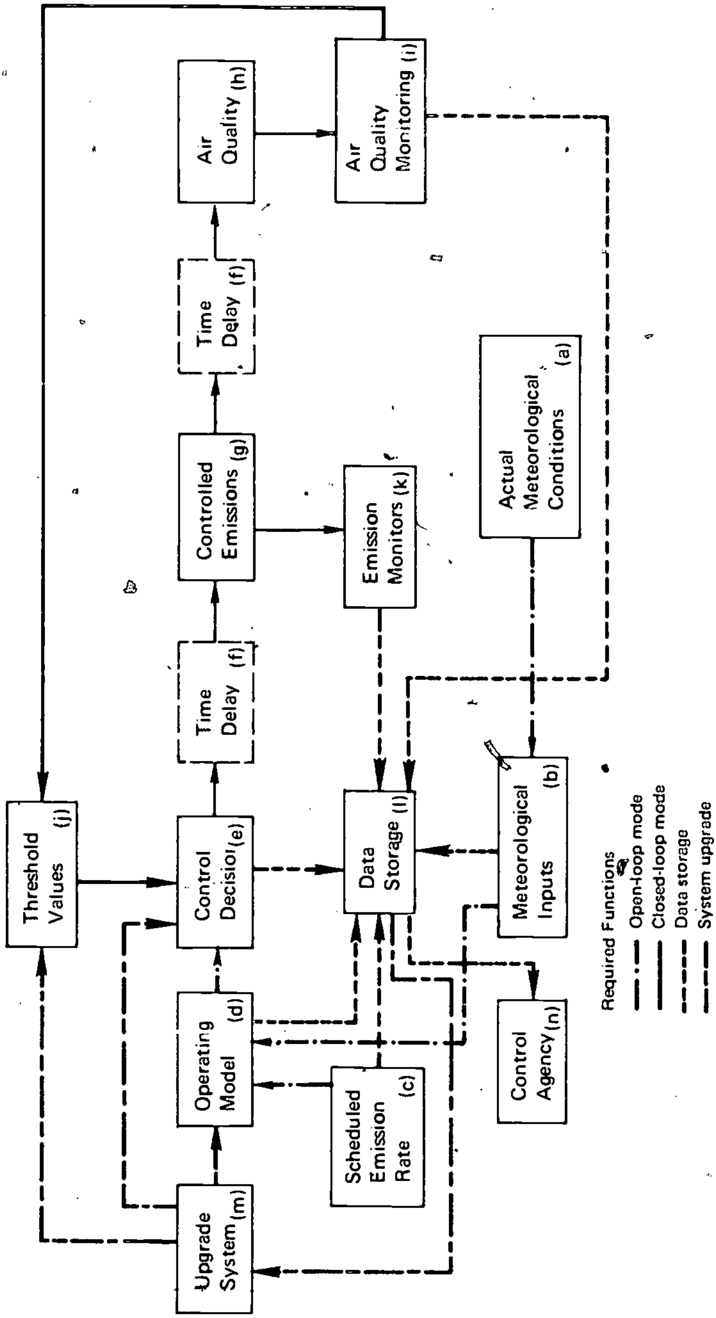

FIGURE 12-2: Elements of an Intermittent Control System (PEDCO 1974)

(4) The ability to control source emissions in a timely manner in order to meet forecast objectives;

(5) The ability to combine elements (1)-(4) into an operational system with adequate flexibility to meet the objectives of the system.

Each of the system requirements can be achieved with some probability of success; however, the likelihood of attaining a given degree of system performance will depend upon the ability to satisfy all of the system requirements concurrently. Typically, a factor of safety must be invoked to insure adequate system response. This factor of safety is usually embodied within the threshold values of the system; i.e., those forecasted levels of sulfur dioxide which necessitate control measures. The upgrade system is then applied as part of an ongoing effort to select proper forecast values, so that curtailment of emissions are invoked often enough to meet the ambient standards, but are not put into effect unnecessarily often. System performance and reliability are clearly related to the number and severity of forecasted sulfur dioxide levels estimated to occur in excess of the standards; the ICS approach has the greatest chance for success where the required number of curtailments of emissions is low.

LEGAL BACKGROUND

The suggestion that tall stacks be used with or without intermittent controls to meet sulfur dioxide standards is a highly controversial one. On the one hand, these techniques have been shown to be effective, at relatively low costs, for reducing ground-level concentrations of sulfur dioxide in the vicinity of power plants burning high sulfur coal (Frankenberg 1970, Smith and Frankenberg 1974, Montgomery et al. 1973, Montgomery and Frey 1974, TVA 1974). On the other hand, the application of these techniques normally provides, over an extended length of time, only a negligible reduction in the amount of pollutants emitted. Primarily for this reason EPA "...considers constant emission reduction techniques, such as flue gas desulfurization, far superior to dispersion techniques and has proposed regulations that limit the use of such dispersion techniques to situations where constant emission reduction controls are not available" (EPA 1974). EPA further claims "...the concept is not compatible with the Clean Air Act requirement that constant emission limitations be used whenever possible" (EPA 1974b).

In the most important legal decision on this subject, Natural Resources Defense Council v. EPA, the 5th Circuit Court of Appeals held that the Clean Air Act requires Georgia to attain national ambient air standards primarily through actual emission reductions rather than dispersion enhancement techniques; application of dispersion techniques is allowed only if exclusive reliance on emission control is infeasible (48a F.2d. 390, 1974). In reaching this decision, the court determined that the section of the 1970 Clean Air Act amendments which requires that state implementation plans "include emission limitations, schedules, and timetables for compliance with such limitation, and such other measures as may be necessary to insure attainment and maintenance of such primary or secondary standards" (42 USC 1857c-5(a)(2)b) indicates a Congressional intent that emission reduction is the preferred method of meeting ambient air quality standards. Therefore, Georgia's tall stack strategy for controlling power plant, sulfur oxide emissions could be included in its implementation plan "only (1) if it is demonstrated that emission limitation regulations included in the plan are sufficient, standing alone, without the dispersion strategy, to attain the standards; or (2) if it is demonstrated that emission limitation sufficient to meet the standard is unachievable or infeasible, and that the state has adopted regulations which will attain the maximum degree of emission limitation achievable."

This decision suggests that utilities which want to implement tall stack-ICS strategies rather than flue gas desulfurization (FGD) must convince state control agencies that FGD technology is commercially or technically infeasible. TVA is presently bringing a suit (TVA v. EPA) in the 6th Circuit Court of Appeals which involves this issue as well as the issues of fact and law considered in the Georgia decision. If the 6th Circuit decision differs from NRDC v. EPA, ultimately the Supreme Court may have to resolve the conflict.

Two previous cases, Appalachian Power Co. et al. v. EPA and Pennsylvania v. Pennsylvania Power Co., reached opposite conclusions regarding FGD technology. In the first of these, the U.S. Circuit Court of Appeals for the District of Columbia rejected a claim by several power companies that technology was not available to meet EPA's new source performance standards. The court, ruling in September 1973, said the evidence "...convinces us that the systems proposed are adequately demonstrated, that cost has been taken into consideration, and that emission standards are achievable." The court added, however, that EPA's consideration of the sludge disposal problem was insufficient, and remanded the record in the case to EPA for further explanation of sludge effects; EPA has not yet responded with the information. In the Pennsylvania Power case, a Pennsylvania state court upheld a county court opinion that adequate scrubbing technology does not exist. In a February 1974 decision, the court concluded that "flue gas scrubbing devices...are available in theory only, and are not available and proven from an operational or practical viewpoint." A Lawrence County, Pennsylvania court had previously determined that "the only conclusion to date...is that the most feasible present method is high stack control."

Recently, a September 1974 Ohio EPA hearing examiner's report favored the tall stack-intermittent control approach, stating that "flue gas desulfurization is not a presently available, technologically feasible method of sulfur dioxide control..." and that "tall stacks, either alone or in combination with supplementary control systems, are a technologically feasible and economically reasonable means of meeting ambient air quality standards for sulfur dioxide" (Ohio EPA 1974).

However, the hearing examiner's report served only as a recommendation to the Director of Ohio EPA; subsequent (December 12) findings and orders issued by the Director differed substantially from the recommendations of the examiner. Specifically, FGD technology was found by the Director to satisfy criteria for feasibility and reliability. However, he did rule that it would be inappropriate to require the use of FGD systems in Ohio until recent air quality data from power plants were collected and analyzed, and after further examination of the efficacy of scrubber technology takes place.

Each of the judicial or administrative decisions referred to above interprets requirements of the Clean Air Act amendments of 1970. More recent legislation, the Energy Supply and Environmental Coordination Act of 1974 (ESECA) (PL 93-319; enacted by Congress June 24, 1974), provides further insight into Congressional intent regarding the implementation of FGD and tall stack-ICS technologies. According to the provisions of ESECA, utilities permitted by EPA to convert to coal can be granted either short-term suspensions of emission limitations until as late as June 30, 1975 or compliance date extensions until as late as January 1, 1979 for meeting emission standards. Plants allowed an extension, however, must prepare a plan for achieving compliance before expiration of the extension with "the most stringent degree of emissions reduction" required by the state implementation plan in force at the time conversion was ordered. Under present EPA regulations this would include contractual commitments for obtaining long-term supplies of low-sulfur coal or installing continuous emission controls. Use of ICS is permitted only under certain circumstances; for example, these systems can be used to comply with interim requirements under emission standard extensions, but only in those cases where ICS utilization is directly enforceable by EPA.

For purposes of discussion, the following presentation assumes that tall stack-ICS technology is in itself a legally valid method of sulfur dioxide control, even though current EPA regulations and ESECA suggest otherwise.

For the moment, however, tall stacks and ICS measures are assumed to provide an important technological option, at the very least for reasons pointed out in EPA's proposed regulations (FR 1973):

- These measures provide alternative means for attaining and maintaining sulfur dioxide AAQS, rather than relying solely on FGD systems or low sulfur fuels.
- Implementation of these measures can result in attainment of AAQS in a shorter time than would otherwise be possible.
- These measures can be important interim techniques, given limitations on availability of FGD systems and low sulfur fuels over the next few years, given difficulty in retrofitting some facilities with FGD systems, and given distribution and allocation problems associated with low sulfur fuels.

ASSESSMENT OF THE TECHNOLOGY

Four aspects of tall stack-ICS technology are assessed here, including system performance, availability, cost, and secondary environmental impacts.

Performance

ICS measures are currently being implemented at a small number of installations on the continent, including (1) a number of TVA po[illegible] plants; (2) other power plants operated by Ontario Hydro (Canada) and Long Island Lighting Company (New York); (3) a number of western smelters operated by American Smelting and Refining Company (ASARCO), Kennecott Copper Corporation, and others; (4) the Dow Chemical plant at Midland, Michigan. Only limited data are available to assess performance of these systems.

TVA-Paradise Steam Plant

Data for the TVA Paradise steam plant are most extensive, and the distribution of 3-hour and 24-hour sulfur dioxide concentrations above AAQS levels before and after initiation of ICS measures are indicated in Table 12-2[3] (TVA 1974). Standards were exceeded prior to September 1960 with a two-unit plant (two 704 MW generating units, each with a 600-foot stack); capacity was increased by 82 percent on September 19, 1969 by installation of an 1150 MW unit with an 800-foot stack, with ICS measures also initiated to control high ground-level concentrations. No violations of 3- or 24-hour AAQS have occurred at the monitoring network since initiation of the ICS program, although four occasions in which sulfur dioxide concentrations were above the 3-hour standard level have been noted; details concerning possible violations at non-monitored locations are not available. At Paradise, plant generation reductions are instituted during adverse conditions for plume dispersion; these periods are predicted through on-site meteorological measurements. If nine meteorological and plume dispersion criteria (Table 12-3) fall within a critical range of values, the magnitude of sulfur dioxide emission reduction needed is determined with a dispersion model, and a load reduction takes place. Emission or load reductions are implemented independent of wind direction or monitor readings; the monitoring network is used for validation of the dispersion model and for checking system performance.

ASARCO-Tacoma Smelter

Data are also available which demonstrate the degree of effectiveness of ICS measures initiated at ASARCO's smelter in Tacoma, Washington (Tables 12-4, 12-5) (Nelson et al. 1973, Welch 1974). The data obtained from 5 ASARCO monitoring stations in continuous operation since 1969 indicate substantial and consistent improvement in the performance of the

TABLE 12-2

Sulfur Dioxide Concentrations Above Ambient Air Quality Standard Levels Before and After Implementation of ICS Program for TVA's Paradise Steam Plant (TVA 1974)

Time Period	Above 3-hour Standard Level (0.5 ppm)	Above 24-hour Standard Level (0.14 ppm)
4/28/63 - 9/18/69 (Before Control)	10*	8*
9/19/69 - 3/31/74 (After Control)	4	0

* Include violations of ambient air quality standards, which allow abrogation of standard level only once per year per location.

TABLE 12-3

Meteorological Criteria for ICS Measures at TVA's Paradise Steam Plant (TVA 1974)

No.	Criteria	Parameter Value	Remarks
1.	Potential temperature gradient between stack top, 183 m, and 900 m	$\frac{\Delta\theta}{\Delta Z} \geq 0.64^{o}K/100m$	Identifies the mean atmospheric stability throughout the layer involved in the plume height calculation
2.	Potential temperature gradient between stack top, 183 m, and 1,500 m	$\frac{\Delta\theta}{\Delta Z} \geq 0.51^{o}K/100m$	Identifies the mean atmospheric stability throughout the layer involved with the thermally induced mixing of the plume effluent
3.	Difference between daily minimum and maximum surface temperature	$T_{max} - T_{min} \geq 6^{o}K$	Identifies the potential magnitude of insolation as it affects the rate of development of thermally induced mixing
4.	Maximum daily surface temperature	$T_{max} \leq 298^{o}K$	Identifies the temperature used in calculation of the maximum mixing height
5.	Maximum mixing height	$MMH \leq 2{,}000$ m	Identifies the maximum level of the mixing height when SO_2 concentrations might exceed the ambient standard levels
6.	Maximum mixing height and plume centerline heights	$MMH \geq H_e$	Identifies the related height at which thermally induced mixing will penetrate the plume and uniformly mix the effluent to the surface

Table 12-3 (contd)

7.	Time for mixing height to develop from plume centerline to critical mixing height	$T \geq 3,960$ sec.	Identifies the potential for persistence of concentrations above the ambient standard levels. The critical mixing height (CMH) is the upper limit of the thermally induced mixing layer when concentrations could exceed the ambient standard levels
8.	Mean wind speed between stack top, 183 m, and 900 m	$2.5 \leq \bar{u} \leq 8$ m sec^{-1}	Identifies the wind speed used in plume rise and CMH calculations
9.	Cloud cover	CC < 80%	Identifies the availability of insolation which affects the thermally induced mixing

TABLE 12-4

Sulfur dioxide Concentrations in Violation of National Ambient Standard Levels at ASARCO Smelter (5 Monitoring Stations), Tacoma, Washington (Nelson et al. 1973, Welch 1974)

Time Period	3-hour Standard Violations (0.5 ppm)		24-hour Standard Violations (0.14ppm)		Annaul Standard Violations (0.3 ppm)	
	Number	Maximum	Number	Maximum	Number	Maximum
1969	26	0.72 ppm	9	0.250 ppm	0	0.019 ppm
1970	13	0.85	2	0.160	0	0.013
1971	3	0.53	0	0.116	0	0.012
1972	0	0.45	0	0.096	0	0.021
1973	0	0.45	0	0.100	0	0.015
1974 (Jan-Sep)	0	0.57	0	0.092	0	0.010

ICS program. National AAQS have not been exceeded since March 1971 at either the ASARCO stations or stations of the Puget Sound Air Pollution Control Agency (PSAPCA). However, more stringent local AAQS (Table 12-5) have been repeatedly violated, although with decreasing frequency since 1969.* ICS control at this location represents a greater challenge than at the TVA-Paradise plant due to the greater emission rate, moderate stack height, and more limited plume rise (565-foot stack, 15 feet/second plume exit velocity), significant terrain effects (local maximum elevations within 250 feet of stack elevation) and complications introduced by the maritime climate meteorology. Plant curtailments are typically initiated in the morning on the basis of meteorological forecasts; during May-October, operations must be curtailed to some degree almost every day. Forecasts are continuously updated on a short-term basis; monitoring station data (both ASARCO and PSAPCA) are then used to further modify curtailment actions. On the basis of controlling ambient sulfur dioxide concentrations, the Tacoma ICS can be considered a success. However, the system is designed to meet local standards as well; here reliability is not adequate. Initiation of improved continuous emission controls have recently dropped SOx emissions an additional 40 percent at full load; this continuous emission reduction may help bring about attainment of local as well as national AAQS for sulfur dioxide.

ASARCO -El Paso Smelter

Further insight into ICS performance is provided by a case study of ASARCO's ICS program at its El Paso smelter. The case study was recently completed by Environmental Research and Technology, Inc. (ER&T) for EPA, as part of a larger effort to develop procedures to analyze ICS programs quantitatively (Gaertner et al. 1974). The El Paso smelter represents a difficult location for ICS control techniques due to severe terrain effects. Emissions from copper and lead smelting operations are

TABLE 12-5

Number of Violations of Local Ambient Standards
at ASARCO Smelter (5 Monitoring Stations), Tacoma, Washington (Welch 1974)

Time Period	1.0 ppm, 5 minutes*	0.4 ppm, 1-hour+	0.25 ppm, 1-hour#	0.1 ppm, 24-hr+	Total Violations
1969	364	273	370	28	1035
1970	257	175	173	8	613
1971	31	29	29	1	90
1972	25	16	5	0	46
1973	10	10	4	0	24
1974 (Jan-Sep)	5	6	2	0	13

* Allowed one exceedance in 8-hour period.
\+ Not to be exceeded.
\# Allowed two exceedances in 7 days.

separately released (828-foot and 611-foot stacks, respectively) with stack tops corresponding to sea-level elevations of approximately 4,600 and 4,400 feet. To the northwest of the smelter, the Franklin Mountain Range (peaks at 6,000-6,600 feet) extends north-south, and acts as a barrier to the ASARCO plume during southwesterly wind flows; the Sierra del Christo Rey peak rises to a 4,700-foot elevation to the west of the smelter and interferes with plume transport to the northwest. ER&T analyzed ICS reliability for the 36-day period March 5-April 9, 1971, evaluating system performance in meeting national AAQS as well as a state variance standard calling for 1-hour concentrations ≤ 0.5 ppm. The following conclusions are apparent from the study:

•ICS measures were effective in reducing the number of excess concentrations east of the plant, but were less effective for concentrations northwest of the plant.

•Without meteorologically-based controls, both long and short-term sulfur dioxide concentrations would have been dramatically higher during the 36-day period. Percent reductions in average sulfur dioxide concentrations at the 17-station ASARCO monitoring network due to use of ICS controls ranged from 27.9 to 65.2 percent.

•As shown by Table 12-6, frequency distributions of concentrations measured by the monitoring network shifted toward the lower end of the spectrum, with the greatest shifting at the highest concentrations.

Although these performance characteristics indicate a substantial improvement in air quality due to ICS controls, system reliability in meeting all AAQS during the 36-day period was not adequate:

•The state variance standard (1-hour, 0.5 ppm standard) was violated on 9 occasions (Note that Table 12-6 indicates 17 violations, but 8 of these occurred at stations in New Mexico where the Texas standard was not a control objective).

•Violations of the 3-hour, 0.5 ppm national AAQS ocurred at one station.

•Violations of the 24-hour, 0.14 ppm national AAQS ocurred at two stations.

TABLE 12-6.

Distribution of Hourly SO_2 Concentrations for 17-Station ASARCO Monitoring Network ASARCO/El Paso ICS Program (March 5 - April 9, 1971) (Gaertner et al. 1974)

Case	Number of 1-hour SO_2 Concentrations										
	Total	>0.1 ppm	>0.2 ppm	>0.3 ppm	>0.4 ppm	>0.5 ppm	>0.6 ppm	>0.7 ppm	>0.8 ppm	>0.9 ppm	>1.0 ppm
Full Load Operation	14508	1187	599	360	225	142	88	62	45	37	31
ICS Control	14508	660	249	98	42	17	7	5	2	2	1
% Reduction due to ICS Control	-	44.4	58.4	72.8	81.3	88.0	92.0	91.9	95.6	94.6	97.0

Meteorological evaluations of five separate periods of high sulfur dioxide concentrations during the 36-day period did reveal methods by which system reliability could be improved, principally through improved forecasting and modeling of terrain disturbances.

Further evidence of the difficulty in meeting national AAQS with the El Paso ICS is provided by Table 12-7 (Nelson 1974). These data, derived from 13 stations in the ASARCO monitoring network which have operated continuously since 1970, indicate continuing violations of national AAQS.

Boston AIRMAP Network

System reliability in any ICS program is closely related to the accuracy of air quality forecasts. Unfortunately, no direct comparisons of air quality forecasts with observed air quality levels are available for operating ICS programs. Information of this type is available, however, from ER&T for their Boston AIRMAP network, an 11 station, real-time sulfur dioxide monitoring and forecasting system in operation since December 1971 (Gaut 1973, Gaut 1975). In Figure 12-3 the percentages of time for which differences between forecasted and observed values are within specified absolute limits (≤0.005 ppm, ≤0.010 ppm, ≤0.020 ppm) are indicated. Predictions are within 0.010 ppm between 80-95 percent of the time. ER&T concludes that these statistics indicate a forecasting skill better than the minimum required for a reliable ICS program. These forecasts are applicable for a multi-source, urban area; forecasts for regions dominated by single sources may be even more reliable. However, specific terrains, source configurations, and local meteorologies can pose signficant difficulties in air quality forecasting, as earlier examples show. Also, typical 24-hour sulfur dioxide concentrations in the Greater Boston area are relatively low (peak concentrations are in the range of 0.05-0.10 ppm); thus, percent error rather than absolute

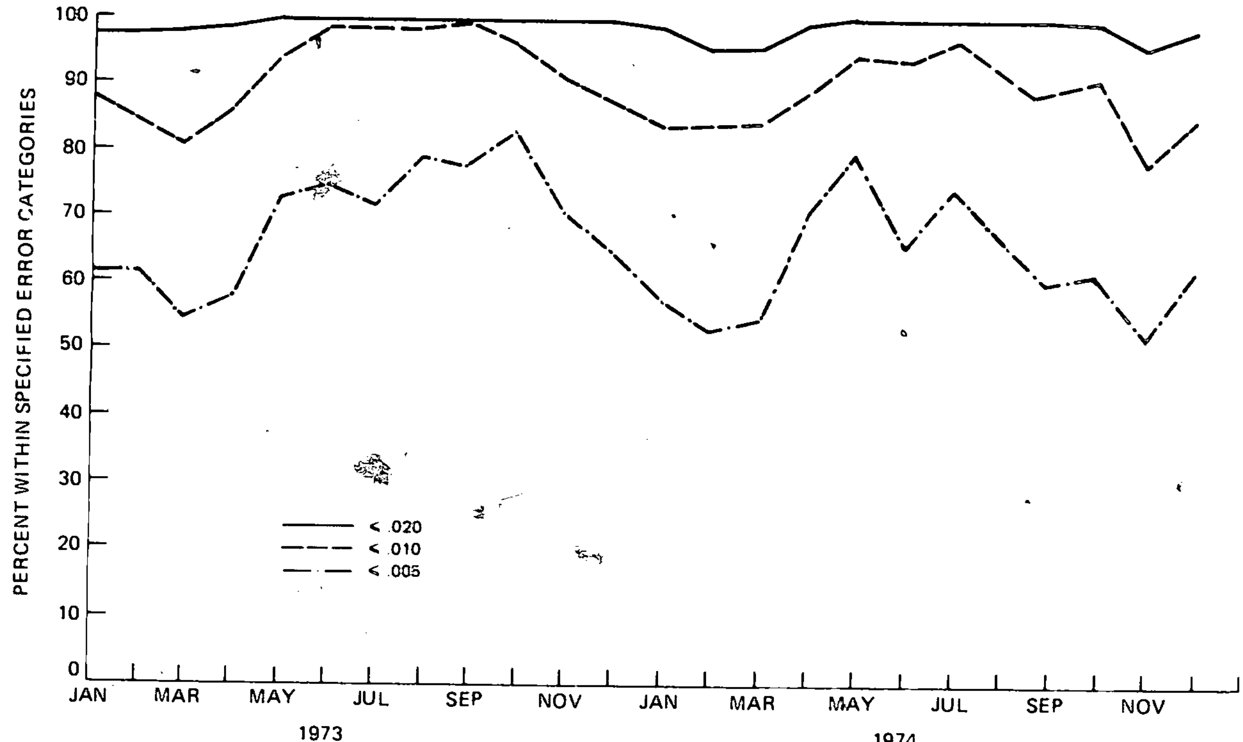

FIGURE 12-3: Absolute 24-Hour SO_2 Concentration Forecasting Errors (ppm), Greater Boston AIRMAP Network (Gaut 1975)

TABLE 12-7

Number of SO_2 Concentrations above Ambient Air Quality Standard Levels for 13 Station ASARCO-El Paso Monitoring Network (Nelson 1974)

Year	Number of SO_2 Concentrations 0.14 ppm (24 Hour Standard)	0.5 ppm (3 Hour Standard	Total
1970	1	8	9
1971	2	1	3
1972	12	1	13
1973	3	9	12
1974 (Jan.-Nov.)	0	1	1

error comparisons of the ER&T data would be less impressive.

Recapitulation

Only limited data are available to assess performance of operating ICS technology. Those data which are available pertain to individual sources of sulfur dioxide, either power plants (TVA) or smelters (ASARCO). System performance is varied; a "typical" ICS cannot be described based on these data. Indeed, ICS performance seems to be highly dependent on local meteorology, terrain and source characteristics, and particular system operating procedures. In general, currently operating systems provide significant reductions in peak ambient sulfur dioxide concentrations measured near the emission source. However, all relevant ambient air standards are still not met in the vicinity of several of the sources which have implemented ICS technology, particularly the short-term local standards which are more stringent than the present Federal standards.

This review has demonstrated that a need exists for a fully-documented analysis of an operating ICS system. Such a study should generate sufficient records of source data, meteorological data, and actual as well as forecasted air quality data to permit a definitive estimate of ICS reliability under various conditions. A 120-day study of a power plant burning eastern coal and applying ICS measures is suggested.

Availability

Two separate issues regarding availability of the tall stack-ICS approach are considered here:

(1) Availability of the generating unit itself, due to meteorologically-based load reductions, including constraints imposed by load shifting or fuel switching.

(2) Availability of trained personnel and/or firms with requisite forecasting and

monitoring capabilities to implement ICS controls.

Generating Unit Availability

The first of these issues can be partially assessed with TVA data which forecast anticipated load reductions at 9 power plants if ICS measures are imposed through load shifting alone (Table 12-8) (TVA 1973). Without construction of 5 new tall stacks at 3 plants (Kingston, Shawnee, Widows Creek), power output drops of 346,000 MW-hours per year are forecast; this represents 0.25 percent of the generating capacity of the 9 plant network (0.4 percent at an average load factor of 0.61). With construction of new stacks, reductions of 107,500 MW-hours/year are anticipated (0.08 percent of full load or 0.13 percent of average load). Predicted and actual performance can be compared for the Paradise plant, for the first 39 months of ICS control (September 1969-November 1972) (Montgomery et al. 1973). Reductions would have been required 106 days during this time (33 days/year) if the plant were at or near full capacity; actual load reductions totaled 41 days (13 days/year) with an average length 3.6 hours and size 454 MW (22 percent reduction).

Several points associated with this topic deserve further explanation:

(1) Although average load reductions required in the TVA system are low, reductions required for other plants may not be in this range. For example, the two ASARCO smelters considered earlier represent much more difficult applications of ICS technology, and require correspondingly greater average reductions in plant output (15-17 percent yearly average curtailment at Tacoma, 27-35 percent at El Paso).

(2) Additional reserve capacity requirements are implied if ICS control is exercised through load shifting rather than fuel switching. For example, preliminary studies by TVA indicate that additional reserve requirements on the order of 3-4 percent of peak

TABLE 12-8

Anticipated Load Reductions at TVA Power Plants due to ICS Measures (TVA 1973)

Plant	Stack Height Number	Stack Height Height (ft.)	Estimated Frequency of Reductions Days/Year	Estimated Frequency of Reductions Hours/Day	Estimated Frequency of Reductions %Hours/Year	Equivalent MW/Hours/Year
Allen	3	400	1	6	0.1	2,000
Colbert	4 1	300 500	24	5.5	1.5	25,000
Cumberland	2	1000	10	3	0.3	15,000
Gallatin	2	500	7	4.5	0.4	9,000
Johnsonville	8	273-400	7	6	0.5	12,000
Kingston (Current)	4 5	250 300	55	7	4.4	100,000
Kingston (Planned)	2	1000	0	0	0.0	0
Paradise	2 1	600 800	13	3.5	0.5	22,000
Shawnee (Current)	10	250	42	6	2.9	87,000
Shawnee (Planned)	2	800	3	3	0.1	4,500
Widows Creek (Current)	6 2	270 500	25	6	1.7	75,000
Widows Creek (Planned)	2 1	500 1000	12	3	0.4	18,000

power system load would be needed if fuel switching were not utilized, and if load switching were confined to the TVA network.

(3) The ability to load switch is limited if large scale adverse meteorology affects a large region rather than only one plant and one locality; the frequency of occurrence of this condition differs in various parts of the country. For example, in the Tennessee Valley, TVA has noted 100 occasions since September 24, 1973 when the meteorological conditions appropriate for generation reduction occurred at one or more of the following plants: Allen, Cumberland, Gallatin, Paradise, and Widows Creek. (These plants are located in a three-state area within the Tennessee Valley.) During this period of time, the potential need for simultaneous generation reduction at two plants existed on 17 days; because of plant operating conditions, simultaneous reductions were necessary on only 3 of the 17 days. The potential need for simultaneous generation reductions at 3 plants existed on one day; curtailments at all three plants were not necessary on that day due to operating conditions (Montgomery 1974).

(4) Load shifting for environmental reasons is currently not practiced in the nation's power pools, with the exception of plants in the Los Angeles basin, where minimum NOx dispatch[5] is practiced (i.e., power plants are brought on line to provide power with minimum total NOx emissions in the basin) (Schweppe 1974). Dispatch of power based on air quality constraints is potentially available, but time would be needed to work out new pool agreements, and some increases in computer control costs and power costs could be expected. However, an individual plant's ability to load shift for ICS purposes would depend on its particular situation (location, power system, and pool arrangement). Some systems, notably American Electric Power Service Corporation, do not operate within a pool, and plants within that system could shift independent of pool agreements. Some plants within power pools could arrange interim load shifts easily, if the pool were willing to treat an environmental

outage as a forced power outage. In other cases, however, interim or long-term pool arrangements to allow ICS load shifting could be more difficult to bring about.

(5) It is important to note that the practicality of implementing ICS actions which demand load reductions can be questioned, since the desire to meet AAQS may conflict with the desire of utilities to maintain an adequate power supply. For example, TVA has stated "...There are two power system operational conditions in which the Paradise Steam Plant would not reduce generation load below a minimum level during a designated sulfur dioxide emission limitation period. These restrictive conditions became effective when further limitation in load would cause power system instability and risk a failure in the supply of firm power to customers" (Montgomery et al. 1973). These two competing objectives--reliable sulfur dioxide control and reliable power--could presumably clash if load reductions are implemented on a large scale in ICS programs.

Most of the constraints suggested above are applicable to ICS approaches which depend on load shifting as a means for controlling emissions during adverse meteorological periods.

Constraints are also applicable for fuel switching (PEDCO 1974):

(1) Many coal-fired plants require major modifications in coal handling and feeding systems before they have a fuel-switching capability based on low sulfur coal. These modifications include provision of separate storage stockpiles of low sulfur fuel, erection of conveyers to move coal from storage areas, and construction of separate or partitioned bunkers for feeding the low sulfur coal supply to the boiler.

(2) The time required to effect a fuel switch to low sulfur coal can prevent the attainment of the federal 3-hour AAQS, or other short-term local standards. Fuel switching times can be expected to vary from 20 minutes (with partitioned bunkers) to 3 or more hours (when no bypass exists for high sulfur coal in the bunkers).

(3) Operating problems can be encountered in units not designed for low sulfur coal when this fuel is burned for prolonged periods of time. Such problems are associated with the higher moisture content, lower heating value, and different ash characteristics of low sulfur coals; the problems include (a) increased emissions of particulate matter and reduced collection efficiency of electrostatic precipitators, (b) reduced capacity of generating units and associated mechanical equipment such as pulverizers and coal bunkers, and (c) increased slagging.

(4) Burning low sulfur fuel oil as an alternate fuel in units with dual firing capabilities eliminates the constraints mentioned above. However, this alternate firing capability (with storage and piping facilities) in units not so equipped will cost from $12-$40/kw, depending on the size of the unit. In addition, assured supplies of low sulfur oil may not be available.

Availability of Personnel and Firms with ICS Capabilities

A number of different potential ICS applications exist, with individual applications of this technology requiring different forecasting, modeling, and monitoring efforts and capabilities. At one end of the spectrum are plants located in relatively flat terrain, equipped with tall stacks, and burning fuels with low to intermediate sulfur content. Without other complicating factors, these plants could presumably implement ICS programs which would feature relatively few emission reductions per year. Forecasting, modeling, and monitoring problems, although significant, would be minimized. At the other end of the spectrum would be plants located in areas where there are severe meteorological conditions or where plume patterns are significantly affected by disturbances of air patterns created by nearby structures or terrain. Implementation of ICS programs in these situations would feature relatively large numbers of emission reductions per year, and forecasting-modeling-monitoring problems would be significantly increased.

The technical efforts exerted to initiate and operate an ICS program at either extreme are significant, but of a different order of magnitude. If the more complex ICS systems were implemented for many sources, current capabilities of competent, trained personnel and firms which can deliver such services could be inadequate. Even more significant constraints, applicable to all ICS applications, could be hardware needs (specifically ambient air quality and emissions monitoring equipment and telemetry) and time requirements to reach full operation of an ICS program.[6]

Costs

Costs for tall stack-ICS programs can be expected to vary considerably from plant to plant, with an obvious trade-off existing between amount spent for tall stack construction and fuel or load switch costs. The amount TVA spent to develop its tall stack-ICS program at nine plants could be illustrative of average plant costs, and is shown in Table 12-9 (Montgomery and Frey 1974, TVA 1974, Frey 1974).

The TVA program includes construction of five new tall stacks (800-1000 feet in height) at three plants (replacing 25 stacks with heights between 250-300 feet), construction of coal switching capabilities at three plants, and development of ICS programs and monitoring at all nine plants. Fuel switching will be practiced at Colbert, Cumberland, and Johnsonville plants; load switching at Allen, Gallatin, Paradise, Shawnee, and Widows Creek plants; Kingston will rely on tall stacks alone. Operating costs presented in Table 12-11 include annualized capital charges, replacement power or low sulfur fuel charges for an equivalent of 107,500 MW-Hours of electrical power, and operation-maintenance expenses.

TVA average costs of approximately $4/kw capital and approximately 0.15 mills/kwh operating cost appear to be low estimates in at least two respects, if these expenditures are to be considered accurate for all situations. Reserve capacity of the TVA system would need to be increased if load switching were undertaken without purchase of replacement power from other utilities; capital and operating costs cited could be at least doubled if reserve requirements on the order of 3-4 percent of system load were required. Also, the per-kilowatt costs may be too low an estimate for other plants due to the large average size (about 1750 MW) of the TVA plants included in the nine plant network; tall stack costs and ICS program-monitoring costs should be relatively independent of plant capacity. For example, note that per plant costs shown in Table 12-9 are capital costs, $6.7 million; annual costs, $1.2 million. If these costs are associated with a 1000 MW plant operating at 65 percent load factor, average costs are increased to $6.70/kw for capital costs and 0.2 mills kw-hour for annual costs.

Hence, costs for operating a tall stack-ICS program at an average power plant can best be stated in terms of a relatively broad range, accounting both for the TVA experience and possible cost increments due to increased reserve requirements and plant size effects.

Capital Costs:	$4-10/kw
Operating Costs, including annualized capital charges:	0.15-0.04 mills/kwh

These costs can be compared to previous estimates by MacDonald (1973) (capital costs, $5/kw; operating costs, 1 mill/kwh) and Jimeson et al. (1974) (operating costs in the range of 0.02-1.4 mills/kwh, dependent on whether load or fuel switching is used).[7]

Secondary Environmental Impacts

Much of the controversy surrounding use of the tall stack-ICS technology is not specifically associated with control of ambient sulfur dioxide concentrations, but rather with possible secondary environmental impacts of the technology. Since the technique essentially relies on dispersion (i.e., dilution) to reduce ambient concentrations, long-term sulfur dioxide pollutant loadings vented to the atmosphere are usually only negligibly reduced. Possible environmental effects associated with these atmospheric loadings include:

•impact of suspended sulfate aerosol concentrations, including possible health effects (increased morbidity and mortality), damage to materials and plants, and aesthetic effects (visibility reduction, climate and weather changes).

•impact of acid rainfall, due to acidification of soils and water.

Unfortunately, most of these impacts are unquantified at present, or are clouded with scientific uncertainty. Acceptance of the tall stack technology, therefore, depends on a subjective decision that the risks and uncertainties associated with potential environmental impacts due to these effects are outweighed by the incremental cost of continuous emission controls.

EPA Viewpoint

EPA suggests concentrations of suspended sulfates in the 6-10 ugm/m³ range (24 hour or longer exposure) are associated with adverse human health effects (Finklea 1974). Altshuller's analysis of National Air Surveillance Network (NASN) data for 1964-1968 indicated that such low levels of sulfate concentration were widespread, and correlated with correspondingly low levels of sulfur dioxide concentrations in urban areas. Altshuller also suggested that high sulfate concentrations at eastern and midwestern non-urban locations were attributable to the chemical conversion of sulfur dioxide to sulfate during transport downwind from urban areas, with sulfate levels of at least 5 ugm/m³ carried into urban areas. Assuming this analysis was correct, Altshuller concluded that, for eastern and midwestern portions of the United States, "...a uniformly large reduction in sulfur dioxide is necessary...to secure substantial decreases in sulfate concentration levels at many urban sites and at all non-urban sites (Altshuller 1973). In a later analysis of NASN data for the period 1964-1970, Frank modified the earlier, general sulfur dioxide-sulfate correlation, finding "sulfate concentrations...are not clearly related to ambient sulfur dioxide concentrations at individual locations and can be better described by the concentration of total suspended particulate matter and the general geographic locality of the monitoring station." Frank's analysis did indicate that a broad geographic similarity exists between overall ambient concentration levels of sulfur dioxide, the sulfate fraction of total suspended particulate matter, and area-wide sulfur dioxide emission densities; in addition, his analysis tended to substantiate the existence of a higher background of sulfate concentrations in the eastern United States. Since most sulfate particulate formation probably results from atmospheric reactions of sulfur dioxide, Frank concluded that "...the influence of more distant sulfur dioxide emissions on sulfate concentrations could satisfactorily explain both the general geographic similiarity of sulfate levels and the apparent differences in ambient sulfur dioxide and sulfate concentrations" (Frank 1974).

Finklea has recently sharpened this argument, pointing out that sulfur dioxide emissions in major cities decreased by 50 percent between 1960-1970, while nationwide sulfur dioxide emissions increased, with 80 percent of these emissions due to power plants east of the Mississippi River. Finklea argues that long-range transport of acid-sulfate aerosols from one region to another is plausible, based on an analysis of turbidity levels, acid rainfall patterns, and high annual average sulfate levels in the eastern United States. However, as Finklea points out, there are significant scientific uncertainties in the sulfate data base: limited historical data; limited monitoring information, especially simultaneous monitoring in urban, suburban, and rural areas; limited understanding of mechanisms and rates of transformation of sulfur dioxide to acid-sulfate aerosols in plumes and in the atmosphere; inadequate dose-response functions for addressing health effects; and lack of precise predictive models showing effects of long-range transport and stack height on ambient pollutant concentrations. Despite these uncertainties, EPA has concluded that: (1) failure to reduce SOx emissions may result in thousands of excess deaths and illnesses; (2) reduction of SOx emissions from urban and rural power plants will be required; (3) conversion of power plants to high sulfur fuels and use of tall stack-ICS approaches will aggravate the sulfate problem (Finklea 1974).

An Opposing Viewpoint

Arguments disputing the EPA sulfate analysis as it applies to tall stack-ICS strategies have been put forth by TVA (1973), by the Federal Energy Administration (1974), and, in a major advertisement series, by American Electric Power Service Corporation. A number of recurrent themes are identifiable in these and other arguments which support tall stack-ICS technology:

(1) This technology does not have the secondary impacts associated with flue gas desulfurization technologies, namely sludge creation and disposal problems or relatively high energy requirements (5-7 percent of output power) to operate the FGD systems;

(2) Tall stack-ICS technology is capable of meeting existing AAQS for sulfur dioxide, even in those situations where current emission regulations will not guarantee meeting ambient standards;

(3) If acid-sulfate aerosols pose dangers to health, welfare, and aesthetics as suggested by EPA, AAQS for this class of pollutants should be established, following the procedures set forth in the 1970 Clean Air Act Amendments. If information regarding sulfates is not sufficient to issue a criteria document, then tall stack-ICS technology should not be rejected on the basis of present information.

(4) EPA's advocacy of oxidation catalysts for vehicles, a control technology which might produce high ambient sulfate concentrations in transportation corridors and in center-city areas, is not consistent with its stance on tall stack-ICS technology. Each technology may exacerbate the sulfate problem, yet EPA is an advocate of oxidation catalyst technology, an opponent of tall stack-ICS technology.

(5) Use of tall stack-ICS methods allows immediate increased combustion of coal, the nation's most plentiful fuel resource. Use of this technology will ease the nation's clean fuels deficit and still allow compliance with sulfur dioxide AAQS.

Recapitulation

Judging the persuasiveness of arguments on both sides of this controversy is a major task of the current NAS/NAE study. The controversy centers about two issues:

(1) What are the health, welfare, and aesthetic effects associated with present and anticipated levels of sulfate aerosols and sulfur dioxide in the atmosphere?

(2) What role do power plants employing ICS technology and/or tall stacks play, both now and in the future, in the relationship?

Decisions regarding these issues may have to rest on subjective as well as objective appraisals. For the tall stack-ICS technology, two separate levels of inquiry appear to be warranted:

(1) Should tall stack-ICS technology be considered as a permanent emission control technique?

(2) If not, is this technology practicable as an interim control technique?

In making these judgments, a potential compromise position can be identified which seems to be within the existing data base. The tall stack-ICS technology could be rejected as a permanent control technique on the basis of the probability of substantial potential risks associated with increased atmospheric loading of sulfur compounds. At the same time, the technology could be accepted for carefully defined situations as an interim control technique, on the basis of the need to:

(1) Select suitable interim sulfur dioxide control techniques during large scale implementation of FGD technology, and

(2) Reduce the clean fuels deficit while allowing compliance with sulfur dioxide AAQS, if not potential sulfate standards. It would appear that the Ford Administration has recently adopted this position, in a November 22 statement of the Energy Resources Council. The Council agreed to support the policy of requiring continuous emission controls for all new and existing power plants, subject to deferral of compliance by those plants where implementation of ICS technology is deemed a feasible and enforceable alternative for meeting primary sulfur dioxide standards. Power plants in some instances could use the ICS approach until as late as January 1, 1985. Ambient air standards would have to be met during the interim, and compliance schedules for installing permanent controls would have to be agreed to before use of ICS controls would be allowed (Environment Reporter 1974).

ENFORCEMENT

An important issue regarding the efficacy of ICS technology concerns the legal enforceability of these systems. Principal attention here has been focused on the question of whether AAQS can be attained without enforcing emission limitations. The issue was pointedly referred to by Senator Edmund Muskie in Senate discussion of ESECA:

> An important clarification in the conference report relates to enforcement of interim procedures to assure compliance. Senate conferrees insisted that the Environmental Protection Agency's determination that emissions from coal converters would not cause primary standards to be

> exceeded must be articulated in emission limitations or other, precise enforceable measures for regulating what comes out the stack. The conference report on this bill underscores the fact that it is not ambient standards which are enforced but emission limitations or other stack related emission control measures. Ambient standards are only a guide to the levels of emission controls which must be achieved by specific sources. In 1970, we recognized that a control strategy based on a determination of ambient air pollutant levels in relation to each individual source would be unenforceable. Existing clean air implementation relies specifically on the application of enforceable controls against specific sources. We have continued that procedure in this law (Cong. Rec. 1974).

Emissions-Based versus Air Quality-Based Enforcement

The relative lack of enforceability of AAQS compared to the relative simplicity of enforcing compliance emission standards is not in question. In the first situation (AAQS), the contribution of a single pollutant source to resulting ambient air quality is difficult to determine; in the other case (emission standards) specific standards are applicable to individual source. The key to effective enforcement of ICS control, then, appears to be rooted in enforcement of specific emission limitation violations rather than (or in addition to) enforcement of AAQS violations. In ICS enforcement based on emission standards, predetermined emission rates (calculated by forecasting meteorology and modeling air quality, and monitored by in-stack measurements) form the basis for determination of individual source compliance; in ICS enforcement based on air quality standards, measured ground-level ambient concentrations are the sole information source relied upon for enforcement.

EPA Proposed Regulations

EPA's proposed regulations (FR 1973) for ICS systems, as well as the agency's recently proposed rule-makings (FR 1974a, 1974b) for ICS controls for certain western smelters, recognize the importance of emission-based controls for practicable enforcement. Specifically, the following measures are provided:

(1) Air quality violations are defined as "non-compliance with stated and agreed upon emission curtailment conditions and procedures" as well as "a single ambient concentration that exceeds the standard at any point in the area significantly affected by the source emissions" (FR 1973).

(2) The emission source must "establish, maintain, and continuously operate monitors for detecting the pollutant emission rate, air quality, and meteorological variables." Furthermore, the agency must be granted "continuous access to all emission, air quality, and meteorological data collected by the source," as well as "authority to inspect, test, and calibrate all sensors, recorders, and other equipment operated by the source to collect these data" (FR 1973).

(3) The emission source is required to assume liability for violation of AAQS within a "designated liability area," i.e., the geographic area within which emissions from a source may significantly affect the ambient air quality."

(4) Before approval of ICS controls will be granted, the emission source must submit to the EPA Administrator a comprehensive report of a study which demonstrates the capability of the ICS, in conjunction with other control measures, to meet ambient air standards (FR 1974b). The study must include a test period of at least 120 days, during which ICS controls are developed and operated. The report must:

•describe and identify various elements of the ICS (monitoring, forecasting, determination of emission rates);

•estimate the frequency of necessary emission rate reductions;

•include data and results from objective reliability tests; and

•demonstrate that implementation of ICS and other control measures will result in attainment and maintenance of AAQS.

(5) Before approval of ICS controls can be granted, the emission source must submit to the Administrator an operational manual (FR 1974b) for the ICS which:

•specifies number, type, and location of ambient air quality monitors, in-stack monitors, and meteorological instruments to be used;

•describes techniques, methods, and criteria for forecasting, as well as methods for systematically evaluating and improving the reliability of the sytem;

•specifies maximum emission rates for all probable meteorological and air quality situations so that AAQS will not be exceeded in the designated liability area; and

•describes specific curtailment actions which will be taken when meteorological conditions demand and/or when specified air quality deterioration occurs.

(6) The source is required to follow certain recordkeeping procedures, including submission of monthly reports describing and analyzing actual ICS performance compared to the approved operations manual.

Assessment of Enforceability

Enforcement based on these factors should be considerably more effective than enforcement based on violations of AAQS alone, for the following reasons (Gaertner et al. 1974):

(1) Workable enforcement procedures can be established prior to the control agency's authorization of an ICS.

(2) Justification for the ICS and operational details of the ICS are subject to evaluation and approval before authorization of

the system. In particular, the agency can demand use of conservative diffusion modeling techniques and data interpretations to help insure that emission reductions will be instituted at least as often as required to maintain ambient standards.

(3) With the ICS application well defined by preliminary documents, enforcement should principally involve:

(a) Determining whether the emission source is operating according to the approved operational manual. Emission rate reductions provided for in the manual and not actually instituted would constitute violations of standards; surveillance of emission rates would detect these violations.

(b) Deciding when violations of AAQS occur. Accurate monitoring of air quality would detect these violations.

Tacit recognition of the accuracy of this assessment appears in the text of a recent EPA decision concerning a Nevada smelter (FR 1974b):

"By allowing the use of [ICS], the Administrator is acknowledging that [ICS] can incorporate design and enforcement features that will provide a reliable means to attain and maintain national sulfur dioxide standards."

Costs of Monitoring and Enforcement

The monitoring and enforcement effort required of regulatory agencies to supervise implementation of ICS technology is greater than that required for continuous emission controls. Hence, agency monitoring and enforcement costs are expected to be higher, although it is difficult to predict the exact level of added resources which will be required.

Estimates of the amount of resources expended by PSAPCA in enforcement of control of emissions from the ASARCO-Tacoma smelter have been referred to in previous evaluations of ICS controls (Cong. Rec. 1974, Slater 1974). Costs per year to the agency for activities associated with control of all smelter emissions are variously estimated as $100,000, $160,000-

$200,000, and $200,000; the current annual total budget of the agency is $1,000,000 (Dammkoehler 1974). At the upper limit, these costs are of the same order of magnitude as those required by ASARCO to operate the ICS, if lost production output is not charged to ICS control. At the lower limit, these costs (about 10 percent of the agency budget) appear reasonable in view of the significance of ASARCO emissions compared to total emissions for the area.

Whatever PSAPCA costs may have been for the administrative, technical, and legal efforts involved in regulating this source, it would appear that costs for future activities by other control agencies cannot easily be estimated from this example:

(1) PSAPCA costs are not easily disaggregated on a source-by-source basis, accounting for the relatively wide disparity in cost figures cited above.

(2) Enforcement at Tacoma has principally been of AAQS, rather than of emission limitations specified by an operational manual. Costs involved in these two types of enforcement methodologies may differ widely.

(3) Many of the enforcement provisions described in the previous discussion have not been applied in Tacoma.

Agency costs for monitoring and enforcement of ICS systems represent a hidden cost of control, not included in Table 12-9. Proposed EPA regulations (FR 1973) suggest that the additional monitoring and enforcement costs involved can be defrayed by licensing of ICS programs and imposition of fees. This suggestion appears equitable, and will increase costs shown in Table 12-9.

STATEMENT OF FINDINGS AND CONCLUSIONS

1. Although current EPA regulations and the Energy Supply and Environmental Coordination Act of 1974 suggest a different interpretation, tall stack-ICS technology is considered here as a potentially important technological option for control of ambient sulfur dioxide concentrations. At the very least, it provides

TABLE 12-9

Estimated Capital and Annual Costs for ICS Programs at Nine TVA Coal-Fired Power Plants (Montgomery and Frey 1974, TVA 1974, Frey 1974)

Capital Costs		Amount
Development, installation of ICS programs and monitoring networks at nine plants[a]		$ 5 million
New tall stacks at three plants[b]		47 million
Coal switching capability at three plants[c]		8 million
Additional reserve capacity[d]		--
Total Capital Cost		$ 60 million
		$3.80/KW[1]
Annual Costs		
Capital charges, $60 million at 9.64 per cent		$ 5.8 million
Capital charges, additional reserve capacity[e]		-
Operation and Maintenance		3.0 million
Monitors & ICS Programs	$2.1 million	
Stacks	0.9 million	
Replacement power charges[f]		1.1 million
Incremental costs for low-S coal		0.6 million
Fuel costs[g]	$0.3 million	
Transportation & handling	0.3 million	
Total Annual Costs		$10.5 million
		0.13 mills/KW-Hr[h]

[a] Total 9-plant capacity, 15713 MW.
[b] Kingston (2 @ 1000'); Shawnee (2 @ 800'); Widows Creek (1 @ 1000').
[c] Colbert, Cumberland, Johnsonville plants.
[d] Option not selected by TVA and costs not estimated.
[e] Five plants, 55.5 x 10^3 MWH @ 20 mills/KWH, purchased.
[f] Three plants, 52 x 10^3 MWH.
[g] Average load factor, 0.61.

an alternate means of attaining and maintaining existing AAQS, an important capability given uncertainties or capacity limits regarding other control strategies.

2. Tall stack dispersion is closely related to ICS control, since increased stack height decreases the need for intermittent emission reductions, assuming control of ambient sulfur dioxide concentrations is the sole objective.

3. In the most important legal decision concerning the use of dispersion techniques to meet AAQS, Natural Resouces Defense Council v. EPA, the 5th Circuit Court of Appeals ruled that emission reduction is the preferred method of meeting ambient standards. A case involving similar issues of fact and law (TVA v. EPA) is now being litigated in the 6th Circuit.

4. A spectrum of potential applications of ICS technology exist, with individual situations requiring different forecasting, modeling, and monitoring efforts and capabilities. The frequency and extent of emission reductions necessary to maintain national AAQS with ICS control appear to be simple indicators which demonstrate the degree of difficulty associated with a given use of this technology.

5. Tall stacks and ICS methods have generally proven effective in reducing the number and extent of excess concentrations of sulfur dioxide in the vicinity of single isolated sources. Reliability (i.e., ability to meet AAQS) of existing systems is as follows:

(a) At TVA's Paradise Steam-generating Plant, ICS reliability appears good; national AAQS have been met since initiation of ICS controls in 1969. Emission reductions are necessary at Paradise 0.5 percent of the year.

(b) At ASARCO's Tacoma smelter, system reliability is inadequate to meet local AAQS, although performance has been improving. However, national AAQS have not been exceeded since March 1971 at this installation. Average yearly load curtailment at Tacoma due to ICS control is 15-17 percent of full output.

(c) At ASARCO's El Paso smelter, based on operating results for a 36-day period in 1971, performance characteristics indicated a substantial improvement in air quality due to ICS controls, but system reliability was inadequate to consistently meet national or local AAQS. Average yearly load curtailment at El Paso due to ICS control is 27-35 percent of full output.

6. System reliability in an ICS program is a function of the accuracy of air quality forecasts. One set of forecasted air quality predictions for a multi-source, urban area (Greater Boston) appears relatively accurate. Other comparisons of forecasted and actual air quality are not available.

7. A need exists for a fully-documented study of an operating ICS system. Such a study should examine information about source data, meteorological data, and actual as well as forecasted air quality measurements in order to permit a definitive analysis of system reliability under various conditions. A 120-day study of a power plant burning high sulfur eastern coal and applying ICS measures is suggested.

8. This concept, if adopted for eastern power plants with ICS controls, would severely limit potential applications of the technology. A recent EPA memo indicates only 18 plants in 15 eastern states have emissions which constitute 90 or more percent of total emissions within their restricted geographical area (U.S. Environmental Protection Agency 1974). TVA suggests the designated liability area concept should be applied as an alternative to emissions-based enforcement procedures, rather than as an additional requirement (Montgomery 1975).

9. Use of the ICS approach with load shifting to reduce emissions during adverse meteorology has a number of implications:

(a) Additional reserve requirements are needed; preliminary assessments by TVA indicate these requirements may be as high as 3-4 percent of peak power system load.

(b) The efficacy of load shifting can be limited when large-scale, adverse meteorology affects more than one plant in a given region. Over a one-year time period, TVA noted the potential need for simultaneous generation reduction at two of five plants on 17 of 100 days. Because of plant operating conditions, simultaneous reductions were necessary on only three of the 17 days.

(c) The ability of an individual plant to load shift for ICS purposes depends on its particular situation, i.e., location, power system, and pool arrangement. For some plants, no serious constraints exist; for other plants, interim or long-term power pool arrangements to allow load shifting for ICS purposes could be difficult to bring about.

(d) The practicality of implementing control actions which demand load reductions can be questioned. Two competing objectives--reliable sulfur dioxide control and reliable power--could presumably conflict if large scale load reductions are needed for implementation of programs.

These constraints do not apply if emission reductions occur through fuel switching as part of ICS programs.

10. The constraints which are applicable to ICS systems using fuel switching to reduce emissions during adverse meteorology include the following:

(a) Many coal-fired plants will require major modifications in coal handling and feeding systems in order to provide a fuel switching capability based on low sulfur coal.

(b) The time required to effect a fuel switch to low sulfur coal may prevent the reliable attainment of short-term AAQS in some situations.

(c) Operating problems with low-sulfur coal may be encountered in units not designed for this fuel. These problems include increased particulate emissions, reduced generating capacity, and increased slagging.

(d) Burning of low sulfur fuel oil as an alternate fuel in units with dual firing capabilities eliminates these problems. However, converting units without this capability to a dual firing capability is costly ($12-$40/kw), and assured supplies of low sulfur oil may not be available.

These constraints do not apply if emission reductions occur through load shifting.

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11. Technical efforts to initiate and operate an ICS program are significant for all systems, but of differing orders of magnitude depending on the complexity of the system. The following constraints exist:

(a) If ICS programs are installed in highly compex situations (e.g., ASARCO-El Paso situation) for many sources, trained personnel and firms needed to deliver ICS services may be in short supply.

(b) More significant constraints, which are applicable to all ICS programs, could be hardware needs (monitoring equipment) and the time requirements for full operation of the ICS program.

12. Costs for implementing the tall stack-ICS approach are significantly less than comparative costs for FGD systems, both in terms of capital and annual expenditures. Based on TVA experience, with an expanded range of costs to account for increased reserve requirements and application to smaller plants, costs can be expected to be as follows:

Capital Costs:	\$4-10/kw
Operating costs, including annualized capital charges:	0.15-0.4 mills/kwh

Regulatory agency expenses for monitoring and enforcement of ICS programs represent hidden costs of control, not included in the above estimates. Defraying these costs by licensing and imposition of fees appears equitable.

13. Much of the controversy surrounding implementation of the tall stack-ICS approach is associated with the impact of this technology on acid-sulfate aerosol formation, and effects of these aerosols on health, welfare, and aesthetics. Major disagreements exist in interpreting the limited data base available for understanding this issue.

14. A potential compromise position regarding public policy toward implementation of tall stack-ICS technology can be identified. The technology could be rejected as a permanent control technique, on the basis of the proba-

bility of substantial potential risks associated with increassed atmospheric loadings of sulfur dioxide. At the same time, the technology could be accepted for carefully defined situations as an interim control technique, because of its ability to meet AAQS during implementation of FGD systems and because of its role in reducing the current clean fuels deficit.

15. An important issue regarding implementation of ICS technology concerns the legal enforceability of these systems. Regulations proposed recently by EPA for ICS control emphasize enforcement of prescribed emission limitations included within an approved operational manual for each ISC system, in addition to enforcement based on AAQS. Enforcement based on this dual strategy should be more effective than enforcement based on AAQS alone, since most of the critical elements of the enforcement procedure can be established by the regulatory agency prior to approval of the ICS installation.

FOOTNOTES

1 These systems are variously referred to as intermittent control systems, closed-loop systems, dynamic emission control systems, and sulfur dioxide emission limitation systems. ICS is used here as a generic term for these concepts.

2 These plume transport conditions are briefly described as follows: (2) Coning: Near neutral stability conditions, moderate-to-high wind speeds, generally occurring on cloudy and windy days or windy nights.
Inversion breakup or fumigation: Stable atmosphere, transport of plume downwind with minimum vertical dispersion, followed by uniform dispersion to ground by thermally induced vertical mixing.
Trapping or limited mixing layer dispersion: Subsidence inversion or stable air aloft, uniform dispersion to surface.

3 Locations and number of monitoring stations have not remained fixed. Data are compared on the basis of equal numbers of observations at different locations before and after implementation of the ICS program. The monitoring network has included from 4 to 14 sulfur dioxide monitors, located in a 22.5 degree area northeast of the plant, at distances ranging from 3 to 17 kilometers from the plant; currently 9 monitors are in operation.

4 Until mid-1974, ASARCO operated under a variance which allowed violations of the 1ppm-5 minute standard. Civil penalties have been repeatedly assessed for violations of the 1-hour standards (24 penalties in 1972 for violations at 4 PSAPCA stations; 16 penalties in 1973 for violations at 4 PSAPCA stations) (Dammkoehler 1974).

5 Dispatch of power refers to the scheduling of power generation from an interconnected network of plants. Normally, plants are scheduled to operate so as to minimize the total cost of producing electricity.

6 TVA estimates a system for one of their plants would require 16-18 months to become fully operational, including field study, design, and installation phases (Montgomery et al. 1974). PEDCO quotes a 12-24 month period for full operation, with some systems then requiring 1-2 years to achieve reliability (PEDCO 1974).

7 Jimeson et al. (1974) cite costs for a 1000-MW plant of $0.88, and 7.6 x 10^6/year, using three ICS strategies. An operating factor of 0.65 was assumed to compute costs per kwh.

8 This concept, if adopted for eastern power plants with ICS controls, would severely limit potential applications of the ICS technology. For example, a recent internal EPA memo indicates only 18 plants in 15 eastern states have emissions which constitute 90 or more percent of total emissions within their "liability area" (EPA 1974).

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PART TWO

Section 3: Analysis of Alternative Emission Control Strategies

CHAPTER 13

ANALYSIS OF ALTERNATIVE EMISSIONS CONTROL STRATEGIES

(Chapter 13 was written by D. Warner North and M.W. Merkhofer under the general supervision of the committee, which reviewed the work at several stages and suggested modifications that have been incorporated. While every committee member has not necessarily read and agreed to every detailed statement contained within, the committee believes that the material is of sufficient merit and relevance to be included in this report.)

INTRODUCTION AND SCOPE

Sulfur oxide and particulate emissions have adverse consequences for human health and welfare, but the means for controlling these emissions entail considerable expense. This section of the report presents a quantitative framework for comparing alternative strategies for emissions control from stationary sources. The primary focus of attention will be on emissions from coal fired steam electric power plants in the eastern United States. The method of approach to be used in this section is easily adapted to other sources and other regions of the country.

The Clean Air Amendments of 1970 required that by July 1, 1975 specified levels of ambient air quality for sulfur oxides must be met. On the basis of these ambient standards the State Implementation Plans (SIP) were developed that set sulfur oxide emission limitations. The national primary ambient air quality standard for sulfur oxide is $80 \mu g/m^3$ for the annual arithmetic mean and $365 \mu g/m^3$ for a maximum 24-hour concentration. These values were selected to protect human health, with a margin of safety. No separate standard presently exists for suspended sulfate levels.

When the Clean Air Act was passed and during the period when State Implementation Plans were being developed, there was still no obvious indication that natural gas would be in very short supply for industrial and utility users or that the U.S. would be unable to rely on imported oil to supply a fuel that was lower in sulfur than the indigenous coals that were being burned. Many utilities converted their coal-fired facilities to low sulfur oil or gas as quickly as they were able, and by 1974 23,600 MW of capacity was burning oil (although these facilities may be reconverted to burn coal). The emerging energy shortages culminated with the Arab embargo of oil to the U.S. in October 1973. This event in turn has motivated an energy policy that puts increased emphasis on reducing oil imports to a level low enough that the U.S. economy can continue to function satisfactorily even if the imports are again embargoed.

The shift to low sulfur fuels was made because it appeared to the electric utility industry to be the best way to meet the new sulfur oxide standards. Tall stacks and intermittent control systems facilitate compliance with sulfur dioxide ambient standards, but they do not reduce the total amount of sulfur oxides released into the atmosphere. The commercial feasibility of stack scrubbing devices, the other viable option for reducing sulfur oxide emissions, has been a matter of sharp dispute between the utility industry and the Environmental Protection Agency (EPA). It is obvious that the sulfur oxide levels that were to be achieved by July 1, 1975 (under the terms of the Clean Air Amendments of 1970) cannot be met now even if stack scrubbing technology were ready to be used routinely at power plants, because of the time required to build and install the scrubbing devices.

During the last decade, emissions and ambient sulfur dioxide concentrations in urban areas have decreased, while nationwide emissions from electric power plants have nearly doubled. Ambient concentrations of sulfate particles in urban areas have not decreased but have remained almost constant from 1957 to 1970. This persistence of high urban sulfate levels despite the decline in urban sulfur oxide emissions may be the result of the increased emissions from remotely located electric power plants. Sulfate levels approaching the level of urban concentrations have been observed in rural areas of the Northeast with no local sources of sulfur oxide emissions and very low sulfur dioxide ambient levels.[1]

Sulfur oxide and particulate emissions from power plants may pose a serious health hazard. Sulfur oxides and suspended particulate matter may act to impair health by a variety of possible mechanisms following inhalation and retention in the human respiratory tract. It will require further investigation to elucidate these mechanisms. In past epidemiological studies devoted to examining the health effects of air pollution, the pollution parameters of sulfur dioxide concentration and total suspended particulate matter concentration have been utilized for correlation with effects. These parameters are probably only indicators of the toxic potential of the pollution mix and not causal agents. Thus, the particulate phase is known not to be a single agent, but a complex mixture of particles of different size, shape, density and chemical composition. The CHESS studies suggested that particulate sulfates, rather than either of the above two parameters, may be a better indicator of the toxic potential of the polluted atmosphere.[2]

Laboratory studies which utilized animals have also suggested that certain particulate sulfates ($Zn(NH_4)SO_4$, $ZnSO_4$ and H_2SO_4) are potent bronchoconstrictors, far more effective than sulfur dioxide in air at concentrations comparable to the particulate sulfate concentration. The bronchoconstriction capacity of the particulate matter increased as particle size decreased in the guinea pig assay method, which utilized pulmonary flow resistance as the indicator of toxic potential.

For the above reasons, and with full recognition of the large uncertainties which still remain to be resolved, this analysis of the social costs of sulfur oxides and particulate pollution will focus on the impact of control strategies for particulate sulfate concentrations in air. The uncertainties in this approach are not only associated with the effects of sulfates on health and the ecosystem, but with the measured and predicted concentrations of sulfate in air; analytical methods to determine particulate sulfates in air are not yet reliable. As will be seen, the range of uncertainty on many of the factors in the analysis is therefore large.

ALTERNATIVES FOR EMISSIONS CONTROL

This portion of the report will examine the costs for various control methods that might be adopted, together with the reduction in emissions that each method might achieve[3]. The analysis will address the choice among alternatives that can be implemented by 1980. Promising technologies are under development to remove sulfur before or during combustion, and to improve efficiency, thereby reducing the quantity of fuel needed to generate a given quantity of electricity. However, these technologies cannot be implemented on a large scale until 1985 or later, and the costs may not be significantly lower than the technologies presently available to remove sulfur from stack gases (see Chapter 10). Therefore, the analysis will focus on alternatives that are presently available.

The presently available alternatives for controlling sulfur oxide and particulate emissions are the following:

Tall Stacks and Intermittent Control

A stack height of the order of 100 to 300 meters may be sufficient (depending to some extent on the quantity of sulfur oxides emitted) to disperse the plume of effluent gases over a wide area, permitting ambient concentrations to be held below the levels established as standards. Under some meterological conditions high concentrations that violate standards may develop. A meteorological monitoring system is used to anticipate the outset of these conditions, and an intermittent control is then exercised to reduce emissions by shifting to a cleaner fuel or by reducing the levels of operation of the plants. The net result is that the total quantity of emissions may be reduced slightly or not at all, but ambient concentration in violation of standards may be avoided (see Chapter 12).

Coal Preparation

By pulverizing the coal and washing it prior to combustion it is possible to remove much of the physically bound portion of the sulfur and a large fraction of the ash. The coal washing process is relatively inexpensive, but some of the energy content of the coal is lost (see Chapter 10).

Shifting to Low Sulfur Coal

Much of the coal burned by utilities in the eastern United States has a sulfur content of 2-6 percent by weight. Some eastern coal is available with a sulfur content below 1 percent, but its extent is limited, and much of it is held for metallurgical applications. Low sulfur eastern coal commands a substantial price premium, which might increase if there were additional demand for low sulfur fuel. Low sulfur western coal is abundant, but the mining and transportation capability does not currently exist to provide it in large quantity to the eastern United States. Because of the low BTU and high ash content compared to the eastern coals, it is generally not possible to burn western coal in a boiler designed for eastern coal without extensive retrofitting or derating of the plant capacity (see Chapter 10).

Flue Gas Desulfurization (FGD)

A number of technologies are under development for scrubbing pollutants from the effluent gases before they are released from the stack. The lime scrubbing process appears to be the best developed technology for coal fired power plants. It permits removal of the order of 90 percent of the sulfur oxide, plus much of the fine particulate matter. Both capital cost and operation costs of flue gas desulfurization are high, but it is the most effective means of removing sulfur oxides and other pollutants from the emissions into the atmosphere (see Chapter 11).

Demand Modification

Since sulfur oxides and other pollutant emissions from power plants are a by-product of electricity generation, one alternative for reducing these emissions is to reduce demand growth for electric power. The relationship between growth and emission levels is not a simple one, however. Demand is allocated among plants in an electric power system so as to meet demand with acceptable reliability at the least total cost. In practice, the newest and most efficient plants are used almost continuously to meet the base load, while the oldest plants are used to meet the peak loads and to furnish reserve capacity. The allocation of demand for electricity from an electrical system is summarized by the total energy demand made on each plant. This total demand is usually expressed by the loading (load factor) for the plant: the equivalent number of hours (percentage of time) the plant must operate during the year at its rated capacity in order to provide that amount of energy. A slowing of demand growth that leads utilities to delay construction on new plants with low emissions levels may have little effect on total pollutant emissions from the system, if older plants with high emissions continue to be used at high load factors (see Chapter 8).

Nuclear power plants provide an economical means of producing base load power without emitting any sulfur oxides or particulates. By accelerating the construction of nuclear power plants, the loading, and consequently, the sulfur oxide emissions from coal fired plants can be reduced. (Of course, there are other environmental problems associated with nuclear power that should be assessed in considering it as an alternative to coal fired plants.)

A continued national effort toward domestic self-sufficiency may result in shifts from oil and gas to coal as a fuel for electric power generation. This effort may involve shifting from oil or gas to coal for many existing power stations that have the capability to burn coal, conversion to coal burning capability for fossil steam plants now being planned or under construction, and higher loading for existing coal fired plants as oil or gas fired plants are taken out of operation or reduced in loading.

METHODOLOGY

The approach to be taken in comparing alternative control strategies is to assess their economic impact on the costs associated with generating electricity and their effect in reducing emissions. A judgment must then be made to evaluate this tradeoff: What increment in increased electricity costs is justified by a given level of emission reduction? We shall assess the benefits of emission reduction by modeling the effect of the emissions on ambient air quality levels and on the deposition of pollutants, then modeling the effects on human health, materials damage, ecological changes, and aesthetic degradation. Through an assessment of costs and benefits the analysis can provide guidance to policymakers in assimilating the complex array of factors that impact on the tradeoff between the consequences of the pollution and the costs associated with emissions control.

Considerable uncertainty may characterize the costs of the alternatives, the emissions reductions that may be achieved, the relationship between emissions and ambient air quality levels, and the health, materials damage, and environmental consequences of given levels of pollutant concentration. The degree to which these uncertainties will impact on the decision among alternative control strategies may be identified by sensitivity analysis. If changing a factor within its range of uncertainty will change the preferred decision alternative, it will be useful to quantify the uncertainty by assessing a probability distribution over the range of values the uncertain factor could assume. The value of resolving the uncertainty can then be computed from the decision context (see Howard 1966, 1968; North 1968, and Tribus 1969 on the use of probability in decision analysis; see Spetzler and Stael von Holstein 1972 on methods to encode probability distributions). The scope of the present report does not permit an extensive application of these methods. The approach will be illustrated on the most important uncertainties; the analysis could be expanded to include other uncertainties.

In the context of a public policy question such as controlling emissions from power plants, the assessment of overall costs and benefits may need to address issues of equity and of distribution: different people may receive the benefit than those who pay the costs. Cost-benefit analysis of public policy decisions usually assumes implicitly that the parties to the decision may be persuaded to make their choice on the basis of maximizing the overall net benefits to society. The question of how to implement the socially optimal alternative may well be the most difficult aspect of the problem. While a cost benefit analysis may be useful in identifying the best alternative from society's point of view, considerable further effort may be needed to determine what is the best alternative for practical and effective public policy. The public policy decision makers must understand how the various concerned private parties to the decision will react to a new policy initiative, and they must choose with care the means by which the private parties are to be motivated to act in society's best interest.

There are two ways that a private party may be motivated to act in the public interest when it is at variance with his own immediate objectives: (1) his decision alternatives may be limited by regulations or standards imposed on him by public authority (2) his values may be shifted toward the overall values of the society by economic means: incentives, taxes, penalties, fees; or by non-economic means such as persuasion that his action will gain him the good will (or enmity) of his fellows. The legalistic approach places the responsibility for planning on the public authority, which must assimilate a complex array of economic and technical factors in order to establish the standard. Once established, a standard is difficult to change. If new information indicates that the standard is not appropriate, the planning exercise must be redone and the concerned private and public parties convinced that the change in standards is justified.

The use of economic incentives has been advocated by virtually every economist who has written on pollution, but it has rarely been used as a way of controlling emissions*. It has the advantage of flexibility: by delegating the social cost of the pollution as a direct cost to the private party making the decision, the public authority provides him with the incentive to make decisions that are optimal from the viewpoint of the public authority. Planning is therefore decentralized, and the detailed knowledge possessed by the private parties can be used to improve the decision process. Flexibility is much easier to achieve: If the public authority determines that levels of pollution are too high, it raises the cost associated with emissions, providing an incentive to the private parties to reduce them.

THE EMISSIONS CONTROL DECISION FOR A REPRESENTATIVE ELECTRIC POWER PLANT

The public policy decision on emissions control will involve setting standards or implementing a system of incentives and fees. The actual resource allocations to implement an emissions control alternative will be made by the electric utility. The decision problem on emissions control is ultimately whether the owners of a power plant shall modify their operations by such means as installing a flue gas desulfurization (FGD) process, switching to a low sulfur content fuel, or installing a taller stack and intermittent control system. The adoption of the emission control strategy will result in higher costs to the owners of the power plant, and these higher costs will generally be passed on as higher prices to the consumers of electricity. The benefits from adopting the emissions control strategy come from the change in amount (and timing) of emissions of sulfur oxide and other materials into the atmosphere that may adversely affect human health, cause damage to other living organisms or material property, and result in effects, such as visibility reduction, that are aesthetically undesirable. A decision between alternative strategies requires a balancing of the additional cost imposed on the generation of electric power against the value of emissions reduction.

The analysis will focus on sulfur oxide emissions from coal fired steam power plants in the northeastern United States. As described in other sections of this report, most of the sulfur is emitted as sulfur dioxide rather than as sulfur trioxide or sulfate, but subsequent atmospheric chemical processes may oxidize the sulfur dioxide to sulfuric acid aerosol and suspended particles of ammonium sulfate and other sulfate salts. Recent epidemiological data have indicated that these sulfates may give rise to serious and widespread health effects (EPA 1974). Damage to material property from atmospheric sulfur oxides has been estimated to cause hundreds of millions of dollars in annual losses (Waddell 1974). "Acid rain" resulting from atmospheric sulfur oxides may lead to retarded growth in forests, deleterious effects on lakes and streams, damage to agricultural crops, and damage to building materials, statues and other material property (see Section 1 of Part Two). Aesthetic effects from degraded visibility may be another substantial problem. While sufur dioxide is invisible, sulfate particles do absorb and scatter visible light[5]. Degraded visibility in areas with high real estate or environmental values is a substantial public concern that should be appropriately reflected in the values associated with sulfur emissions (Randall et al. 1974).

There are many distinctions that must be made between power plants in different locations. Ideally, a detailed model for assessing costs and benefits should be developed at each power plant for which a decision on emissions control is to be made. This scale of effort is not possible in the present study. We shall not attempt to address in detail the decision at a particular plant, but rather do _illustrative_ calculations that are chosen to be _representative_ of different types of plants and different locations in the northeastern United States. Specifically, we shall consider the following as representative cases:

(1) An existing coal fired plant in a remote rural location.
(2) An existing plant capable of burning coal in an urban or near-urban location.
(3) A new coal fired plant in a remote rural location.

A fourth category, a new coal-fired plant in an urban or near-urban location could be added, but for this category the decision would seem relatively obvious: an efficient sulfur removal system would almost surely be required under existing state and local air quality regulations. Federal New Source Performance Standards (NSPS) will require flue gas desulfurization on new coal-fired plants beginning in 1975[6]. (This requirement also holds true for remotely sited plants, but since some utilities have alleged that application of these standards to such plants serves little or no social purpose, consideration of that case was included in this analysis.)

The approach in carrying out the cost-benefit analysis will be to evaluate alternative strategies for representative power plants in each category by assessing the economic costs of electricity generation and the costs associated with sulfur oxide pollution effects. Because of the suspected hazards of sulfate, the emphasis will be on regional effects caused by long range pollutant transport, rather than local violations of air quality standards in the immediate vicinity of the plant. The intention is to assess the marginal benefit of pollution control strategies for this plant as it affects regional ambient levels of sulfur dioxide, sulfate, and acid rain, and to compare the consequences associated with these levels with the marginal increases the strategy will impose on the cost of electricity generation. Both the consequences of the sulfur oxide emissions and the cost of abatement methods will vary, depending on local and regional conditions. These local and regional factors should be taken into account in the decision among alternative emissions control strategies.

AN OVERVIEW OF THE ASSESSMENT OF COSTS AND BENEFITS FOR A REPRESENTATIVE PLANT

Figure 13-1 presents an overview of a model for assessing costs and benefits, which will be applied for each alternative strategy for pollution control. The framework is a general one that could be applied to any stationary source, but the concern here is a coal fired steam electric plant. This plant meets demand from the power system for electricity: The effect of demand on an individual plant is summarized by its loading, the number of hours the plant is operated.

In steam power plants the sulfur content of the fuel is oxidized in the combustion process and mixes with other combustion products that are exhausted from the boiler. The heating value and sulfur content of the fuel and the efficiency of the power plant determine the

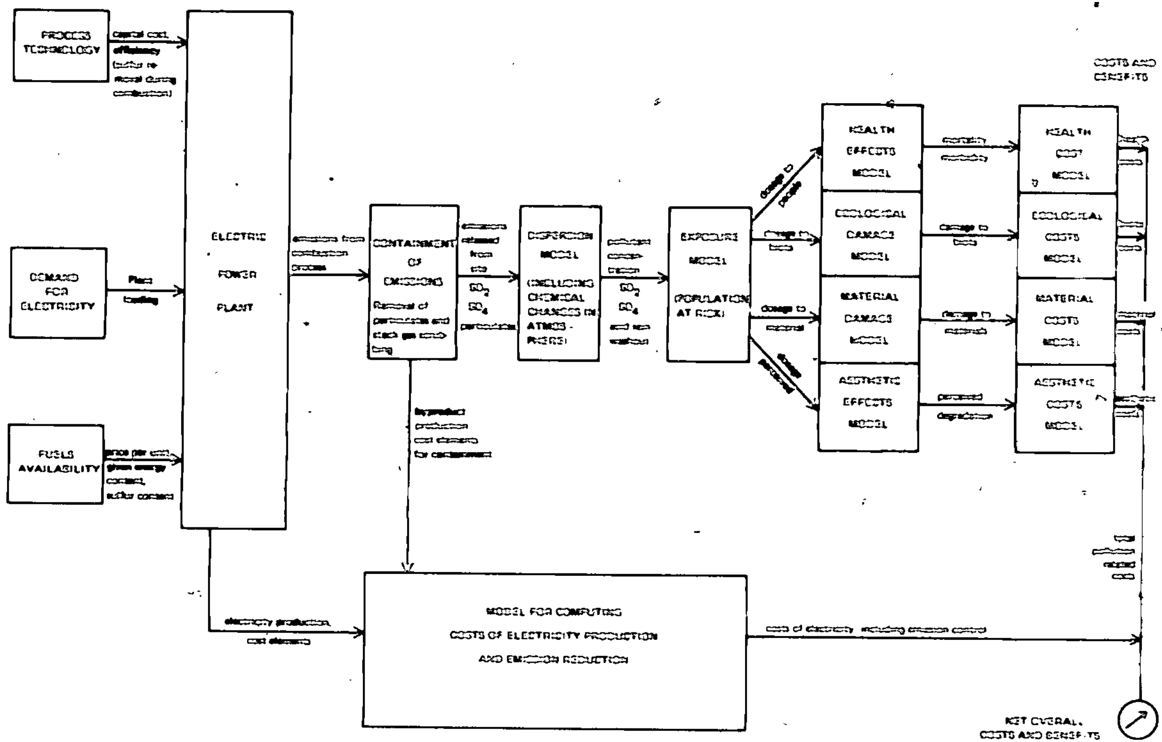

FIGURE 13-1: Overview: Sulfur Oxide and Fine Particulate Pollution From a Stationary Source

amount of sulfur oxides produced per kilowatt hour (kwh) of electricity generated. Sulfur oxide emissions from the combustion process may be removed by stack gas scrubbing devices, and particulates may be removed by the scrubing devices or electrostatic precipitators. The costs of these technologies are assessed and included in the cost of electricity generation.

The price of the fuel, e.g., the number of dollars per ton of coal, varies by its energy content and its sulfur content. For example, a coal with 24 million BTU's heating value per ton and 3 percent sulfur content (by weight), is assumed for this analysis to be available at $24/ton delivered to the power station. Lower sulfur content coal will generally be more expensive: for example $32/ton for coal with 0.9 percent sulfur and the same heating value. The supply of low sulfur coal is limited, and a large shift from high sulfur to low sulfur coals for power plant fuel would result in an increase in their relative prices. The supply-demand-price relationship may be critical in the evaluation of an overall national policy intended to cause a shift to lower sulfur content coal, but changes in the price relationship will not be discussed further. If the price increase is sufficiently high, the situation becomes equivalent to not having low sulfur coal available. We shall examine how the choice among alternative strategies is affected if eastern low sulfur coal is not available.

From the fuel prices, capital costs, plant efficiency, and loading, the costs of electricity generation may be computed. We can then compare alternative strategies for emission control on the basis of two numbers: the cost per kilowatt-hour of electricity generated, and the sulfur oxide emissions produced per kilowatt-hour of electricity[7]. For example, a representative existing power plant burning 3 percent sulfur coal produces electricity for a cost of 17.2 mills per kilowatt-hour and emits 0.026 lbs of sulfur per kilowatt-hour generated. If higher priced (eastern) low sulfur (0.9 percent) coal is used, the cost of electricity increases to 20.6 mills per kilowatt-hour, but emissions are reduced to 0.0078 lbs of sulfur per kilowatt hour generated.

A choice between these two alternatives implies assessing the tradeoff between reducing the sulfur emissions and increasing the cost of electricity. A simple way in which to assess the tradeoff is by placing a value on each pound of sulfur emission, and then examining the total cost of electricity generation plus the cost attributed to sulfur emissions. We shall carry out this calculation on an incremental or marginal basis for the individual power plant, rather than national implementation of the emission control alternatives. That is, we shall assess the benefits from reducing emissions by a relatively small amount relative to existing pollution levels, and compare them to the costs based on existing price levels for the emission control alternatives. Benefits from emissions reduction may depend on ambient levels, and costs for emission control alternatives may depend on the demand for them. For example, if health effects from ambient sulfate were significant only above a threshold level, then the pollution cost per pound of sulfur emitted would drop substantially once ambient sulfate levels are reduced below this threshold level. Likewise, increased use of low sulfur coal to reduce power plant sulfur oxide emissions will cause the price premium over high sulfur coal to rise. In our calculations we assume only small changes in pollution levels (and costs) as a result of implementing an alternative strategy at a particular power plant. As a consequence, the pollution cost per pound of sulfur is assumed to remain constant for this power plant regardless of the amount of sulfur emissions removed. However, the pollution cost per pound of sulfur removed will depend on many factors (such as ambient pollution levels) that differ for different plants in different locations. The benefits of a control strategy per pound of sulfur removed may be substantially different at different power plants.

Figure 13-2 shows the calculation of a representative existing power plant that can burn 3 percent sulfur coal or a more expensive

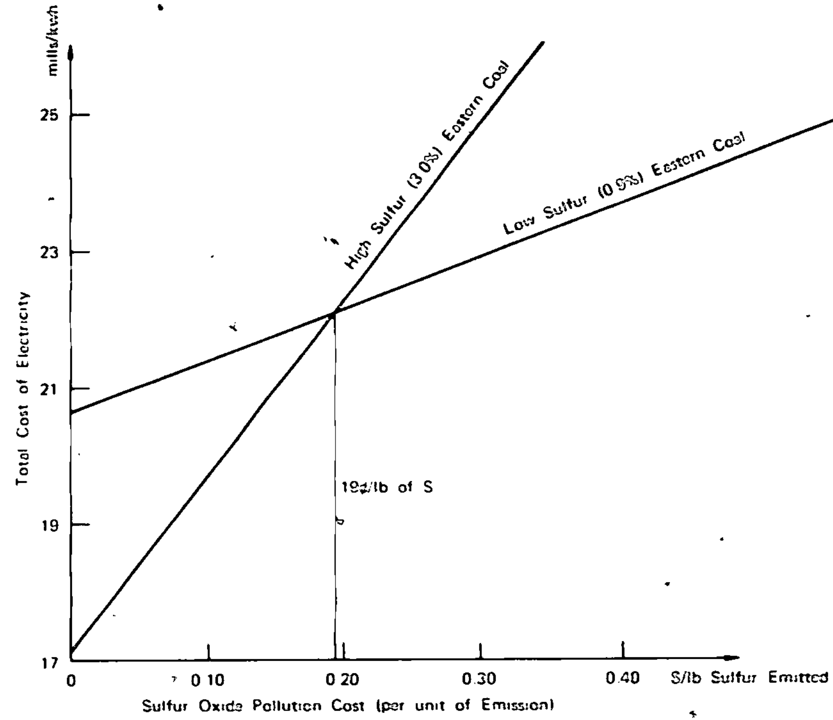

FIGURE 13-2: Total Cost of Electricity, Given Social Cost per Unit of Sulfur Oxide Emission

0.9 percent sulfur coal. If no cost is attributed to sulfur emission the total cost for the two alternatives is 17.2 and 20.6 mills per kilowatt hour as before. Suppose instead a cost of $0.30 is attributed to each pound of sulfur emitted into the atmosphere. Then the total cost for generating a kilowatt-hour using high sulfur coal is 25 mills per kilowatt-hour, 17.2 for the cost of producing the electricity plus 0.026 x 300 mills (= $.30) for the cost attributed to the sulfur emissions. The total cost of generating a kilowatt-hour using low sulfur coal is 23 mills/kwh, 20.6 for the cost of producing the electricity plus 0.0078 x 300 mills (=$.30) for the sulfur emissions. The lowest total cost is now achieved using low sulfur coal, whereas when emissions were assigned a zero cost, high sulfur coal resulted in the lowest total cost. The total cost for producing electricity plus the cost of the emissions associated with producing the electricity is shown plotted against the cost assigned to a pound of sulfur emissions in Figure13-2 for each of the alternatives. It may be seen that the graph of total cost for each alternative is a straight line, since the cost increases linearly with the pollution cost per unit emitted. The slope of the line is given by the pounds of sulfur emitted per kilowatt-hour generated. The point at which the two lines cross gives the cost per pound of sulfur emitted ($.19) at which the total costs of the alternatives are equal: If the cost attributed to sulfur emission is greater than 19 cents per pound of sulfur, the best alternative is the low sulfur coal; and if the cost attributed to sulfur emissions is less than 19 cents per pound of sulfur, the best alternative is the high sulfur coal. <u>A judgment that the low sulfur coal alternative is better than the high sulfur coal alternative is equivalent to a judgment that the social cost of sulfur emissions are judged to exceed 19 cents per pound of sulfur emitted, for this is the equivalent increase in the price of electricity that must be accepted in return for the lower emission levels.</u> Similarly, a judgment to use 3 percent sulfur coal in this plant instead of the more expensive low sulfur coal implies a judgment that the social cost of sulfur emissions is less than 19 cents per pound of sulfur emitted; the reduction in emissions from a switch to low sulfur coal is judged not worth the increased electricity generation costs.

If the dollar value of the consequences of sulfur emissions could be accurately assessed, it would be a simple matter to carry out this type calculation to determine what emissions control alternative is best in the sense of providing a kilowatt-hour of electricity at the least total cost to society*. But the consequences of the emissions are complex and uncertain, and the assessment process is a difficult one. However, it must be kept in mind that any decision to impose a particular alternative technology, or a requirement that emissions shall be held below a specified level, implies a value judgment on the cost of pollution. A decision among emissions control alternatives is required: either the existing fuels and control technology will be used, or a switch will be made to another type of fuel and/or emission control technology. Since this decision will implicitly require judgments on the value of reducing emissions, the assessment process cannot be avoided. The value of emissions reduction must be assessed, either explicitly or implicitly.

An explicit method for assigning values to the social cost of pollution is represented by the right hand side of Figure 13-1. The left hand side may be regarded as a model for calculating the emissions released into the environment and electricity generating costs for a representative electric power plant. The boxes shown on the right hand side represent the models used to evaluate the social costs to be assessed on the emissions from that power plant:

- o A Dispersion Model to relate sulfur oxide and particulate emissions to ambient concentrations of sulfur dioxide, sulfate, and particulates, and to acid rain washout.
- o An Exposure Model of the population, biota, and material property that may be impacted by the pollutant

concentrations. The output of this stage is the dosage of pollutant received.

- Models for the effect of a given dosage to a given population on human health, on vegetation and other ecological systems, on material property, and consequences that are aesthetically undesirable, such as visible smog. The output of this stage is a description of the physical consequences of the pollutant concentration: for example, morbidity and mortality, reduced growth in vegetation, eroding of galvanized steel, and reduction in visibility.
- The last stage is the assessment of values on these physical consequences of pollution so that they may be compared with the costs of emissions control strategies. Values assigned to health, vegetative damage, materials damage, and aesthetic degradation become the basis for valuing the effects caused by a given level of emissions, and the model structure can be used to compute a value per pound of pollutant emitted for the evaluation of alternative strategies as illustrated in Figure 13-2.

The need for making a decision motivates the need for the value assessment process. <u>The models for the emission to ambient relationship, the effect of ambient air quality on human health, the effect of acid rain on vegetation, etc., are summaries of the information available as a basis for decision making.</u> There may be substantial uncertainties in some of the factors or relationships used in these models, and there may be disagreements on the valuing, for example, of health effects. In carrying through the assessment process, attention and effort should be focused on those uncertainties and value judgments that affect the decision.

Now that the methodology has been described, we turn our attention to the calculation of the electricity generation costs and emissions levels for representative plants, which will be summarized in a series of diagrams constructed in the same manner as Figure 13-2. We then develope dispersion models for the emissions to ambient relationship for these representative plants. Finally, models for the effects on health, ecological systems, material property, and aesthetics are used to evaluate the consequences of pollution and assess a social cost per unit of emissions. Important uncertainties on pollution consequences are reflected in a probability distribution over a range of values for this pollution cost, and the impact of the uncertainty on the emissions control decision is examined. Since resolving the uncertainty leads to an improvement in the decision compared to a choice made on the basis of presently available information, a value of resolving the uncertainty will be calculated.

CALCULATION OF TOTAL SOCIAL COSTS FOR VARIOUS SULFUR EMISSION REDUCTION ALTERNATIVES

Calculations are now presented for the electricity generation costs and emission levels that might be expected if various decision alternatives are chosen. The alternatives considered are:

1. High sulfur coal, perhaps with tall stacks and intermittent control.
2. Removal of sulfur from fuel before combustion (coal preparation)
3. Flue gas desulfurization (FGD)
4. Shift to low sulfur fuel (eastern or western low sulfur coal)

Since the best strategy for controlling sulfur emissions may differ for different plants, three representative cases are considered:

1. An existing coal fired plant in a remote non-urban location.
2. A coal fired plant planned for construction in the near future in a remote non-urban location.
3. An oil burning plant, originally designed to burn coal, which may be reconverted to coal. This plant is presumed to be located in an urban area of the East Coast.

Detailed assumptions for the specific plants are summarized in Table 13-1. Since an additional alternative might be to construct a nuclear plant instead of a new coal fired plant, data for a base load nuclear plant is also included in the table for a point of reference.

Tables 13-2, 13-3, and 13-4 give the cost and sulfur emissions data assumptions made in the evaluation of each alternative. Coal preparation is assumed to require a small capital investment and to result in coal heating value losses of the order of 10 percent. The reduction in sulfur content achievable by coal cleaning depends on the particular process used. Conventional techniques now available will reduce the sulfur content of high sulfur coal by approximately 40 percent[10]. Flue gas desulfurization, represented by the lime scrubbing process, is expensive: there is a sizeable capital cost, additional operating costs, a loss of plant capacity, and an associated energy loss. The price of low sulfur western coal depends critically on transportation costs. A higher price is assumed for plants located on the East Coast as opposed to the mideastern portion of the country. For older plants with boilers not designed for lower BTU coal, capacity derating will result from the burning of the lower heating value western coal.

Use of tall stacks and intermittent control is equated to the base case, which we henceforth denote by "high sulfur coal". Such techniques may be useful in meeting ambient standards for sulfur dioxide in the immediate vicinity of the plant, but it is not anticipated that their costs or contributions to the overall reduction in sulfur oxide emissions would be significant relative to those of the other strategies being considered[11]. An efficient particulate removal system is also assumed. The importance of particulates in oxidizing sulfur dioxide to sulfates will be discussed in the next section, below.

Capital charges, operating and maintenance, and fuel costs for each plant type under each alternative were summed to obtain an estimated production cost per kilowatt-hour of electri-

TABLE 13-1

Economic and Technical Factors for Representative Power Plants.[a]

CASE	TECHNOLOGY				FUEL				ECONOMICS		
	Plant Size (MW)	Average Load (hrs/yr)	Load Factors %	Efficiency %	Fuel	Sulfur Content %	MMBTU/ton	Cost including trans.	Capital Cost ($/kw)	Capital Amortization %	Operating Cost (mills/kwh)
Existing coal fired plant (non-urban)	620	6000	69	33	Coal	3	24	$24/ton $1.00/MMBTU	250	14	1.0
New coal fired plant (non-urban)	612	7000	80	38	Coal	3	24	$24/ton $1.00/MMBTU	500	17	.5
Old coal fired plant re-converted from oil to coal (urban)[b]	620	6000	69	33	Coal	3	24	$24/ton $1.00/MMBTU	261[e]	14	1.0
Nuclear plant	1000	7000	80	32	U	-	-	$.18/MMBTU	800	17	.4

[a] Sources: Chapter 10 and personal communications, Harry Perry.

[b] plant assumed to be located on East Coast.

[d] plant is assumed to be equiped with a particulate collection system.

[e] includes $7/kw conversion charges plus $4/kw to upgrade the particulate collection system. The latter is not charged if FGD is employed.

TABLE 13-2

Coal Preparation Cost Factors [a]

CASE	Capital Costs	Added Fuel Costs/ton [b]	% Energy Loss	% Reduction Sulfur Content
Existing coal fired plant (non-urban)	$6/ton yr.	$2.25	10	40
New coal fired plant (non-urban)	$6/ton yr.	$2.25	10	40
Old coal fired plant re-converted from oil to coal (urban)	$6/ton yr.	$2.25	10	40

[a] Source: Chapter 10.

[b] Includes amortization of capital costs.

TABLE 13-3

Flue Gas Desulfurization Cost Factors[a]

CASE	Capital Costs/kw	% Amortization[b]	Operating Costs/kwh[c]	Sludge Disposal Cost/kwh[d]	% Capacity Derating[e]	% Energy Consumed	% Reduction of Sulfur Oxides
Existing coal fired plant (non-urban)	$125	17	0.6m	0.5m	6	6	90
New coal fired plant (non-urban)	$100	17	0.5m	0.3m	6	6	90
Old coal fired plant reconverted from oil to coal (urban)	$125	17	0.6m	0.9m	6	6	90

[a] Costs based on lime scrubbing. Source: Chapter 11 and personal communications, Leigh Short.

[b] Harry Perry, personal communication.

[c] labor, maintenance, and supplies cost. Capital amortization, sludge disposal, and energy loss included separately.

[d] Ponding of sludge is assumed for the non-urban site. For the urban sites it is assumed that sludge will have to be disposed of by other means.

[e] capacity valued at $500/kw.

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TABLE 13-4

Low Sulfur Coal Cost Factors[a]

Case	Fuel Costs/.on	Fuel Heating Value/ton	% Fuel Sulfur Content	Plant Efficiency	% Capacity Derating	
Existing coal fired plant (non-urban)	$30[b]	21 MMBTU	.7	No Change	10	LOW SULFUR WESTERN COAL
New coal fired plant (non-urban)	$30[b]	21 MMBTU	.7	No Change	0	
Old coal fired plant re-converted from oil to coal (urban)	$36[c]	21 MMBTU	.7	No Change	10	
Existing coal fired plant (non-urban)	$32	24 MMBTU	.9	No Change	0	LOW SULFUR EASTERN COAL
New coal fired plant (non-urban)	$32	24 MMBTU	.9	No Change	0	
Old coal fired plant re-converted from oil to coal (urban)	$32	24 MMBTU	.9	No Change	0	

[a] Source: Chapter 10 and Harry Perry, personal communication.

[b] plant located in mideastern portion of the U.S.

[c] plant located on East Coast.

city. The sulfur emissions in pounds of sulfur per kilowatt-hour of electricity were also calculated. Table 13-5 summarizes these results. Details of the cost calculation for the base case (high sulfur coal) and for flue gas desulfurization are given in Appendix 13-D.

Total social cost will be the production cost plus the product of sulfur emissions and pollution cost per unit of sulfur emission. For this reason, the plots of total social cost versus pollution cost, illustrated in Figures 13-3, 13-4, and 13-5, are straight lines. We may easily identify the most desirable emission reduction alternative as that straight line relationship that, for a given assignment for pollution costs, yields the lowest value of total social costs. Inspection of the figures show that for pollution costs below 19 cents per pound of sulfur emitted the best alternative is to burn high sulfur coal. As pollution costs are increased above 19 cents per pound of sulfur emitted the best alternative becomes eastern low sulfur coal. We note that there is little difference in the location of this crossover point among the three cases. For the cost elements given in Tables 13-1 through 13-4 the alternatives of coal preparation and switching to western low sulfur coal do not give as large a marginal reduction in sulfur emissions compared to their marginal cost. For pollution costs above 19 cents per pound, the eastern low sulfur coal provides an additional value from reducing sulfur emissions more than sufficient to offset the 33 percent premium in fuel costs, which is reflected in the cost of electrical generation.

As pollution costs are raised still higher, the flue gas desulfurization (FGD) alternative becomes the best alternative. Although it is the most expensive (excepting low sulfur western coal) the FGD alternative permits overall emissions reductions approaching 90 percent wheras low sulfur eastern coal gives only a 70 percent reduction. The added cost for the additional sulfur removal may be substantial, so that the crossover points where FGD drops below low sulfur eastern coal are high: 53 cents per pound of sulfur for the retrofit non-urban plant

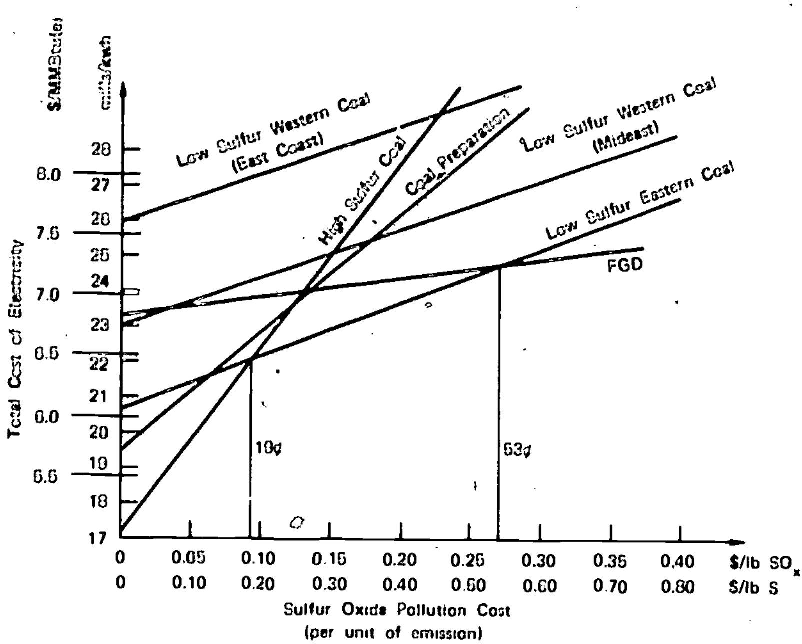

FIGURE 13-3: Total Social Cost Versus Pollution Cost, Existing Coal Fired Plant (Nonurban)

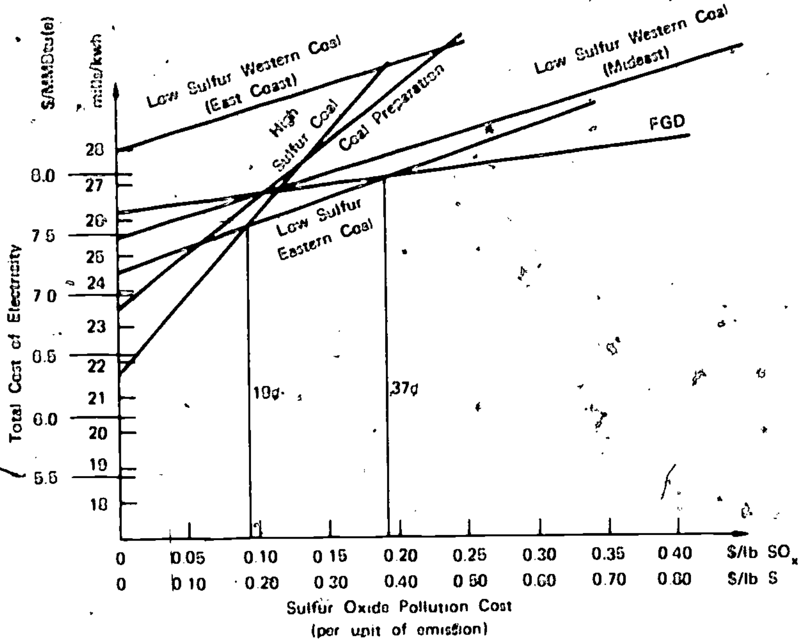

FIGURE 13-4: Total Social Cost Versus Pollution Cost, New Coal Fired Plant (Nonurban)

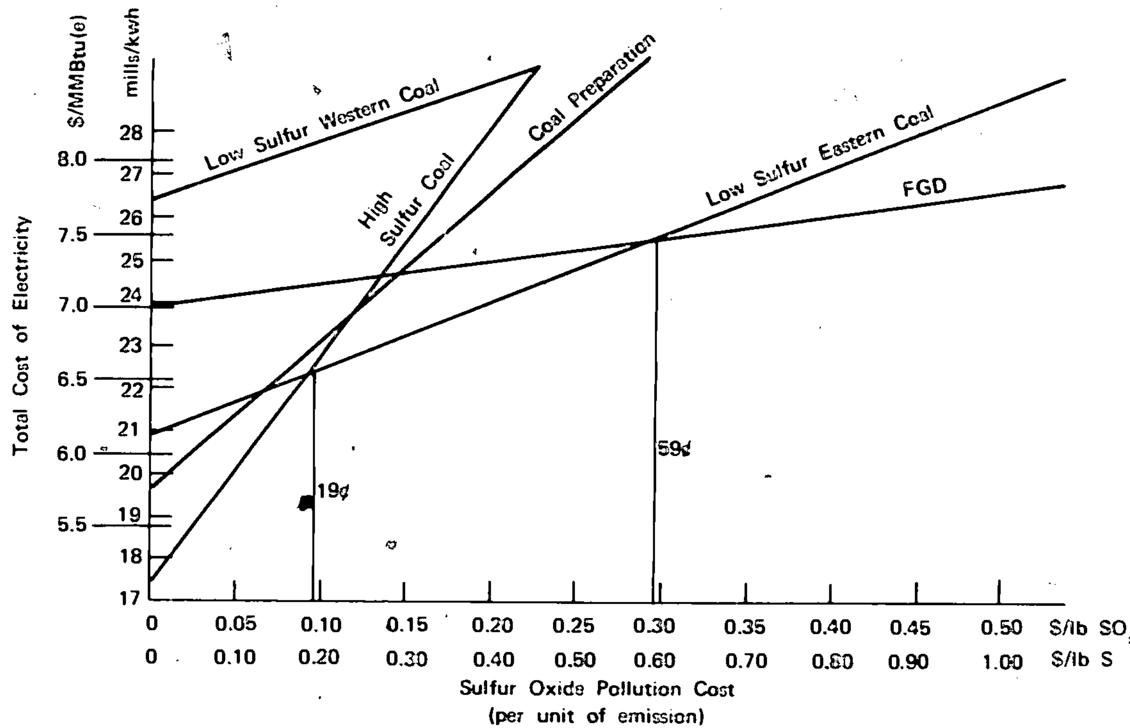

FIGURE 13-5: Total Social Cost Versus Pollution Cost, Old Coal Fired Plant Reconverted to Coal ~~(Urban)~~

(Figure 13-3), 37 cents for the new non-urban plant (Figure 13-4), and 59 cents for the retrofit urban plant (Figure 13-5). The latter case includes a cost of 0.9 mills/kwh to sludge disposal, which corresponds to about $4/ton of sludge removed. At this cost level recycling FGD systems such as Wellman Lord or magnesium oxide should be almost competitive. However, there is less experience in applying these systems to high sulfur coal than is the case for lime scrubbing.

The economics of sludge disposal or material recycling may be a dominant consideration for FGD installations in urban areas. Where sludge can be ponded, the sludge disposal costs are moderate, of the order of 0.15 to1.0 mills/kwh (see Chapter 11). However, for existing urban or suburban power plant locations ponding of sludge may not be feasible because of the absence of a suitable site. If sludge must be disposed of by hauling it off, the costs are estimated to be much higher: in the range of 0.6 to 1.5 mills/kwh, or possibly even higher (Leigh Short, personal communication). We have taken 0.9 mills as a nominal value for the representative urban plant calculation. The sludge material is about two thirds water, even after settling, so the volume to be removed is of the order of 50-100 percent of the coal needed to fuel the plant. Given the environmental restrictions and aesthetic considerations that may apply in some urban sites, lime or limestone scrubbing may be more expensive because of sludge disposal. For these sites the use of low sulfur coal or other scrubbing technologies will be indicated.

Low sulfur eastern coal is available only in limited supply. For some power plants it may not be available except at a price premium well in excess of the 33 percent we have assumed in the calculations above, especially if large numbers of other plants have shifted from high sulfur to low sulfur coals. In this case the line on Figures 13-3, 13-4 and 13-5 for low sulfur eastern coal will be shifted up to where it lies above high sulfur coal, coal preparation, and FGD for all values of pollution cost. With low sulfur coal no longer a competitive alternative the values of pollution cost at which flue gas desulfurization becomes advisable drop sharply. For an existing plant in a remote rural location, lime scrubbing is preferred to burning high sulfur coal if the pollution cost per pound of sulfur emitted is above 26 cents (Figure 13-3). If coal preparation is considered as the alternative to scrubbing, then the cost per pound of sulfur at which scrubbing becomes preferred is 27 cents. For a new plant (Figure 13-4) the value is substantially lower: above 23 cents per pound of sulfur removed lime scrubbing will be the preferred alternative in the absence of available low sulfur eastern coal. Coal preparation is not quite marginally competitive: the marginal cost of sulfur removal for a new plant is about 27 cents per pound of sulfur removed, and this alternative permits only 33 percent sulfur removal (including the effect of energy losses), compared to about 89 percent for lime scrubbing. For plants located in mideastern as opposed to east coast locations, western low sulfur coal may be a competitive alternative if it is available at a cost somewhat lower than as assumed in Table 13-4.

Retrofit installations of flue gas desulfurization systems on oil burning plants in urban areas may involve higher costs for sludge disposal. If low sulfur coal is available, the pollution cost justifying lime scrubbing instead of low sulfur eastern coal is 59 cents per pound of sulfur removed. If low sulfur coal is not available, the crossover point drops to 29 cents per pound (where lime scrubbing is preferred to coal preparation) or 28 cents per pound if the gas desulfurization is compared to burning high sulfur coal (Figure 13-5).

The values of the crossover points are sensitive to the emissions levels and costs of electrcity given in Table 13-5. If different values are used the crossover points will change. The crossover between low sulfur coal and flue gas desulfurization is particularly sensitive; a change of 1 mill per kilowatt-hour in the cost of flue gas desulfurization chnages the crossover point by 23.2 cents for a new plant, and 19.8 cents for a retrofit

TABLE 13-5

Electricity Production Costs and Sulfur Emissions for Three Representative Power Plants

ALTERNATIVE	CASE: EXISTING COAL FIRED PLANT (non-urban)		NEW COAL FIRED PLANT (non-urban)		OLD COAL FIRED PLANT RE-CONVERTED FROM OIL TO COAL (urban)	
	Cost per unit of electricity (mills/kwh)	Sulfur Emitted ($x10^{-3}$ lbs/kwh)	Cost per unit of electricity (mills/kwh)	Sulfur Emitted ($x10^{-3}$ lbs/kwh)	Cost per unit of electricity (mills/kwh)	Sulfur Emitted ($x10^{-3}$ lbs/kwh)
High sulfur coal with tall stacks and intermittent control	17.2	25.9	21.6	22.5	17.5	25.9
Coal preparation	19.4	17.2	23.6	15.0	19.7	17.2
Flue gas desulfurization	23.3	2.75	26.2	2.4	23.9	2.75
Switch to low sulfur Western coal	23.0 (mideastern) 26.0 (East Coast)	6.9	25.5 (mideastern) 28.0 (East Coast)	6.0	26.3	6.9
Switch to low sulfur Eastern coal	20.6	7.8	24.6	6.7	20.9	7.8
Nuclear	-	-	New Nuclear Plant 21.7	0	-	-

installation. If the comparison is between high sulfur coal and flue gas desulfurization, the sensitivity is rather low: a change of 1 mill per kilowatt-hour in the cost of flue gas desulfurization causes a change in the crossover point by 5.0 cents for a new plant and 4.3 cents for a retrofit installation. The crossover point between high sulfur and low sulfur coal changes by 6.3 cents for a 1 mill increase in the cost for low sulfur coal for a new plant, and 5.5 cents for an existing plant. These are both equivalent to 0.57 cents increase in the crossover point for a 1 cent per million BTU change in the price differential of low sulfur coal over high sulfur coal.

THE RELATION BETWEEN SULFUR OXIDE EMISSIONS FROM A SINGLE POWER PLANT AND AMBIENT INCREASES IN SULFUR DIOXIDE AND SULFATE LEVELS

The relationship between the emissions of sulfur oxide and ambient levels of sulfur dioxide and sulfate is not well understood at present (see Part Two, Section 1). The major source of difficulty lies in the lack of knowledge on the oxidation rate of sulfur dioxide to sulfate. There are a variety of chemical mechanisms by which the oxidation reaction can occur. In clean air, photooxidation of sulfur dioxide may proceed rather slowly: smog chamber experiments indicate rates of the order of 0.1 to 0.2 percent per hour (Bufalini 1971). Impurities act to catalyze the oxidation reaction at a much faster rate: photo-induced radicals, soot or metallic oxide particulates, and various organics or oxidants may lead to oxidation rates of the order of 5-10 percent per hour. Rates of this magnitude have been calculated for observations of sulfate in the photochemical smog of the Los Angeles Basin (Roberts and Friedlander 1974), and similar rates are consistent with recent airborne measurements of sulfur oxides in England (Smith and Jeffrey 1975). Rates in excess of 5 percent per hour have been observed in direct measurements of sulfate levels in plumes from electric power plant and smelter plumes (Gartrell 1963, Weber 1970, Newman et al. 1975a). However, recent measurements of a power plant plume from a coal fired plant with an efficient electrostatic precepitation system (99.5 percent by weight particulate removal) indicate a lower rate, below 5 percent cumulative oxidation during the period (about two hours) for which the plume could be observed before sulfate levels were too close to background for accurate measurement (Newman 1975b).[12]

Sulfur dioxide may leave the atmosphere by being adsorbed on vegetation, soil, or water before it is oxidized to sulfate. The rate of sulfur dioxide removal has been extensively studied in England. The rate of removal of a volume of gas by a surface area is generally expressed as a deposition velocity (volume per unit time divided by area has the dimensions of velocity). A deposition velocity of 0.8 cm/sec was judged appropriate as an average value for the English countryside (Garland 1974). The deposition rate for removal of sulfates (in the absence of precipitation) has also been studied, but there is less agreement for this figure. The OECD study (OECD 1974) is using 0.4 cm/sec for sulfate deposition, although the other data indicates that this value may be high.[13]

Precipitation is relatively efficient in removing sulfate particles from the air. There is evidence that high relative humidities that accompany precipitation increase the oxidation rate of sulfur dioxide to form sulfate, and some sulfur dioxide is removed directly by being absorbed into the water droplets. European data indicate that rain will remove nearly all the sulfur oxide from the air within 60-100/km of the point of emission (Miller and DePena 1973, Hogstrom 1973a,b; Brosset 1973). The resulting precipitation is highly acidic, and this acid rain may have deleterious effects on vegetation, fisheries, and materials (see Chapter 7).

To carry through our analysis we need to assess the effect of emissions from a power plant in changing the ambient levels of sulfur dioxide and sulfates in downwind areas. Our concern is with average annual levels, not peak concentrations, and with large areas relatively remote from the plant location. Motivation for this emphasis comes from the health and material damage assessments to follow: It is believed that very low level increases in ambient sulfate levels may cause significant health effects and damage to material property.

In the absence of available quantitative relationships between sulfur oxide emissions and ambient sulfate levels, we have developed a highly simplistic model for the emissions to ambient relationship[14]. The details of this model are given in Appendix 13-A. The model is not intended as a predictive device, but rather a summary of the limited available understanding of the relationships involved. An important purpose of the model is to clarify the need for further data and research on the emissions to ambient relationships. For decisions on emissions control alternatives for specific power plants, this model is clearly inadequate. Data on sulfur dioxide and sulfate levels and detailed information on regional meterology could be acquired at a modest cost and this information should be used as the basis for a much more extensive analysis than we have been able to carry out in this study.

The model is based on the following assumptions:

In a period shortly following the emission, the sulfur oxides become uniformly distributed from the ground to a mixing layer height. The height of the plume then remains constant.

The emissions are uniformly distributed over an arc of constant size, so that the width of the plume expands in direct proportion to the time since emission, or (with constant wind velocity) in direct proportion to the distance downwind traveled by the plume.

We are assuming that the emissions travel down the plume uniformly distributed in a "box" whose length is the distance traveled by the wind per unit time[15], whose height is the height of the mixing layer, and whose width is the distance perpendicular to the wind direction subtended by an angle of constant size. Thus, the width of the box grows in direct proportion to time, and the concentration of pollutants decreases inversely with time.

We make the following assumptions about the chemical reactions of the sulfur oxides.

Sulfur dioxide to sulfate oxidation takes place according to a first order rate reaction. The rate may change considerably between comparatively clean rural air and pollutant-laiden urban air.

Sulfur dioxide is removed to the ground at a constant deposition velocity, beginning at the time of emission. (This assumption will overestimate the amount of sulfur dioxide removal to ground for a plume from a tall stack that travels many miles before touching ground, and it will underestimate the sulfur dioxide removal from a shorter stack where the plume is in contact with the ground for substantial time before the plume is dispersed uniformly up to the inversion or mixing layer height).

Suspended sulfate is removed to the ground at a constant deposition velocity, beginning with the time of emission.

Precipitation removal can be ignored in calculating reaction rates. In rainy weather it is reasonable to assume that virtually all of the ambient sulfate is removed over a distance of the order of 100 km. We shall assume later that a fraction of the sulfate is removed by precipitation between the time of its formation and the time it reaches the area when ambient levels are to be measured.

The relationships in the model are shown in Figures 13-6 and 13-7. Figure13-6 indicates the chemical changes and removal mechanisms involved and Figure 13-7 shows the geometry assumed.

We shall use the model to examine two representative situations:

(1) a plant located in a remote rural area, approximately 500 km upwind of a major metropolitan area,

(2) a plant located in a metropolitan area, with urban settlement extending 40-80 km downwind from the plant.

We shall use the first to examine an existing coal fired plant and a new coal fired plant in a remote location, and the second to

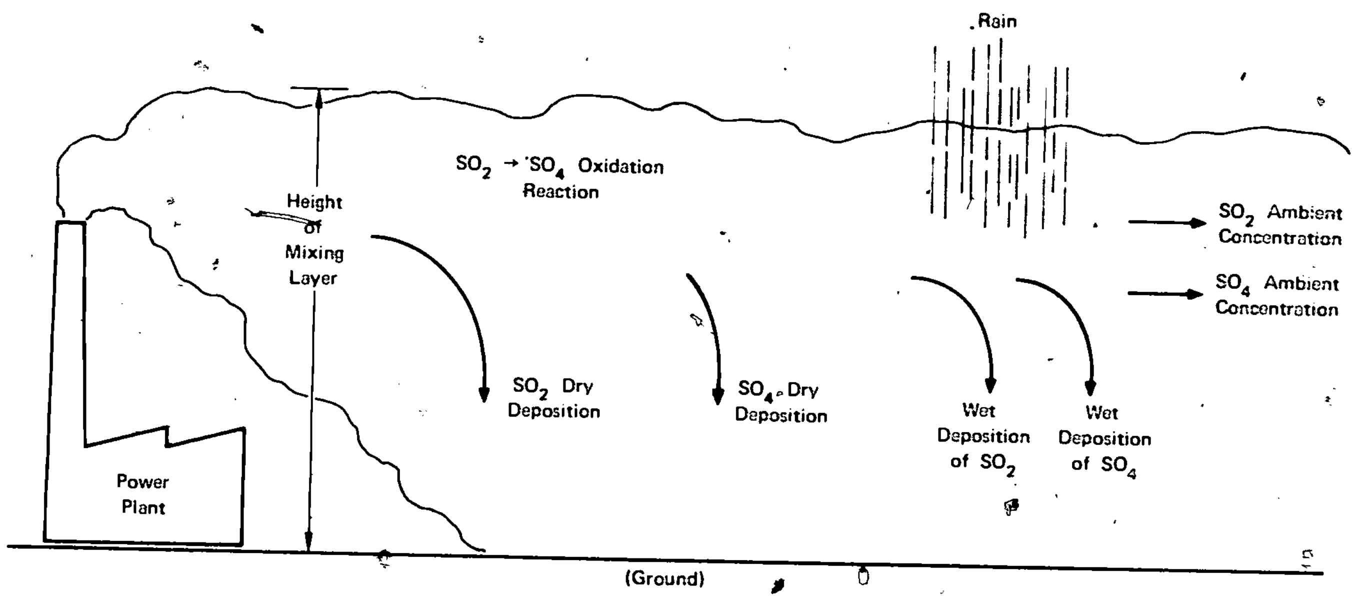

FIGURE 13-6: Model for Emission to Ambient Relationship for Sulfur Oxide Emissions from a Representative Power Plant

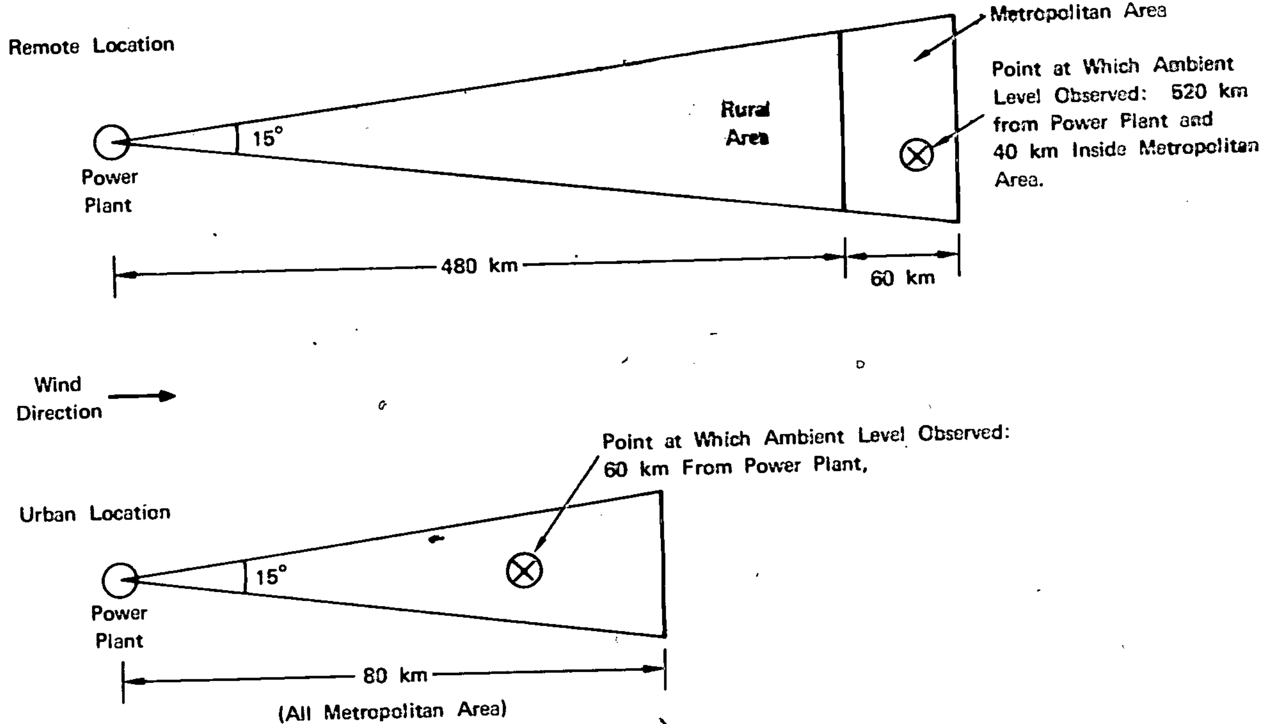

FIGURE 13-7: Geometry of Representative Cases

examine an existing plant in an urban location that could be reconverted from oil to coal.

We use the following values as inputs to the calculation:

(1) The wind is a constant 20 kilometers/hour.

(2) The angle subtended by the plume is 15°.

(3) The height of the mixing layer above ground is 1,000 meters.

(4) The depostion velocity of sulfur dioxide to the ground is 0.8 cm/sec, giving a removal rate of 2.88 percent per hour with a mixing layer height of 1,000 meters above ground.

(5) The deposition velocity of SO_4 to the ground is 0.4 cm/sec, giving a removal rate of 1.44 percent per hour with a mixing layer height of 1,000 meters.

(6) The oxidation rate of sulfur dioxide to form sulfates is 0.5 percent per hour in rural air outside of a metropolitan area, and 5.0 percent when the air has passed over a metropolitan area and contains particulates, oxidants, and hydrocarbons from urban emission sources.

(7) All sulfur oxides emitted from the power plant are emitted as sulfur dioxide rather than as sulfates. Assuming 1 to 2 percent of SO_4 is emitted as sulfate, the error introduced by this approximation is negligible.

We now examine the solution obtained for these numbers. (The equations will be found in Appendix 13-A.) First, let us examine the case of a remotely located power plant of about 600 MW, burning 3 percent sulfur coal. Its average emission level (including the effect of plant loading) is computed to be 10^4 kilograms of sulfur dioxide per hour. Table 13-6 shows the marginal addition to the ambient concentrations of sulfur dioxide and sulfate from this plant, assuming that the pollution does not encounter pollutant laden metropolitan air (oxidation rate of SO_2 to SO_4: 0.5 percent/hour).

TABLE 13-6

Incremental Contributions to Ambient Levels of Sulfur Dioxide and Sulfate Resulting from the Emissions of a Single 600 MW Power Plant, Rural Air (Oxidation Rate of 0.5 percent per hour Assumed).

Time, hours since emission	3	6	12	18	24	50
Distance from plant, km	60	120	240	360	480	1000
Increase in SO_2 concentration ($\mu g/m^3$)	28.6	12.9	5.27	2.87	1.76	0.35
Increase in SO_4 concentration ($\mu g/m^3$)	0.66	0.62	0.53	0.46	0.40	0.22

500-307 O - 75 - 40

Now we repeat the calculation assuming that the emissions occur in pollutant laden urban air, for which an oxidation rate of 5.0 percent per hour is assumed (see Table 13-7).

We now give the results of calculations for the representative situations. First, let us examine the situation in which the power plant is remotely located, but the emissions encounter polluted urban air in a metropolitan area 480 kilometers (300 miles) downwind after 24 hours. At this time we assume the oxidation rate changes from 0.5 percent to 5 percent per hour. We compute the incremental contributions to ambient sulfur dioxide and sulfate levels as shown in Table 13-8 and plotted in Figure 13-8.

A detailed assessment of health, materials, damage, and other consequences would include the spatial variation in the ambient levels of sulfur dioxide and sulfate. We shall avoid this level of detail and use representative single values for the incremental contribution to ambient sulfur dioxide and sulfate levels resulting from the emissions of the power plant. For the remotely located plant, we take as representative for computing pollution consequences the values after two hours of oxidation in urban air (following 24 hours in rural air to give a total of 26 hours since emission from the power plant). For the urban plant, we take as representative the values three hours after emission, assuming, assuming oxidation in urban air during this time (see Table 13-9).

Of the effect of changes in the inputs, the most uncertain is the oxidation rate. A sensitivity analysis is given in Table 13-10.

The dominant effect of the oxidation rates in determining the ambient sulfate concentration shows up strongly in Table 13-10: Depending on whether we use high or low values, we get about an <u>eightfold</u> change in the contribution to ambient sulfate levels from the power plant. Note we would have even higher sulfate levels from the remotely located plant if we had used a uniform 2 percent per hour oxidation rate for both urban and rural air. (A forthcoming paper by Eliassen and Saltbones [1975] uses a trajectory model with conversion assumptions similar to ours to examine sulfate observations

TABLE 13-7

Incremental Contributions to Ambient Levels of Sulfur Dioxide and Sulfate Resulting from the Emissions of a Single 600 MW Power Plant, Urban Air (Oxidation Rate of 5.0 percent per Hour Assumed).

Time, hours since emission	3	6	12	18	24	50
Distance from plant, km	60	120	240	360	480	1000
Increase in SO_2 concentration ($\mu g/m^3$)	25.0	9.9	3.1	1.3	0.6	0.04
Increase in SO_4 concentration ($\mu g/m^3$)	6.2	5.4	4.2	3.3	2.6	1.03

TABLE 13-8

Incremental Contributions to Ambient levels of SO_2 and Sulfate from the Emissions of a Single Power Plant: Representative Calculation for 600 MW Plant 300 miles Upwind of Urban Area.

Time, hours since emission	3	6	12	18	24	25	26	27
Distance from plant, km	60	120	240	360	480	500	520	540
Oxidation rate, % per hour	0.5	0.5	0.5	0.5	5.0	5.0	5.0	5.0
Increase in SO_2 concentration ($\mu g/m^3$)	28.6	12.9	5.27	2.87	1.76	1.56	1.39	1.23
Increase in SO_4 concentration ($\mu g/m^3$)	0.66	0.62	0.53	0.46	0.40	0.50	0.58	0.65

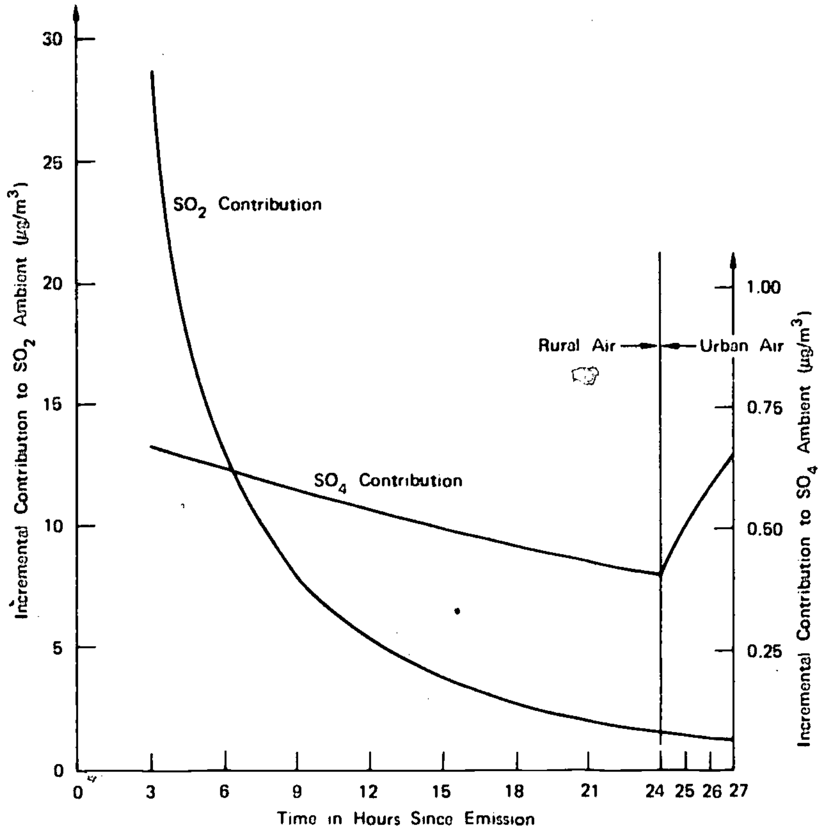

FIGURE 13-8: Incremental Contributions to Ambient Levels of SO_2 and Sulfate from the Emissions of a Single Power Plant, Representative Calculation for 600 MW plant 300 Upwind of Urban Area

TABLE 13-9

Emissions to Ambient Calculation for Representatives Power Plants (Emissions Rate Assumed is 10^4 Kilograms of Sulfur Dioxide per Hour)

Case	Increase in Ambient Concentration, $\mu g/m^3$ SO_2	Increase in Ambient Concentration, $\mu g/m^3$ Sulfate	Location of Measurement	Oxidation Rate Assumption
Remotely located plant (Existing or New Plant)	1.39	0.58	Urban Area 26 Hours (540 km) Downwind	24 hours at 0.5%/hr. then 2 hours at 5%/hr.
Urban Plant (Existing)	25.0	6.2	Urban area 3 hours (60 km) Downwind	3 hours at 5%/hr.

TABLE 13-10
Sensitivity Analysis, Emissions to Ambient Relationship

	Remote Plant, 480 Km from (after 26 hours)		Urban Plant (after 3 hours)	
	SO_2 conc, $\mu g/m^3$	SO_4 conc, $\mu g/m^3$	SO_2 conc, $\mu g/m^3$	SO_4 conc, $\mu g/m^3$
Nominal Values	1.39	0.58	25.0	6.20
Low Oxidation Rate 0.1% per hour in rural air 1.0% per hour in urban air	1.6	0.13	28.2	1.32
High Oxidation Rate 1.0% per hour in rural air 10.0% per hour in urban air	1.1	1.06	21.5	11.6
With Constant 2% Oxidation Rate	1.03	1.30	27.3	2.59
SO_2 Deposition Rate				
Low: 0.4 cm/sec	2.0	0.74	26.1	6.33
High: 1.6 cm/sec	0.7	0.38	22.9	5.95

TABLE 13-10 (Cont.)

SO_4 Deposition Rate				
Low: 0.03 cm/sec	1.4	0.67	25.0	6.32
High: 0.8 cm/sec	1.4	0.51	25.0	6.07
Height of Mixing Layer				
Low: 500 meters	1.3	0.64	45.3	11.6
High: 1500 meters	1.2	0.48	17.2	4.27
Angle Subtended by Plume				
Low: 10^o	2.1	0.88	37.6	9.33
High: 22.5^o	0.9	0.39	16.5	4.10

in Northern Europe and to compute rates for sulfur dioxide and SO4 oxidation. The mean of their computed rate values is 0.8 percent per hour. This rate gives essentially the same answer for the incremental increase in ambient sulfate as our representative rural plant calcuation using a rate of 0.5 percent for 24 hours and 5 percent for two hours.)

The sensitivity values shown in Table 13-10 have been chosen rather subjectively by the authors as representing a set of reasonable extreme values. As a rough approximation for assessing the uncertainties involved, we will assume that each set represents approximately the 5 percent and 95 percent points on a cumulative probability distribution assigned to the quantity (e.g., the probability is judged to be 90 percent that the uncertain quantity would lie in the interval between the low and high values used in the sensitivity analysis, rather than outside the interval). In addition, uncertainties are assumed independent, except for the rural and urban oxidation rates which are assumed totally dependent (e.g., if one is high then the other will be high also, and visa versa). A sketch of the resulting probability distribution on ambient sulfate levels is given in Figures 13-9 and 13-10. These curves are meant to illustrate the great uncertainty on the incremental change in ambient sulfate levels resulting from the emissions from a power plant located upwind from an urban area in which there are substantial health and material property values at risk. More refined models, formal probability assessment procedures, and formal probability processing could be used to improve the degree to which these curves summarize present knowledge on the emissions to ambient relationship for sulfates. The curves shown may be taken as rough summaries of the present state of knowledge, and they are subject to revision as further information is obtained.

RECONCILING THE MODEL FOR AMBIENT INCREASES FROM A SINGLE PLANT TO EMISSIONS DATA AND AMBIENT SULFUR OXIDE LEVELS FOR THE NORTHEASTERN UNITED STATES

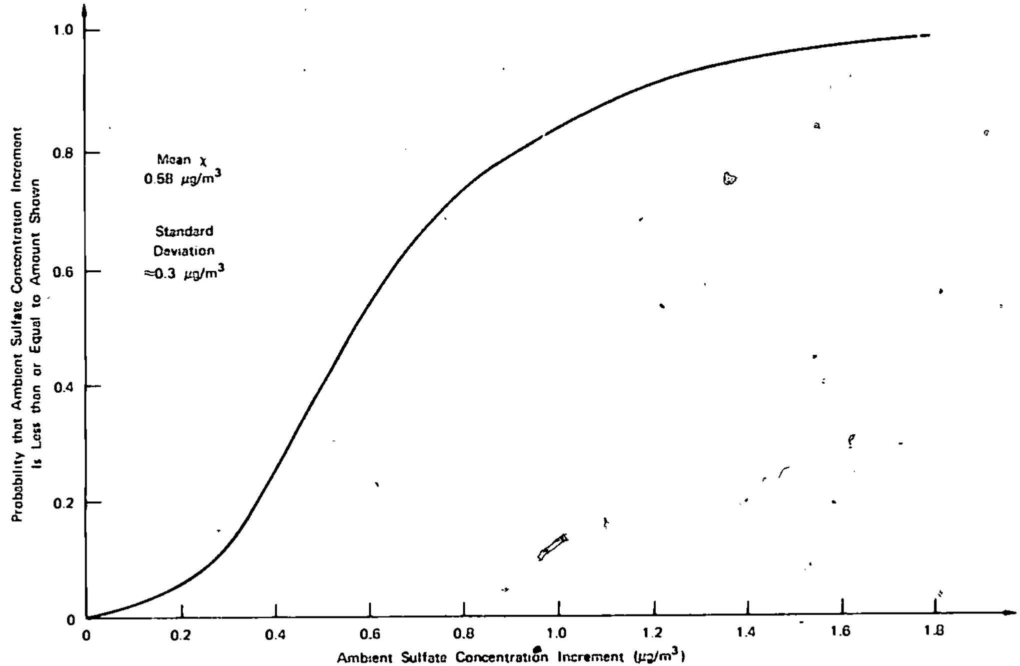

FIGURE 13-9: Sketch of Probability Distribution Illustrative of Present Uncertainty on Increment of Ambient Sulfate Concentration in Urban Area Approximately 300 Miles (500 km) Downwind of Rural Power Plant Emitting 10^4 kg of Sulfur Oxides Per Hour

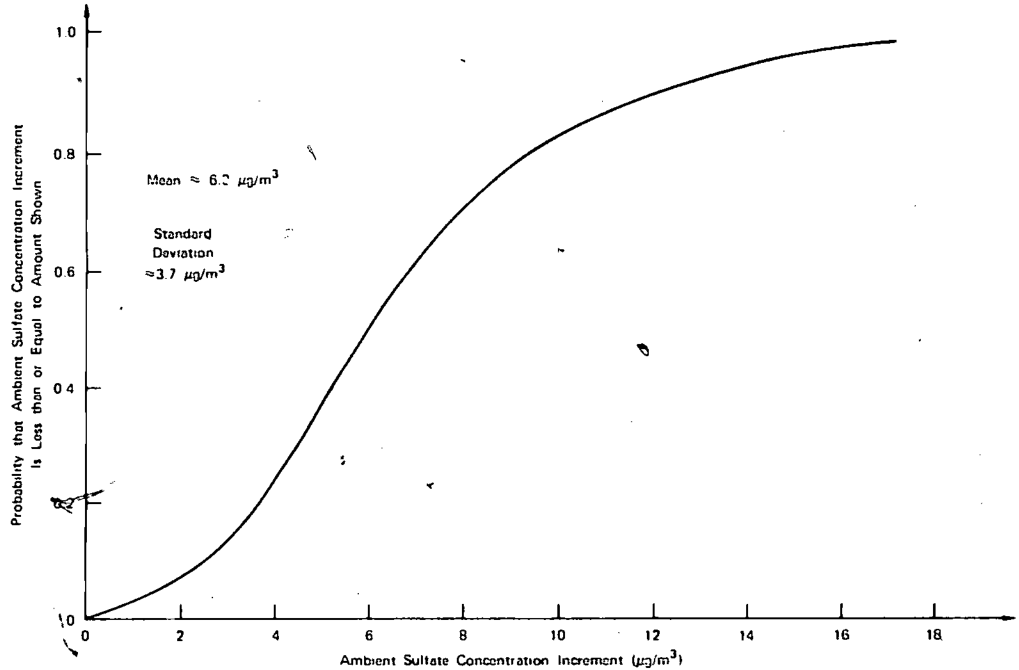

FIGURE 13-10: Sketch of Probability Distribution Illustrative of Present Uncertainty on Increment of Ambient Sulfate Concentration in Urban Area 60 km Downwind of (Urban) Power Plant Emitting 10^4 kg of Sulfur Oxides Per Hour

The calculations in the last section give us an indication of the relationship that may exist between sulfur oxide emissions from a single power plant and the ambient levels of sulfur dioxide and sulfate in downwind areas. The relationship must be regarded as tenuous, and subject to substantial change when regional models of sulfur oxide transport become available. Nonetheless, the objective set forth for this portion of the report is to carry out an analysis of alternative strategies based on the limited information presently available. We will therefore attempt to use the preceding calculations, after suitably modifying them to account for observed sulfate levels, as a basis for assessing the social cost per pound of sulfur emitted. The framework for this calculation is easily adapted to improved models relating power plant emissions to observed ambient sulfate levels, where such improved models are developed. The model used in the last section (and described in more detail in Appendix 13-A) is essentially a "back-of-the-envelope" calculation carried out by the authors when we were apprised that there was no model for long range sulfur oxide transport available for our use in a cost-benefit analysis. We believe that an essential step in a cost benefit analysis of alternative abatement strategies must be a quantitative relationship between the emission level and the pollutant concentrations at the location where the pollutants cause adverse consequences.

In order to assess the degree that our representative cases of the previous section were representative of the power plants presently emitting sulfur oxide, we compiled the map shown in Figure 13-10.** The data on power plant emissions were obtained from Federal Power Commission records for 1972 and dots were placed in the Air Quality Regions corresponding to plant locations. Each dot corresponds to power plant emissions of 50,000 tons of sulfur dioxide per year. For the representative plant calculations of the last section we assumed emissions of 10,000 kilograms of sulfur dioxide per hour, or about 96,500 tons of sulfur dioxide per year. This quantity corresponds to just less than two dots on the map. One dot is quivalent then to about 300 MW of capacity burning 3 percent sulfur coal. Lists of large power plants burning coal and oil fired plants that might be reconverted to burn coal are given in Appendix 13-B.

Let us examine the map of Figure 13-10a. We note a large concentration (about 70 dots) in Eastern Ohio - Western Pennsylvania - Maryland - West Virginia area. This amount corresponds to more than 10 percent of the total sulfur oxide emissions in the nation. Another large concentration is found in the Southern Illinois-Indiana-Kentucky area.

A look at the surface wind maps of Figures 13-11 and 13-12 indicates the direction of the wind, and Figure 13-13 shows us the location of the population concentrations. The implication of these maps is grim: The sulfur oxides emitted by these rural power plants tend to blow into the populous urban Northeast corridor.

Let us focus our attention on those 70 dots that represent the concentration of power plants in the Pennsylvania-Ohio border area about 500 km upwind of Metropolitan New York. Recall that for the representative rural plant we calculated a sulfate concentration of 0.66 ug/m³ (after 3 hours or 60 km), 0.40 ug/m³ (after 24 hours or 480 km) and then an increase to 0.58 ug/m³ after 2 hours exposure to urban air. The observed data of sulfate levels for rural locations in Pennsylvania and Virginia is an annual average level of about 9 ug/m³ (see Appendix 13-C). In these locations there are no local sources of sulfur oxide emissions, and observed sulfur dioxide levels are very low, of the order of ten micrograms per cubic meter. The average annual sulfate levels observed in greater New York are in the range of 12-20 ug/m³. While 12.5 ug/m³ is the average for urban areas in the Eastern United States, the urban areas of New York, Pennsylvania, New Jersey and New England are somewhat higher, in the range of 16 ug/m³ (see Appendix 13-C).

We must reconcile these observations with the ambient increases computed from the model in the last section. To do this, two additional factors must be considered that were not

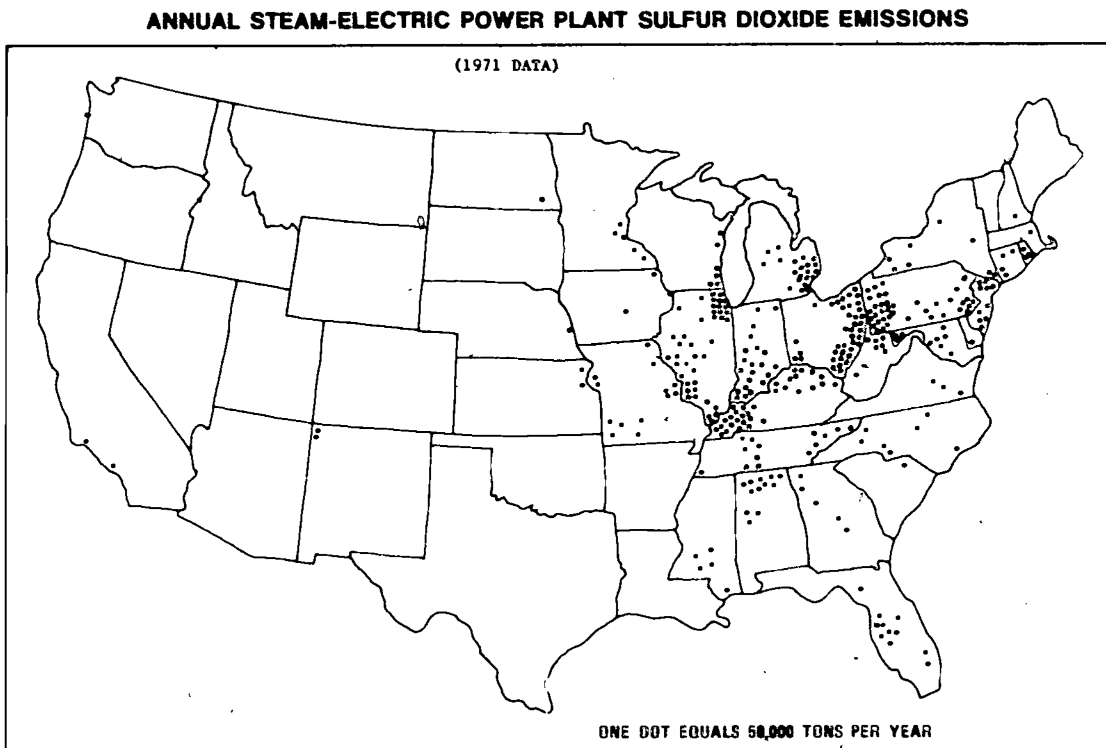

FIGURE 13-10a: Annual Steam-Electric Power Plant Sulfur Dioxide Emissions (1971 Data)

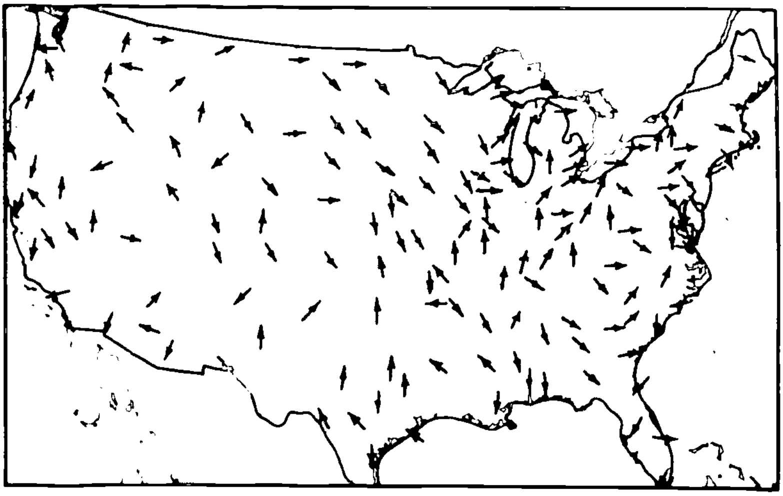

FIGURE 13-11: Normal Surface Wind Directions in January.

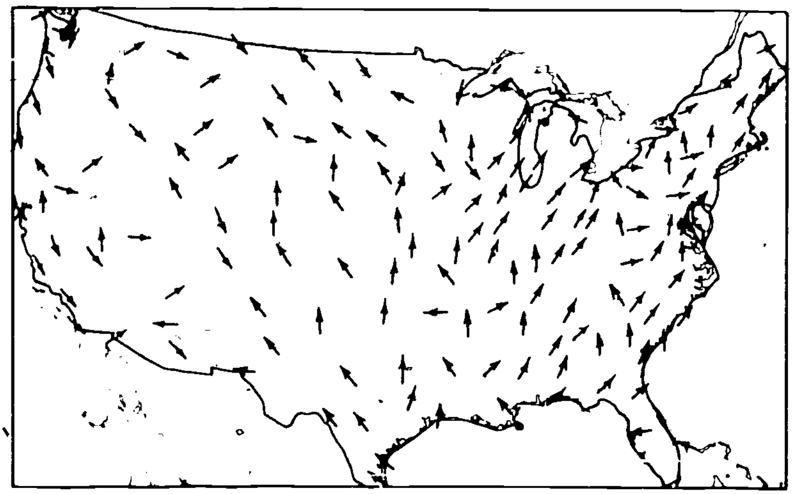

FIGURE 13-12: Normal Surface Wind Directions in July.

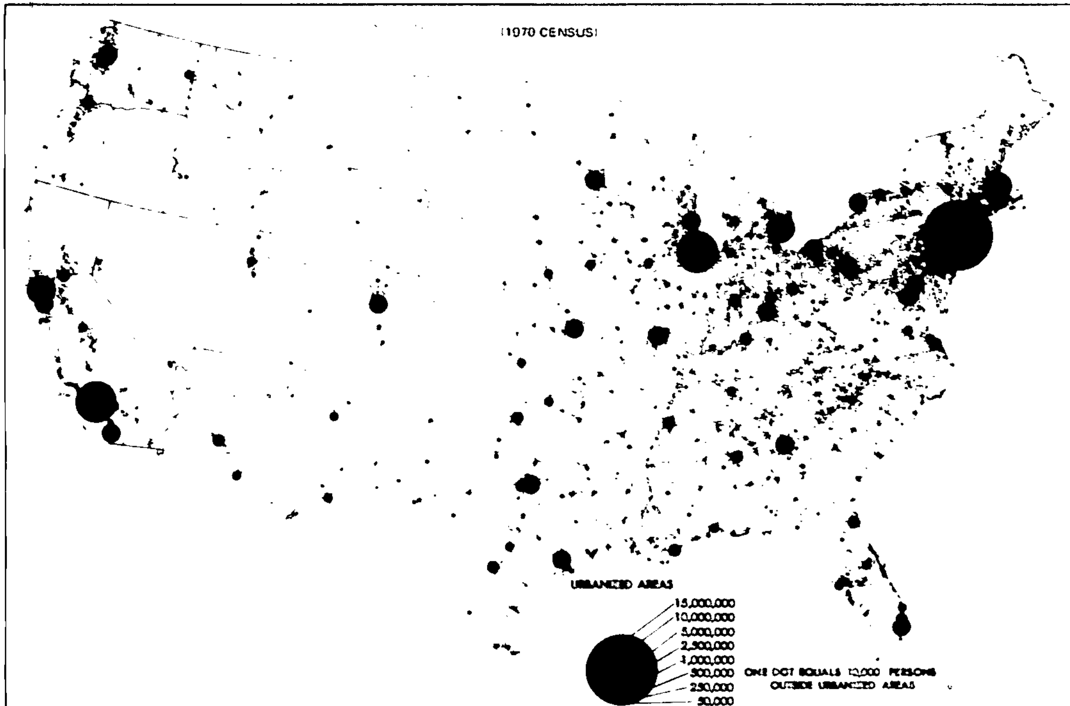

FIGURE 13-13: Population Distribution.

incorporated in the model: (1) variations in wind direction, and (2) loss due to wet deposition of sulfur dioxide and sulfate.

Whereas 15° may be an appropriate value for the angle subtended by the plume on a particular day, during the year the wind will vary in direction and the sulfur oxide emissions will be distributed over a much wider sector. We shall assume that during the year the variations in wind direction cause the sulfur oxide to be distributed over a sector of 45°, three times the 15° sector we assumed for the power plant plume. This will reduce average annual sulfate (and sulfur dioxide) ambient increments to one third the values shown in Table 13-9. To take account of wet deposition losses from rain, we will assume that 25 percent of the sulfur dioxide and sulfate is removed by precipitation prior to observation[16]. This will further reduce the average annual ambient increments by 25 percent. The average annual ambient increments contributed by a single representative plant would then be 1/4 the values shown in Table 13-9. The adjusted annual average increments are shown in Table 13-11.

The 70 dots in the Pennsylvania/Ohio Border-West Virginia area on the map, Figure 13-10, indicate the equivalent of 36 representative plants of 600 MW burning 3 percent sulfur coal. Using the values of Table 13-11(and the sulfate value for 12 hours from Table 13-6 adjusted for precipitation and plume width) we would then expect to see an increase of 36 x 0.25 x 0.53 = 4.8 ug/m³ in rural areas about half way to New York and an increase of 36 x 0.25 x 0.58 = 5.2 ug/m³ in the urban New York area. If we assume a background level of sulfates from other natural and remote man-made sources of 4-5 ug/m³ and a contribution from local sources near New York of about 3-10 ug/m³ of sulfate, we see that the numbers are in general agreement with observations. Obviously, the assumptions (1) that the sulfate resulting from emissions in the Pennsylvania/Ohio border-West Virginia area is distributed over a 45° sector, and (2) that 25 percent of the sulfate is removed by rain within 12 to 24 hours can and should be subjected to considerable refinement using meteorological

TABLE 13-11

Summary of Judgment for Average Annual Ambient Increases of Sulfur Oxide and Population at Risk Assumptions for Representative Power Plant Case Analyses

Representative Power Plant with Emissions of 10^4 kg of SO_2/hour or 96,500 tons of SO_3 per year	Ambient Increase from Plant Emissions for a 15° Angle Plume, from Table 13-9		Average loss from Wet Deposition of Sulfur Oxides	Average Annual Ambient Increases from Plant Emissions Assuming Distribution over a 45° Sector		Population at Risk Assumed with 45° Sector for Representative Case
	SO_2 ($\mu g/m^3$)	Sulfate ($\mu g/m^3$)		SO_2 ($\mu g/m^3$)	Sulfate ($\mu g/m^3$)	
Remote Location (Ambient Increase measured 26 hours or 520 km downwind)	1.39	0.58	25%	0.35	0.145	50 million
Urban Location (Ambient Increase measured 3 hours or 60 km downwind)	25.0	6.2	10%	7.5	1.86	11.5 million

information for the area on air trajectories and precipitation frequencies.

It may be useful to note the following implication for the numbers above: If sulfur oxide emissions from power plants on the Pennsylvania/Ohio border-West Virginia area were doubled, we would expect an increase in rural sulfate levels from 9 to 13.8 $\mu g/m^3$, an increase of about 50 percent; and an increase in the urban sulfate level in the Boston-New York-Washington D.C. urban complex from about 16 $\mu g/m^3$ to 21.2 $\mu g/m^3$, an increase of about 30 percent. These numbers are of course extremely rough calculations based on many simplifying assumptions, not the least of which is the assumption of first order rate reactions. If oxidation is limited by the availability of pollutants that act as catalysts, then the increase in urban sulfates from a doubling of sulfur oxide emissions would be less. An overall estimate for the increase in urban sulfate in the Boston-New York-Washington urban corridor from doubling power plant emissions in the Ohio-Western Pennsylvania-West Virginia area might be taken to be in the range of 15-45 percent[17].

Let us now consider the population at risk for a plant in the Pennsylvania/Ohio border West Virginia area. If we assume the emissions to be distributed over a 45° arc in the direction of the prevailing winds, the area affected will include the New England states, New York, Pennsylvania, New Jersey, Maryland, Delaware, and the District of Columbia. The combined population of these states in 54 million (1970 census) of which 47 million live in urban areas[18]. A substantial population in Southeastern Canada may also be affected by the emissions. We shall summarize these considerations by assuming a population at risk of 50 million persons impacted by the increase in sulfate levels from a representative rural plant located in the area approximately 500 km west of the Boston-Washington metropolitan corridor.

We will make similar assumptions for the urban location representative plant. Let us assume that on an average annual basis, the emissions from the power plant are distributed through a 45° arc in the direction of the prevailing winds. Let us further assume that about 10 percent of the sulfate is removed by precipitation prior to impacting on the population at risk (since the time assumed between emission and measurement is 3 hours for this case instead of 26 hours, we would expect less loss from rain). The net computation of average annual ambient increments is equivalent to multiplying the values in Table 13-9 by 0.3: a reduction of a factor of 3 caused by variations in wind direction, and a further reduction of 10 percent caused by washout and rainout from precipitation. For the population impacted by the emissions from the urban location representative plant, we will rather arbitrarily take 11.5 million, the population of the greater New York Metropolitan area. The calculated values for SOx levels for the urban location case are summarized in Table 13-11.

ESTIMATED HEALTH COSTS OF ELEVATED AMBIENT LEVELS OF SULFUR OXIDES

A comprehensive evaluation of a pollution reduction strategy must compare the dollar costs of implementing that strategy with the health and other costs sacrificed by not implementing it. In terms of our present problem, this means that we must convert a given level of sulfur oxide pollution to a dollar health cost. Placing a dollar value on morbidity and mortality is a difficult task, one that we all, understandably, are reluctant to undertake. However, if the comparisons of monetary and health costs are not made explicitly in the analysis, they will be made implicitly elsewhere. The importance of the assessment of the health effects of sulfur oxide emissions dictates their explicit consideration in our analysis (see Part One).

Our discussion in this section will consist of (1) a review of some of the health effects which appear to be associated with sulfur oxide air pollution, (2) an illustration of the sort of quantitative model which is needed for the evaluation of pollution reduction strategies, (3) an application of that model which will indicate the sensitivity of health effects to changes in the level of sulfate pollution, and (4) a method for converting specific health effects to aggregated health costs for society.

The Relationship Between Adverse Health Effects and Ambient Levels of Sulfur Oxides

In order to choose logically among sulfur oxide pollution reduction strategies we need to have some understanding of the dose response relationship between adverse health effects and ambient levels of sulfur oxides. Unfortunately, this relationship is not accurately known at present.

In general, epidemiological studies have indicated an association between ambient sulfur dioxide levels and adverse human health effects, yet controlled human exposure studies show that considerably higher than existing sulfur dioxide levels are incapable of producing an acute response in man. The apparent paradox is explainable by the possible toxicity of sulfur dioxide by-products present in polluted air. This contention is supported by recent studies carried out as part of the Community Health and Environmental Surveillance System (CHESS 1974) which indicated that health effects seem to be more closely associated with exposure to suspended atmospheric sulfate than with exposure to other pollutants.

The mechanisms by which sulfur oxides and suspended particulate matter may act to impair health are not well understood. The fine sulfate particles, particulrly acid sulfate aerosols, are believed to irritate the membranes lining the respiratory tract, making breathing more difficult and impeding the normal protective and self-cleansing mechanisms of the lung. There is some data indicating that the bronchoconstricting potency of particulate sulfates is related both to physical properties of the particles, such as size[19] and to chemical properties, such as pH (Lewis et al. 1972, Amdur 1969). The biological reactivity of the particles also seems to depend on such atmospheric conditions as humidity and temperature and there may be important synergistic effects with other atmospheric agents (McJilton and Frank 1973). The most effective control strategy for sulfur oxides will most likely depend critically on the exact nature of the sulfate-biologic response relationship in man. Therefore, it would appear that there is a very high value to clarifying these biological issues before any substantial resource commitment is made to a particular method for atmospheric ambient level sulfate control.

Adverse Health Effects Attributable to Elevated Levels of Atmospheric Suspended Sulfates.

The stated objective of the epidemiologic studies of the Environmental Protection Agency's Community Health and Environmental Surveillance System (CHESS) program is to establish data to aid the development of dose-response relationships between short-term and long term pollutant exposures and adverse health effects. As stated above, more research is required before any definite quantitative relationship may be established. However, progress is being made in this area, and it is useful to demonstrate the manner in which dose-response curves should be utilized in the context of the sulfur oxide pollution control decision problem. In the next section we shall present possible forms for the dose-response curves for some of the pollutant-disease associations that have been observed in recent epidemiological studies. A description and assessment of studies linking various adverse health effects with sulfur oxide pollution appears in Chapter 4.

Short term exposures to elevated levels of sulfur oxides, especially acid-sulfate aerosols, seem to aggravate asthma and pre-existing heart and lung disorders. Results of a study of asthmatics in the New York area showed higher rates of asthmatic attacks, which are characterized by marked hyperconstriction of the trachea and bronchi leading to sneezing and gasping for breath, on days with the highest suspended sulfate levels. The average threshold for observed aggravation by sulfates was calculated as 7.3 ug/m³ on warmer days with a minimum temperature above 50 F and 11.9 ug/m³ on warmer days with a minimum temperature lying between 30 and 50 F (Finklea et al. 1974). A study of cardiopulmonary patients in the New York Metropolitan area found elevated levels of suspended sulfates to be consistently associated with symptom aggravation, particularly shortness of breath, cough, and increased production of phlegm (Goldberg et al. 1974).

Elevated short term exposures to acid sulfate aerosols have frequently been shown to accompany perceptable increases in daily mortality. Older individuals with pre-existing heart and lung disease are particularly at risk. However, it is not known whether severe episoede of air pollution tend to shorten only the life span of the severely ill, or if the life span of a much larger group of the population is affected (see the section on chronic bronchitis and emphysema in Part One).

Epidemiological studies appear, in general, to indicate that repeated short term peak exposures or annual average elevations of suspended sulfates accompany excess acute lower respiratory disease in children, excess risk for chronic respiratory disease symptoms in adults, and decreased ventilatory function in children. The Salt Lake Basin studies of the CHESS program indicated, for example, an increased incidence of lower respiratory infection, croup, and bronchitis in children living in communities with higher annual average levels of suspended sulfates. Similarly, chronic bronchitis, which was characterized by the presence of cough and phlegm on most days for at least three months each year, was found to have a higher incidence rate among adults living in the more polluted communities (Finklea 1974, Galke 1974 a,b).

Dose-Response Relationship between Atmospheric Sulfate Level and Health Effects

Summarizing, epidemiological studies seem to indicate that short term exposures to elevated levels of sulfur oxides

1. aggravate pre-existing heart and lung disorders in elderly patients,
2. aggravate asthma,
3. perceptably increase daily mortality.

Repeated short-term exposures or elevations of annual average exposures seem to

4. increase the incidence of lower respiratory disease in children,
5. increase the risk for chronic respiratory disease in adults.

These effects were observed in community studies where levels of sulfur dioxide, acid sulfate aerosols, and suspended particulate matter were usually but not always simultaneously elevated. Accurate measurement of total suspended sulfate was in many cases not available. The data, therefore, is seriously limited by the lack of accurate dose information. Similar problems are involved in the evaluation to the response side of the dose response relationship. The studies often differ in their locale and their methods of ascertainment of illness. Different analytical and statistical approaches have been utilized to evaluate the available data, making it difficult to compare studies performed by different investigators.

Despite these difficulties, and in reconition of them, an attempt has been made at a preliminary quantification of these health effects in the form of dose-response curves[20]. As a crude approximation let us assume that each of the above health effects is characterized by a linear threshold relationship. The approximation is that no adverse health effect results below some threshold level of suspended sulfate concentration. Above this level, the percent of cases of the adverse health effect which may be attributed to the sulfate (the "percent excess" is assumed to increase linearly with concentration. By plotting the results of various epidemiological studies on a graph relating the percent excess of health effects to levels of suspended sulfates, the "best fit" for such a linear-threshold relationship may be obtained. The results of such a curve fitting exercise and the studies upon which they were based are summarized in Table 13-12.

Obviously there is a good deal of arbitrariness in the definition of these curves. Consequently, the analysis that follows is meant more to illustrate method than to accurately quantify health costs. The uncertainty in the magnitude of the health effects from a given increase in ambient sulfate is of the order of a factor of 20, as will be described below.

Health Effects of Suspended Sulfates in the New York Metropolitan Area

Once dose-response curves have been agreed upon, they may be combined with expected frequency distributions of suspended sulfate concentrations to obtain an estimation of the health effects expected from an increase in the current ambient levels of suspended sulfates.

We illustrate the reasoning with an analysis of the health effects of suspended sulfates in the New York Metropolitan area. Total population for this area and the population of various age groups (of interest as populations at risk) are summarized on page 604.

TABLE 13-12

"Best Judgment" Dose-Response Functions.*

ADVERSE HEALTH EFFECT	EXPOSURE DURATION	BEST JUDGMENT THRESHOLD FUNCTION THRESHOLD ($\mu g/m^3$)	SLOPE**
INCREASED DAILY MORTALITY	24 HOURS OR LONGER	25	.252
AGGRAVATION OF HEART AND LUNG DISEASE	24 HOURS OR LONGER	9	1.41
AGGRAVATION OF ASTHMA	24 HOURS OR LONGER	6	3.35
EXCESS LOWER RESPIRATORY DISEASE IN CHILDREN	Up to 10 YEARS	13	7.69
EXCESS RISK FOR CHRONIC RESP. DISEASE IN ADULTS***	Up to 10 YEARS	12	11.1

*These dose response relationships were developed in an unpublished study for the U.S. Environmental Protection Agency. The "best judgment threshold functions" represent subjective approximations to data, not precise mathematical fits. The studies upon which the estimates were based are as follows: Mortality; Lindeberg (1968), Martin and Bradley (1960), Lawther (1963), Glasser and Greenburg (1965), Brasser et al. (1967), Watanabe and Kaneko (1971), Nose and Nose (1970), Buechley et al. (1973). Aggravation of heart and lung disease; Carnow et al. (1970), Goldberg et al. (1974). Aggravation of asthma; French, Sugita et al. (1970), Finklea et al. (1974a), Finklea et al. (1974c). Excess lower respiratory disease in children; Nelson et al. (1974) Finklea et al. (1974b), Douglas and Waller (1966), Lunn et al. (1967), Love et al. (1974), Hammer (1974). Excess chronic respiratory disease; Burn and Pemberton (1974), Goldberg et al. (1974), House et al. (1973), Hayes et al. (1974), Yashizo (1968), House (1974), Galke and House (1974a), Galke and House (1974b).

**Change in percent excess over base rate for population, per $\mu g/m^3$ change in suspended sulfate level.

***For chronic respiratory disease, difficulties with available data necessitated the unit of measurement to be excess risk rather than direct incidence of illness. Actually, in its originally calculated form, separate dose response functions were assessed for cigarette smokers and nonsmokers. The function described in the table is a weighted linear average based upon the average prevalence of cigarette smoking in the adult population at risk.

NEW YORK METROPOLITAN AREA[20]

- Population 11.5 million
  2.12 million children age 0-13
  7.27 million adults
  1.24 million elderly over age 65

Daily concentrations of ambient suspended sulfate concentrations are assumed to follow a lognormal frequency distribution with an annual average of 16 ug/m³ and a standard deviation of 5.6 ug/m³. This assumption is consistent with the New York studies of the CHESS report. The objective of the analysis will be to answer the following two questions:

- What are the health effects of a 1 ug/m³ increase in the annual average level of suspended sulfate concentration?
- Suppose the concentration for the top 1 percent peak days increases by 10 ug/m³.

What are the health effects?

The answers to these questions will tell us something about the sensitivity of the health effects to changes in levels of sulfate concentration. This information, in turn, should help us to evaluate the health impacts of various emission reduction alternatives.

For increased daily mortality, aggravation from heart and lung disease, and asthmatic attacks, which are affected by daily levels of suspended sulfates, the expected effect of 1 ug/m³ increase in sulfate levels may be estimated by calculating the expected number of cases of each health effect under current ambient concentrations and under current ambient levels plus 1 ug/m³ sulfate concentration. More precisely, if $f(x;m,\sigma)$ represents the (normalized) frequency distribution[21] for 24 hour sulfate concentrations and $y(x)$ is the dose-resonse curve, the additional percent of adverse health cases due the unit increase in sulfate concentrations is given by

Additional Percent Cases =

$$\int_x y(x)\,f(x;m+1,\sigma)\,dx - \int_x y(x)\,f(x;m,\sigma)\,dx$$

Lower respiratory disease in children and chronic respiratory disease in adults are more closely related to yearly average levels and so the percent excess cases may be read directly from the annual average dose-response curves. Comparing, category by category, the percent excess cases with the total number of observed cases, estimates of the number of cases attributable to the suspended sulfate concentration are obtained.

The results of such calculations are summarized in Table 13-13. Since annual average concentrations may differ from 16 $\mu g/m^3$, depending upon exact location, the calculations were also performed assuming annual average concentrations of 12.5 and 20 $\mu g/m^3$. The last column of Table 13-13 gives the expected number of additional cases resulting from a 1$\mu g/m^3$ annual average sulfate concentration increase.

The effects of an increase of 10 $\mu g/m^3$ in the top 1 percent days of peak concentration may be obtained by altering the shape of the 24 hour sulfate concentration frequency distribution. Specifically, shifting the upper 1 percent "tail" of the distribution by 10 $\mu g/m^3$ will give us the desired mathematical expression of the effect. The results are given in Table 13-14, the last column of which again gives the additional number of cases to be expected from the concentration increase. Of the five health effects, only premature mortality, heart and lung disease aggravation, and asthmatic attacks are significantly affected by short term elevations of sulfate concentrations. Due to the assumed linearity of the dose-response curves above threshhold levels, these entries are independent of average concentration. However, for high levels of concentrations, non-linearities become increasingly important and may imply significant deviations from these values. Nevertheless, the indication is clear that, with the exception of premature mortality, a 1 $\mu g/m^3$ average increase is considerably more significant in terms of average health effects than is an occasional 10 $\mu g/m^3$ increase on high pollution days. This result is of interest for the comparison of control measures which are directed to annual average concentration levels

TABLE 13-13

Estimates of Adverse Health Effects Attributable to a 1 g m[3] Increase of Average Suspended [illegible] in the New York Metropolitan Area

HEALTH EFFECT	AVERAGE NUMBER OF CASES* MILLION	CASES ATTRIBUTABLE TO SUSPENDED SO_4 CONCENTRATION					
		AT AVERAGE CONCENTRATION			AT AVERAGE CONCENTRATION + 1 g m[3]		
		[illegible]	PERCENT OF CASES	NUMBER OF CASES	PERCENT OF CASES	NUMBER OF CASES	ADDITIONAL EXPECTED CASES
		[illegible]	[illegible]	44.4	[illegible]	[illegible]	[illegible]
[illegible] MORTALITY (premature deaths per yr.)	[illegible] Deaths year	[illegible]	[illegible]	94.2	[illegible]	[illegible]	[illegible]
		[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
AGGRAVATION OF HEART AND LUNG DISEASE (million person-days per yr.)		[illegible]	[illegible]	1.4[illegible]	[illegible]	[illegible]	[illegible]
	24.4	[illegible]	[illegible]	2.41[illegible]	11.5[illegible]	[illegible]	[illegible]
		[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	.431
		[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
ASTHMATIC ATTACKS ([illegible] per year)	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
		[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
LOWER RESPIRATORY DISEASE IN CHILDREN ([illegible] per year)		[illegible]			[illegible]	[illegible]	[illegible]
	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
		[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
CHRONIC RESPIRATORY DISEASE SYMPTOMS ([illegible], point prevalence)		[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
		[illegible]	[illegible]	.3333	102.10	[illegible]	[illegible]

TABLE 13-13 (continued)

*The figures cited under "Average Number of Cases" are estimates based upon data from the Statistical Abstract of the U.S., 1970, and information on health effect prevalence rates. The prevalence rates used are as follows: (1) annual death rate (per 1000); 10.2, (2) prevalence of heart and lung disease; .27, average number of aggravated days per day person; .20, (3) asthma prevalence rate; .03 (Prevalence of Selected Chronic Respiratory Conditions U.S.-1970), attacks per day per asthmatic; .02, (4) annual incidence rate for lower respiratory disease in children; .06 (Acute Conditions-Incidence and Associated Disability U.S. 1971-72), (5) chronic respiratory disease prevalence rate; .02 for nonsmokers (62%) and .10 for smokers (38%) (Prevalence of Selected Chronic Respiratory Conditions 1970, Archives of Environmental Health, Vol. 27, Sept. 1973).

TABLE 13-14

Estimates of Adverse Health Effects of a 10 $\mu g/m^3$ Increase in Suspended Sulfate on the Top 1 percent Peak Concentration Days

HEALTH EFFECT	AVERAGE NUMBER OF CASES*	CASES ATTRIBUTABLE TO SO_4 WITH INCREASE		ADDITIONAL CASES DUE TO INCREASE
		PERCENT OF CASES	NUMBER OF CASES	
INCREASED DAILY MORTALITY (Premature deaths total per year)	.118 million deaths a year	.0483	57.2	7.70
AGGRAVATION FROM HEART AND LUNG DISEASE (Million man-days per year)	24.4	5.865	1.431	.00895
ASTHMATIC ATTACKS (Millions per year)	2.51	22.38	.5617	.000439

*See footnote for Table 13-13.

versus intermittent controls which are designed only to reduce sulfate concentrations on days of extremely high ambient levels.

Estimation of Health Costs

To complete our estimation of the health costs resulting from increases in the ambient levels of suspended sulfates in the New York Metropolitan area we need to assign estimates of the costs to society appropriate to each incidence of an adverse health effect. The nominal dollar values which we have chosen to employ are summarized below.

ESTIMATION OF PER CASE HEALTH COSTS

One Premature Death	=	$30,000
One Day Aggravation From Heart and Lung Disease Symptoms	=	$20
One Asthma Attack	=	$10
One Case Child's Lower Respiratory Disease	=	$75
One Case Chronic Respiratory Disease	=	$250

Ideally, these values should be assigned so as to reflect what society would (or should) just be willing to pay in dollars to prevent the losses sustained when a typical individual suffers one of the possible health effects (see Lave and Seskin 1970). We believe that medical cost and lost productivity should not be the only basis for assessing these values, as was done in their paper). Since there may be considerable disagreement on what the dollar assignments should be, we will need to check the sensitivity of our results to these values.

Although expressing health losses in terms of the dollar is convenient for our purposes, other common units of measurement are possible. One other popular numeraire for estimating health damages is the "equivalent day of restricted activity (EDRA)" (Jacoby et al. 1973: 189). The Public Health Service defines a day of restricted activity as one on which a person substantially reduces the amount of activity normal for that day because of a specific illness or injury (PHS 1971: 53). If we assume a person-day of restricted activity to imply health costs of between $1 and $10 (Jacoby et al. 1973, NAS 1974: volume 4), then our nominal health costs are equivalent to the following assessment of EDRA's:

one day aggravation from heart and lung disease	= 2 - 20 EDRA's
one asthma attack	= 1 - 10 EDRA's
one case child's lower respiratory disease	= 7.5 - 75 EDRA's
one case chronic respiratory disease	= 25 - 250 EDRA's

These ranges compare well with the estimations of Jacoby et al. (1973: Table 8-4). For premature death we have used a value of $30,000. Most of the deaths occur among chronically ill elderly people, and the amount by which their lives are reduced may be only a matter of days or weeks. It is not known whether sulfate levels have any correlation with a general reduction in life expectancy for persons of average age and health. Rather, the effect observed is a statistical increase in number of deaths recorded on days of high pollution levels versus days of low pollution levels. For this reason we have used a value of $30,000 per life rather than the value of $200,000 used in highway safety and other applications for the value of life for a representative individual in the population (NAS 1974). Sensitivity analysis indicates that our results are not dependent on the value assigned to premature death.

Multiplication of per-case health costs times the number of cases given in Table 13-13 yields the societal health costs resulting from various levels of suspended sulfate concentration. Our sample values yield the health costs shown in Table 13-15. Observe that the major contribution to health costs from a unit increase in sulfate level appears to be due to

TABLE 13-15

Estimation of Health Costs for Various Levels of Ambient Suspended Sulfate (Population at Risk = 11.5 million)

Health Effect	Annual Average Ambient Sulfate Concentration, $\mu g/m^3$	Health Effect, (number)	Health Cost (million $)	Additional Change in Sulfate Concentration Increased $1 \mu g/m^3$ effect (number)	cost (million $1)
Excess Mortality (number of deaths) per year	12.5	49	1.5	11	0.33
	16.0	99	3.0	23	0.68
	20.0	227	6.8	51	1.53
Aggravation of Heart-Lung Disease Symptoms (million person-days of aggravation per day)	12.5	1.4	28.4	0.26	5.2
	16.0	2.4	48.3	0.41	8.1
	20.0	3.7	73.9	0.43	8.6
Asthmatic Attacks (thousands of attacks) per year	12.5	560	5.6	80	0.81
	16.0	840	8.4	84	0.84
	20.0	1180	11.8	84	0.84
Lower Respiratory Disease in Children (thousands of cases per year)	12.5	0	0	4.9	0.36
	16.0	29	2.2	9.9	0.74
	20.0	69	5.1	9.9	0.74
Chronic Respiratory Disease (thousands of cases, point prevalence)	12.5	28.4	7.1	40.6	10.2
	16.0	171	42.6	40.6	10.2
	20.0	333	83.3	40.6	10.2
	12.5		42.6		16.9
Total Cost	16.0		104.5		20.5
	20.0		180.9		21.9

the expected increase of 40,600 cases of added risk for chronic respiratory disease, valued at \$250 per case, and 262 thousand additional person-days of aggravated heart lung disease symptoms, valued at \$20 each.

Sensitivity of Health Costs

We now investigate the sensitivity of our estimate of the health costs of a 1μg/m^3 increase in average sulfate concentration to various assumptions used in the analysis. The calculations are in principle identical to those performed in the section on the relationship of sulfur dioxide emissions to ambient sulfate levels. Rather than fixing each quantity in the analysis at a nominal value, we vary that quantity over a range of possible values and observe the effect on the output value, which in this case is total health cost. The results of the analysis appear in Table 13-16.

The sensitivity values shown in Table 13-16 have been chosen somewhat arbitrarily as representing a set of reasonable extreme values. The low and high values for the change in incidence of each health effect, per unit change of ambient sulfate, were taken to be 10 percent and 200 percent of the nominal values respectively. This judgment was communicated to us by representatives from the Assembly of Life Sciences Committee that authored for Part one of this report.

We see from the sensitivity results that health costs of a 1μg/m^3 increase in ambient sulfate are most sensitive to (in order of magnitude) the estimated health cost per case of chronic respiratory disease, the health cost per day of aggravation from heart and lung disease symptoms, the number of additional cases of chronic respiratory disease, and the additional number of days of aggravation from heart and lung disease symptoms.

TABLE 13-16

Sensitivity Analysis: Total Health Cost of a 1 $\mu g/m^3$ Increase in Average Suspended Sulfate Concentration in the New York Metropolitan Area

	Total Health Cost per $\mu g/m^3$ SO_4 (million $)
Nominal values	20.5
Additional premature deaths	
low: 2.3	19.9
high: 46	21.2
Additional days of aggravated heart and lung disease	
low: 0.041 mil./yr.	13.2
high: 0.82 mil./yr.	28.6
Additional asthmatic attacks	
low: 8.4 thous./yr.	19.7
high: 168 thous./yr.	21.3
Additional cases lower respiratory disease in children	
low: .99 thous./yr.	19.8
high: 19.8 thous./yr.	21.2
Additional cases chronic respiratory disease	
low: 4.06 thous./yr.	11.3
high: 81.2 thous./yr.	30.7
Cost per premature death	
low: $3,000	19.9
high: $120,000	22.5
Cost per day aggravation from heart and lung disease	
low: $2[illegible]	13.2
high: $80	44.8
Cost per asthma attack	
low: $1	19.7
high: $40	23.0
Cost per case child's lower respiratory disease	
low: $7.[illegible]	19.8
high: $300	22.7
Cost per case chronic respiratory disease	
low: $25	11.3
high: $10[illegible]	51.1

Uncertainty in Assessed Health Costs

The indication from the sensitivity analysis is that the most critical quantities in the assessment of the total health cost of a 1 ug/m^3 increase in suspended sulfate concentration are the number of additional cases of chronic respiratory disease, and the number of additional person-days of aggravated heart-lung disease symptoms[22]. As a rough approximation we shall neglect all uncertainties in health costs but those arising from these two most critical variables. To proceed with a quantitative description of the uncertainty we shall assume that the range of uncertainty in the dose-response relationship (of 10 percent to 200 percent of the values given in Table 13-12) can be treated as having a probability of 90 percent. The uncertainties in the number of additional cases of chronic respiratory disease and the number of additional days of aggravation from heart and lung disease symptoms, are judged to be such that there is only once chance in ten that each of these quantities lies outside the interval defined by the low and high values used in the sensitivity analysis. Specifically, we assume that there is only a 5 percent chance that the additional number of cases of chronic respiratory disease from a 1 ug/m^3 increase in SO_4 is less than 4060, and a 5 percent chance that this number is greater than 81,200.[23] Further, these uncertainties are assumed to be totally dependent (e.g. if the additional number of cases of chronic respiratory disease is high then the number of additional days aggravation from heart and lung disease will also be high, and visa versa). Similarly, we assume that there is a 5 percent chance that the number of additional days of aggravation from heart and lung disease is less than 41,000, and a 5 percent chance that this number exceeds 820,000. Finally, in order to facilitate the calculations, we shall assume the probability distribution characterizing the resulting uncertainty in the total health cost is a lognormal distribution. The shape of this distribution makes it a good approximation, however, greater accuracy could be obtained by formally assessing the subjective distributions of experts in the area of health effects. Procedures for such probability assessments do exist (Spetzler and Stael von Holstein 1972).

Using the nominal values of $250 per case of chronic respiratory disease and $20 per person day of aggravated heart-lung disease symptoms and the uncertainty on the dose-response relationship for these two health effects as characterized above, we can compute a probability distribution on the total health cost for a 1 ug/m³ increase in average annual ambient sulfate concentration. As has been the case previously, the calculation is done for an average ambient sulfate level of 16 ug/m³ and a population at risk of 11.5 million persons. This probability distribution shows a 5 percent chance that the total health cost will lie above $39 million and 5 percent chance that the total health cost will lie below $4 million, with a 90 percent chance that the total health cost will lie between these values. A lognormal distribution with these properties has an arithmetic mean of $16 million and a standard deviation of $12.3 million[24]. This broad distribution characterizes the great uncertainty on the magnitude of the health effects caused by suspended sulfate. The distribution is plotted as Figure 13-14.

SULFUR OXIDE DAMAGE TO MATERIALS

A number of studies have indicated that sulfur oxide air pollution accelerates the degradation of many materials (Waddell 1974). Spence and Haynie conclude that sulfur dioxide plays an important role in the chemical deterioration of modern day exterior paints. Based upon a corrosion study of metal structures, Fink et al. (1971) estimate that the accelerated corrosion of zinc by sulfur oxides accounts for most of the damage to metals. They estimate that more than 90 percent of the national economic costs of air pollution corrosion are due to the effect of sulfur oxides on zinc. As part of an economic evaluation of the damages of air pollution, based primarily on a review of

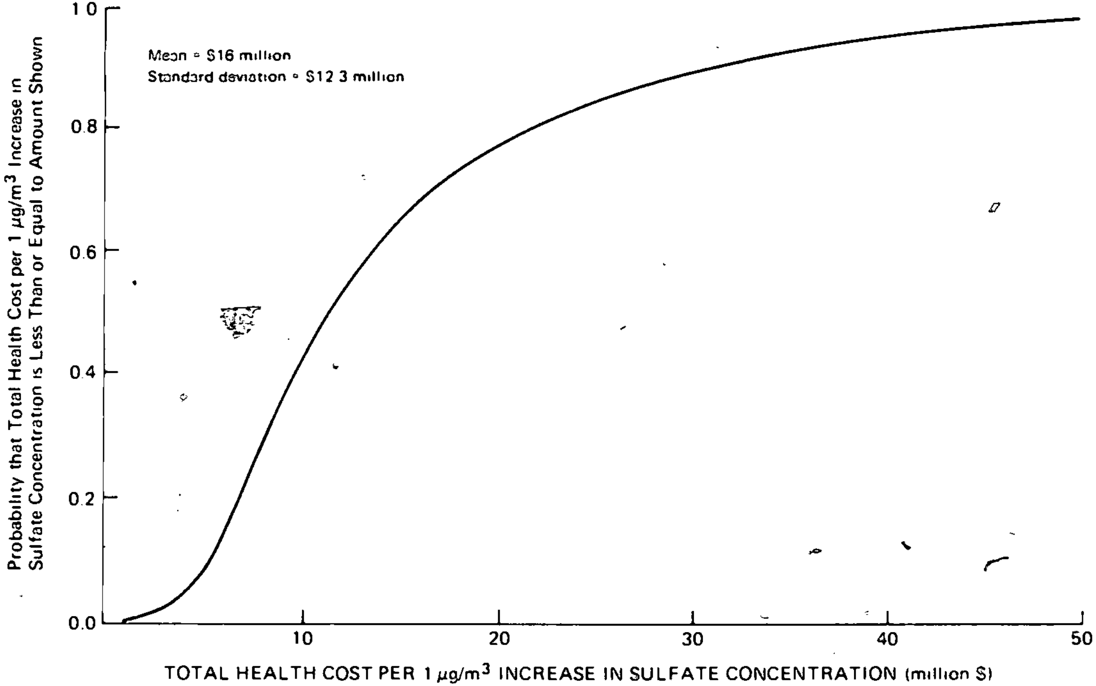

FIGURE 13-14: Probability Distribution Illustrative of Present Uncertainty on Total Health Cost due to a 1μg/m^3 Increase (from an Ambient Level of 16μg/m^3) in Annual Average Suspended Sulfate Concentration for the New York Metropolitan Area. Population at Risk: 11.5 million

the literature, Waddell (1974) assesses the national SOx-related material damage costs in 1970 to lie between \$.4 billion and \$.8 billion. These figures are arrived at by summing past estimates of damages to specific material categories. Consequently, there may be some double counting, and it is possible that some significant categories of materials have not been taken into account.

A Simple Materials Damage Model

Although the total national sulfur oxide pollution damage is a useful number, it does not help us much if we are trying to judge between various pollution reduction strategies. What we really need to know is the sensitivity of pollution damage to pollution levels. Therefore, our objective in this section will be to estimate the reduction in pollution damage which will occur for a given reduction in ambient sulfur oxide concentration.

Unfortunately, little is known about pollution material damage dose-response relationships. In fact, the mechanism by which pollution contributes to the degradation of concrete and metal building materials, paints, and fibers, which account for most of the estimated damage, is not entirely clear. Rates of deterioration appear to be functions of sulfate accumulation implying that there is no ambient sulfate level below which no effect will occur [25]. The relative significance for damage of the various atmospheric sulfur compounds are not known.

We shall summarize our available information into a crude dose-response model. Assume that most material damage occurs in Northeastern urban areas and let us take as the average Northeastern urban concentrations 45 $\mu g/m^3$ sulfur dioxide (Gillette 1974) and 16 $\mu g/m^3$ suspended sulfate (see Appendix 13-C). The model consists of a linear approximation to material costs about these concentrations. Let f be the fraction of the estimated materials damage cost which is due to exposure to sulfur dioxide and let the remaining fraction 1-f, be the result of exposure to suspended sulfates. A doubling of sulfur oxide pollution levels is assumed to roughly double costs (see Appendix 13-C). The net change in urban material damage costs, denoted MD, for a given change k1 in average sulfur dioxide concentration and a given change k2 in average sulfate concentration, is then given by

$$MD = \left[f \frac{k_1}{45 \mu g/m^3} + (1-f) \frac{k_2}{16 \mu g/m^3} \right] X \text{ Total SOx Material Cost for the area.}$$

Following the method of analysis in the section preceding on health costs, we wish to estimate the effect of a 1 $\mu g/m^3$ increase in ambient sulfate concentration will have on material damage costs. In the absence of more detailed information we shall proceed as follows. We begin with an estimate of $600 million for annual damage caused by sulfur oxides to material property, which applies to the nation as a whole (Waddell 1974: a high value of $800 million and a low value of $400 million are given in addition to a "best" value of $600 million). We shall assume that 90 percent of this damage occurs in the northeastern United States, 60 percent in the New England states, New York, Pennsylvania, Maryland, New Jersey, Delaware, and Greater Washington, D.C. and 30 percent elsewhere east of the Mississippi river. In the above mentioned states we shall assume that the materials damage from sulfur oxides for an area is proportional to its population; most of the population in these states is in urban areas and sulfur oxide damage to materials such as paints and metals will occur primarily in urban areas. We assume as earlier that the population at risk for the remotely located plant (assumed in the Pennsylvania/Ohio border-West Virginia area) is 50 million, and that total sulfur oxide materials damage in area affected by this plant (the states mentioned above) is 60 percent of $600 million or $360 million dollars per year. For the urban representative plant, the population at risk is assumed to be 11.5 million, so the total sulfur oxide materials damage is assumed to be (11.5/50) x 60 percent x 600 = $83 million per year.

The calculated values for the change in materials damage per 1ug/m³ change in ambient sulfur dioxide and sulfate levels for the representative case analysis are given in Table 13-17. In the absence of better information we assume that fifty percent of the damage is caused by sulfur dioxide and 50 percent by sulfate. The numbers of Table 13-17 must be regarded as extremely crude estimates.

A review of the information on material property damage by a member of the Committee suggests that Waddell's estimate for material property damage caused by sulfur oxides might be low by a factor of four or five (see Appendix 13-E). The implication of this increase in material property damage will be addressed in the concluding section.

AESTHETIC COSTS

Damages to the aesthetic quality of the environment by air pollution are a major public concern. Aesthetic costs include the reduction of long distance visibility and light transmittal of the atmosphere, noxious odors, soiling, and the deterioration of materials of historic or artistic significance.

Public perception seems to be that the magnitude of these costs are substantial. A study by the New Mexico State University Agricultural Experiment Station recently assessed the aesthetic environmental damage of the Four Corners Power Plant at roughly $25 million per year in 1970. Respondents to questionnaires used in this study considered reduced visibility to be the most serious aesthetic effect (Randall et al. 1974).

The visibility-restricting air pollutants are particulates and nitrogen dioxide. The extent of visibility restriction caused by suspended particulate depends upon the chemical composition of the particle, its size, shape, and concentration. It is likely that sulfates may significantly contribute to visibility

TABLE 13-17

Materials Damage Increment from a 1 μg/m^3 Change in Ambient SOx Levels

Fraction of Damage Caused by	Change in Materials Damage for North Central and New England States* per 1 μg/m^3 change in		Change in Materials Damage for New York Metropolitan Area** per 1 μg/m^3 change in	
SO_2	SO_2	Sulfate	SO_2	Sulfate
	(million $)	(million $)	(million $)	(million $)
1.0	6.0	0	1.4	0
0.5	3.0	11.3	0.7	2.6
0	0	22.5	0	5.2

*Maine, Vermont, New Hampshire, Rhode Island, Massachusetts, Connecticut, New York, Pennsylvania, New Jersey, Delaware, Maryland, Washington, D.C. Population at risk approximately 50 million.

**Population at risk 11.5 million.

reduction by increasing the particulate loading (see Chapter 5). Oxides of sulfur are also thought to be responsible for the accelerated decay of art and statuary (Waddell 1974).

One method that has been used to estimate aesthetic costs of air pollution is by market studies of real estate values (see Waddell 1974). Most investigators agree that costs associated with organoleptic effects (effects pertaining to sight and smell) as well as soiling-caused cleaning and maintenance expenditures are capitalized in real estate market values (Waddell 1974:27). By applying classical least squares regression analysis, the existence of a relationship between air pollution and property values can be tested. In most applications a statistically significant inverse relationship between air pollution and property values (or rents) has been identified.

There are at least two major criticisms of the property value approach. First, for the method to have validity the housing market must adequately internalize aesthetic pollution costs into its pricing structure. It may be argued that this is not the case. Second, all significant multi-collinearities between the pollution variables and other explanatory variables for property values must be removed. Since many unfavorable neighborhood attributes will likely be correlated with air pollution (e.g., automobile congestion) there is considerable danger of overstating the pollution coefficients.

Waddell gives an estimate of $5.8 billion as the annual national cost of aesthetic and soiling effects, of which 50 percent is allocated to sulfur oxides and 50 percent is allocated to particulates (Waddell [1974], Table 13-21). The allocation seems misplaced if visibility and odors are the major factors. Photochemical smog and particulates may account for much of the local visibility effects and local concentrations of industrial emissions are responsible for most of the noxious odors. Sulfur dioxide is not detectable by its odor in concentrations below 80 $\mu g/m^3$.

Suspended sulfates may play a significant role in reducing visibility on a regional scale (see Chapter 5). These visibility effects due to sulfate deserve further investigation. Sulfate particles are concentrated in the submicron size range that causes the largest visibility reduction affects. Other fine particulates may also be important. Electrostatic precipitators remove only about half of the submicron particles, and flue gas desulfurization should remove a substantially larger proportion. The effect on aesthetics of sulfur oxide emissions from power plants may be most pronounced in rural areas with high recreational or aesthetic values, such as national parks. If most particulates are removed by efficient electrostatic precipitators, it is not likely that appreciable sulfate oxidation will occur until the plume is dispersed, so the reduction in visibility may be small in the neighborhood of the plant.

The aesthetic effects of sulfur oxide emissions may be substantial for some particular power plants. It is difficult to assess values to the aesthetic effects on a regional basis. We will retain an aesthetics cost term, and for a value we will arbitrarily take 20 percent of Waddell's 5.8 billion per year figure for aesthetic damages, $1.16 billion as being due to SO_4, and prorate this over 33.9 million tons of sulfur dioxide, or 33.9 x 10^9 pounds of sulfur, emitted per year. The resulting value of 3.4 cents per pound of sulfur emissions is too low to be of major significance compared to health effects, but we shall keep this in the analysis for sensitivity since in a specific instance it might be considerably larger.

ACID RAIN: EFFECTS ON SOILS, FORESTS, AND FISHERIES

Acid rain resulting from sulfur oxide and nitrogen oxide emissions is now widespread over Northern Europe and the northeastern United States. (See further discussion in Chapter 7.) The consequences may include acidification of soils, loss of growth in forests, death of fish from low pH in lakes and streams, and damage to outdoor statues and building facades. These effects are difficult to assess in economic terms, but they do not seem significant compared to the potential health effects discussed above. In Chapter 7 the first three effects are discussed, and it it is roughly estimated that each might involve incremental costs of the order of 1 cent per pound of sulfur emitted with the sulfur emissions levels that might be achieved by 1980 in the absence of control. However, not all of the costs would be felt immediately, and direct evidence for the extent of the damage is limited.

We will therefore choose arbitrarily a value for the overall costs associated with acid rain of $500 million annually, which is equivalent to about 1.5 cents per pound of sulfur emitted (with 1970 emission levels). As with aesthetic effects this value will be too low to be of significance compared to the potential health effects. Sensitivity analysis will indicate that the costs ascribed to aesthetics and acid rain have little effect on the overall result for the total pollution cost per pound of sulfur emitted.

EVALUATION OF SULFUR OXIDE POLLUTION COSTS FOR THE REPRESENTATIVE CASES

We shall now summarize our assessment of the costs associated with sulfur oxide emissions from the types of plants we have taken as representative, and compare these to generating costs and emission quantities to identify the best strategy. It is worth reiterating that the calculations rest on very limited information, and many assumptions have been made rather arbitrarily in order to carry these calculations out. The purpose of the analysis is not to achieve numerical answers, but by using a quantitative framework to achieve and understanding of the complex issues involved. An analysis of this kind does not resolve uncertainty; it may be useful in determining the importance of uncertainties in the context of a decision. As we shall see, the uncertainty on the pollution cost to be attributed to a pound of sulfur emission is large, and it would be worth a great deal to resolve this uncertainty in the context of decisions among the alternative strategies for sulfur oxide emissions control on power plants.

Remotely Located Plant

We have assumed for this case that the population at risk of 50 million, roughly that of the North Central and New England states. The plant is assumed to be located in the Western Pennsylvania-Ohio-West Virginia area, about 500 kilometers upwind of the urban concentrations in the Northeast corridor. We have assumed an oxidation rate of 0.5 percent per hour for the 24 hours when the plume is traveling through rural air, then 5 percent per hour for two hours after the plume has encountered a pollutant-laden urban air mass. Assuming that over a year the emissions are dispersed over a 45° sector and 25 percent is removed by rain before reaching the urban area, we compute an increase in average annual ambient concentrations of 0.145 $\mu g/m^3$ of sulfate and 0.35 $\mu g/m^3$ of sulfur dioxide, as resulting from 10^4 kilograms per hour of SOx emissions. This level of emissions equals 96.5 million pounds of sulfur emitted per year. It corresponds to the emissions from 620 MW of generating capacity with an efficiency of 33 percent burning 3 percent sulfur coal at a load factor of 6000 hours per year (representative existing plant), or 612 MW of generating capacity with an efficiency of 38 percent burning 3 percent sulfur coal at a load factor of 7000 hours per year (representative new plant).

The nominal value for the pollution cost described to these emissions is computed in Table 13-18. The total emission cost is seen to be $20.3 million, or $0.21 per pound of sulfur emitted. We can place these costs on a mills per kilowatt hour basis by dividing them by the numbers of kilowatt hours produced annually. The existing plant produces 3.72 x 10^9 kwh per year, so the pollution cost computes out to about 5.5 mills/kwh. The more efficient new

TABLE 13-18

Cost of Sulfur Oxide Emissions Representative Calculation for Remote Plant Emitting 10,000 kg of Sox Per Hour (96.5×10^6 Pounds of Sulfur Per Year)

Costs computed based on $0.145 \mu g/m^3$ ambient increase in sulfate and $0.35 \mu g/m^3$ ambient increase in sulfur dioxide in metropolitan areas with a population of 50 million

Health effects (computed at ambient level of $16 \mu g/m^3$)	
25,600 cases of chronic respiratory disease x $250 per case	$ 6.4 million
256,000 person-days of aggravated heart-lung disease symptoms x $20	5.1
53,000 asthma attacks x $10 each	0.5
6,200 cases of children's respiratory disease x $75	.5
14 premature deaths x $30,000	.4
Total Health Costs	12.9
Materials damage	
$11.3 million per $\mu g/m^3$ of SO_4 x 0.145	1.6
$3.0 million per $\mu g/m^3$ of SO_2 x 0.35	1.1
Aesthetics ($0.034 x 96.5×10^6 lbs)	3.3
Acid Rain ($0.015 x 96.5×10^6 lbs)	1.4
Total Emissions Costs	$20.3 million
Emissions Cost Per Pound of Sulfur	21¢

plant produces 4.28 x 10^9 kwh in producing the same amount of sulfur emission, so the pollution cost computes out to 4.7 mills/kwh. The reader may wish to verify on Figures 13-3 and 13-4 this correspondence: Adding a pollution cost of 21 cent per pound of sulfur emitted raises the total social cost of generating electricity in an existing plant burning high sulfur coal from 17.2 mills per kilowatt hour to 22.7 mills per kilowatt hour, and similarly from 21.6 mills per kilowatt hour to 27.4 mills per kilowatt hour for a new plant. For both cases we see that a better alternative is to burn low sulfur eastern coal. The reduction in sulfur pollution costs achieved by switching to a low sulfur coal slightly more than offset the higher fuel price for this fuel. The increase needed before flue gas desulfurization becomes the best alternative is the order of a factor of two: to 53 cents per pound of sulfur for an existing rural plant, to 59 cents per pound for an existing urban plant, and to 37 cents per pound for a new plant.

The pollution cost is very sensitive to many of the input values assumed for the analysis. A list of sensitivity calculations is given in Table 13-19. The two most important health effects are chronic respiratory disease and aggravation of symptoms for persons suffering from chronic heart or lung disease conditions. Both the costs ascribed to these health effects and the dose-response relation for increases in ambient sulfate concentration may cause changes of the order of a factor of two on the pollution cost value. Oxidation rates are also very important: For example, a constant oxidation rate of 2 percent for both rural and urban air causes the pollution cost to rise to 39.4 cents, nearly a 100 percent increase.

Examination of the sensitivity analysis indicates that changes in input values such as oxidation rate, health costs, and the dose response relation for health effects could lead to a pollution cost above 37 cents, at which point flue gas desulfurization becomes the preferred alternative for a new plant, and possibly even above 53 cents, the value at which flue gas desulfurization becomes the preferred

TABLE 13-19

Selected Sensitivity Analysis Pollution Cost Per Pound of Emitted Sulfur for Representative Remote Plant

Nominal value	21¢
Cost of chronic respiratory disease increased to \$1000/case (nominal value = \$250/case)	40.9¢
Cost per person-day of aggravated heart-lung disease symptoms increased to \$80/day (nominal value = \$20/day)	36.9¢
Cost per premature death increased to \$200,000 (nominal value = \$30,000)	23.5¢
All health costs increased by a factor of four	61.1¢
All health costs decreased by a factor of ten	9.0¢
Incidence of chronic respiratory disease per $\mu g/m^3$ of SO_4 increased by a factor of two	27.7¢
Incidence of chronic respiratory disease per $\mu g/m^3$ of SO_4 decreased by a factor of ten	15.1¢
Dose response for aggravated heart-lung disease symptoms increased by a factor of two	26.3¢
Dose response for all health effects increased by a factor of two	34.4¢
Dose response for all health effects decreased by a factor of ten	9.0¢
Materials damage increased by a factor of four	29.4¢
Materials damage decreased by a factor of four	18.9¢

TABLE 13-19 (continued)

Aesthetics costs increased by a factor of four	30.9¢
Acid Rain costs increased by a factor of four	25.5¢
Oxidation Rates assumed doubled to 1% per hour in rural air and 10% per hour in urban air	33.2¢
Low Oxidation rates 0.1% per hour in rural air and 1.0% per hour in urban air	9.5¢
Oxidation rate constant at 2% per hour	39.4¢

alternative for a retrofitted (rural) plant in our representative plant calculations. The uncertainty is such that although low sulfur eastern coal looks like the best alternative for the representative remote plant, neither burning high sulfur coal nor flue gas desulfurization can be ruled out. With better information to resolve the uncertainty, either could become the preferred alternative.

Urban Plant

The population at risk here is assumed to be 11.5 million, the population of the greater New York City area. The plant is assumed to be located about sixty kilometers upwind of the city, and an oxidation rate of 5.0 percent per hour has been assumed for sulfur dioxide in this metropolitan area. The increase in average annual ambient sulfate levels is computed on the assumption that the emissions are dispersed over a 45° sector and that 10 percent of the sulfur is removed by rain before impacting on the population or values at risk. As for the remotely located plant, the level of emissions assumed is 96.5 million pounds of sulfur per year, or an average of 10^4 kilograms of sulfur dioxide per hour.

The nominal value for the pollution cost ascribed to these emissions is computed in Table 13-20. The total annual emission cost is $53.1 million, or $0.55 per pound of sulfur emitted.

Looking at the total social cost calculations for Figure 13-5, we see that this value lies well above the crossover point of 19 cents at which low sulfur coal is preferred to the alternative of burning high sulfur coal, and well below the crossover point of $1.00 per pound of sulfur where flue gas desulfurization becomes the least cost alternative.

Another point to be mentioned here is that reduction of other particulate emissions accomplished by flue gas desulfurization has not been included in this analysis. Reduction in emissions of other toxic particulates, such as trace metals, may be an important social benefit for the flue gas desulfurization alternative as

TABLE 13-20

Cost of Sulfur Oxide Emissions Representative Calculation for Urban Plant Emitting 10,000 kg of SOx per hour (96.5 x 10^6 pounds of sulfur per year)

Costs computed based on 1.86 $\mu g/m^3$ increase in sulfate and 7.5 $\mu g/m^3$ increase in sulfur dioxide concentrations in metropolitan areas with population of 11.5 million.

Health effects from increase: (computed at ambient level of 16 $\mu g/m^3$)

75,500 cases of chronic respiratory disease x $250 per case	$18.9 million
755,000 person days of aggravated heart-lung disease symptom x $20 per day	15.1
156,000 asthma attacks at $10 each	1.6
18,400 cases of children's lower respiratory disease at $75	1.4
42 premature deaths at $30,000	1.3
Total Health Costs	$38.3 million
Materials damage	
Materials damage	
2.6 million per $\mu g/m^3$ of SO_4 x 1.86	4.8
0.7 million per $\mu g/m^3$ of SO_2 x 7.5	5.3
Aesthetics ($0.034 x 96.5 x 10^6 lbs)	3.3
Acid Rain etc. ($0.015 x 96.5 x 10^6 lbs)	1.4
Total Emissions Costs	$53.1 million
Emissions Costs per Pound of Sulfur	55¢

TABLE 13-21

Selected Sensitivity Analysis Pollution Cost per Pound of Emitted Sulfur for Representative Urban Plant

Nominal value	55¢
Cost of chronic respiratory disease increased to $1000/case (nominal value = $250/case)	113.8¢
Cost per person-day of aggravated heart-lung disease symptoms increased to $80/day (nominal value = $20/day)	102.0¢
Cost per premature death increased to $200,000 (nominal = $30,000)	62.4¢
All health costs increased by a factor of four	174¢
All health costs decreased by a factor of ten	19.3¢
Incidence of chronic respiratory disease per $\mu g/m^3$ of SO_4 increased by a factor of two	74.6¢
Incidence of chronic respiratory disease per $\mu g/m^3$ of SO_4 decreased by a factor of ten	37.4¢
Dose response for aggravated heart-lung disease symptoms increased by a factor of two	70.7¢
Dose response for all health effects increased by a factor of two	94.7¢
Dose response for all health effects decreased by a factor of ten	19.3¢
Materials damage increased by a factor of four	86.4¢
Materials damage decreased by a factor of four	47.2¢

TABLE 13-21 (cont.)

Aesthetics costs increased by a factor of four	64.9¢
Acid rain costs increased by a factor of four	59.4¢
Oxidation rate assumed doubled (10%/hour)	93.1¢
Oxidation rate decreased to one fifth nominal value (e.g., 1%/hour)	20.6¢
Oxidation rate assumed 2%	29.5¢

opposed to the use of low sulfur eastern coal. This consideration ought to be included in making the decision on emissions control alternatives for a particular power plant, especially one that is located in a metropolitan area.

As in the case of the rural plant we have carried out sensitivity analysis to indicate how the pollution cost per pound of sulfur would change if different inputs were assumed for the calculation. The results, shown in Table 13-21, show that the pollution cost per pound of sulfur emitted could differ by a factor of two or more if different assumptions were used for the cost attributed to the most significant health effects (chronic respiratory disease and aggravation of heart-lung disease symptoms), the increased prevalence of these effects per unit increase in average ambient sulfate concentration, and the oxidation rate for sulfur dioxide to form sulfate.

UNCERTAINTY ON POLLUTION COSTS AND THE VALUE OF RESOLVING THIS UNCERTAINTY

In previous sections we have used sensitivity values as a basis for sketching probability distributions to represent the uncertainty on the ambient increase in urban sulfate resulting from the emissions of a single power plant (Figures 13-9 and 13-10) and the health effects of a given ambient sulfate increase on incidence of chronic respiratory disease and aggravation of heart-lung disease symptoms (Figure 13-14). The product of these two uncertain factors is the dominant term in establishing the pollution cost per pound of sulfur emitted; other terms such as materials damage, aesthetics, and effects caused by acid rain are judged to be small by comparison. Thus, the probability distribution on the product provides a first cut assessment of the overall uncertainty on pollution cost.

An overall probability distribution on pollution cost per pound of sulfur is given in Figure 13-15 for the representative remote

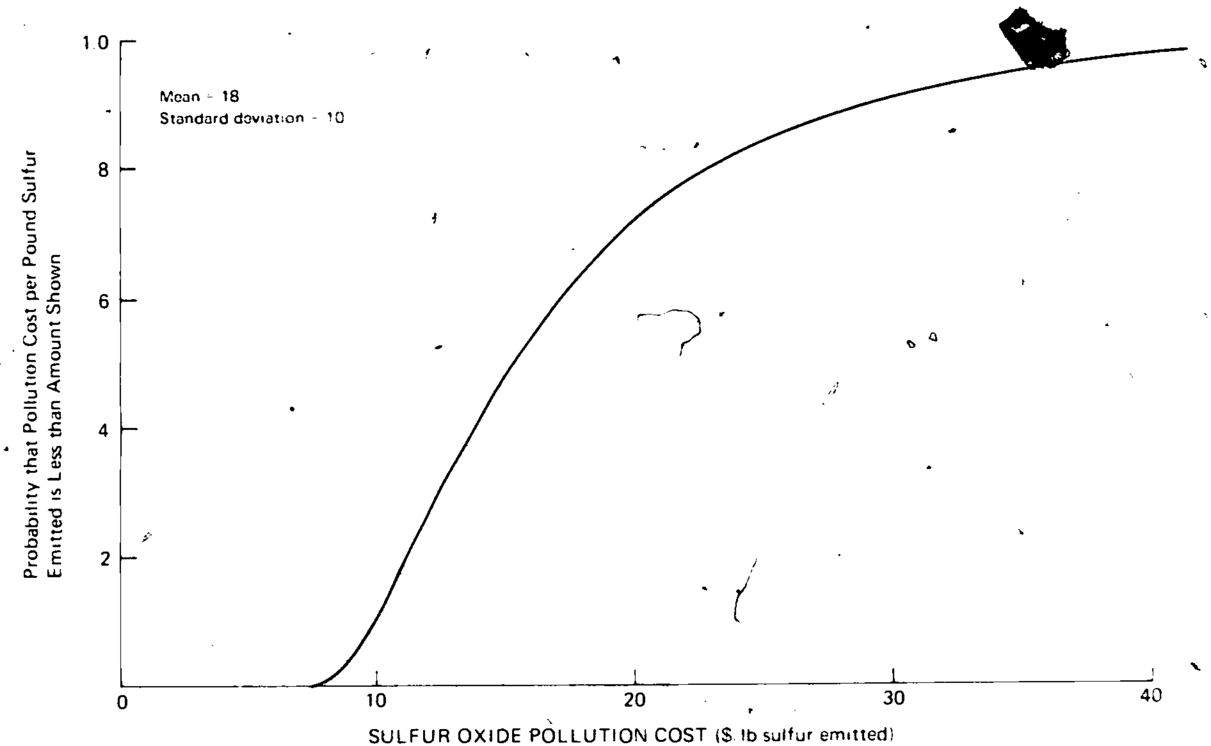

FIGURE 13-15: Probability Distribution Illustrative of Present Uncertainty on Pollution Cost per Pound of Sulfur Emitted from Rural Power Plant

plant, and Figure 13-16 for the representative urban plant.

These curves should be regarded as sketches indicating the present state of information. They are computed from the probability distribution on the emissions to ambient relationship (Figures 13-9 and 13-10) and the probability distribution on health effects (Figure 13-14), by assuming that each of these distributions could be approximated as lognormal. Since the product of lognormal distributions is lognormal, the the distributions shown in Figures 13-15 and 13-16 are of the lognormal family. It should be noted that only uncertainties in the emissions to ambient relation and the magnitude of the health effects listed in Table 13-12 are included in these distributions. Other health effects, property damage, visibility reduction, and possible climatic changes all would introduce additional uncertainty that would serve to broaden these probability distributions.

It might be noted also that values to health effects (e.g., $250 per case of chronic respiratory disease) have not been treated as uncertain. Changes in these values would change the width of the probability distributions. For example, if health value were increased by a factor of four, the probability distribution in Figure 13-15 would spread from about 8 cents to about $1.36 instead of 40 cents.

Let us recall the pollution costs corresponding to the cross-over points between the alternative strategies which are shown in Figures 13-3, 13-4, and 13-5. For the remote rural plant the probability that the pollution cost would be below 19 cents, were the uncertainties in the health effects and emission to ambient relationship resolved, is about 65 percent. The probability that the cost would be above 37 cents is 5 percent and above 53 cents, of the order of one percent. We can similarly read from Figure 13-16 the corresponding values for the probability that the pollution cost will be above 59 cents, the crossover pont for flue gas desulfurization, case where sludge could not be ponded sludge disposal about 19 percent. It

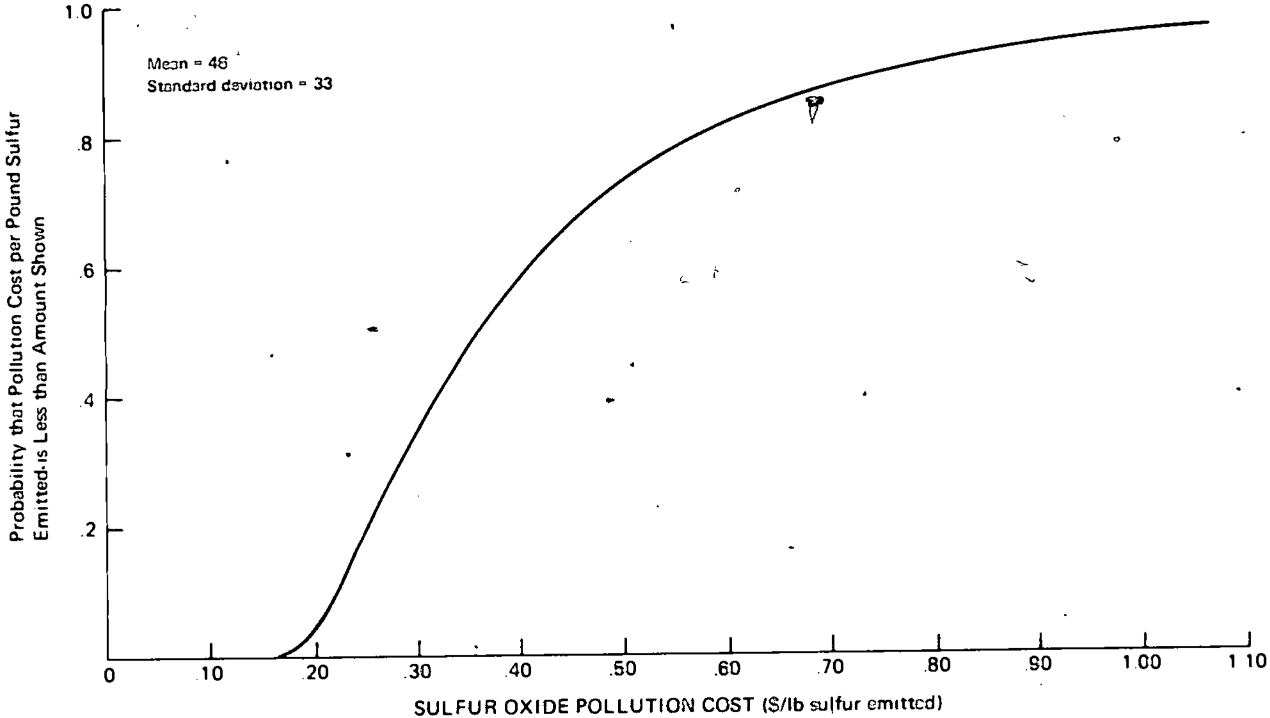

FIGURE 13-16: Probability Distribution Illustrative of Present Uncertainty on Pollution Cost per Pound of Sulfur Emitted from Urban Power Plant

should be recalled that these probabilities are very rough assessments.

The Figures 13-17, 13-18, and 13-19 are copies of Figures 13-3, 13-4, and 13-5, showing the total social cost per kilowatt hour for the representative plant as a function of the cost per pound of sulfur emitted. We show the probability distribution on pollution cost plotted as a probability density function. The height of the curve is the likelihood of various values of the pollution cost. The area under the curve between any two values for the pollution cost corresponds to the probability that the pollution cost would lie in that interval were the uncertainty to be resolved.

The Value of Resolving Uncertainty

Perhaps the most important consequence of the decision analysis formulation is that we can now place a value on what it would be worth to resolve the uncertainty. The value of resolving uncertainty is the difference between the value of the decision situation where information will be made available to resolve the uncertainty before the decision is made, and the value of the decision situation where the decision must be made in the face of uncertainty. (The concept of the value of information is a basic and rather subtle idea in modern decision theory. See for example Howard (1966,1968), North (1968), Raiffa (1961, 1969), Tribus (1969) for a detailed explanation of the concept and the computation). To illustrate this concept, let us pose a hypothetical question. Suppose a clairvoyant were available who knew the emissions to ambient relationship, and the health effects of sulfate. From his information we would be able to compute the pollution cost of sulfur and resolve the uncertainty shown in Figures 13-15 and 13-16. In the context of the decision between alternative strategies, how much would we pay him for this information? He could help us avoid expensive mistakes. For example, if we had planned to use low sulfur Eastern coal in an existing rural plant but he then told us the pollution cost was only 10 cent

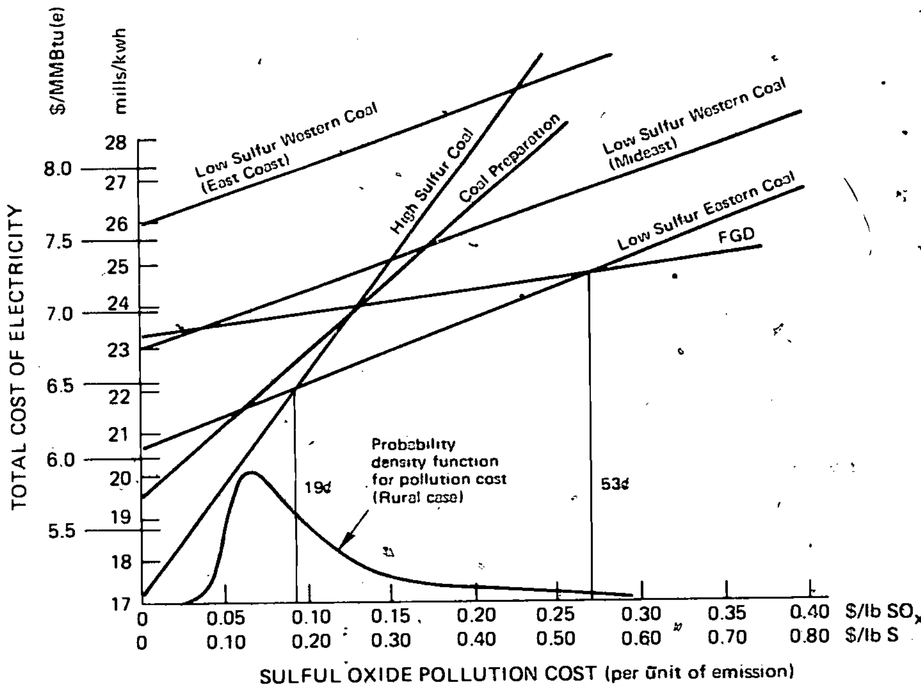

FIGURE 13-17: Total Social Cost Versus Pollution Cost, Existing Coal Fired Plant (Nonurban), Showing Probability Distribution on Pollution Cost per Pound of Sulfur.

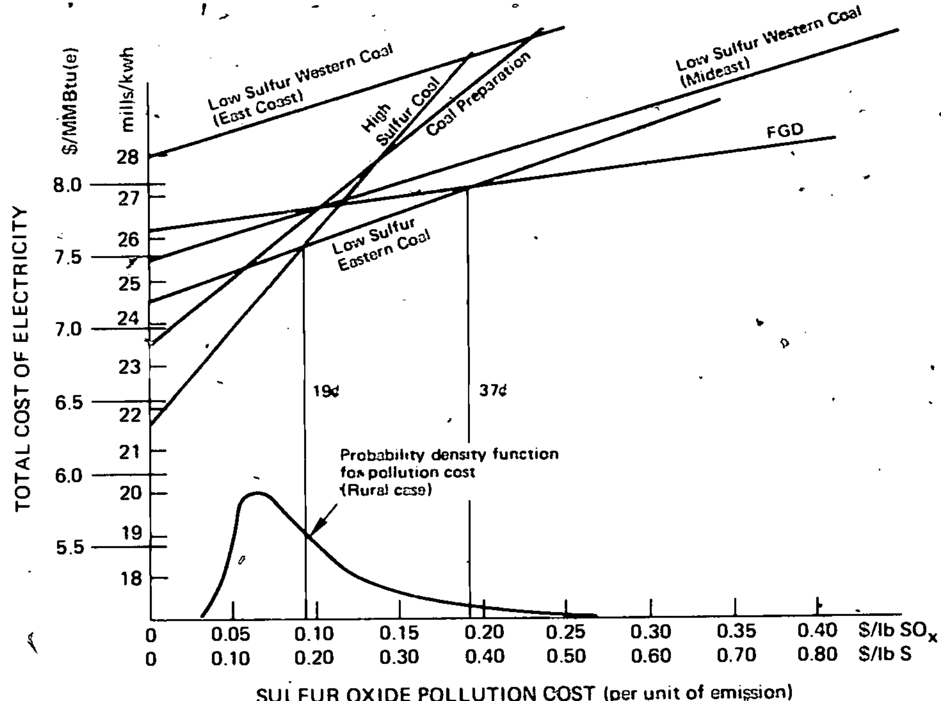

FIGURE 13-18: Total Social Cost Versus Pollution Cost, New Coal Fired Plant (Nonurban), Showing Probability Distribution on Pollution Cost per Pound of Sulfur

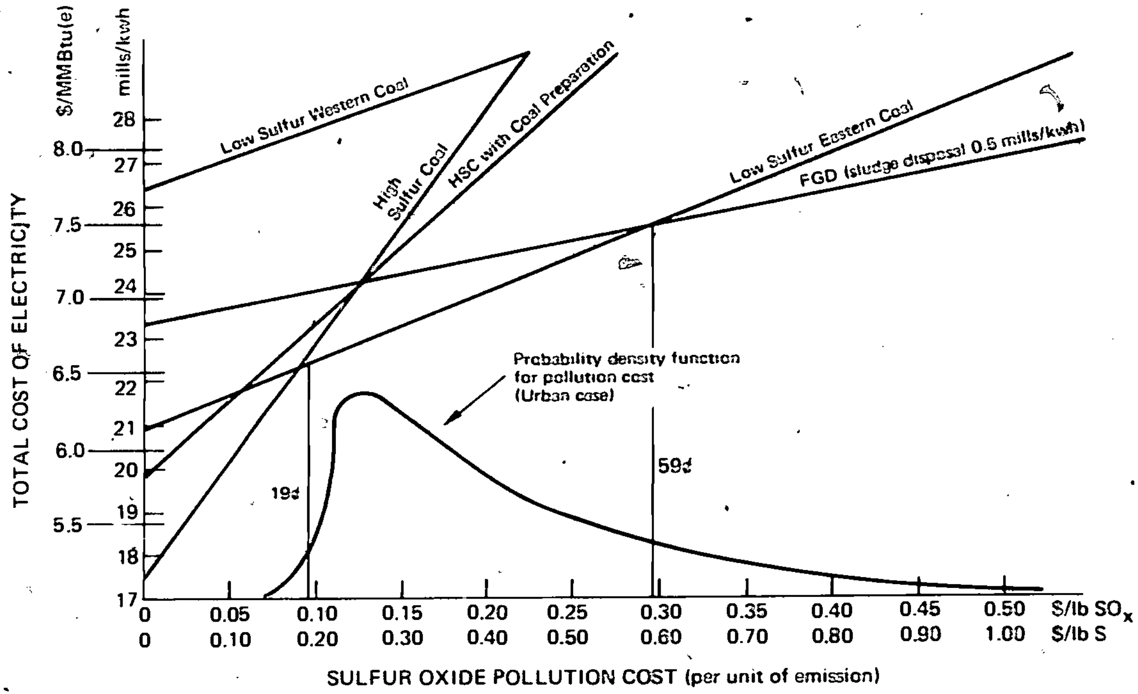

FIGURE 13-19: Total Social Cost Versus Pollution Cost, Old Coal Fired Plant Reconverted to Coal (Urban), Showing Probability Distribution on Pollution Cost per Pound of Sulfur.

per pound, we would switch to high sulfur coal and save for an existing plant, 20.6 + .78 - (17.2 + 2.59) = 1.59 mills per kilowatt hour, or about $6 million a year in social cost for a 620 MW plant running 6000 hours per year.

We know what we would have done without the information, and given the information, we can compute how much we would save by switching the decision. We compute an expected value over the probability distribution on pollution cost of the savings we would get by switching the decision from the one we would have made without the clairvoyant's information to resolve the uncertainty. The probability distribution on pollution cost is of course exactly the same as a probability distribution on what the clairvoyant will tell us.

For the existing rural plant, the decision is close between burning high sulfur and low sulfur Eastern coal. It is unlikely but possible that resolution of the uncertainty would indicate that flue gas desulfurization is the best alternative. The value of resolving the uncertainty in this situation is about 0.53 mills/kwh, or $2 million a year for the plant. The new rural plant has a similar answer. It is a bit more likely that resolving the uncertainty will result in a switch to flue gas desulfurization, but the result of the calculation is about the same, .48 mills/kwh or $2 million a year for a new 612 MW plant operating 7000 hours a year. If low sulfur eastern coal is not available at the rural plant location the value of resolving uncertainty drops by about one half, to about $1.2 million per year for an existing plant and $1.6 million for a new plant.

For the urban plant the decision is very close between low sulfur coal and flue gas desulfurization. With sludge disposal cost at 0.9 mills/kwh for F&D for the urban site, the value of resolving the uncertainty on pollution cost per pound is about 0.25 mills/kwh, or $900,000 per year for an existing plant of 620 MW running 6000 hours per year.

Judging from the list of large coal burning plants in Appendix 13-B and the sulfur emissions data in Figure 13-10, there are at least the equivalent of about one hundred of our representative rural plants (60,000 MW) burning coal of the order of 3 percent sulfur in the Northeastern region of the United States. In addition there are about 60,000 MW of new coal plants planned or under construction, and the order of 20,000 MW of oil fired plants that might be converted to coal. If we scale up the result of the value of information calculation according to these numbers, we reach a value of about $250 million per year as a rough estimate of what it might be worth to resolve uncertainty about the emissions to ambient relationship and health consequences of sulfate.

Relation to the costs of a Research Program to Resolve Uncertainty on Health Effects and the Emission to Ambient Relationship

Some estimates for the costs and time to obtain results in resolving the uncertainties on the health effects of sulfates and the emissions to ambient relationship are contained in a paper by David P. Rall, of EPA, which is in the Volume 2 of the National Academy of Sciences report on Automobile Emissions 1974 p. 427-431. The estimated cost for the research program recommended in this document is approximately $8-10 million per year, with significant results expected from most of the program elements in two to five years. The program funding is about equally divided between research on monitoring and understanding sulfate in the atmosphere, and achieving a better understanding of the health effects of sulfates. In the light of the preceding calculation, which shows a value of the information the order of twenty-five times higher than the cost of carrying out this research, such a research program would seem like an excellent idea.

CONCLUSIONS AND OBSERVATIONS

The analysis of this section is intended as a means for organizing and placing in perspective the information made available in this study to serve as a basis for decisions on sulfur oxide control. This task is an ambitious one, and the authors are well aware of the limited way in which they were able to deal with various issues under limitations of time and resources. We regard the analysis as a framework and not a finished product, and we hope that diligent review and criticism of our work will provide a better basis for decisions.

The major thrust of the analysis has been to relate the consequences of emissions levels to the costs imposed on electric power generation. We carried out calculations for three representative power plants in which we examined the effect of alternative strategies on the sum of the cost of electricity plus the social costs attributable to sulfur oxide emissions. Whereas the calculation of electricity cost is relatively straightforward, the social cost of sulfur oxide emission is both uncertain and judgmental, so its assessment is difficult. Yet a decision on emissions control strategy requires that this assessment be made, explicitly or implicitly. We have shown a way of carrying through the assessment explicitly, and we have illustrated how the preferred alternative for each representative plant changes with the assessment of pollution cost and with changes in the added costs of electricity imposed by that alternative.

In Table 13-22 we have summarized the main analytical results for the representative plant calculations. We show only the alternatives of burning high sulfur coal, switching to low sulfur eastern coal, or installing flue gas desulfurization. (Coal preparation and use of western low sulfur coal were found in these calculations to be somewhat inferior in sulfur removal for the additional cost, but it should be cautioned that for some power plant situations one of these alternatives could be preferred.) The values of pollution cost corresponding to the crossover points between high sulfur coal, low sulfur coal if available, and flue gas desulfurization are shown in the first section, and below we summarize the change in the crossover point per increment of additional cost in the abatement alternatives.

TABLE 13-22

Summary of Major Results for Representative Plants

Crossover values of pollution cost (cents per pound of sulfur) at which alternative strategy changes:

	Switch from high sulfur to eastern low sulfur coal	Switch to FGD with low sulfur coal available	Switch to FGD without low sulfur coal available
new plant (rural)	19	37	23
existing plant (rural)	19	53	26
oil plant to be reconverted (urban) to coal	19	59	28

Sensitivities: Change in crossover values for a 1 mill/kwh increment in capital cost of FGD (1 cent/MMBTU increment in price premium for eastern low sulfur coal)

new plant (rural)	(0.57)	23	5.0
existing plant (rural)	(0.57)	20	4.3
oil plant reconverted (urban)	(0.57)	20	4.3

Pollution costs per pound of sulfur emitted

	Normal value	Point on Probability Distribution* 5%	Mean	95%
rural case (new and existing)	21	9	18	37
urban case (oil plant reconverted)	55	21	46	100

Expected Value of Resolving Uncertainty*, in mills/kwh (million $/year, each representative plant)

	If low sulfur coal Available	Not available
new plant (rural)	0.48 (2.1)	0.38 (1.6)
existing plant (rural)	0.53 (2.0)	0.34 (1.2)
oil plant reconverted (urban)	0.19 (0.9)	0.40 (1.5)

*The probability distributions (Figures 13-15, 13-16) are a rough summary of judgment regarding uncertainties in the emissions to ambient relationship and in a number of cases adverse health effects given in Table 13-12. Other uncertainties have not been included in these distributions or the value of information calculation.

Assessment of the pollution cost was found to depend most critically upon the sulfur dioxide-ambient sulfate relationship, the dose-response relation between health effects and ambient sulfate, and the social cost ascribed to the health effects. Material property damage was the next most important factor, and the other effects were judged to have less importance. Calculation for the representative cases was carried out using nominal values for these input factors, and uncertainty from the emissions-to-ambient and dose-response relations was characterized by probability distributions. These probability distributions were then used to calculate the expected value of resolving the uncertainty before the choice of emissions control strategy is made.

High Sulfur Coal: A Poor Alternative for the Urban Representative Case

The results for the urban case analysis indicate that burning high sulfur coal in a plant close upwind of a major metropolitan area is a poor strategy relative to low sulfur eastern coal or flue gas desulfurization. With a population at risk of 11.5 million (e.g., Metropolitan New York) and health values of $250 per case of chronic respiratory disease, $20 per day of aggravated heart lung disease symptoms, the nominal pollution cost was computed at 55 cents, far above the cross-over points of 19 cents (from high sulfur coal to low sulfur coal) and 28 cents (from high sulfur coal to FGD, assuming low sulfur coal is not available). Resolving the uncertainty on emissions to ambient and dose-response is very unlikely to bring the pollution cost below 19 cents, although a significant chance (29 percent) is computed that this resolution would bring the cost below 28 cents.

Would it be worth waiting to resolve this uncertainty before acting to abate the emissions? The following argument indicates that the waiting is not advisable. Let us consider the case where low sulfur coal is not available, and assume the decision is between high sulfur coal and flue gas desulfurization. Suppose the decision is delayed a year, during which high sulfur coal is burned: the expected net social cost of this choice (pollution cost plus electricity cost) is about 4 mills/kwh, or about $15 million/year. The expected value of resolving the uncertainty is 0.4 mills/kwh, a tenth as much. Even discounting this value forward for the remaining life of the plant, the expected improvement in the decision does not compensate for the loss incurred by a year of delay. The argument for continued use of high sulfur coal if low sulfur coal is available is even weaker: the cost of waiting is high and the chances are very low that further information would show that sulfur removal was not worth at least 19 cents per pound.

Other Comparisons Among Strategies: The High Value of Further Information

The size of the uncertainties and the importance of judgment on health values make it difficult to draw additional implications in comparing other strategy alternatives. For the urban case low sulfur eastern coal and flue gas desulfurization are close, and for the rural case the best strategies appear to be low and high sulfur coal. The difference between these alternatives is slight, and the value of resolving uncertainty is large. An estimate for the country as a whole based on the representative plant calculations is $250 million per year to resolve the uncertainty on the sulfur oxide emissions to ambient sulfate relationship and the dose response relation for health effects. This indicates the great need for near term research results in these areas.

The values assigned to health effects play an equally important role in the calculations. If the values for chronic respiratory disease of $250 per case and $20 per day for aggravated heart-lung disease symptoms are increased a factor of four, the range of pollution costs is shifted up to where flue gas desulfurization appears desirable for both the urban and rural case. If further information confirms oxidation rates and dose response relationships of approximately the present estimates, the decisions will hinge largely on these difficult value judgments: how society shall make tradeoffs between reduction in these health effects and reduction in the cost of electricity. In this situation equity and distributional issues would undoubtedly assume much greater importance. On the other hand, further information may indicate that the consequences of sulfates on human health are sufficiently great (or small) so that the decisions are much less sensitive to these value judgments.

Other Needed Information

Appendix 13-F contains a brief review of the materials damage literature, with the implication that the materials damage value used in this analysis may be low by as much as a factor of four or five. One implication of this change would be to make emissions abatement for the urban case justified on the basis of alleviating material property damage alone. It should be recalled, however, that many of the assumptions used in the materials damage calculation were extremely arbitrary, and clearly more clarification on the materials damage issue would be very welcome.

There are a number of important potential consequences of sulfur oxide emissions that were not included in the analysis: the possibility of more serious health effects such as cancer or emphysema, climatic changes induced by sulfates, or catastrophic effects on living systems from high levels of acid rain. These areas should receive substantial research attention, and information on these areas should be included in subsequent analysis on sulfur oxide emissions control strategy. Information that suflates contribute substantially to serious respiratory disease could motivate a strategy of very high emissions reduction wherever possible. Since the cost of additional land permitting a scrubber to be added at a later time is low, perhaps in the range of a few thousand dollars annualized cost per year, it would seem prudent for all new electric coal burning power stations to allow for this possibility.

Using the Methodology on a Case by Case Basis

The variation in circumstances from one power plant to another makes it highly advisable that the decision among alternative strategies be examined on a case by case basis. A possible problem in this approach arises from the economic and air quality relationships between the plants. The main economic relation is the limited near term supply of low sulfur eastern coal. If there is more demand for it, its price premium will increase relative to high sulfur coal until it is effectively no longer available. We have avoided this difficulty by examining the two extreme cases, low sulfur coal available at about the present price premium, and ~~not~~ available at all. The relation through air quality is that if a large number of plants install emissions abatement, the pollution damage per pound of sulfur may decrease below the crossover point to install abatement. Given the present uncertainties this relation does not seem a serious concern: there is little evidence to indicate a sharp threshold for damages as emissions (or ambient levels) are changed.

We believe that an important use for the type of methdology illustrated in this section is to determine where limited resources (such as flue gas desulfurization construction capacity, available low sulfur coal) can do the most to alleviate pollution damage in the near term. This approach may be most helpful in setting priorities among types of plants regionally or among several plants in one locality, and it could be extended to case by case analysis of a number of power plants in a large region.

FOOTNOTES

1 Ambient levels of sulfur dioxide and suspended sulfates and their relation to power plants and other emission sources are discussed in Section 1 of Part Two.

2 Further discussion on the relationship between health effects and ambient levels of sulfur oxides and particulates is found in Part 1.

3 Since other pollutants act as catalysts to accelerate the oxication of sulfur dioxide to sulfate, a possible additional control strategy to reduce ambient sulfate levels is to reduce the emissions of these other pollutants. In some local situations this approach might be very effective. For example, the difference in observed oxidation rates in plumes (Gartrell 1963, (Newman et al. 1975a,b) may be largely due to the presence of particulates, especially trace metals such as vanadium. For the northeastern region of the U.S. taken as a whole the effectiveness of this approach appears much less promising. The chemistry of the oxidation is complex and not well understood, and there are many materials believed to act as catalysts. Some of these may be impossible or extremely expensive to eliminate from the atmosphere: for example, terpenes and other hydrocarbons given off naturally by vegetation, soot and smoke particles, and photochemical smog resulting from automobile emissions. While further investigation of a catalyst control approach appears warranted, the best approach for reducing sulfate levels appears to be reduction of the quantity of sulfur oxide emitted into the atmosphere. We shall not include a catalyst control alternative among the strategies considered in the analysis.

4 An exception is the toxic materials sections (307 and 311) of the Federal Water Pollution Control Act Ammendments of 1972, Public Law 92500, 92 Congress, S. 2770, October 18, 1972, specifying a fine schedule depending on the amount spilled for toxic substances

discharged into the nation's waterways. Excellent discussions of the merits of the incentives approach are to be found in Solow (1971) and Freeman et al. (1973).

5 Other possible consequence of ambient sulfate levels include changes in regional radiation balances through the increase in atmospheric turbidity (Source: George Hidy, personal communication) and influence on regional precipitation through cloud nucleation effects (Source: Myron Tribus, personal communication). These meterological effects deserve further investigation.

6 Or the use of very low sulfur coal. The standard of 1.2 lbs. of SOx per million BTU could be met with 0.7 percent sulfur eastern coal or 0.5 percent sulfur western coal. The availability of such coal is very limited at present, but may increase as western coal resources are developed.

7 Only sulfur oxides are being counted here in the assessment of pollution costs. This restriction is not necessary from a methodological point of view, rather it is made to increase the clarity of the presentation. Any other significant pollutants should also be included in the assessment process in the same way. For example, if substantial water pollution might result from the sludge generated by a FGD process, the costs of the water pollution consequences should be assessed and included in the total social cost. Since closed-loop operation and careful disposal practices are expected to reduce potential water pollution from sludge to a low level, no such cost has been included. Other potential deterious consequences from FGD would include environmental damage from limestone extraction and lime processing. To the degree that these effects are of substantial magnitude and not already included in the cost of the FGD alternative, they should be brought into the analysis. Coal preparation may also cause environmental damage from water pollution; this damage

should be assessed and if significant included in the cost of coal preparation. Other fine particulates beside sulfate may have substantial health effects, but there is virtually no detailed information now available on these effects. In the absence of more information sulfate is used as an indicator of the toxic potential of particulates. It should be noted that FGD results in a reduction of about 50 percent in fine particulate emissions over the levels obtainable with particulate removal technologies alone, and switching to low sulfur coal might actually increase fine particulate emissions from a power plant unless particulate removal technologies are carefully optimized for the new coal. Some credit for lowering fine particulate emissions should be given to FGD relative to the low sulfur coal alternative. In view of the great uncertainties in the health effects area we have not attempted to assess the magnitude of this credit.

8 While the discussion has dealt with sulfur oxide pollution, effects of other pollutants and any other important externalities should be included in the assessment of the least total cost to society. See footnote7.

9 Pollution externalities associated with nuclear power have not been included in the cost of the nuclear alternative. The magnitude of nuclear externalities should be assessed and included in a comparison between nuclear and coal fired generation. Health effects in the mining and processing of fuels may be as significant as the pollutants released from nuclear and coal fired plants. See, for example, Sagan (1972), (1974).

10 More advanced techniques of coal preparation are discussed in Chapter 10. These techniques are considerably more expensive for the additional amount of sulfur removed.

11 As stated earlier, the focus of this analysis is the hazards from low levels of sulfur dioxide and sulfate, and not the reduction in violations for the sulfur dioxide primary ambient standard. We assume that such violations will be eliminated either by using tall stacks and intermittent control,

or by one of the alternative strategies being considered in this analysis for reducing sulfur oxide emissions. Although benefits from eliminating violations of the primary sulfur dioxide standards might be significant in some cases, we are not including them in the analysis. It is reasonable to assume that these benefits could be obtained with a tall stack-intermittent control system, whose cost of a few tenths of a mill per kwh is small compared to the other alternatives under consideration.

12 A recent airborne measurement by EPA on a coal fired power plant plume indicates an oxidation rate of 1 percent per hour for the first 10 miles, then an increase to a rate of about 3 percent per hour from 10 to 20 miles as more ambient (rural) air is entrained into the plume. Background ozone is suggested as one possible mechanism that might lead to this oxidation rate. (Source: personal communication from William E. Wilson, February 24, 1975.) This result and the recent measurements from Europe (Eliassen and Saltbones 1975) provide evidence for rural oxidation rates on the high side of the range of uncertainty we have assumed: towards average values of 1 percent per hour, or perhaps even higher, rather than 0.1 percent per hour. Even a few more measurements of this type could very substantially reduce the uncertainty in oxidation rate, and even though the cost of airborne measurements is high, the value of resolving this uncertainty is orders of magnitude higher, as will be discussed below.

13 Garland (1974) cites the work of other investigators as a basis for a value of 0.03 cm/sec. AEC practice for submicron particulates is to use a value of 0.15 cm/sec. (Source: C. Gammertsfelder, personal communication).

14 The form of this model is similar to that developed by Dr. Michael Mills of GCA Corporation, Bedford, Massachusetts, for some unpublished work carried out for EPA. Additional references on long range

transport of sulfur oxides include Zeedijk and Velds (1973) and Rodhe (1972).

15 Strictly speaking, the emissions are not uniformly distributed along the length unless the unit of time is small. See Appendix A for more precise description of the model.

16 About 40 percent of the emitted sulfur oxide is returned as sulfate in rain, from calculations in Chapter 10.

17 These results are in general agreement with the results given in Part Two Section 1: the 10-40 percent increase was predicted for urban sulfate levels (for the nation as a whole) as a consequence of doubling sulfur oxide emissions from all power plants throughout the country.

18 Source: 1973 Statistical Abstract, using 1970 census data. 47 million persons lived in standard metropolitan stastical areas in the eleven states plus the Greater Washington, D.C. area.

19 Limited data have shown that, within the range of .3 to 2.5 um, the smaller the particle size the greater the irritant potency. Reference: Amdur (1960).

20 The dose-response relations that we are about to describe were developed in an unpublished study for the U.S. Environmental Protection Agency. Our use of these particular curves is not meant to indicate an endorsement. Rather, we present them as highly simplified illustrations of a set of possible relationships that have not yet been firmly established. Judgment communicated to us by representatives from the Assemble of Life Sciences Committee responsible for Part one of this report was that these numbers were not unreasonable, and that they could be considered the best estimates of the dose response relationships available at this time. Source: Statistical Abstract of the U.S., 1972. The populations listed for the various age groups are estimations based upon the total New York Metropolitan Population (Table 20, pg 20) and the age distribution for New York State (Table 36, pg 31).

21 This distribution is conventially log normal in the air pollution literature. Data given in the CHESS studies indicate that the log normal distribution does give a good fit.

22 As shown above, health costs are also quite sensitive to the dollar costs we associate with each case of a given health effect. However, rather than interpret these as uncertain quantities, we take the point of view that the dollar cost for a health effect is a critical quantity which society should assign as a basis for decision-making rather than ask analysts to estimate. Those responsible for setting public policy should reflect on what amount of money should be allocated to alleviate the suffering caused by disease, e.g., chronic respiratory disease and aggrevation of heart-lung disease symptoms in the context of the decision addressed here. Sensitivity analysis indicates how the pollution cost, and hence the alternative strategy having the lowest social cost, will change depending on the value judgments used. Although the values used in this analysis are subjective, they are not inconsistent with some crude measures of individuals' willingness to pay to alleviate suffering, as for example, assessed by Jacoby and Steinbrenner (1973).

23 The reader should recall that the population at risk from which these numbers are calculated is that of the New York Metropolitan area, 11.5 million.

24 The mean is below the nominal value of $20.5 million because of the highly asymmetric nature of the range: from 10 percent to 200 percent of the nominal value to the two most serious health effects.

25 However, as Gillette (1973) points out, while physical deterioration of materials may occur at relatively low pollution levels, for the effects to be economically important (1) the normal service life of the material must be reduced, (2) the frequence of maintenance tasks must be increased, or (3) the quality of the services rendered by the product must be diminished. For this

reason, while a threshold may not exist for physical damage, economic loss from physical damage may exhibit such a threshold. Threshold effects may be introduced when converting physical material damage to economic loss. Our analysis ignores this effect, possibly causing us to underestimate marginal material damage costs. More accurate dose-response information ought to be applied in the analysis as it becomes available.

APPENDIX 13-A

A MODEL RELATING SULFUR OXIDE EMISSIONS TO AMBIENT SULFATE LEVELS

Appendix 13-A presents the mathematical relationships and calculations for the emissions to ambient sulfate model. As described in the text, the model is a simple "box" model in which the width of the box grows proportionally with time. SO_2 removal, SO_2 oxidation to sulfate, and sulfate removal all proceed according to a first order rate reaction. The geometry of the model is illustrated in Figures App. 13-A1 and App. 13-A2.

The geometric relationships in the model are as follows. We assume a constant wind velocity of u kilometers per hour, and assume that the plume subtends an angle θ in the direction downwind of the plant. That direction is assumed to remain constant. In a small time increment Δt a quantity Δx of SO_2 is emitted. We assume this to remain uniformly distributed between ground and the mixing layer at height h above ground, across a width (perpendicular to the direction of the wind) of $w(t)=2ut \tan(\theta/2)$, and through a length (parallel to the direction of the wind) of $u\Delta t$. Then the assumption for the box model is that at time t the concentration of sulfur dioxide per unit volume, y_o, is uniform within this box, which has a volume of

$$u\Delta t \times ut(2\tan \theta/2) \times h = A(t)\, u\Delta t \qquad (1)$$

where A(t) is the rectangular area in the crosswind vertical plane. If we assume an emissions rate $\Delta x/\Delta t = q$ then the concentration of SO_2

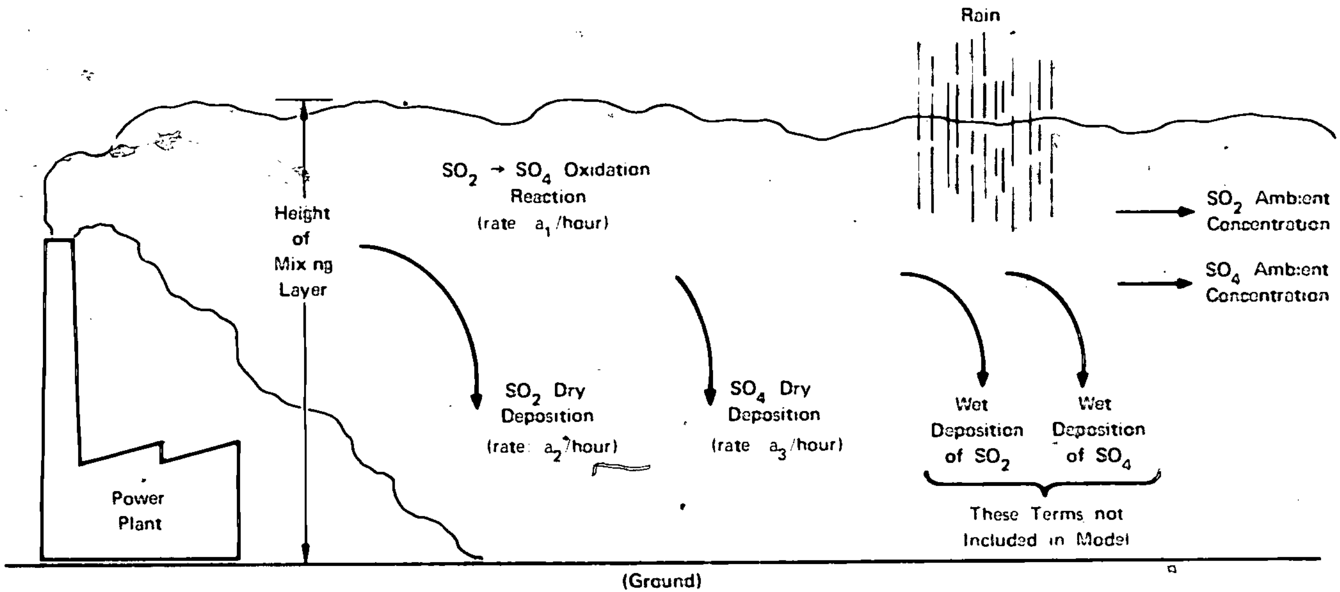

FIGURE 13A-1: Model for Emission to Ambient Relationship for Sulfur Oxide Emissions from a Representative Power Plant

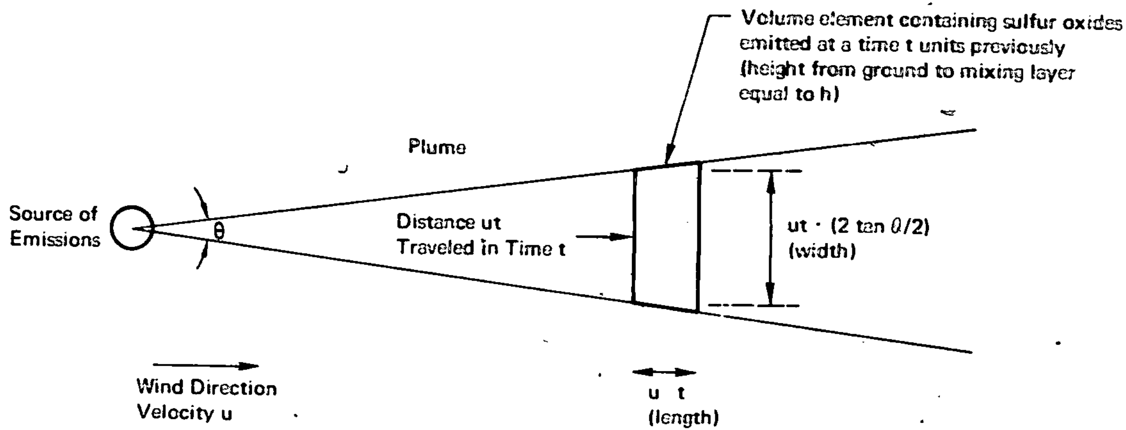

FIGURE 13-A2: Geometric Relations for Emissions to Ambient Model

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from the power plant at a time t after emission (a distance of ut downwind), assuming no oxidation or removal has occurred, is given by the quantity emitted $\Delta x = q\Delta t$ divided by the volume $A(t)\,u\Delta t$ of the volume element:

$$y_o(t) = \frac{q\Delta t}{A(t)u\Delta t} = \frac{q}{2u^2ht\tan(\theta/2)} \qquad (2)$$

We assume that there are no further sources of SO_2, and we ignore any SO_2 present before the air reached the power plant. Our concern is just the SO_2 emitted from the power plant, and y_o is the incremental SO_2 concentration from that power plant, to be added to the ambient levels from other sources. Likewise, in the discussion below, we treat incremental quantities of SO_2 and sulfate resulting from the emissions of the power plant source.

Let us consider the volume element $\Delta V(t)=A(t)u\Delta t$ to contain an amount of sulfur dioxide $Y(t)$ and an amount of sulfate $Z(t)$. No new sulfur oxide enters this volume element after it leaves the power plant, and sulfur dioxide and sulfate are assumed to be removed only through dry deposition at given deposition velocities. The sulfur oxides are assumed to be uniformly distributed throughout the volume element $\Delta V(t)$ at all times. We now write the differential equations for the changes in the quantities contained in $\Delta V(t)$. Then from equation (1) we can compute the concentrations of SO_2 and SO_4 by dividing the quantities by the volume $\Delta V(t)$ in the same manner as for equation (2).

Let us first take the oxidation reaction. Of the quantity $Y(t)$ of SO_2 contained in $\Delta V(t)$, a proportion of a_1 is lost through oxidation to sulfate per unit of time:

$$\frac{dY(t)}{dt} = -a_1Y(t) \qquad \text{(oxidation)} \qquad (3)$$

A loss also occurs through dry deposition. Of the quantity $Y(t)$ of SO_2 contained in the volume element, a proportion is lost per unit time equal to v_1 (the dry deposition velocity for SO_2) times the area of contact with the ground, which is the $u\Delta t \times 2ut \tan(\theta/2)$, times the concentration, which is $Y(t)/hu\Delta t \times 2ut \tan(\theta/2))$. The proportion of SO_2 lost through dry deposition per unit time is then

$$\frac{dY(t)}{dt} = \frac{-v_1 \times u\Delta t \times 2ut \tan(\theta/2) \times Y(t)}{h\, u\Delta t \times 2ut \tan(\theta/2)}$$

$$= \frac{-v_1\, Y(t)}{h} \quad \text{(dry deposition)} \qquad (4)$$

We shall define $v_1/h = a_2$, the rate of loss due to dry deposition of SO_2. Since loss through oxidation and loss through dry deposition are the only mechanisms by which the quantity of SO_2 in the volume element is changed, we combine (3) and (4) into a single differential equation for $Y(t)$, the quantity of SO_2 in the volume element:

$$\frac{dY(t)}{dt} = -a_1\, Y(t) - a_2\, Y(t) \qquad (5)$$

This equation has the solution

$$Y(t) = Y_o e^{-(a_1+a_2)t} \qquad (6)$$

where Y_o is the initial quantity of SO_2 in the volume element. By dividing the quantity $Y(t)$ by the volume of the element $A(t)\, u\Delta t$ we obtain

$$y(t) = \frac{qe^{-(a_1+a_2)t}}{2u^2ht \tan\theta/2} \qquad (7)$$

We go through a similar reasoning process to arrive at a solution for the sulfate concentration. If oxidation of one gram of SO_2 produces k grams of sulfate ion, then the quantity of sulfate $Z(t)$ in the volume element is given by

$$\frac{dZ(t)}{dt} = ka_1 Y(t) \quad \text{(sulfate formation through oxidation)} \tag{8}$$

since $a_1Y(t)$ grams of SO_2 are oxidized per unit time in the volume element at time t (from equation(3)). We also must account for the loss of sulfate through dry deposition. The quantity lost from the volume element per unit time is the dry deposition velocity (v_2) times the lower surface of the volume element, the area in contact with the ground, times the concentration of sulfate in the volume element. The area of contact with the ground is (as before) $u\Delta t \times 2ut \tan(\theta/2)$, and the concentration of sulfate is the quantity $Z(t)$ of sulfate in the volume element divided by the volume of the element: $Z(t)/hu\Delta t \times 2ut \tan(\theta/2))$. The proportion of sulfate from the volume element lost per unit of time is then

$$\frac{dZ(t)}{dt} = \frac{-v_2 \times u\Delta t \times 2ut \tan(\theta/2) \times Z(t)}{hu\Delta t \times 2ut \tan(\theta/2)}$$

$$= \frac{-v_2 Z(t)}{h} \quad \text{(dry deposition)} \tag{9}$$

We shall define $v_2/h = a_3$, the rate of loss due to dry deposition of sulfate. Since formation through oxidation and loss from dry deposition are the only means by which the quantity of sulfate in the volume element is changed, we can combine (8) and (9) into a single differential equation for the quantity of sulfate $Z(t)$:

$$\frac{dZ(t)}{dt} = ka_1Y(t) - a_3Z(t) \tag{10}$$

The solution to this differential equation is given by

$$Z(t) = Z_o e^{-a_3 t} + \left(\frac{a_1 k\ Y_o}{a_1 + a_2 - a_3}\right)\left(e^{-a_3 t} - e^{-(a_1+a_2)t}\right) \tag{11}$$

where Z_o and Y_o are initial quantities of sulfate and SO_2, respectively, in the volume element. As with sulfur dioxide, we can divide the quantity $Z(t)$ by the size of the volume element to obtain the concentration $z(t)$. If the initial quantity of sulfate Z_o is zero,

$$z(t) = \frac{q}{2u^2ht\tan\theta/2} \times \frac{ak_1(e^{-a_3t} - e^{-(a_1+a_2)t})}{(a_1+a_2-a_3)} \qquad (12)$$

It is also possible to solve these equations (6) and (11) for rates that change discontinuously from one time period to another. One computes the solution for the first time period, uses this to establish the quantities Y_o and Z_o as initial conditions for the second time period, then one solves equations (6) and (11) for the second time period. Of course, when the solutions are expressed as concentrations these must be computed by dividing the quantities of SO_2 and sulfate by the volume $\Delta V(t)$ of the volume element, and this volume is increasing in direct proportion to time.

In solving this model to obtain concentration of SO_2 and sulfate we shall use the following values for the inputs:

(1) The wind is a constant 20 kilometers/hour.

(2) The angle subtended by the plume is 15^o.

(3) The height of the mixing layer above ground is 1000 meters.

(4) The dry deposition velocity for SO_2 to the ground is 0.8cm/sec, giving a loss rate a_2 of 2.88 per cent per hour with a mixing layer height of 1000 meters.

(5) The dry deposition velocity of SO_4 to the ground is 0.4cm/sec, giving a loss rate a_3 of 1.44 percent/hour with a

mixing layer height of 1000 meters above ground.

(6) The oxidation rate a_1 of SO_2 to form sulfate is 0.5 percent per hour in rural air outside of a metropolitan area, and 5.0 percent per hour when the air has passed over a metropolitan area and contains particulates, oxidants, and hydrocarbons from urban emission sources.

(7) We neglect the 1-2 percent of sulfur oxide emitted initially as sulfate and consider that all of the 10^4 kilograms per hour of sulfur dioxide is emitted as SO_2. This assumption is of course easily relaxed, but since the initial sulfate is small compared to the sulfate formed subsequently by the oxidation, we may neglect it in the calculation without introducing a significant error.

Calculations are given in Table 13-6 of the main text for incremental increases in the ambient concentrations of SO_2 and sulfate over time for an oxidation rate of 0.5 percent per hour and in Table 13-7 of the text for an oxidation rate of 5 percent per hour. Table App. 13-A1 shows the results if a 2 percent oxidation rate is used.

Table App. 13-A1

Incremental Contributions to Ambient Levels of SO_2 and Sulfate Resulting from the Emissions of a Single 600MW Power Plant (Oxidation Rate of 2.0 percent per hour assumed.)
hour assumed.)

Time, hours since emission	3	6	12	18	24	50
Distance from plant, km	60	120	240	360	480	1000
Increase in SO_2 concentration ($\mu g/m^3$)	27.3	11.8	4.4	2.2	1.23	0.17
Increase in SO_4 concentration ($\mu g/m^3$)	2.59	2.36	1.96	1.64	1.37	0.66

Table 13-8 of the main text and Figure 13-8 give results for the case in which an oxidation rate of 0.5 per hour is assumed for 24 hours (480 km) then the rate increases to 5 as the sulfur dioxide oxidation reaction is catalyzed by the presence of urban pollutants. Tables App. 13-A2, 3, and 4 show the effect of varying the time (distance) between the plant and the metropolitan area. The values after two hours (40 km) of exposure to urban air are shown in boxes for comparison to the values of 1.39ug/m^3 of SO_2 and 0.58ug/m^3 of SO_4 for the ambient increments from Table 13-8.

Table App. 13-A2

Incremental Contributions to Ambient Levels of SO_2 and Sulfate from a Power Plant in a Remote Location (10^4 kg/hour SO_2 emissions, 120 km from plant to city).

Time, hours since emission	3	6	7	8	9
Distance from plant, km	60	120	140	160	180
Increase in SO_2 concentration ($\mu g/m^3$)	28.6	12.9	10.2	8.3	6.8
Increase in SO_4 concentration ($\mu g/m^3$)	0.66	0.62	1.32	1.78	2.1
Location of plume	rural air	edge of city		metropolitan area	
Oxidation Rate % per hour	0.5	5.0	5.0	5.0	5.0

Table App. 13-A3

Incremental Contributions to Ambient Levels of SO_2 and Sulfate from a Power Plant in a Remote Location (10^4 kg/hour SO_2 emissions, 240 km from plant to city).

Time, hours since emission	3	6	12	13	14	15
Distance from plant, km	60	120	240	260	280	300
Increase in SO_2 concentration ($\mu g/m^3$)	28.6	12.9	5.3	4.5	3.9	3.3
Increase in SO_2 concentration ($\mu g/m^3$)	0.66	0.62	0.53	.83	1.06	1.23
Location of plume	rural air		edge of city		metropolitan area	
Oxidation Rate % per hour	0.5	0.5	5.0	5.0	5.0	5.0

Table App. 13-A4

Incremental Contributions to Ambient Levels of SO_2 and Sulfate from a Power Plant in a Remote Location (10^4 kg/hour SO_2 emissions, 1000 km from plant to city).

Time, hours since emission	6	12	18	24	50	51	52	53
Distance from plant, km	120	240	360	480	1000	1020	1040	1060
Increase in SO_2 concentration ($\mu g/m^3$)	12.9	5.3	2.87	1.76	0.35	0.32	0.29	0.26
Increase in SO_4 concentration ($\mu g/m^3$)	0.62	0.53	0.46	0.40	0.22	0.24	0.25	0.26
Location of plume	rural air				edge of city		metropolitan area	
Oxidation Rate % per hour	0.5	0.5	0.5	0.5	5.0	5.0	5.0	5.0

APPENDIX 13-B

THE REPRESENTATIVENESS OF THE ILLUSTRATIVE CASES

Ideally, the analysis of alternative emission control strategies should be carried out for each individual power plant, but such an effort was obviously not possible for the present study. Our analysis has, instead, focused on two illustrative cases: a rural plant 300 miles upwind of a metropolitan area, and a plant in or adjacent to an urban area.

Tables App. 13-B1,2, and 3 indicate the number and nature of the various power plants that our illustrative examples represent. The data is illustrative, not definitive: in some cases it is out of date or in need of qualification. The reader is directed to the referenced source documents for further details.

Table App.13-B1 lists some of the larger existing coal fired plants, their locations, and some relevant technical and economic characteristics. Table App. 13-B2 lists some new coal fired plants and those units planned for construction in the near future. Table App. 13-B3 contains a tabulation of existing oil fired plants deemed convertible to coal. (The determination of which oil burning plants could be converted to burn coal was carried out by the Oil Savings Work Group, Office of Energy Conservation, Federal Energy Agency. All of those plants listed were originally designed to burn coal.)

TABLE App. 13-B1

EXISTING LARGE COAL FIRED POWER PLANTS, 1972[a,b]

Region State City	Company	Plant	Capacity (MW)	Cost Per KW of installed Capacity ($)
NEW ENGLAND				
New Hampshire				
Bow	Public Service Co of New Hampshire	Merrimack	459	132
MIDDLE ATLANTIC				
New York				
Dunkirk	Niagara Mohawk Power Corporation	Dunkirk	620	139
Tonawanda		C R Huntley	920	149
Pennsylvania				
Springdale	Duquesne Light Company	Cheswick	565	209
Elrama	"	Elrama	510	176
South Heights	"	F. Phillips	411	161
Portland	Metropolitan Edison Company	Portland	427	149
Shawville	Pennsylvania Electric Company	Shawville	640	139
Homer City	"	Homer City	1,319	166
W. Pittsburgh	Pennsylvania Power Company	New Castle	426	128
York Haven	Pennsylvania Power & Light Co.	Brunner Island	1,559	100
Sunbury	"	Sunbury	410	209
Shelocta	"	Keystone	1,872	101
Indiana Co.	"	Conemaugh	1,872	119
Strawberry Ridge	"	Montour	823	176
Eddystone	Philadelphia Electric Company	Eddystone	707	226
Courtney	"	Mitchell	469	160
Masontown	"	Hatfield's Ferry	1,728	139
EAST NORTH CENTRAL				
Illinois				
Bartonville	Central Illinois Light Company	E.D. Edwards	725	179
Coffeen	Central Illinois Public Service Co.	Coffeen	1,006	163
Joliet	Commonwealth Edison Company	Joliet	1,787	118
Kincaid	"	Kincaid	1,319	107
Pekin	"	Powerton	1,213	182
Waukegan	"	Waukegan	933	160
Lockport	"	Will County	1,269	133
Joppa	Electric Energy, Incorporated	Joppa	1,100	152
Baldwin	Illinois Power Company	Baldwin	623	188
Venice	Union Electric Company	Venice #2	500	144
Indiana				
Madison	Indiana-Kentucky Electric Corp	Clifty Creek	1,304	115
Lawrenceburg	Indiana & Michigan Electric Company	Tanners Creek	1,100	127
Petersburg	Indianapolis Power & Light Company	Petersburg	733	117
Dune Acres	Northern Indiana Public Service Co	Bailly	616	149
Gary	"	D.H. Mitchell	529	164
Cayuga	Public Service Co of Indiana, Inc	Cayuga	1,018	161
New Albany	"	Gallagher	637	152
Terre Haute	"	Wabash River	881	130
Michigan				
West Olive	Consumers Power Company	J.H. Campbell	650	129
Muskegon	"	B.C Cobb	511	134
Essexville	"	D E Karn	530	158
Monroe	Detroit Edison Company	Monroe	817	224
E. China Twp	"	St Clair	1,905	137
Ohio				
New Richmond	Cincinnati Gas & Electric Co	W.C Beckjord #1-5	761	145
New Richmond	"	W C Beckjord #6	461	132
Ashtabula	Cleveland Electric Illuminating Co	Ashtabula	640	NA
Avon Lake	"	Avon Lake	1,275	143
Eastlake	"	Eastlake	1,257	150
Cleveland	"	Lake Shore	514	131
Conesville	Columbus & Southern Electric Co	Conesville	434	129
Aberdeen	Dayton Power & Light Company	J.M Stuart	1,831	146
Shadyside	Ohio Edison Company	R.E Burger	546	157
Stratton	"	W.H Sammis #1-7	1,304	153
Brilliant	Ohio Power Company	Cardinal	1,231	112
Beverly	"	Muskingum	1,530	120
Philo	"	Philo	500	115
Cheshire	Ohio Valley Electric Corporation	Kyger Creek	1,086	124
Oregon	Toledo Edison Company	Bay Shore	640	165
Wisconsin				
Oak Creek	Wisconsin Electric Power Company	N Oak Creek	500	169
Port Washington	"	Port Washington	400	121
Oak Creek	"	S Oak Creek	1,192	116
WEST NORTH CENTRAL				
Minnesota				
Oak Park Hts	Northern States Power Co (Minn.)	A S King	598	137
Missouri				
Henry Co	Kansas City Power & Light Company	Montrose	563	117
Sibley	Missouri Public Service Company	Sibley	519	151
Labadie	Union Electric Company	Labadie	1,665	179
St. Louis	"	Meramec	800	153
West Alton	"	Sioux	978	113
Randolph Co.	Associated Electric Cooperative	Thomas Hill	456	138

Annual Generation (Million KWH)	Heat Rate (BTU/KWH)	Operating Costs, Exclusive of Fuel (Mills/KWH)	Coal Consumption (1000 tons)	Avg Heat Content (Million BTU/ton)	Cost per Ton as Burned ($)	Avg Sulfur Content (%)
2,964	10,381	69	1,099	27 18	13 01	2 33
1,266	9,764	1 02	1,286	25 39	12 04	2 54
3,450	10,142	1 36	1,300	25 30	14 30	2.20
3,262	9,494	89	1,307	22 00	8 31	2 10
2,008	10,635	1 17	1,403	21 80	8 32	2.09
2,109	11,781	1 47	1,094	22 69	8 26	1 89
2,530	9,718	1 43	974	24 89	13 36	2 53
4,101	10,484	1 15	1,700	25 20	8 10	2 80
5,772	10,181	1 73	2,479	23 46	9 69	2 10
2,128	11,432	1 34	1,031	22 94	7.71	3 48
8,305	9,836	80	3,238	25 04	12 11	2.04
2,518	12,374	1 60	1,333	24 17	8 57	2 40
8,341	9,789	1 48	3,476	23 33	8 10	2 23
9,957	9,349	1 28	3,559	23 22	9 47	2 59
3,464	9,889	75	1,358	25 29	11 73	
4,374	8,897	1 16	1,978	26 09	13 82	4 11
2,771	10,544	74	1,147	25 50	7 80	2 59
8,857	9,440	46	3,386	24 70	7 11	3 04
2,840	9,877	69	1,339	20 83	6 74	2 83
1,969	10,369	1 17	1,110	18 38	5 77	4 48
8,060	10,444	1 36	3,819	20 92	6 95	2 95
5,669	10,503	1 36	2,982	19 37	6 29	4 02
1,798	13,176	1 51	1,114	21 26	7 71	3.90
4,438	10,261	1 24	2,131	20 91	8 12	2 45
4,606	10,273	61	2,561	18 47	10 79	2 58
6,999	9,986	1 22	3,102	22 75	7 98	2 71
3,796	9,969	55	1,842	19 92	4 25	4 02
2,055	13,025	2 60	1,116	22 53	7 88	2.34
9,530	9,390	59	4,108	21 60	6 94	3 56
5,974	9,179	1 01	2,501	21 76	7 24	3.10
4,369	9,525	57	1,880	21 90	6 00	3 41
2,714	10,022	1 22	1,186	22 36	7 12	3 60
3,274	9,967	74	1,396	21 16	6 23	3 15
4,582	9,744	80	2,140	21 30	5 27	2 33
3,257	[illegible],568	1 11	1,508	22 41	6 67	3 56
4,731	10,170	1 49	2,187	21 85	5 86	2 72
3,971	9,071	80	1,497	24 00	9 10	3 39
2,874	10,736	1 27	1,190	25 89	12 10	2 85
3,490	9,161	72	1,290	24 71	11 61	2 10
3,521	9,590	1 64	1,298	25 38	10 85	2 82
12,386	9,110	59	4,508	23 84	10 39	3 06
3,243	10,042	1 15	1,767	21 08	7 81	NA
2,247	9,881	44	1,027	21 16	7 79	NA
1,811	10,370	1 87	808	22 13	9 82	3 27
5,957	9,869	48	2,448	23 45	9 69	2 58
5,491	9,853	84	2,379	23 62	10 62	3 04
2,597	11,338	1 15	1,228	23 98	11 14	2 84
[illegible]	10,425	1 64	[illegible],229	21 23	7 02	4 51
9,061	9,328	44	3,676	22 46	8 30	1 54
2,642	10,828	1 47	1,243	[illegible] 19	9 03	3 23
12,735	9,449	45	5,247	22 96	8 [illegible]	2 68
7,180	9,088	64	2,861	22 70	7 46	2 96
8,903	9,404	73	4,044	20 65	8 00	4.91
1,103	12,002	1 78	655	20 1	6 33	3 90
[illegible]	9,405	63	1,351	22 88	7 07	3 93
4,192	9,301	85	1,610	24 33	10 81	2 11
3,381	10,212	1 60	1,555	22 98	10 16	2 09
1,517	11,575	2 51	720	24 38	11 65	2 98
5,365	9,567	1 36	2,207	23 35	10 16	2 00
3,310	9,608	70	1,476	21 54	8 68	3 12
3,346	10,586	77	1,851	19 11	5 02	5 52
2,053	10,638	1 47	918	23 79	7 08	3 67
4,665	9,971	1 25	2,045	22 44	6 20	2 99
5,021	10,260	1 27	2,208	23 26	8 75	2 46
3,712	10,252	2 04	1,729	21 95	6 92	3 30
3,195	9,643	36	1,539	20 01	9 75	4 25

TABLE App. 13-B1 (cont.)

EXISTING LARGE COAL FIRED POWER PLANTS, 1972[a,b]

Region State City	Company	Plant	Capacity (MW)	Cost Per KW of Installed Capacity ($)
SOUTH ATLANTIC				
Florida				
Tampa	Tampa Electric Company	Big Bend	446	162
Tampa	"	F.J. Gannon	1,270	101
Georgia				
Bartow County	Georgia Power Company	Bowen	1,595	115
Floyd County	"	Hammond	953	106
Milledgeville	"	Harlee Branch	1,746	90
Maryland				
Aquasco	Potomac Electric Power Company	Chalk Point	728	127
North Carolina				
Moncure	Carolina Power & Light Company	Cape Fear	621	96
Goldsboro	"	H F Lee	403	117
Roxboro	"	Roxboro	1,065	82
Skyland	"	Ashville	616	129
Belmont	Duke Power Company	Allen	1,155	107
Spencer	"	Buck	740	NA
Cliffside	"	Cliffside	781	160
Terrell	"	Marshall	2,000	103
Mount Holly	"	Riverbend	631	102
South Carolina				
Rockland City	South Carolina Electric & Gas Company	Wateree	772	132
Virginia				
Carbo	Appalachian Power Company	Clinch River	713	166
Glen Lyn	"	Glen Lyn	403	112
Alexandria	Potomac Electric Power Company	Potomac River	515	130
West Virginia				
St. Albans	Appalachian Power Company	Amos	1,633	166
Glasgow	"	Kanawha River	439	137
Graham	"	Phillip Sporn	1,106	NA
Maidsville	Monongahela Power Company	Fort Martin	1,152	120
Captina	Ohio Power Company	Kammer	713	128
Moundsville	"	Mitchell	1,633	139
Mt. Storm	Virginia Electric & Power Company	Mt. Storm	1,161	113
EAST SOUTH CENTRAL				
Alabama				
Bucks	Alabama Power Company	Barry	1,771	108
Gorgas	"	Gorgas	1,546	109
Demopolis	"	Greene County	569	96
Wilsonville	Southern Electric Generating Company	E.C. Gaston	1,061	110
Pride	Tennessee Valley Authority	Colbert A	847	116
Pride	"	Colbert B	550	122
Stevenson	"	Widows Creek A	853	100
Stevenson	"	Widows Creek B	1,125	120
Kentucky				
Louisa	Kentucky Power Company	Big Sandy	1,097	139
Burgin	Kentucky Utilities Company	E.W. Brown	726	126
Drakesboro	Tennessee Valley Authority	Paradise A	1,408	138
Drakesboro	"	Paradise B	1,150	137
Paducah	"	Shawnee	1,750	123
Sebree	Big Rivers Electric Coop. Corp.	Coleman	521	138
Tennessee				
Clinton	Tennessee Valley Authority	Bull Run	950	168
Gallatin	"	Gallatin	1,255	110
Johnsonville	"	Johnsonville	1,485	110
Kingston	"	Kingston	1,700	112
Rogersville	"	John Sevier	823	122
WEST SOUTH CENTRAL				
Texas				
Fairfield	Dallas Power & Light Company	Big Brown	1,187	NA
Colorado				
Denver	Public Service Company of Colorado	Cherokee	801	128
MOUNTAIN				
New Mexico				
Farmington	Farmington Arizona Public Service Company	Arizona Public Four Corners #4-5	636	102
Wyoming				
Glen-Rock	Pacific Power & Light Company	D. Johnston	750	169

[a] Plants which in 1972 (1) derived at least 95% of their BTU energy consumption from coal, (2) possessed at least 400MW of installed capacity, and (3) consumed at least 500,000 tons of coal. Source: Steam-Electric Plant Factors, 1973 Ed. [ ].

[b] Source: Steam-Electric Plant Construction Cost and Annual Production Expenses, 1972 Supplement [ ].

Annual Generation (Million KWH)	Heat Rate (BTU/KWH)	Operating Costs, Exclusive of Fuel (Mills/KWH)	Coal Consumption (1000 tons)	Avg. Heat Content (Million BTU/ton)	Cost per Ton as Burned ($)	Avg. Sulfur Content (%)[a]
1,977	10,501	00	929	22 51	8 95	3 30
5,136	10,460	1 40	2,421	22 19	8 70	1 40
4,192	9,072	55	1,770	23 01	8 41	1 26
3,093	11,126	00	1,094	22 03	11 22	2 40
7,418	9,726	65	2,979	24 23	10 85	1 17
3,403	9,655	1 35	1,297	26 96	13 01	2 12
2,119	9,959	66	850	26 65	[d] 12 63	1 40
2,171	10,094	1 10	820	25 66	12 71	1 00
6,425	9,461	50	2,430	24 00	10 25	1 40
2,303	9,505	76	902	26 66	10 70	1 36
8,089	9,332	35	3,230	23 37	10 69	1.09
2,256	10,016	1 26	1,020	26 18	11 16	94
3,607	10,271	45	1,490	26 76	9 68	1 11
14,227	8,670	20	5,206	22 90	10 07	91
3,105	10,605	91	1,395	23 96	10 55	1 10
3,709	8,078	.65	1,300	24.63	11.59	1 50
5,405	8,959	37	2,069	26 06	7 13	71
2,150	9,766	78	846	24 19	9 19	89
2,511	10,143	1 11	967	26 11	15 76	98
8,675	8,959	37	3,115	26 73	9 61	1 00
3,161	9,106	56	1,239	23 41	8 72	89
6,343	9,126	90	2,618	21 96	8 76	1 30
7,617	9,056	55	2,763	26 26	6 96	1 07
3,982	9,065	80	1,629	23 90	7 99	4 00
6,092	9,461	69	2,750	23 37	7 82	3 16
6,261	10,153	71	2,753	22 59	8 36	2 16
7,319	10,037	84	3,031	23 84	9 77	2.61
4,096	10,601	80	2,262	23 32	8 60	1 43
3,057	9,595	.80	1,228	23 96	7.87	1.08
6,045	9,462	46	2,795	23 18	9 67	1 07
4,017	9,850	1 02	1,770	22 36	7 73	4 17
1,451	9,910	1 61	641	22 32	7 57	4 19
3,631	10,800	1 16	1,721	22 70	7 32	2 40
4,960	9,730	1.06	2,157	22 36	7 28	3.21
6,720	9,476	69	2,661	23 29	8 10	1 04
2,796	10,431	50	1,231	23.60	9 55	2 28
8,030	9,680	1 06	1,763	20 12	3 64	NA
6,314	9,100	80	2,071	20 38	3 59	NA
9 798	9,078	.65	4,610	20 82	6 05	2.81
3,160	10,200	50	1,476	21 51	5 28	1 85
5,082	9,100	63	2,025	22 66	6 23	1 64
5,782	9,440	82	2,514	21 60	7 49	3 13
6,006	10,610	97	2,802	22 28	6 64	3 72
9,156	9,560	67	4,095	21 82	8 27	2 05
3,797	9,570	75	1,579	22 98	8 72	1 77
2,661	10,610	1 89	1,790	16.00	2 94	60
5,160	10,417	52	1,599	21 71	6 23	.50
9,046	9,766	82	5,262	18 03	2.71	65
3,284	10,577	.89	2,336	16.86	1.53	.62

[a] Sulfur content data is for the year 1971 and hence cannot be directly compared with the other data. Source: Steam-Electric Plant Air and Water Quality Control Data, 1971 [ ]

[d] New plant initially reported in 1972.

TABLE APP.13-B2

LARGE COAL FIRED POWER PLANTS-NEW PLANTS OR UNITS PLANNED OR UNDER CONSTRUCTION, 1973-1979 [a]

Region / State / City	Company	Plant	Capacity MW	In Service Date (19--)
MIDDLE ATLANTIC				
Pennsylvania				
Indiana County	Pennsylvania Electric Co.	Conemaugh#3	640	75
Homer City		Homer City#3	640	76
Shipping Port	Pennsylvania Power Co.	Bruce Mansfield#1&2	880/830	75/76
Strawberry Ridge		Montour#2	750	
EAST NORTH CENTRAL				
Illinois				
Canton	Central Illinois Light Co.	Duck Creek#1	400	76
Newton	Central Ill. Public Service Co.	Newton#1	600	77
Pekin	Commonwealth Edison Co.	Powerton#5&6	340/340	73/75
Baldwin	Illinois Power Co.	Baldwin#2&3	605/600	73/75

[a] 400 MW and larger. Source: Steam Electric Plant Factors, 1973 Ed.. [ ].

Indiana				
Indianapolis	Indianapolis Power & Light Co.	E.W. Stout#7	450	73
Petersburg		Petersburg#3-4	515	77
Michigan City	Northern Indiana Pub.Ser.Co.	Michigan City#12	504	73
Unassigned		Northwest Ind.#15	500	73
Wheatfield		R.M.Schahfer#14	500	75
Nr. Princeton	Public Service Co.of Indiana	Gibson#1-4	650/650	75/76
Michigan				
West Olive	Consumers Power Co.	Campbell#3	800	77
Monroe	Detroit Edison Co.	Monroe#2-4	1,578/786	73/74
Ohio				
North Bend	Cincinnati Gas & Electric Co.	Miami Fort#7&8	500/500	75/77
Conesville	Columbus & South Ohio Elec.Co.	Conesville#4-6	842/403	73/77
Brilliant	Ohio Power Co.	Cardinal#3	615	76
Cheshire		Gavin#1&2	1,500/1,300	74/75
Aberdine	Dayton Power & Light Co.	J.M.Stuart#4	580	74
Wisconsin				
Portage	Wisconsin Power & Light Co.	Columbia#1	527	75
WEST NORTH CENTRAL				
Iowa				
Council Bluffs	Iowa Power & Light Co.	Council Bluffs#3	600	79
Sioux City	Iowa Public Service Co.	Neal#3	520	76

TABLE APP.13-B2 (continued)

Region / State / City	Company	Plant	Capacity MW	In Service Date (19--)
Kansas				
Linn County	Kansas Gas & Electric	La Cygne#1&2	840/630	73/77
Topeka	Kansas Power & Light Co.	Unassigned	700	73
Minnesota				
NA	Northern States Power Co.	Base Load#1	800	79
Becker		Sherbourne#1&2	680/720	76/77
Missouri				
New Madrid	Associated Electric Corp.	New Madrid#2	600	77
Labadie	Union Electric Co.	Labadie#4	582	73
Crystal City		Rush Island #1&2	592/592	75/76
Nebraska				
NA	Nebraska Public Power District	Gentleman	600	77
North Dakota				
Center	Minnkota Power Coop Inc.	Milton Young	400	77
Stanton	Basin Electric Power Corp.	Leland Olds#2	460	75

South Dakota				
Grant	Otter Tail Power Co.	Big Stone#1	430	75
SOUTH ATLANTIC				
Florida				
Pensacola	Gulf Power Co.	Crest#7	516	73
Tampa	Tampa Electric Co.	Big Bend#2	425/425	73/75
Georgia				
Centerville	Georgia Power Co.	Bowen#3&4	910/910	74/75
Carrollton		Wansley#1&2	910/952	76/77
Newman		Yates#6&7	725	74
Maryland				
Morgantown	Potomac Electric Power Co.	Morgantown#3	550	79
North Carolina				
North Winston	Duke Power Co.	Belews Creek#1&2	1,143/ 1,143	74/75
Roxboro	Carolina Power & Light Co.	Roxboro	720/72	73/76
Virginia				
Yorktown	Virginia Electric & Power Co.	Yorktown#3	845	74

TABLE APP.13-B2 (continued)

Region State City	Company	Plant	Capacity MW	In Service Date (19--)
West Virginia				
Scary	Appalachian Power Co.	J.E.Amos#3	1,300	73
Haywood	Monongahela Power Co.	Harrison#2&3	650/650	73/75
Mt. Storm	Virginia Elec.& Power Co.	Mt. Storm	575	73
EAST SOUTH CENTRAL				
Alabama				
Jefferson Co.	Alabama Power Co.	West Jefferson #1&2	880/880	73/79
Wilsonville	So. Electric Generating Co.	Gaston#5	910	74
Kentucky				
Maysville	East Kentucky Rural Elec. Coop.	Ohio River#1	300	76
Sebree	Henderson Municipal Utilities	No.2#1&2	334	73
Ghent	Kentucky Utilities Co.	Ghent#1&2	500/500	74/77
Louisville	Louisville Gas & Elec.	Mill Creek#2-4	336/425	74/77

Mississippi				
Gulfport	Mississippi Power Co.	Watson#5	520	73
Tennessee				
Cumberland City	Tennessee Valley Authority	Cumberland#2	1,275	73
WEST SOUTH CENTRAL				
Louisiana				
NA	Central Louisiana Elec. Co.	Unassigned	550	78
Oklahoma				
Muskogee	Oklahoma Gas & Elec.	Muskogee#4&5	550/550	77/73
Texas				
Cason	Southwestern Elec.Power Co.	Cason#1&2	530/528	77/73
Tatum Rusk Co.	Texas Utilities Co.	Martin Lake #1&2	750/750	76/77
Mt. Pleasant		Monticello #1&2	575/575	74/75
MOUNTAIN				

TABLE APP. 13-B2 (continued

Region / State / City	Company	Plant	Capacity MW	In Service Date (19--)
Arizona				
Page	Salt River(AIP) District Project	Navajo#1-3	750/750/750	74/75/76
Montana				
Colstrip	Montana Power Co.	Colstrip#1-4	700/700	73/79
Nevada				
NA	Nevada Power Co.	Arrow Canyon #1&2	500/500	77/78
New Mexico				
Farmington	Public Service Co.of N.M.	San Juan#1-3	500	73
Utah				
Huntington	Utah Power & Light Co.	Huntington Canyon #1&2	430/400	74/74
Wyoming				
Rock Springs	Idaho Power Cp	Bridget#1-3	500/500/500	74/75/76

Washington / Centralia	Pacific Power & Light Co.	Centralia#1&2	700	73

TABLE App.13-B3

OIL FIRED PLANTS CONVERTIBLE TO COAL [a]

Region / State / City	Company	Plant	Capacity Convertible MW
NEW ENGLAND STATES			
Connecticut			
Devon	Connecticut Light & Power Co.	Devon	273
Montville		Montville	162
Norwalk		Norwalk Harbor	325
Middletown	Hartford Electric Light Co.	Middletown	422
Hartford	"	South Meadow	105
MAine			
Wiscasset	Central Maine Power Co.	Mason	69
Massachusetts			
Holyoke	Holyoke Water Power Co.	Mt. Tom	136
Somerset	Montaup Electric Co.	Somerset	325
Somerset	New England Electric Co.	Brayton Point	1,162
Salem		Salem Harbor	200
W.Springfield	Western Mass.Electric Co.	W.Springfield	209
New Hampshire			
Portsmouth	Public Service Co. of N.H.	Schiller	100
Rhode Island			
Providence	Narragansett Electric Co.	South Street	54

[a] Source: Draft Report of the Oil Savings Work Group [ ].

MIDDLE ATLANTIC STATES

New Jersey			
Deepwater	Atlantic City Electric Co.	Deepwater	202
Beesleys		England	299
Sayreville	Jersey Central Power & Light Co.	Sayreville	248
S. Amboy		Werner	60
Holland TWP		Gilbert	126
Ridgefield	Public Service Electric & Gas Co.	Bergen	650
Burlington		Burlington	481
Jersey City		Hudson	455
Kearney		Kearney	294
Sewaren		Sewaren	431
Vineland	Vineland Electric Util.	Down	61
New York			
New York	Consolidated Edison Co.	Arthur Kill	826
New York		Astoria	1,455
New York		Ravenswood	1,000
Island Park	Long Island Lighting Co.	Barrett	187
Bethlehem	Niagara Mohawk Power Co.	Albany	400
Tomkins Cove	Orange & Rockland Util.	Lovett	495
Pennsylvania			
W. Norristown	Philadelphia Electric Co.	Barbadoes	148
E. Pikeland		Cromby	230
Philadelphia		Delaware	272
Peach Bottom		Southwark	378

TABLE App. 13-B3 (continued)

Region State City	Company	Plant	Capacity Convertible MW
SOUTH ATLANTIC STATES			
Delaware			
Delaware City	Delmarva Power & Light Co.	Delaware City No.3	38
Wilmington		Edge Moor	389
Georgia			
Brunswick	Georgia Power Co.	McManus	140
Maryland			
Baltimore	Baltimore Gas & Electric Co.	Crane	400
Baltimore		Wagner	268
Vienna	Delmarva Power & Light	Vienna	67
Aquasco	Potomac Electric Power, Co.	Chalk Point	728
Newburg		Morgantown	1,114
North Carolina			
Wilmington	Carolina Power & Light Co.	Sutton	671
Virginia			
Chester	Virginia Electric Power Co.	Chesterfield	1,485
Chesapeake		Portsmouth	650
Dumfries		Possum Point	491
Yorktown		Yorktown	375

OTHER STATES

Colorado			
Denver	Pub.Serv.Co.of Colorado	Zuni	38
Illinois			
Stickney	Commonwealth Edison Co.	Ridgeland	690
Kansas			
Lawrence	Kansas Power & Light Co.	Lawrence	87
Tecumseh		Tecumseh	231
Michigan			
River Rouge	Detroit Edison Co.	River Rouge	283
E. China Twp.		St. Clair	358

APPENDIX 13-C

TABLE APP.13-C1

NATIONAL AIR SURVEILLANCE NETWORKS

URBAN SULFATE OBSERVATIONS

ANNUAL AVERAGE SULFATE CONCENTRATION, $\mu g/m^3$

Station	Year 1969	1970
Bridgeport, Conn.	11.9	11.6
Hartford, Conn.	12.1	13.9
New Haven, Conn.	20.0	20.9
Waterbury, Conn.	13.6	15.1
Newark, Del.	n.a.	13.0
Wilmington, Del.	19.2	13.7
Washington, D.C.	13.9	--
Baltimore, Md.	13.9	19.8
Portland, Maine	--	17.0
Boston, Mass.	15.3	--
Fall River, Mass.	12.4	14.9
Springfield, Mass.	8.5	12.5
Worchester, Mass.	13.0	15.5
Concord, N.H.	6.5	9.2
Burlington Co., N.J.	13.2	10.3
Camden, N.J.	22.0	17.2
Elizabeth, N.J.	11.5	13.2
Glassboro, N.J.	13.4	12.6
Hamilton, N.J.	9.9	--
Jersey City, N.J.	12.1	14.5
Newark, N.J.	11.2	11.8
Paterson, N.J.	12.7	12.7
Perth Amboy, N.J.	13.3	9.9
Trenton, N.J.	12.8	15.0
Albany, N.Y.	9.4	--
Buffalo, N.Y.	11.4	16.9
New York City, N.Y.	19.1	22.2

Station	Year 1969	1970
Niagara Falls,N.Y.	12.5	18.1
Rochester, N.Y.	14.1	14.9
Syracuse, N.Y.	16.3	9.3
Utica, N.Y.	7.7	9.0
Yonkers, N.Y.	--	12.3

TABLE App. 13-C2

NASN SULFATE OBSERVATIONS - NONURBAN STATIONS

Station	AVERAGE ANNUAL SULFATE CONCENTRATION, $\mu g/m^3$		
	1968	1969	1970
Kent County, Delaware	9.5	n.a.	n.a.
Monroe County, Indiana	9.7	7.9	7.5
Park County, Indiana	6.9	8.5	13.5
Acadia Nat. Park, Maine	5.8	4.9	7.0
Calvert County, Maryland	14.1	9.8	n.a.
Coos County, New Hampshire	7.3	3.4	6.5
Jefferson County, New York	10.0	9.1	9.4
Clarion County, Pennsylvania	9.8	9.1	12.4
Washington County, Rhode Island	10.5	9.1	7.7
Orange County, Vermont	7.5	5.8	8.0
Shenandoah Nat. Park, Virginia	8.1	13.3	9.2

Source: 1968 data: U.S. EPA publication, "Air Quality Data from the National Air Surveillance Networks and Contributing State and Local Networks."

1969-1970 data: Air Quality Data for Nonmetallic Inorganic Ions, 1969 and 1970, National Air Surveillance Networks, APTD-1466, U.S. Environmental Protection Agency, June 1973.

TABLE APP.13-C2(Cont.)

NASN URBAN SITES

ANNUAL AVERAGE SULFATE CONCENTRATION $\mu g/m^3$

Station	1969	1970
Canton,Ohio	11.9	16.7
Cincinnati,Ohio	15.1	12.4
Cleveland,Ohio	15.9	18.0
Columbus, Ohio	14.4	--
Dayton, Ohio	15.6	11.9
Toledo, Ohio	10.2	12.9
Youngstown, Ohio	11.8	16.8
Allentown, Pa.	16.8	20.4
Altoona, Pa.	10.2	30.2
Bethlehem, Pa.	11.2	21.0
Erie, Pa.	15.6	16.5
Harrisburg, Pa.	10.2	16.0
Hazelton, Pa.	9.9	--
Johnstown, Pa.	15.3	16.9
Philadelphia, Pa.	--	21.9
Pittsburgh, Pa.	--	17.9
Reading, Pa.	--	18.6
Scranton, Pa.	--	13.9
Warminster, Pa.	--	10.9
Wilkes-Barre, Pa.	--	13.7
York, Pa.	--	14.3
East Providence, R.I.	15.5	12.0
Providence, R.I.	11.9	12.9
Burlington, Vt.	10.9	12.0
Charleston, West Va.	25.8	25.0
So. Charleston, West Va.	--	15.5

Source: Air Quality Data for Nonmetallic Inorganic Ions, 1969 & 1970, National Air Surveillance Networks, APTD-1466, U.S. Environmental Protection Agency, June 1973.

APPENDIX 13-D

COST CALCULATIONS FOR ELECTRIC POWER GENERATION WITH AND WITHOUT FLUE GAS DESULFURIZATION

Since calculations have not been included elsewhere in the report, this appendix is provided to give a detailed basis for the cost of flue gas desulfurization using the lime scrubbing process. These costs are used in Table 13-5 of this chapter, in the Executive Summary, and in Part Two in Brief. The calculations assume the use of coal of approximately 3 percent sulfur and a plant of moderate size, approximately 600 MW. Fixed charges, including amortization, of 17 percent on invested capital are assumed for new investment, and fixed charges on existing plant capacity are computed at 14 percent. Other needed assumptions are as specified in Tables 13-1 and 13-3. All numbers are rounded to the nearest 0.1 mill, so some columns do not sum exactly.

TABLE App. 13-D1

Cost Calculation for New Plant

Without flue gas desulfurization:

capital cost: \$500/kw x 0.17 ÷ 7000 hours	12.1 mills/kwh
fuel cost: heat rate of 8,982 x \$1/MM BTU of fuel	8.9
other operating costs:	0.5
Cost of power generation without scrubber	21.6 mills/kwh

Added cost of lime scrubbing process:

capital cost: \$100/kw x 0.17 ÷ 7000 hours	2.4 mills/kwh
operating cost: labor, chemicals, etc.	0.5
sludge disposal	0.3
Subtotal	3.2 mills/kwh
energy loss of 6% of plant output needed for scrubber operation:	0.6
capacity derating of plant resulting from 6% loss in output capacity:	0.7
Subtotal	1.3
added cost of lime scrubbing process	4.5 mills/kwh

Cost of power generation with scrubber: 26.2 mills/kwh

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TABLE App. 13-D2

New Plant: Sensitivity Calculations on Added Cost from Scrubber

Low case:	
$60/kw; 0.15 mills/kwh for sludge disposal	2.1 mills/kwh
5% energy loss and capacity derating	1.1
Added cost of scrubber, low case:	3.2 mills/kwh
High case:	
$130/kw; 1.0 mills/kwh for sludge disposal	4.7 mills/kwh
7% energy loss and capacity derating	1.5
Added cost of scrubber, high case:	6.2 mills/kwh

TABLE App. 13-D3

Cost Calculation for Retrofit to Existing Plant, Rural Location

Without flue gas desulfurization:	
capital cost $250/kw x 0.14 ÷ 6000 hours	5.8 mills/kwh
fuel costs: heat rate of 10,342 x $1/MM BTU of fuel	10.3
other operating costs	1.0
Cost of power generation without scrubber	17.2 mills/kwh
Added cost of lime scrubbing process:	
capital cost of $125/kw x 0.17 ÷ 6000 hours	3.5 mills/kwh
operating cost: labor, chemicals, etc.	0.6
sludge disposal	0.5
Subtotal	4.6 mills/kwh
energy loss, 6% of plant output needed for scrubber operation	0.7
capacity derating of 6% (replacement at $500/kw, 17% fixed charge)	0.8
Subtotal	1.5
added cost of lime scrubbing process	6.1 mills/kwh
Cost of power generation with scrubber:	23.3 mills/kwh

TABLE App. 13-D4

Cost Calculation for Retrofit Installation, Existing Oil Burning Plant Reconverted to Coal, Urban Location

Without flue gas desulfurization:

capital cost \$250/kw x 0.14 ÷ 6000 hours	5.8 mills/kwh
conversion charges of \$7/kw plus \$4/kw for particulate removal upgrade	0.3
fuel cost: heat rate of 10,342 x \$1/MM BTU of fuel	10.3
other operating costs	1.0
Cost of power generation without scrubber	17.5 mills/kwh

Added cost of lime scrubbing process:

capital cost of \$125/kw x 0.17 ÷ 6000 hours	3.5 mills/kwh
less credit of \$4/kw for particulate upgrade	- .1
operating cost: labor, chemicals, etc.	0.6
sludge disposal	0.9
Subtotal	4.9 mills/kwh
energy loss, 6% of plant output needed for scrubber operation	0.7
capacity derating of 6% (replacement at \$500/kw, 17% fixed charge)	0.8
Subtotal	1.5
added cost of lime scrubbing process	6.4 mills/kwh

Cost of power generation with scrubber: 23.9 mills/kwh

APPENDIX 13-E

COMMENTS ON ESTIMATES OF MATERIAL DAMAGE

(Written by I.C.T. Nisbet, a member of the Review Committee on Air Quality and Power Plant Emissions.)

In their discussion of damage to man-made materials attributable to sulfur dioxide and sulfates, North and Merkhofer (Chapter 13) relied for numerical estimates upon the review by Waddell 1974. The following comments are offered not in criticism of the work of Waddell, North and Merkhofer, who used the best data available, but to point out that the available data are severely limited and that Waddell's estimates of damage may be very conservative.

Waddell (1974, pp. 90-91, 128-130) estimated that the total damage caused by sulfur oxides to material property in 1970 was about $600 million (with a possible range of $400-800 million). This estimate was based primarily on studies by Gillette (1973) of damage to galvanized and painted steels, by Spence and Haynie (1972) of damage to other painted surfaces, and by Salmon (1970) of damage to non-ferrous metals and miscellaneous materials.

DAMAGE TO GALVANIZED AND PAINTED STEELS

Gillette's (1973) estimates were based on the assumption that corrosion rates are proportional to average ambient concentrations of sulfur dioxide, and in the case of galvanized steels are also dependent on the relative humidity. Accordingly Gillette deduced that most of the damage to these materials was taking place in urbanized regions of the northeastern and north-central states, and that damage has been decreasing rapidly as ambient sulfur dioxide levels have been reduced. His estimates of total annual damage in the United States fell from $909 million in 1968 to $397 million in 1970 and to $75 million in 1972 (Gillette 1973, Table IV).

However, it is probable that some of the damage should be attributed to acid sulfate particulates and/or to sulfuric acid in rain. As pointed out by Gillete himself (1973, p. 4), the agent primarily responsible for damage is probably sulfuric acid, either formed by oxidation in situ after absorption of sulfur dioxide onto moist surfaces, or deposited in acid particulates or acid precipitation. If even half of the damage were attributable to sulfate particulates or acid rain, Gillette's deduction of a drastic decrease in damage between 1968 and 1972 would be incorrect and his estimate for 1970 would be too low. Moreover if acid rain were a significant cause of corrosion, significant damage would also be predicted to occur to rural areas and in smaller towns, where Gillette's damage function predicts negligible damage because of low ambient sulfur dioxide levels.

It should be noted also that Gillette's estimate of about $300 million for total damage to galvanized materials in the U.S. in 1970 is much lower than estimates made independently by Salmon (1970: $778 million) and by Fink et al. (1972: $1,350 million). A study in Sweden (Bolin et al. 1971) gave rise to estimates more similar to those of Gillette -- about $16 million per year, or $2 per capita -- for galvanized steel. However, the Swedish study yielded estimates of damage to painted steels substantially larger than those derived by Gillette -- about $25 per capita annually in Sweden, of which roughly 10 percent was attributable to sulfur oxides (Bolin et al. 1971: Tables 4.7 and 7.4), versus Gillette's estimate of $0.88 per capita for the U.S.

DAMAGE TO OTHER PAINTED MATERIALS

Waddell (1974, p. 128) used a figure of $200 million for damage caused by air pollutants to other painted materials, based on a study by Spence and Haynie (1972). However, Spence and Haynie actually estimated total losses as $700 million, of which $540 million was attributable to deterioration of household exterior paints (Waddell 1973, Table 14). Hence Waddell appears to have overcorrected for overlap between this and Gillette's study, which considered only painted metal surfaces.

Moreover, Spence and Haynie's estimates of damage appear to be based on very low figures for the total value of paintwork at risk from air pollution. Their table indicates that the total in-place value of exterior paints in the categories considered (including the labor value of painting) was only $2.5 billion, of which household exterior paints accounted for only $1.5 billion. This contrasts with a figure of $23.9 billion used by Salmon (1970) for the in-place value of exposed paints in the United States, and a figure of $2.9 billion for the total expenditure of residential property owners on maintenance painting in 1972 alone (Stat. Abstract U.S. 1973).

In addition, Waddell's attribution of half the total damage to sulfur oxides and half to particulates appears somewhat arbitrary. Apart from soiling and staining, most particulate matter would not be expected to damage paint, except insofar as it retains materials such as acid sulfates and nitric acid and thereby helps to maintain their contact with painted surfaces.

Waddell's final estimate of $100 million for the total amount of damage caused by sulfur oxides to painted surfaces corresponds to costs of no more than $0.80 per capita per year, even if all the damage is supposed to occur in the northeastern states. This is less than the estimates derived from the Swedish study, in which the total cost of deterioration of painted woodwork was estimated as $23 per capita in 1970, of which at least 10 percent was associated with exposure to sulfur oxides (Bolin et al. 1971: Tables 4.6 and 7.4).

DAMAGE TO NON-FERROUS METALS

Based on the study by Salmon (1970), Waddell (1974, pp. 90-91) adopted a figure of $400 million for the cost of air pollution damage to other materials, primarily non-ferrous metals. Of this, he ascribed one-quarter to sulfur oxides, one quarter to particulates, and one-half to oxidants (Waddell 1974, pp. 128-130). This division appears questionable because others have attributed accelerated corrosion of metals primarily to sulfur oxides and acids (Salmon 1970, Bolin et al. 1971). Moreover Waddell excluded damage to copper, which is classically associated with air pollution.

DAMAGE TO AUTOMOBILES

None of the studies cited above explicitly considered damage to motor vehicles, except that Spence and Haynie (1972) included a figure of $88 million per year for the excess cost of repainting automobiles. However, it is questionable that the cost of repainting is an adequate measure of the economic damage caused by accelerated corrosion. The resale value of automobiles is substantially influenced by exterior appearance, so that accelerated deterioation of paint or metal-plated trim contributes disproportionally to depreciation. The number of motor vehicles in the area most exposed to sulfur oxides and acid rain (the northeastern U.S. and southeastern Canada) is in excess of 60 million; their current value is greater than $75 billion, and rates of depreciation are of the order of 20-30 percent annually If air pollution caused even 1 percent of the depreciation in value, this would represent a substantial economic cost not included in the other surveys.

SUMMARIZING COMMENTS

The above discussion illustrates the large uncertainties involved in making estimates of economic damage caused by air pollution from the very limited data available. Waddell's estimate of total damage, although based on a thorough and comprehensive review, is among the lowest of a range that could be obtained from the same data; an independent review suggests that a figure 4 or 5 times higher would be more consistent with the original sources. The uncertainty is further compounded by our lack of knowledge of exposure-response relationships, and of the relative importance of sulfur dioxide, suspended acid sulfates, and acid precipitation. Clearly this is another area in which the value of resolving uncertainties would exceed the cost of the research required to do so.

DAMAGE TO WORKS OF ART AND HISTORIC BUILDINGS

Sulfur oxides and acid rain also cause damage to works of art, historic buildings and monuments. The damage is almost impossible to assess in economic terms because such objects have historic or artistic value far greater than the cost of replacement; many historic objects have no market value at all. Information supplied by curators of museums and buildings suggests that the materials at greatest risk are paper (rare books, manuscripts, and prints), building stone (limestone and marble), and outdoor sculptures (bronze, steel, and stone). Although it is possible in principle to alleviate damage by protective measures -- such as air-conditioning, cleaning and restoration, or protective coatings -- in practice insufficient money is available for such measures and many important buildings, monuments, and collections are unprotected. Unless a method can be devised to finance the restoration and preservation of these historic and cultural objects, increased emissions of sulfur oxides are likely to speed their deterioration. Their loss should be regraded as a major intangible cost of sulfur oxide emissions, to be weighed in conjunction with the more tangible costs and benefits.

APPENDIX 13-F

INCOME REDISTRIBUTION AND EQUITY[1]

As noted briefly by North and Merkhofer (See Chapter 13), the assessment of alternative strategies for pollution control may need to address issues of equity and distribution. The burden imposed by pollution falls unequally on individuals within society, and the various strategies under consideration would redistribute this burden in different ways. Some -- although not all -- of these redistributional effects can be predicted, and should be taken into account in choosing between the alternative strategies. If the strategy which is identified as socially optimal (on the basis of a comparison between total costs and total benefits to society) involves predictable adverse effects on identifiable individuals or groups within the population, some compensatory actions may be desirable. Alternatively, it may be justified to choose a strategy which is less than socially optimal in the strict economic sense, in order to minimize social inequities. Such questions of social policy are outside the purview of this committee, but it is nevertheless important to point out what is known about redistributional effects.

The adverse effects of sulfur oxides on human health are expected to fall primarily on residents of urban areas in the northeast, where the highest ambient concentrations of both sulfur dioxide and suspended sulfates are found (Chapters 1-4, 6). Similarly, the costs imposed by damage to materials are believed to fall primarily on property owners

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[1] By I.C.T. Nisbet

in northeastern cities (Appendix Chapter 13-E). Except for localized damage to vegetation close to some point sources, effects in rural areas appear to be generally smaller. However, the exact magnitude of the urban-rural difference in impact is difficult to specify, because of our lack of knowledge of exposure-response relations. For example, if the threshold level for health effects of suspended sulfates is low or zero, substantial effects might be occurring in rural areas of the northeast where ambient concentrations of sulfates are moderately high (Chapter 6). Similarly, if a substantial fraction of the damage to materials is caused by acid rain, it would be taking place in rural areas as well as in cities (Chapter 7).

It is not clear whether the costs of pollution would fall disproportionately on different income groups. To the extent that health and material damage effects fall on residents of cities, they might fall disproportionately on the poor. Also, many of the health effects fall on individuals who are old or chronically sick. However, these differentials are offset by the facts that suburban residents are also exposed to high pollution levels in some areas, and that the rural poor are relatively unaffected. More information is required to resolve this question.

In addition to the urban-rural differences in impact discussed above, there are probably major regional inequities. Chapters 6 and 7 show that sulfur oxides emitted in the east-central and midwestern states contribute significantly to airborne sulfate particulates and acid rain in the northeastern states. A special case of this regional distribution problem is the likely impact of U.S. emissions on eastern Canada. The urbanized region between Windsor and Montreal lies downwind from major emitting areas in the north-central U.S.; it is probable that the high concentrations of sulfates in rain in this region of Canada are attributable in large part to U.S. emissions, since Canadian emissions are comparatively modest (with the exception of the Sudbury smelting complex well to the northwest) (See Chapter 7). Likewise the prospective impact of acid rain on forestry would be most likely to be economically significant in eastern Canada (Chapter 5).

A general increase in sulfur oxide emissions (as would accur if a Tall Stack-Intermittent Control strategy were widely adopted) is expected to lead to a general increase in ambient concentrations of sulfur dioxide and suspended sulfates , and an increase in impacts in rural areas. However, the models in Appendix A to Chapter 13 indicate that the highest concentrations of suspended sulfates would continue to occur in urban areas, even if emissions upwind were confined to rural sites. Accordingly the urban-rural difference in impacts would probably be maintained without major change. The regional differences would probably be maintained similarly.

Under an emission control strategy (low sulfur fuels and/or flue gas desulfurization) the increase in total exposure to sulfur oxides and sulfates would be limited, and ultimatly reversed if the policy were sufficiently widely implemented. The immediate effect would be to pass on the cost of emission control to consumers in the form of higher prices. This would help to offset the existing regional inequities, because consumers in the central states and in rural and suburban areas in the northeast would be assuming part of the social cost of generating their power that would otherwise be borne by residents in cities downwind. However, it would probably have a regressive income distributional effect, because the poor spend a higher fraction of their income on electricity than the wealthy (Ford Foundation 1974), and so would be affected disproportionately by higher prices.

The distributional effects of the adoption of marginal cost pricing for electricity also need to be considered. To the extent that it would lead immediately to higher prices, its effects are expected to be regressive, as discussed in the previous paragraph. However, this would be offset if the pricing policy involved reduction of the higher rate for the initial amount of electricity used, as might be justified on the grounds that demand for electricity in that sector of the market is inelastic. Peak load pricing (or time-of-day metering) might also offer advantages to poorer individuals, because it permits cost savings by those with sufficient incentive to do so.

To summarize, an increase in emissions would lead to an increase in external impacts, probably without major change in the pattern of inequities. Adoption of an emission control strategy would relieve existing urban-rural, regional, and international inequities. However, it would probably have a regressive income distributional effect, unless accompanied by a pricing policy designed to reflect price elasticity of demand.

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PART THREE
CONTROL OF NITROGEN OXIDES
FROM STATIONARY SOURCES

Part Three was prepared under the direction of the Commission on Natural Resources of the National Research Council. The discussions of nitrogen oxide sources in Chapter 14 and of tall stacks and intermittent control for nitrogen oxides in Chapter 15 are based on analyses by John Spengler, Anthony Cortese, and Douglas Dockery of the Harvard School of Public Health. The examination of control techniques in Chapter 15 is based on the work of Adel Sarofim and Richard Flagan of the Massachusetts Institute of Technology.

CHAPTER 14

NITROGEN OXIDE EMISSIONS AND THEIR DISTRIBUTORS

INTRODUCTION

Nitrogen Compounds

Nitrogen, the most abundant gas in the atmosphere, is found in a variety of gaseous and particulate forms. The overwhelming amount in air (79 percent of air by volume or 4.6×10^{12} tons) is present as relatively inert nitrogen gas, N_2. However, the oxidation of nitrogen by bacteria, lightning, organic protein decay, and high temperature combustion and chemical processing causes the appearance of nitrogen in a variety of compounds. The most important, because of health effects and reactivity, are: NO (nitric oxide), NO_2 (nitrogen dioxide), NH_3 (ammonia) and to a lesser extent N_2O (nitrous oxide).

Nitrous Oxide (N_2O)

Nitrous oxide, a colorless and odorless gas, has been used as an anesthetic (laughing gas). It is present in the atmosphere in concentrations averaging about 0.25 ppm (Junge 1963, Bates and Hays 1967). There are no direct pollutant sources of N_2O, although it may be an indirect and minor product of NO_2 photolysis with sunlight and hydrocarbons. The interest in N_2O is not in the troposphere (ground level to 8-15 km) where it is practically inert, but in photodissociation reactions in the stratosphere. Bates and Hays (1967) indicate that the dissociation of N_2O into NO and atomic nitrogen accounts for about 20 percent of the dissociation in the stratosphere. The NO thus formed provides an important sink reaction for ozone.

Ammonia (NH_3)

As an industrial emission, ammonia is produced mainly from coal and oil combustion but natural production from biological generation over land and ocean is many times greater than that from anthropogenic sources (250 to 1). NH_3's importance is the significant role it plays in atmospheric reactions in both the nitrogen and sulfur cycles. Nearly three-fourths of the NH_3 is converted to ammonium ion condensed in droplets or particles. These aerosols are then subject to the physical removal mechanisms of coagulation, washout, rainout and dry deposition.

In general, ambient background concentrations of NH_3 vary directly with the intensity of biological activity. The highest concentrations occur in the summer and in the tropical latitudes. Concentrations, as reported by many investigators, range from 1 to 10 ppb (Strauss 1972).

Nitric Oxide-Nitrogen Dioxide

NO, a colorless, odorless gas, is formed naturally from the nitrates in various materials by bacteria and then is oxidized to NO_2 (Peterson 1956).

Altschuller (1958) and others have reported very hazardous conditions for farm workers near closed silos where $NO \rightarrow NO_2$ bacterial production has resulted in toxic concentrations of several hundred ppm of NO_2.

Organic nitrogen compounds are found in

coal and oil in concentrations of a few tenths to a few percent by weight. Bituminous coal contains 1-2 percent nitrogen, and United States crude oil approximately 0.05-0.5 percent nitrogen (Demski et al. 1973).

Natural gas, while containing up to 4 percent nitrogen gas, does not contain any significant organic nitrogen (Perry et al. 1963). Because organic nitrogen compounds have relatively high molecular weights, they tend to be concentrated in the residual and heavy oil fractions during distillation.

Nitrogen oxides are produced during combustion by the oxidation of organic nitrogen compounds in fossil fuels and by the thermal fixation of atmospheric nitrogen gas, N_2.

The primary sources of NO and NO_2 as pollutants are combustion processes in which temperatures are high enough to fix N in the air and fuel, and in which the quenching of combustion is rapid enough to reduce decomposition back to N_2 and O_2. The predominant product of this high temperature combustion is NO. During combustion, approximately 5-40 percent of the nitrogen in coal, and 20 to 100 percent of the nitrogen in oil is oxidized to nitrogen oxides (see Chapter 15).

NO is subsequently oxidized to NO_2 either in the stack gas or, to a lesser extent, in the diluted plume. Once the NO has been diluted to 1 ppm (1230 $\mu g/m^3$) or less, the direct reactions with O_2 do not contribute significantly to NO_2 formation (USEPA 1971). However, the reaction of NO with tropospheric ambient concentrations of O_3 (ozone) to form NO_2 is rapid. It is believed that the almost everpresent background concentrations of O_3 will yield NO_2 predominance over NO, although some researchers have reported higher NO than NO_2 concentrations in remote areas (Lodge and Pate 1960, Ripperton et al. 1970).

NO_2 is removed from the atmosphere either by further O_3 oxidation to a nitrous salt or by the more favored conversion to HNO_3 in the presence of water vapor. The HNO_3 is then rapidly removed by reactions with NH_3 and absorption by hygroscopic particles (Strauss 1972).

Nitrogen Oxides

Global Emissions

To estimate the total annual emission of oxides of nitrogen (NOx), emission factors have been applied to the fuel usage of several sources (Robinson and Robbins 1968). According to Robinson and Robbins, the annual production in 1967 was about 53 x 10^6 tons with coal combustion contributing the majority, 51 percent, followed by petroleum production and combustion contributing 41 percent. Natural gas combustion on a world-wide basis is comparatively less important (4 percent): However, it should be noted that on a local or regional basis, it could be the major source of NOx. (It should also be noted that these figures include combustion sources only since careful surveys of industrial process losses had not been undertaken at the time these estimates were prepared.) See Table 14-1.

National Emissions

Anthropogenic sources in the United States produce nearly 50 percent of the world's NOx emissions (USEPA 1971). While emissions from human activities amount to far less than the estimated 50 x 10^7 tons of NOx emitted annually from natural sources (USEPA 1971), the spatial concentration of emissions in urban areas leads to concentrations of NOx 10 to 100 times higher than those in non-urban atmospheres.

Fuel combustion is the major cause of anthropogenic NOx emissions in the United States (See Figure 14-12). In 1972, coal, oil, natural gas and motor-vehicle fuel combustion contributed over 86 percent of the estimated 24.6 million tons of NOx emitted in the United States. Stationary area and point sources account for approximately 64 percent of all the NOx. Direct stationary fuel combustion is the largest source

TABLE 14-1

World-Wide Urban Emissions of Nitrogen Dioxide

Fuel	Source	NO_2 Emissions (10^6 Tons)	% Total	Sub %
TOTAL		52.9	100	
COAL			51	100
	Power Generation	12.2	23	47
	Industrial	13.7	26	52
	Domestic/Commercial	1.0	2	3
PETROLEUM			41	100
	Refinery Production	0.7	1	3
	Gasoline	7.5	14	34
	Kerosene	1.3	2	6
	Fuel Oil	3.6	7	16
	Residual Oil	9.2	17	41
NATURAL GAS			4	100
	Power Generation	0.6	1	25
	Industrial	1.1	2	50
	Domestic/Commercial	0.4	·1	25
OTHER				
	Incineration	0.5	·1	
	Wood	0.3	·1	
	Forest Fires	0.5	1	

Source: Modified from Robinson and Robbins (1968).

category (49.7 percent) with coal as the single largest contributor of NOx in this group.

Gasoline powered vehicles are the overwhelming source of transportation-related NOx contributing 32 percent of all NOx and 82 percent of the transportation NOx.

Significant quantities of NOx are emitted from industrial processes, primarily the manufacturing and use of nitric acid and refining of petroleum. On a local scale, electroplating, engraving, welding, and metal cleaning are responsible for industrial NOx emissions which may also be significant. In 1972, industrial process losses accounted for 2.9 million tons of NOx, or 11.7 percent of total nationwide emissions.

Overall, about 77 percent of the total NOx emissions occur in highly populated areas. Eighty percent of stationary source emissions occur in populated areas as do 71 percent of motor vehicle emissions.

The 1972 NOx emissions for the Nation will be defined in greater detail in the following section.

There has been a steady growth of NOx nationwide emissions. The decade of the sixties witnessed a greater increase in emissions than the previous two decades (see Table 14-2).

TABLE 14-2

NITROGEN OXIDES: Estimated Total Nationwide Emissions (10^6 tons) (USEPA 1974)

1940	1950	1960	1970
6.5	8.8	11.4	22.1

Over the past three decades, total nationwide emissions are estimated to have quadrupled. During this period, emissions from motor vehicles have increased at a steady rate of 4.6 to 4.9 percent per year. Emissions from stationary sources, however, have contributed progressively increasing proportions. Total NOx emissions from power plants have increased at an annual rate of 6.9 to 7.4 percent.

Nitrogen Oxide Concentrations

Controversy and uncertainty about NOx measurement methods have made reliable urban NO and NO_2 concentration data almost as scarce as the remote area data. Global remote measurements show NO_2 variations with growing season, latitude, and altitude. Lodge and Pate (1966) reported average dry season values of 0.9 ppb and wet season values at 3.6 ppb NO_2 in Panama. Junge reported in 1966 measured NO_2 concentrations averaging 0.9 ppb in Florida and 1.3 ppb at 10,000 foot high Mauna Kea, Hawaii (Junge 1956). In the continental U.S., several investigators found NO_2 values in the 4 ppb range and NO concentrations about 50 percent lower at 2 ppb (Hamilton et al. 1968, Ripperton et al. 1970).

Based upon the estimated global background levels and the annual emissions rates, the average residence time of NO_2 in the atmosphere is about 3 days and that of NO is about 4 days. Residence times of atmospheric pollutants reflect the action of natural scavenging processes including photochemical reactions. Figure 14-1 provides a flow diagram summary of the atmospheric NO-NO_2 cycle.

The spatial and temporal variations in ambient NO_2 concentrations are great. Not surprisingly, the highest concentrations are found in urban regions. Measurements of NO_2 have been taken since 1961 through the CAMP and SCAN programs of EPA (formerly NAPCA). However, there is now considerable uncertainty regarding ambient levels and trends for NOx. Although an EPA study of CAMP data for 5 cities reported slight increases in ambient NOx for 1962-1971, the data were obtained by the Jacobs-Hochheiser method for NOx analysis, which has been shown to overestimate ambient NOx levels at low concentrations (CEQ 1975). Thus, these NOx data must be viewed with caution. (See Appendix 14-B).

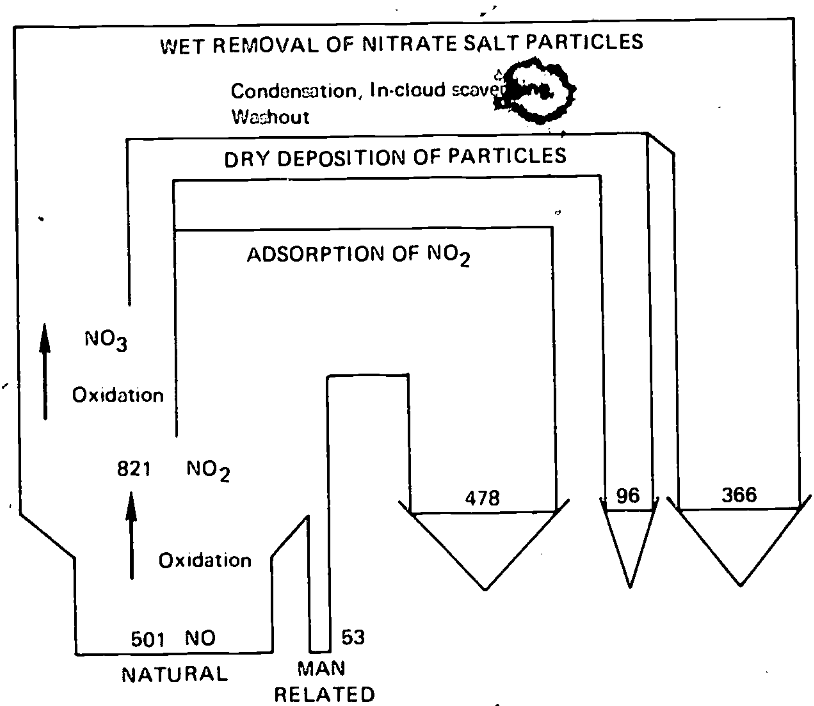


FIGURE 14-1: Nitrogen Oxide Cycle in the Atmosphere (10^6 Tons/Year).

The National Air Surveillance Network (NASN) program for measuring NOx by a modified Jacobs-Hochheiser method was started in 1972 and it will be several years before the program can provide useful trend data on ambient NOx levels.

EPA has suspended all Air Quality Control Regions priority classifications based on NOx. However, regions where the standards for NO_2 are believed to be exceeded are Los Angeles, Chicago, and Baltimore, and possible, New York-New Jersey-Connecticut, Salt Lake City, and Denver.

In spite of the uncertainties in the absolute values, there are NOx data available to indicate the general yearly trends in a few areas. The trends in CAMP observations of nitrogen dioxide are provided in Table 14-3. With the exception of Chicago, annual average nitric oxide concentrations of the period 1967-1971 are consistently higher than those of the period 1962-1966.

Annual nitrogen dioxide (NO_2) averages show a greater variability among cities than do the nitric oxide averages, and the trends for nitrogen dioxide do not parallel those of nitric oxide. It is not clear whether this deviation is a result of instrument variation, or whether it can be attributed to differences in atmospheric conversion rates in various cities (NAS 1974).

Monitoring data for NO and NO_2 in New Jersey cities presented in Figure 14-2 suggest that a pattern of change similar to that observed in the CAMP cities has occurred at these locations. Maximum monthly averages of nitric oxide appear to have increased after 1971, while the levels of NO_2 have remained essentially constant over the same period.

In Los Angeles County there has been a direct relationship between increasing NOx emissions and the reported annual average one hour concentrations of NO_2. Between 1965 and 1972, NOx emissions have increased in L.A. County at an annual average rate of 3.8 percent per year. The annual average of the maximum hourly average total NOx concentrations has increased approximately 3.2 percent per year from 1965 to

TABLE 14-3

Nitrogen Dioxide Trends in CAMP Cities 1962-71 (National Academy of Sciences 1974)

Station	Average NO concentration, ppm		Average NO_2 concentration, ppm	
	1962-66	1967-71	1962-66	1967-71
Chicago	0.10	0.10	0.04	0.05
Cincinnati	0.03	0.04	0.03	0.03
Denver	0.03	0.04	0.04	0.04
Philadelphia	0.04	0.53*	0.04	0.04
St. Louis	0.03	0.04	0.03	0.03

*The unusually high NO concentration for Philadelphia in 1967-71 is discussed in the section of this chapter on U.S. Nitrogen Oxide Emissions.

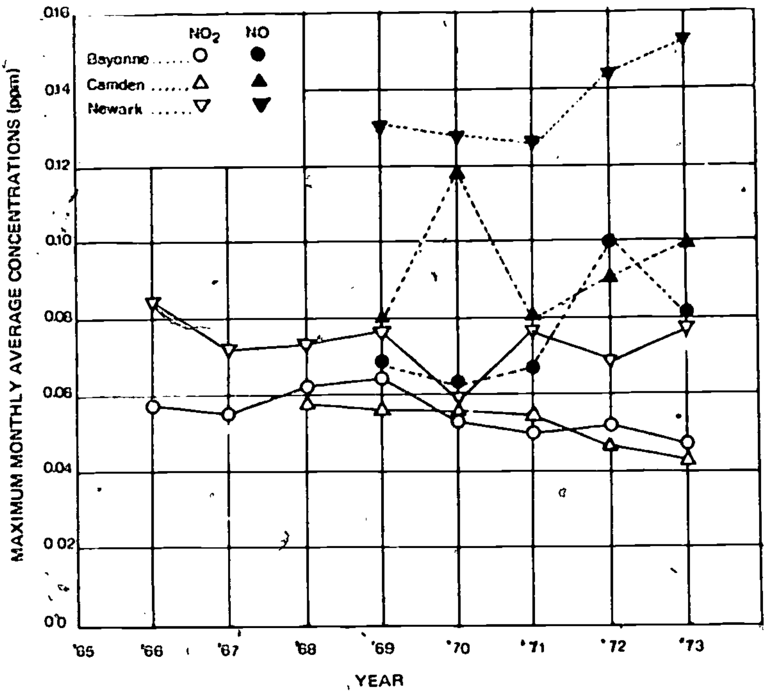


FIGURE 14-2: Maximum Monthly Average Concentrations of Nitrogen Oxides in Three New Jersey Cities (National Academy of Sciences 1974).

1972 at Burbank and downtown L.A. However, Anaheim and Azusa experienced increases in these annual averages of close to 11 percent per year.

There is more detailed discussion of NOx, hydrocarbon, and oxidant trends and relationships presented in Vol. 3, "The Relationship of Emissions to Ambient Air Quality" of the National Academy of Sciences Report, Air Quality and Automobile Emission Control, September 1974.

Diurnal Nitrogen Oxide Concentrations

The concentrations of NO and NOx in the ambient air change during a typical day. While all aspects of the typical diurnal cycle are not completely understood (Comm. of Mass, 1973, 1974; Coffey and Stasink 1975), it is possible to trace the concentrations of NO and NO_2 throughout the day.

On a normal day in a city, ambient NOx levels follow a regular pattern with the sun and traffic. Nocturnal levels of NO and NO_2 are relatively stable and are usually higher than the minimum daily values. During the dawn hours of 6 to 8 a.m., NO begins to increase as a result of automobile emissions. With increasing ultraviolet sunlight to drive the conversion reactions, NO_2 concentrations increase until most of the NO is converted to secondary NO_2. Photochemical oxidants accumulate as NO decreases to low levels (<0.1 ppm) and they reach a peak about midday. Secondary and tertiary reactions involving NO and NO_2 occur leading to complex formations of eye irritants such as PAN (peroxyacylnitrates).

Later in the afternoon, there is decreased mixing, and increased atmospheric stability. Evening rush hour traffic (4 to 7 p.m.) produces another build-up of NO which is not readily converted to NO_2 or more oxidants. The absence of sunlight does not completely halt NO_2 formation, however. The principal oxidant present, ozone (O_3), continues to react rapidly with NO to form NO_2, until the O_3 supply is exhausted, and thus the early evening NO_2 concentration may continue to rise. This condition may be ascribed to meteorological factors and, on cold evenings, to increased emissions from stationary sources.

A smoothed concentration profile for NO, NO_2, carbon monoxide (CO), and O_3 in Los Angeles on July 19, 1965, is displayed in Figure 14-3. The figure shows concentration on only a single day but it graphically displays the diurnal phenomenon described above. The NO_2 peak lags the NO peak. The build-up of O_3 is coincidental with the decrease of NO. The evening increase of NO_2 apparently did not occur on this day.

The classical diurnal trend is also apparent in Figure 14-4, which illustrates the diurnal variation in monthly mean 1-hour average NO_2 concentrations from St. Louis, Philadelphia, and Bayonne, N.J.

Figure 14-5 compares the diurnal patterns of NO for weekend days and weekdays (for Chicago CAMP stations). The Sunday 8 a.m. peak is about one-third of the weekday peak concentration. On some weekends, some locations have peak values, but these occur between 9 and 11 a.m.

The effect of stagnation conditions on the nitrogen oxide- photo-oxidant relationship is graphically exemplified in the diurnal concentrations of NO, NO_2 and oxidants during a stagnating air mass over Washington, D.C. (USDOC 1967). Figures 14-6 through 14-8 compare the diurnal concentration pattern for NO, NO_2 and oxidants that occurred during a four day stagnation in Washington, D.C., October 15 to 19, 1963, with a composite of the normal concentrations without stagnation and inversions. The efficiency of the NO-NO_2 conversion to photo-oxidants is obvious. During midday when solar energy is most intense, the photo-oxidation of NO and NO_2 in the presence of hydrocarbons is so efficient that their concentrations differ little from the norm. Details of these photochemical reactions are discussed in Appendix 14-A.

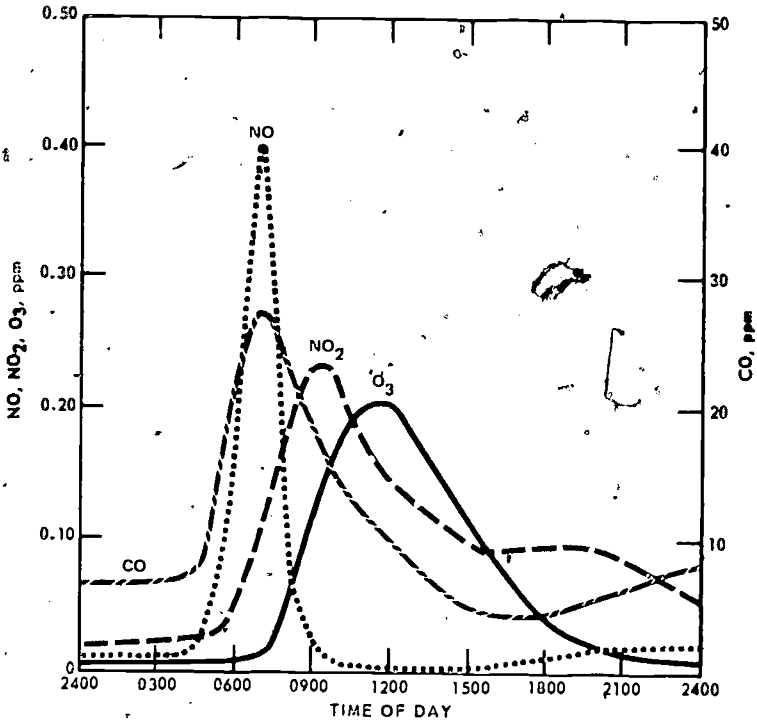


FIGURE 14-3: Average Daily 1-Hour Concentrations of Selected Pollutants in Los Angeles, California, July 19, 1965 (National Academy of Sciences 1974).

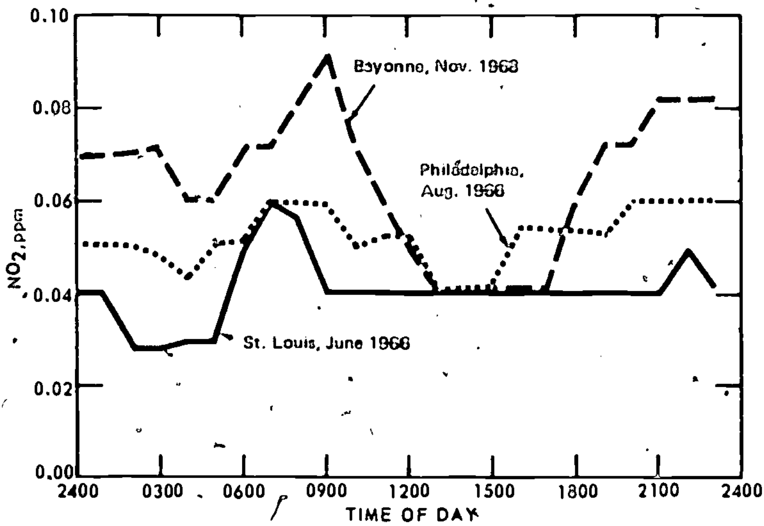


FIGURE 14-4: Diurnal Variation in Monthly Mean 1-Ho average NO_2 concentrations from three urban stations (Systems Development Corporation 1970).

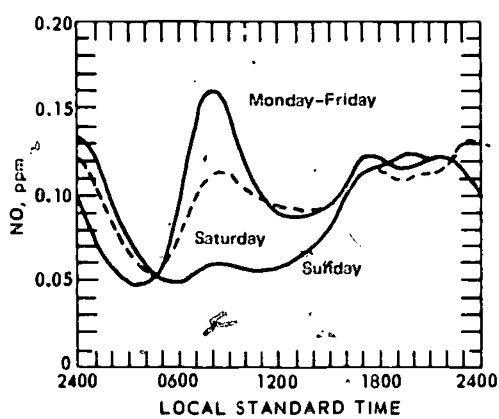


FIGURE 14-5: Weekday and Weekend 1-Hour Average NO Levels in Chicago, 1962 through 1964 (USDC 1967).

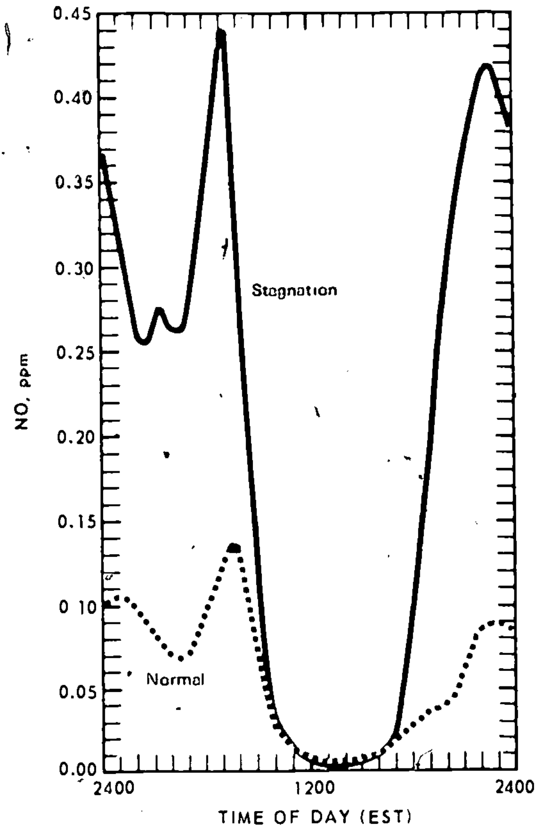


FIGURE 14-6: Diurnal Variation of NO Levels During 1963 Stagnation in Washington, D.C., October 15 through 19, 1963 (USDC 1967).

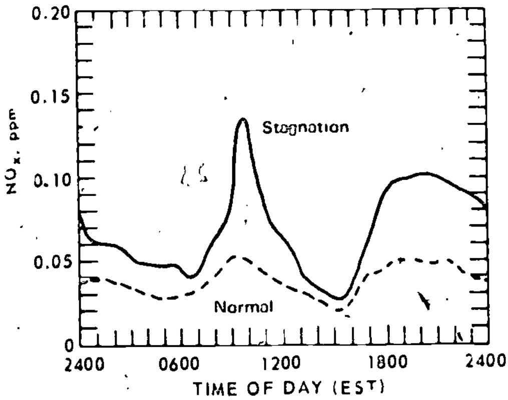


FIGURE 14-7: Diurnal Variation of NO_2 Levels During Stagnation in Washington, D.C., October 15 through 19, 1963 (USDC 1967).

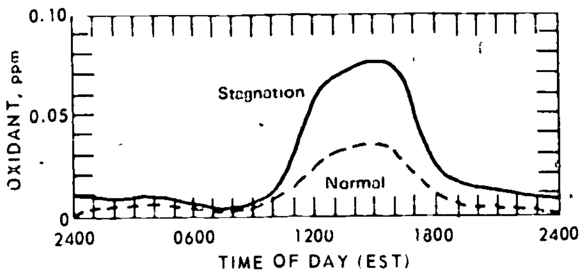


FIGURE 14-8: Diurnal Variation of Oxidant Levels During Stagnation in Washington, D.C., October 15 through 19, 1963 (USDC 1967).

Seasonal Patterns

The concentration of NO displays a seasonal variation while that of NO_2 does not. For NO, higher mean values are observed during late fall and winter months coinciding with decreased atmospheric mixing and generally less untraviolet energy available for the formation secondary products.

Figure 14-9 shows the seasonal patterns in NO by presenting the mean values by month of the year. The higher winter levels are quite distinctive.

The seasonal pattern for NO_2 is less consistent, as seen in Figure 14-10. Thus there appears to be little monthly variation for Denver (1966) and Bayonne, N.J. (1967). Even though a greater amount of NO is converted to NO_2 during the summer months, the actual concentrations of NO_2 are governed by the rate of conversion to oxidant. This can lead to apparently contradictory comparisons between Los Angeles and Chicago monthly trends.

NO_2 is highest during summer months in Chicago and during winter months in L.A. On closer examination of the L.A. aerometric data, one finds an inverse relationship between oxidant levels and NO_2 levels. The presence of oxidants during summer months, when the synoptic weather condition superimposes a persistent inversion lid over the L.A. Basin, and when there is higher solar radiation intensity and higher temperature, effectively scavenges the NO molecules. Figure 14-11.

Indoor Concentrations of Nitrogen Oxides

There is now a very active interest in determining pollutant levels in the home environment. However, very little of the research performed to date has been concerned with NOx indoor levels (USEPA 1973). One of the studies which has examined nitrogen oxide concentrations both indoors and outdoors was performed in Tokyo and found that although particulate matter and sulfur dioxide concentrations in indoor air

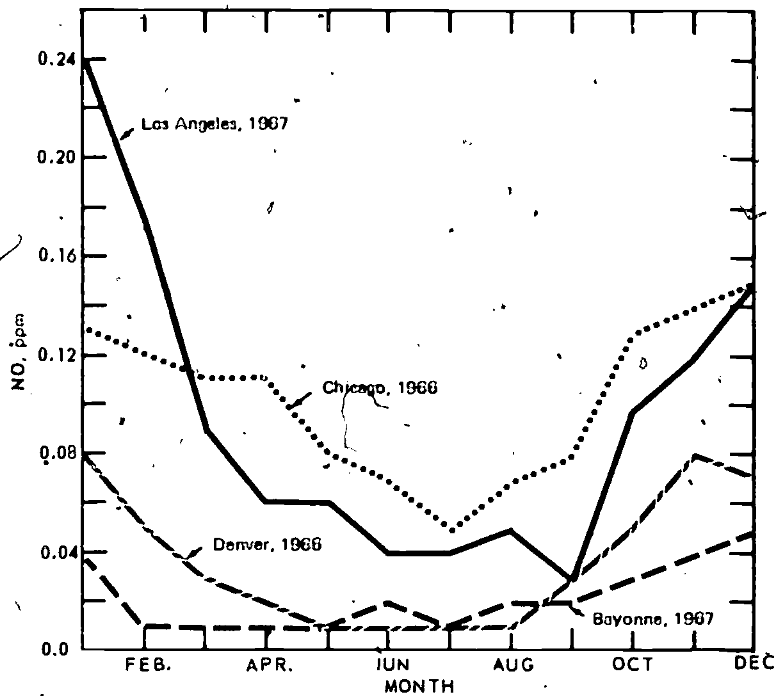


FIGURE 14-9: Monthly Mean NO Concentrations at Four Urban Sites (System Development Corporation 1970).

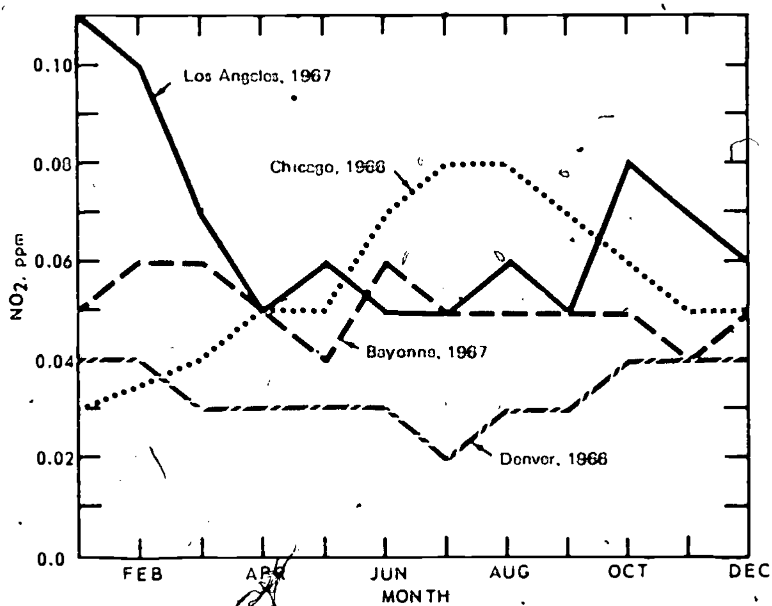


FIGURE 14-10: Monthly Mean NO_2 Concentrations at Four Urban Sites (System Development Corporation 1970).

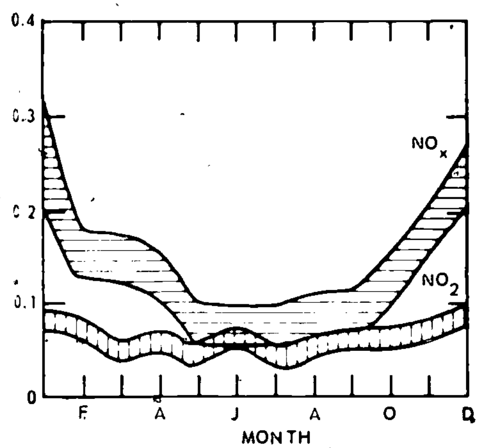


FIGURE 14-11: Average and Standard Deviation of Daily Maximum Oxides of Nitrogen Dioxide Concentrations for Seven Locations in Los Angeles Basin, 1962 (USDC 1967).

were lower than concentrations in outdoor air, there was no such difference for nitrogen dioxide (Miura et al. 1975). The results for NO_2 confirmed the findings of an earlier study in Dushambe, Russia (Berdyer et al. 1967). It has been found that nitrogen oxide concentrations in private homes with gas stoves can exceed 100 $\mu g/m^3$ (Wade et al. 1974). The existence of such pollutant levels indicates that there is clearly a need for further investigation of the sources of nitrogen oxides within the home and of the factors affecting the indoor/outdoor concentration relationship.

U.S NITROGEN OXIDE EMISSIONS

Description of Methodology of Emissions Estimation

Emission data from the Environmental Protection Agency's National Emission Data System (NEDS) can be used to describe the present pattern of nitrogen oxide (NOx) emissions across the United States (USEPA 1972b). NEDS, a computerized emission data system, summarizes emissions by source type, e.g., transportation, electric generation, and by fuel use, e.g., coal, oil, and natural gas for every county, Air Quality Control Region (AQCR) and State in the U.S. Emissions are further summarized by point and area source classification for each source type. Point sources are all industrial process emission sources and stationary fuel combustion sources in urban areas emitting more than 100 tons per year of any one contaminant, and stationary fuel combustion sources in rural areas emitting more than 25 tons per year of any one pollutant. Area sources include all sources other than those defined as point sources.

The 1972 National Emissions Report from NEDS provides the basis for all of the current emission descriptions in this section. The 1972 report contains emission data representative of either 1970, 1971, or 1972 for each political jurisdiction, depending on the base year inventory used in state implementation plans required by the Clean Air Act. Because the 1972 report represents the first such comprehensive emissions summary and because the sophistication of state and local jurisdictions in gathering emissions data varies widely, the accuracy of the data varies from state to state. Such variability can result in apparent data anomalies. For example, according to the 1972 report, the Metropolitan Philadelphia Interstate AQCR accounts for 75 percent of the nationwide industrial process emissions of nitrogen oxides. This is presumed to occur because this AQCR contains the only comprehensive survey of petroleum refinery emissions in the U.S. and seems to indicate that nationwide industrial process emissions are significantly underestimated. However, the emission data for stationary fuel combustion and transportation sources which account for over 85 percent of the nationwide NOx emissions are much more accurate and consistent. In fact, the national emission totals obtained from the 1972 report agree quite closely with independent national emission estimates performed with national fuel use and vehicle usage data (Mason and Shimizu 1974, Cortese 1975).

As new and more accurate emission factors become available, emission estimates and projections will be improved. Currently there is variability in emission estimates resulting from use of different emission factors. Because of recent changes in transportation source emission factors, the 1972 NEDS data overestimates diesel emissions by 590,000 tons per year and underestimates gasoline emissions by 320,000 tons per year (USEPA 1973b, 1974b). Thus total transportation emissions are overestimated by approximately 270,000 tons per year.

Nationwide Nitrogen Oxide Emission Patterns

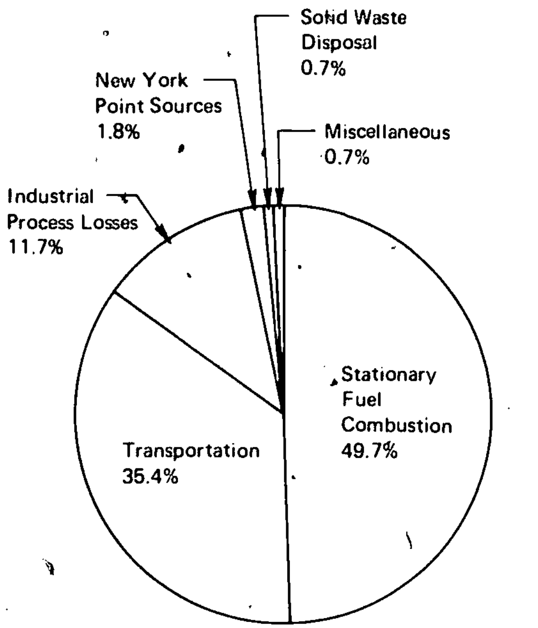
Nationwide NOx emissions for 1972 are summarized in Table 14-4 and Figure 14-12. In 1972, 24.64 million tons of NOx were emitted into the nation's atmosphere; of this total 49.7 percent

TABLE 14-4

Summary of Nationwide NOx Emissions, 1972

Source Type	Emissions (10^6 tons/year)			% of Total Emissions		
	Total	Point	Area	Total	Point	Area
Stationary Fuel Combustion	12.27	10.78	1.49	49.7	43.8	5.9
Transportation	8.72	-	8.72	35.4	-	35.4
Industrial Process Lossess	2.88	2.88	-	11.7	11.7	-
Solid Waste Disposal	0.18	0.03	0.15	0.7	0.1	0.6
Miscellaneous	0.17	-	0.17	0.7	-	0.7
New York Point Sources[a]	0.42	0.42	-	1.8	1.8	-
TOTAL	24.64	14.11	10.53	100.0	57.3	42.7

[a]New York point sources were separately treated since a further dissection of these data by source type was not possible.



Source Type	Emissions (10^6 ton/year)
Stationary Fuel Combustion	12.27
Transportation	8.72
Industrial Process Losses	2.88
Solid Waste Disposal	0.18
Miscellaneous	0.17
New York Point Sources	0.42
Total	24.64

FIGURE 14-12: Nationwide Emissions of Nitrogen Oxides, 1972

or 12.27 million tons were produced by stationary fuel combustion sources and 35.4 percent or 8.72 million tons were produced by transportation sources. All fuel combustion accounted for over 85 percent of the national total. Dissecting the data in a slightly different manner, point sources accounted for 57.3 percent (14.11 million tons) while area sources accounted for 42.7 percent (10.53 million tons) of the total national emissions. The major portion of the point source emissions was contributed by stationary fuel combustion and industrial process losses (97 percent) and the major portion of area source emissions was contributed by transportation sources (83 percent).

Since stationary fuel combustion sources accounted about half of the total national emissions in 1972, it is appropriate to examine the types of stationary sources and fuels which make the major contribution to stationary fuel combustion emissions. As shown in Table 14-5 and Figure 14-13, electric power generation (48 percent) and industrial fuel combustion (44 percent) shared almost equally in the production of over 92 percent of the stationary fuel combustion emissions. However, a look at the type of fuel consumed in electrical power generation and industrial heating discloses differences. Coal accounted for 67 percent of the NOx emissions from electric power generation, but only 14 percent of the industrial fuel combustion emissions. The major contributor to industrial fuel combustion NOx emissions was industrial process gas, such as coke oven gas and refinery gas, which accounts for 48 percent of the industrial fuel combustion emissions. Natural gas was the second largest contributor for each source type, accounting for 30 percent of industrial fuel combustion emissions and 19 percent of electric power generation emissions.

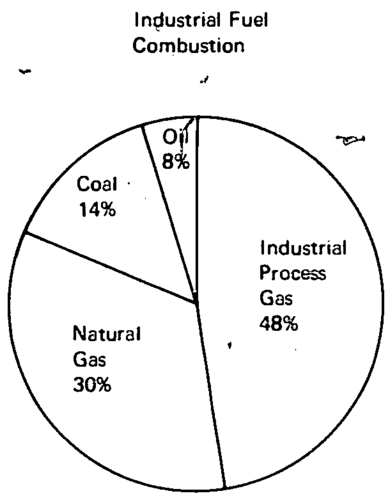
The predominance of coal-related NOx electric power generation emissions is a result of two factors. First, coal is the major fuel used in electric power generation. Secondly, coal has a higher NOx emission rate than oil or natural gas on an equivalent heat production basis as indicated in Table 14-6.

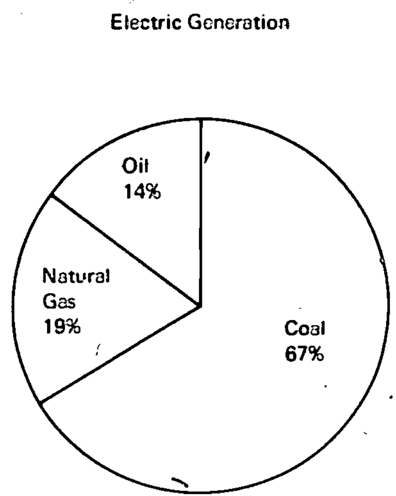
588-587 O - 75 - 50

TABLE 14-5

Summary of Nationwide NOx Emissions by Source Type and Fuel Use, 1972

Source Type	Emissions (10^6 tons/year)			% of Total Emissions		
Stationary Fuel Combustion	12.27			49.7		
Electric Generation		5.94			24.1	
Coal			3.95			16.0
Oil			0.85			3.4
Natural Gas			1.14			4.7
Industrial Fuel Combustion		5.39			21.8	
Coal			0.76			3.0
Oil			0.41			1.6
Process Gas			2.58			10.5
Natural Gas			1.64			6.7
Commercial-Institutional		0.65			2.6	
Residential		0.29			1.2	
Transportation	8.72			35.4		
Gasoline		6.62			26.9	
Diesel		1.90			7.7	
Other		0.20			0.8	
Industrial Process Losses	2.88			11.7		
Solid Waste Disposal	0.18			0.7		
Miscellaneous	0.17			0.7		
New York Point Sources	0.42			1.8		
TOTAL	24.64			100.0		





Source	Emissions (10^6 tons/year)	% of Total Emissions
Electric Generation	5.94	48.4
Industrial Fuel Combustion	5.39	43.9
Commercial-Institutional	0.65	5.3
Residential	0.29	2.4
Total	12.27	100.0

FIGURE 14-13: Nationwide Stationary Combustion Emissions, 1972

TABLE 14-6

Nitrogen Oxide Production in Large Electric Utility Boilers (EPA 1973a)

Fuel	NOx Emissions ($lb/10^6$ BTU)	
	Existing Facilities	New Facilities
Coal	0.72 - 1.2	0.7
Oil	0.7	0.3
Natural Gas	0.3 - 0.7	0.2

Projected increases in energy demand coupled with a desire to convert existing electric utilities from oil and natural gas to coal combustion and to require new electric utilities to burn coal could result in a substantial increase in NOx emissions from electric power generation in the future.

Geographical Nitrogen Oxide Emission Patterns

General Patterns

Geographical nitrogen oxide emission patterns were determined by summarizing NOx emission data for all the AQCR's and states located within the jurisdictional boundaries of each of the Environmental Protection Agency's (EPA) 10 regional offices. A complete listing of the states located within the jurisdictional boundaries of each EPA regional office is given in Table 14-7.

Table 14-8 displays the geographical distribution of 1972 NOx emissions for the United States. The North-east includes all states east of the Mississippi River and north of the Mason-Dixon line, the South includes all states south of the Mason-Dixon line and east of Arizona, and the West includes all of the area west of the Mississippi River and north of Oklahoma and Arkansas. The North-east was responsible for 56 percent of total U.S. NOx emissions with the South and West being nearly equal contributors to the remaining 44 percent. Distribution of stationary fuel combustion emissions followed a similar pattern. Transporation emissions followed a somewhat different pattern more closely following population patterns. The North-east accounted for 42 percent of U.S. NOx emissions from transportation while the South and West contributed 32 percent and 26 percent respectively.

A closer look at the contribution of EPA Regions to total emissions reveals that 27 percent of U.S. total NOx emissions and 39 percent of U.S. stationary fuel combustion emissions

TABLE 14-7

Listing of States Located Within Jurisdictional Boundaries of EPA Regional Offices (EPA 1972)

EPA Region	States and Territories
I	Connecticut, Maine, Massachusetts, New Hampshire, Rhode Island, Vermont
II	New Jersey, New York, Puerto Rico, Virgin Islands
III	Delaware, Maryland, Pennsylvania, Virginia, West Virginia
IV	Alabama, Florida, Georgia, Kentucky, Mississippi, North Carolina, South Carolina, Tennessee
V	Illinois, Indiana, Michigan, Minnesota, Ohio, Wisconsin
VI	Arkansas, Louisiana, New Mexico, Oklahoma, Texas
VII	Iowa, Kansas, Missouri, Nebraska
VIII	Colorado, Montana, North Dakota, South Dakota, Utah, Wyoming
IX	Arizona, California, Hawaii, Nevada, American Samoa, Guam
X	Alaska, Idaho, Oregon, Washington

TABLE 14-8

Geographical Distribution of Nitrogen Oxide Emissions, 1972[a]

Geographical Area (EPA Region)[b]	% Total U.S. Emissions		% Total U.S. Stationary Fuel Combustion Emissions		% Total U.S. Transportation Emissions	
EAST	56		59		42	
V		27		38		19
III		19		12		10
II		7		6		9
I		3		3		4
SOUTH	24		23		32	
IV		14		14		19
VI		10		9		13
WEST	20		18		26	
IV		10		10		12
VII		5		5		7
VIII		3		2		4
X		2		1		3

[a]Percentages have been rounded off

[b]Regions listed in decreasing order of contribution to Area emissions.

were produced in the six Eastern states located in Region V. Regional transportation emissions were consistent with population distribution. Stationary fuel combustion emissions on the other hand, reflected geographical differences in industrialization and electric power requirements.

NOx Emission Patterns in EPA Regions

Tables 14-9 through 14-20 compare the distribution of NOx emissions in each of the ten EPA Regions. The Regional NOx emissions are dissected according to the degree of urbanization is determined by the largest Standard Metropolitan Statistical Area (SMSA) population within an AQCR's are grouped in the following manner: grouped in the following manner:

(1) large urban AQCR's: AQCR's with an SMSA population greater than 1 million;

(2) medium-sized urban AQCR's: AQCR's with an SMSA population of 250,000-1,000,000

(3) small urban AQCR's: AQCR's with an SMSA population of 50,000 - 250,000; and

(4) rural AQCR's: AQCR's not containing an SMSA.

Although it reflects the obvious distribution of population, electric power generation and industrialization, Table 14-9 reveals some interesting points:

-Region VIII had high rural emissions due to electric power generation. Electric generation emissions were 62 percent of rural emissions and 25 percent of the Region's total emissions. This reflects the presence of the Four Corners power plant in Region VIII.

-In Region X the rural emissions were also high. However, 66 percent of the emissions

TABLE 14-9

Effect of Urbanization on Nitrogen Oxide Emissions for Different Geographical Areas[a,b]

Geographical Area (EPA Region)	% of EPA Region Emissions					
	Rural AQCR's	Small Urban AQCR's	Medium-Sized Urban AQCR's	Large Urban AQCR's	Total Urban AQCR's	All AQCR's
I	9	25	43	23	91	100
II	3	3	13	81	97	100
III	7	6	15	72	93	100
IV	14	20	52	14	86	100
V	12	4	21	63	88	100
VI	8	16	34	42	92	100
VII	22	32	3	43	78	100
VIII	58	23	8	11	42	100
IX	12	1	20	67	88	100
X	46	6	3	45	54	100
All U.S.	13	10	24	53	87	100

[a]Urbanization is based on largest SMSA population in an AQCR: Large Urban = AQCR with SMSA population > 1,000,000; Medium-sized Urban = AQCR with SMSA population of 250,000 - 1,000,000; Small Urgan = AQCR with SMSA population of 50,000 - 250,000; Rural = AQCR with no SMSA.

[b]Percentages have been rounded off.

TABLE 14-10

1972 NOx Emissions
EPA Region I

Air Quality Control Regions

Source Category	Rural (7)[a]		Small Urban[b] (5)[a]		Medium-Sized Urban (3)[a]		Large Urban[b] (1)[a]		Total Urban		Total Region	
	Tons	%	Tons	%	Tons	%	Ton	%	Tons	%	Tons	%
Stationary Fuel Combustion	20632	33 100	89116	48 100	158335	51 100	98664	59 100	346115	52 100	366747	50 100
Residential	2790	13	5657	6	8000	5	5918	6	19575	6	22365	6
Electric Generation	3359	16	48298	54	90825	57	47617	48	186740	54	190099	52
Industrial	10785	52	20407	23	19271	12	6581	7	46259	13	57044	16
Commercial-Institutional	3695	18	14754	17	40239	25	38548	39	93541	27	97236	26
Industrial Process Losses	23	0	1041	1	184	0	0	0	1225	0	1248	0
Solid Waste Disposal	1254	2	3816	2	4105	1	1058	1	8979	1	10233	1
Transportation	41654	66 100	90528	49 100	140409	48 100	68830	41 100	307767	46 100	349421	48 100
Light-duty gas vehicles	27778	67	64178	71	102637	69	56075	81	222890	72	250668	72
Other	13876	33	26350	29	45792	31	12755	19	84877	28	98753	28
Miscellaneous	0	0	0	0	0	0	0	0	0	0	0	0
Total	63564	100	184500	100	311032	100	168553	100	664085	100	727649	100

a Number in parentheses indicates the number of AQCR's in that category

b Largest SMSA population within AQCR

c Total urban = emissions from all AQCR's except Rural AQCR's

TABLE 14-11

1972 NOx Emissions
EPA Region II[d]

Air Quality Control Regions

Source Category	Rural (2)[a]			50,000-250,000[b] (1)[a]			250,000-1,000,000[b] (4)[a]			>1,000,000[b] (3)[a]			Total Urban[c]			Total Region		
	Tons	%		Tons	%		Tons	%		Tons	%		Tons	%		Tons	%	
Stationary Fuel Combustion	9516	18	100	31985	58	100	35120	18	100	317296	36	100	384401	34	100	393917	33	100
Residential	1797		19	819		3	7208		21	39896		13	47923		12	49720		13
Electric Generation	824		9	22884		72	0		0	139924		44	162808		42	163632		42
Industrial	713		7	2851		9	0		0	57939		18	60790		16	61503		16
Commercial-Institutional	6181		65	5431		17	27913		79	79538		25	112882		29	119063		30
Industrial Process Losses	0	0		402	0.7		0	0		2333	3		23737	2		23737	2	
Solid Waste Disposal	706	1		240	0.4		3261	2		15523	2		19024	2		19730	2	
Transportation	41843	80	100	22512	41	100	161947	81	100	511706	59	100	696165	62	100	738008	63	100
Light Duty gas vehicles	25126		60	11325		50	96189		59	341373		67	440887		64	474013		64
Other	16717		40	11187		50	65758		41	170333		33	247278		36	263995		36
Miscellaneous		0		0	0		0	0		2145	0		2145	0		2145	0	
Total	52064	100		55139	100		200329	100		870005	100		1125473	100		1177537	100	

a Number in parentheses indicates the number of AQCR's in that category

b Largest SMSA population within AQCR

c Total urban = emissions from all AQCR's except Rural AQCR's

d Does not include 421,000 tons from New York City Point Sources.

TABLE 14-12

1972 NOx Emissions
EPA Region III

Air Quality Control Regions

Source Category	Rural (12)[a]		50,000-250,000[b] (4)[a]		250,000-1,000,000[b] (6)[a]		>1,000,000[b] (4)[a]		Total Urban[c]		Total Region	
	Tons	%	Tons	%	Tons	%	Tons	%	Tons	%	Tons	%
Stationary Fuel Combustion	200902	65 100	219192	73 100	422045	61 100	723466	22 100	1364703	31 100	1565605	34 100
Residential	4515	2	3389	1	11185	3	19096	3	33670	2	38185	2
Electric Generation	156591	78	162769	74	301993	72	496032	69	960794	70	1117305	71
Industrial	35347	18	49738	23	85460	20	146640	20	281838	21	317185	20
Commercial-Institutional	4559	2	3926	1.7	23407	6	61697	9	89030	7	93589	6
Industrial Process Losses	2025	0.6	6210	2	4524	0.6	2175857	65	2186591	50	2188616	47
Solid Waste Disposal	1749	0.5	1259	0.4	1341	0.2	6312	0.1	8912	0.2	10661	0.2
Transportation	103413	34 100	75748	25 100	265678	38 100	439907	13 100	781333	18 100	884746	19 100
Light-duty gas vehicles	59474	58	43950	58	160580	60	266356	61	470886	60	530360	60
Other	43939	42	31798	42	105098	40	173551	39	310447	40	354386	40
Miscellaneous	56	.01	0	0	0	0	4	0	4	0	60	0
Total	308147	100	302410	100	693,587	100	3345546	100	4341543	100	4649690	100

a Number in parentheses indicates the number of AQCR's in that category

b Largest SMSA Population within AQCR

c Total urban = emissions from all AQCR's except Rural AQCR's

TABLE 14-13

1972 NOx Emissions
EPA Region IV

Air Quality Control Regions

Source Category	Rural (16)[a]			50,000-250,000[b] (12)[a]			250,000-1,000,000[b] (15)[a]			>1,000,000[b] (3)[a]			Total Urban[c]			Total Region		
	Tons	%		Tons	%		Tons	%		Tons	%		Tons	%		Tons	%	
Stationary Fuel Combustion	156522	31	100	376839	52	100	950265	52	100	237407	49	100	1564511	51	100	1721033	48	100
Residential	6672		4	8115		2	11984		1	1646		1	21739		1	28411		2
Electric Generation	97421		62	247034		66	689749		73	184154		77	1102937		70	1218358		71
Industrial	43360		28	111834		30	220141		23	45886		19	377861		24	421221		24
Commercial-Institutional	9066		6	9858		3	28393		3	5726		2	43977		3	53043		3
Industrial Process Losses	9072	2		9180	1		67525	4		5539	1		82244	3		91316	3	
Solid Waste Disposal	6359	1		8169	1		12670	1		2332	1		23171	1		29530	1	
Transportation	338750	66	100	329640	46	100	804008	44	100	234761	49	100	1368409	45	100	1707159	48	100
Light-duty gas vehicles	243307		72	199124		60	589073		73	146702		62	934899		68	1178206		69
Other	95443		28	130156		40	214935		27	88059		38	433510		32	528953		31
Miscellaneous	1076	0		0	0		2394	0		1974	0		4368	0		5444	[illegible]	
Total	511776	100		723834	100		1837044	100		482016	100		3042894	100		3554670	100	

a Number in parentheses indicates the number of AQCR's in that category

b Largest SMSA population within AQCR's except Rural AQCR's

c Total urban = emissions from all AQCR's except Rural AQCR's

TABLE 14-14

1972 NOx Emissions
EPA Region V

Air Quality Control Regions

Source Category	Rural (13)[a] Tons	Rural (13)[a] %	50,000-250,000[b] (5)[a] Tons	50,000-250,000[b] (5)[a] %	250,000-1000000[b] (15)[a] Tons	250,000-1000000[b] (15)[a] %	>1,000,000[b] (7)[a] Tons	>1,000,000[b] (7)[a] %	Total Urban[c] Tons	Total Urban[c] %	Total Region Tons	Total Region %
Stationary Fuel Combustion	550994	66 100	136182	46 100	754891	55 100	3375474	80 100	4266552	73 100	4817546	72 100
Residential	9695	2	5694	4	22719	3	37865	1	66278	2	72973	2
Electric Generation	441305	80	53019	39	445422	59	574867	17	1078308	25	1514613	31
Industrial	84982	15	63809	47	234826	31	2693250	80	2991885	70	3076867	64
Commercial-Institutional	15048	3	13657	10	51925	7	69498	2	135080	3	150128	3
Industrial Process Losses	47450	6	236	1	35693	3	51781	1	89835	2	137285	2
Solid Waste Disposal	3399	0.4	3004	1	9946	1	21970	1	34920	1	38319	1
Transportation	231182	28 100	151566	52 100	562855	100	727989	17 100	1442410	25 100	1673592	25 100
Light-duty gas vehicles	121864	53	79263	52	294859	52	438331	60	812453	56	934317	56
Other	109318	47	72303	48	267996	48	289658	40	629957	44	739275	44
Miscellaneous	2	0	0	0	0	0	1	0	1		3	0
Total	832307	100	293103	100	1363388	100	4197602	100	5854093	100	6686400	100

a Number in parentheses indicates the number of AQCR's in that category

b Largest SMSA population within AQCR

c Total urban = emissions from all AQCR's except Rural AQCR's

TABLE 14-15

1972 NOx Emissions
EPA Region VI

Air Quality Control Regions

Source Category	Rural (9)[a]		50,000-250,000[b] (7)[a]		250,000-1,000,000[b] (9)[a]		>1,000,000[b] (3)[a]		Total Urban[c]		Total Region	
	Tons	%	Tons	%	Tons	%	Tons	%	Tons	%	Tons	%
Stationary Fuel Combustion	72887	38 100	157265	40 100	417050	50 100	488012	48 100	1062127	47 100	1135214	47 100
Residential	2510	3	7711	5	6385	2	5527	1	19623	2	22133	2
Electric Generation	38846	53	81637	52	201899	48	301193	62	584729	55	623575	55
Industrial	28789	39	62068	39	194014	47	163377	33	419459	39	448248	39
Commercial-Institutional	2744	4	5838	4	14751	4	17915	4	38504	4	41248	4
Industrial Process Losses	4736	2	28701	7	25989	3	123458	12	178148	8	182884	7
Solid Waste Disposal	2127	1	3396	1	6610	1	8825	1	18831	1	20958	1
Transportation	113393	59 100	207023	52 100	387972	46 100	386572	38 100	981567	44 100	1094960	45 100
Light-duty gas vehicles	55143	49	90624	44	198671	51	205388	53	494083	51	549226	50
Other	58250	51	116399	56	189301	49	181184	47	487484	49	545734	50
Miscellaneous	0	0	0	0	0	0	0	0	0	0	0	0
Total	193143	100	396375	100	837620	100	1012866	100	2246861	100	2440004	100

a Number in parentheses indicates the number of AQCR's in that category

b Largest SMSA population within AQCR

c Total urban = emissions from all AQCR's except Rural AQCR's

TABLE 14-16

1972 NOx Emissions
EPA Region VII

Air Quality Control Regions

Source Category	Rural (10)[a] Tons	Rural (10)[a] %	50,000-250,000[b] (10)[a] Tons	50,000-250,000[b] (10)[a] %	250,000-1,000,000[b] (1)[a] Tons	250,000-1,000,000[b] (1)[a] %	>1,000,000[b] (2)[a] Tons	>1,000,000[b] (2)[a] %	Total Urban[c] Tons	Total Urban[c] %	Total Region Tons	Total Region %
Stationary Fuel Combustion	90687	32 100	195354	46 100	16667	41 100	375130	67 100	587151	57 100	677838	51 100
Residential	4731	5	6176	3	616	4	4916	1	11708	2	16439	2
Electric Generation	54077	60	146372	75	12879	77	327690	87	486941	83	541018	80
Industrial	24567	27	30833	16	1539	9	35877	10	68249	12	92816	14
Commercial-Institutional	7311	8	11975	6	1633	10	6647	2	20255	3	27566	4
Industrial Process Losses	8530	3	11695	3	1354	3	15619	3	28668	3	37198	3
Solid Waste Disposal	2926	1	3991	1	216	0.5	2470	0.4	6677	0.6	9603	1
Transportation	184239	64 100	214704	50 100	21855	54 100	170659	30 100	407218	40 100	591457	45 100
Light-duty gas vehicles	84109	46	111437	52	13191	60	96782	57	221410	54	305519	52
Other	100130	54	103267	48	8664	40	73877	43	185808	46	285938	48
Miscellaneous	745	0.2	356	0.1	80	0.1	0	0	436	.04	1181	0.1
Total	287126	100	426102	100	40172	100	563877	100	1030151	100	1317277	100

a Number in parentheses indicates the number of AQCR's in that category

b Largest SMSA population within AQCR

c Total urban = emissions from all AQCR's except Rural AQCR's

TABLE 14-17

1972 NOx Emissions
EPA Region VIII

Air Quality Control Regions

Source Category	Rural (16)[a] Tons	Rural (16)[a] %	50,000-250,000[b] (5)[a] Tons	50,000-250,000[b] (5)[a] %	250,000-1,000,000[b] (1)[a] Tons	250,000-1,000,000[b] (1)[a] %	>1,000,000[b] (1)[a] Tons	>1,000,000[b] (1)[a] %	Total Urban[c] Tons	Total Urban[c] %	Total Region Tons	Total Region %
Stationary Fuel Combustion	229297	51 100	31125	18 100	21900	38 100	30194	37 100	83419	26 100	312716	41 100
Residential	5390	2	1194	4	1288	6	1441	5	4123	5	9513	3
Electric Generation	187860	82	11147	36	6298	29	18192	60	36337	44	224197	72
Industrial	27406	12	14400	46	10647	49	6696	22	31743	38	59149	19
Commercial-Institutional	8704	4	3681	12	3667	17	3863	13	11215	13	19919	6
Industrial Process Losses	7976	2	9159	51	5095	9	4726	6	101412	32	109388	14
Solid Waste Disposal	2864	1	691	0	329	1	188	0	1208	0	4072	1
Transportation	203745	46 100	54018	30 100	30581	53 100	47469	57 100	132068	41 100	335813	44 100
Light-duty gas vehicles	92125	45	27671	51	18981	62	29631	62	76283	58	168408	50
Other	111620	55	26347	49	11600	38	17838	38	55785	42	167405	50
Miscellaneous	3780	1	431	0	10	0	0	0	444	0	4224	1
Total	447700	100	17805	100	57916	100	82578	100	318553	100	766233	100

a Number in parentheses indicates the number of AQCR's in that category

b Largest SMSA population within AQCR

c Total urban = emissions from all AQCR's except Rural AQCR's

TABLE 14-18

1972 NOx Emissions
EPA Region IX

Air Quality Control Regions

Source Category	Rural (12)[a] Tons	%		50,000-250,000[b] (1)[a] Tons	%		250,000-1,000,000[b] (8)[a] Tons	%		1,000,000[b] (3)[a] Tons	%		Total Urban[c] Tons	%		Total Region Tons	%	
Stationary Fuel Combustion	156693	55	100	5177	42	100	155213	33	100	882293	57	100	1042683	51	100	1199376	52	100
Residential	1915		1	255		5	4164		3	12172		1	1[illegible]591		2	18506		2
Electric Generation	112924		72	4259		82	121888		78	182969		21	309116		30	422040		35
Industrial	39254		25	0		0	19381		12	660586		75	679967		65	719221		60
Commercial-Institutional	2604		2	662		13	9781		7	26568		4	3 011		4	39615		3
Industrial Process Losses	13167	5		1	0		3804	0.[illegible]		62140	4		61945	3		79112	3	
Solid Waste Disposal	2703	1		5	0		7368	2		17189	1		24562	1		27265	1	
Transportation	110240	39	100	7049		100	304801	64	100	589829	38	100	901679	44	100	1011919	44	100
Light-duty gas vehicles	51967		47	4342		62	153512		54	381100		65	548954		61	600921		59
Other	58273		53	2707		38	141289		46	208729		35	352[illegible]25		39	410998		41
Miscellaneous	0	0		0	0		2611	.5		0	0		2611	0		2611	0	
Total	282820	100		12232	100		473794	100		1551453	100		2037479	100		2320299	100	

a Number in parentheses indicates the number of AQCR's in that category

b Largest SMSA population within AQCR

c Total urban = emissions from all AQCR's except Rural AQCR's

TABLE 14-19

1972 NOx Emissions
EPA Region X

Air Quality Control Regions
(Washington, Oregon, Idaho)

Source Category	Rural (8)[a]			50,000-250,000[b] (2)[a]			250,000-1,000,000[b] (1)[a]			>1,000,000[b] (1)[a]			Total Urban[c]			Total Region		
	Tons	%		Tons	%		Tons	%		Tons	%		Tons	%		Tons	%	
Stationary Fuel Combustion	32346	21	100	5192	21	100	4673	47	100	37512	21	100	47377	22	100	79723	22	100
Residential	3283		10	500		10	277		6	4386		12	5163		11	8446		11
Electric Generation	0		0	0		0	0		0	313		1	313		1	313		0
Industrial	27772		86	4328		83	4239		91	28285		75	36852		78	64624		81
Commercial-Institutional	1286		4	363		7	156		3	4519		12	5038		11	6324		8
Industrial Process Losses	13337	9		0	0		0	0		1509	1		1509	1		14846	4	
Solid Waste Disposal	2193	1		329	1		214	2		3325	2		3868	2		6061	2	
Transportation	100035	68	100	19179	76	100	5027	51	100	137280	76	100	161486	74	100	261521	71	100
Light-duty gas vehicles	48796		49	8816		46	2777		55	77714		57	89307		55	138103		53
Other	51239		51	10363		54	2250		45	59566		43	72179		45	123418		47
Miscellaneous	2858	2		483	2		21	.0		2071	1		2575	1		5433	1	
Total	150770	100		25184	100		9934	100		181697	100		216815	100		367505	100	

a Number in parentheses indicates the number of AQCR's in that category

b Largest SMSA population within AQCR

c Total urban = emissions from all AQCR's except Rural AQCR's

TABLE 14-20

1972 NOx Emissions
EPA Region X

Air Quality Control Regions
(Alaska)

Source Category	Rural (1)[a]		50,000-250,000[b] (0)[a]		250,000-1,000,000[b] (0)[a]		>1,000,000[b] (0)[a]		Total Urban[c]		Total Region	
	Tons	%	Tons	%	Tons	%	Tons	%	Tons	%	Tons	%
Stationary Fuel Combustion	20,873	58 100		100		100		100		100	20873	58 100
Residential	1095	5									1095	53
Electric Generation	7022	34									7022	34
Industrial	9513	46									9513	46
Commercial-Institutional	3243	16									3243	16
Industrial Process Losses	0	0									0	0
Solid Waste Disposal	776	2									776	2
Transportation	14459	40 100		100		100		100		100	14459	40 100
Light-duty gas vehicles	3879	27									3879	27
Other	10580	73									10580	73
Miscellaneous	0	0									0	0
Total	36108	100		100		100		100		100	36108	100

a Number in parentheses indicates the number of AQCR's in that category

b Largest SMSA population within AQCR

c Total urban = emissions from all AQCR's except Rural AQCR's

were transporation related. Surprisingly, this represented 27 percent of the total Region's emissions. Further, rural industrial fuel combustion emissions equaled urban industrial fuel combustion emissions in this Region.

- In Region II 81 percent of emissions arose in three large urban AQCR's. The transportation emissions in just these large urban AQCR's represented 43 percent of the Region's total emissions. Stationary fuel combustion in these largest urban AQCR's generated 27 percent of the Region's emissions with only 12 percent coming from electric power generation.
- In Region III the urban emissions were dominated by industrial processing. Forty-six percent of the Region's emissions arose from industrial processing in a single AQCR, the Metropolitan Philadelphia Interstate AQCR. (This is due, at least in part, to the fact that petroleum refinery emissions have been carefully surveyed in the Philadelphia area, as discussed earlier in this chapter.)
- In Region I transportation emissions are dominated by light duty gasoline vehicles (72 percent). For all other Regions, light duty gas line vehicles contribute an average of 57 percent of the transportation emissions.

There are some similarities among Regions in the distribution of emissions which reflect the degree of urbanization and industrialization of the Regions:

- Regions I and IV (New England and the Southeast) had a similar distribution of emissions between rural and urban AQCR's. The majority of the Regional emissions were produced in medium-sized urban AQCR's. However, Southern states had a greater proportion of electric power emissions in rural areas.
- Regions II, III, and V represent major population centers. Sixty-three to 80 percent of the Regions' emissions were produced in large urban areas.

Effects of Degree of Urbanization on Nationwide Nitrogen Oxide Emissions

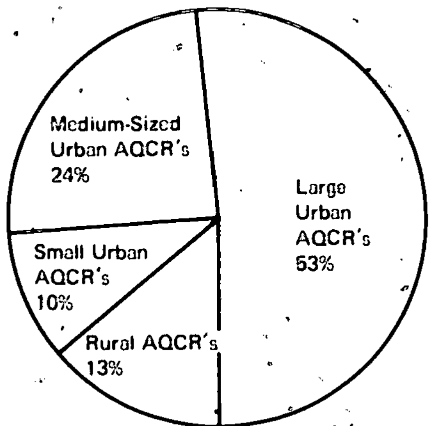
Figure 14-14 displays the effects of the degree of urbanization on nationwide NOx emissions in 1972. Urban AQCR's accounted for 87 percent of the total NOx emissions. Large urban AQCR's alone accounted for 53 percent of total emissions while rural AQCR's contributed only 13 percent of total emissions. Thus, the major portion of the NOx problem as characterized by NOx emissions was in urban areas.

Analysis of the 10 AQCR's with the largest SMSA populations indicates that these AQCR's accounted for 39 percent of the total U.S. NOx emissions. The emissions from these ten AQCR's are shown in Table 14-21. The majority of the emissions in these AQCR's were produced by stationary fuel combustion and industrial process loss sources with transportation sources contributing only 20 percent of the total. This is in marked contrast to the remainder of the country where transportation contributed 45 percent of total emissions.

Nitrogen Oxide Emissions by States

Table 14-22 ranks the states and the District of Columbia according to their total statewide NOx emissions. Table 14-22 also provides another basis for characterizing NOx emissions; it compares emissions per capita and emissions per unit area for each state to the NOx emission density for the contiguous United States. The U.S. NOx emission density is approximately 8.15 tons per square mile and National emissions per capita are approximately 0.112 tons.

The column labeled Ratio Emission Densities (State/U.S.) in Table 14-22 presents a tabulation of each state's NOx emissions per square mile divided by 8.15 tons per square mile, the nationally averaged emission density. The ratios range from 104 for the District of Columbia to .008 for Alaska. The states of Pennsylvania, Michigan, Indiana, New Jersey, Massachusetts, Maryland and Rhode Island had NOx emission den-



AQCR	Emissions (10^6 tons/year)
Large Urban	12.88
Medium-Sized Urban	5.83
Small Urban	2.60
Rural	3.17
Total	24.48

a Urbanization is based on largest SMSA population in an AQCR:
Large Urban = SMSA population > 1,000,000
Medium-Sized Urban = SMSA population 250,000–1,000,000
Small Urban = SMSA population 50,000–250,000
Rural = AQCR containing no SMSA

b Miscellaneous sources accounting for 160,000 tons/year are not included.

FIGURE 14-14: Distribution of 1972 Nationwide Nitrogen Oxide Emissions by Degree of Urbanization[a]

TABLE 14-21

1972 NO_x Emissions from 10 Largest Urban Air Quality Control Regions[a]

AQCR[b] (Name of Largest City)	Total Emissions (10^6 tons/year)	Stationary Fuel Combustion Emissions (10^6 tons/year)	Transportation Emissions (10^6 tons/year)	Other Sources
New York-New Jersey-Connecticut Interstate	1.15	0.69	0.43	0.11
Metropolitan Los Angeles Intrastate	1.20	0.79	0.36	0.05
Metropolitan Chicago Interstate	1.33	1.07	0.23	0.03
Metropolitan Philadelphia Interstate	2.58	0.24	0.18	2.16[c]
San Francisco Bay Area Intrastate	0.28	0.06	0.19	0.03
Metropolitan Detroit-Port Huron Intrastate	2.01	1.85	0.15	0.01
Greater Metropolitan Cleveland Intrastate	0.29	0.15	0.13	0.01
Metropolitan Boston Intrastate	0.17	0.09	0.07	0.01
National Capitol Interstate (Washington, D.C.)	0.18	0.09	0.09	0
Metropolitan St. Louis Interstate	0.43	0.31	0.11	0.01
TOTAL	9.62	5.26	1.94	2.42

[a] AQCR's chosen by SMSA population
[b] AQCR's listed in descending order of SMSA population
[c] Due to industrial process emissions

TABLE 14-22

NOx Emissions by States and the District of Columbia

Rank	State	State's Emissions	Running % U.S. Total	Ratio Emission Densities (State/US)	Ratio Emission Capita (State/US)
1	Pennsylvania	3,326,053	13	9.1	2.5
2	Michigan	2,449,019	22.9	5.3	2.5
3	California	1,033,297	30.9	1.4	.02
4	New York	1,659,019	37.6	2.7	.52
5	Indiana	1,511,526	43.7	5.1	2.6
6	Texas	1,427,195	49.6	.67	1.2
7	Ohio	1,214,163	54.5	3.6	.90
8	Illinois	1,074,061	50.9	2.4	.07
9	Florida	710,764	61.8	1.6	.94
10	South Carolina	574,904	64.1	2.3	1.9
11	New Jersey	539,260	66.3	0.0	.67
12	Missouri	494,166	60.3	.13	2.1
13	Tennessee	470,005	70.2	1.4	1.1
14	Louisiana	466,076	72.1	1.3	1.1
15	Kentucky	462,025	73.9	1.4	1.3
16	North Carolina	454,013	75.0	.11	.79
17	Wisconsin	450,332	77.6	1.0	.09
18	Alabama	437,693	79.4	.03	.04
19	Georgia	407,653	01	.06	.79
20	Massachusetts	360,590	02.5	5.0	.50
21	Virginia	363,000	04	1.1	.69
22	Minnesota	343,730	05.4	.53	.00

TABLE 14-22 (cont.)

Rank	State	State Emissions	Running % U.S. Total	Ratio Emission Densities (State/US)	Ratio Emission Capita (State/US)
23	Maryland	293,337	86.6	7.2	.68
24	Iowa	267,337	87.7	.59	.85
25	Kansas	257,926	88.7	.39	1.0
26	West Virginia	253,088	89.8	1.3	1.3
27	Oklahoma	245,470	90.8	.44	.86
28	New Mexico	219,559	91.5	.22	1.9
29	Washington	207,150	92.2	.38	.54
30	Mississippi	190,170		.49	.76
31	Arkansas	186,278		[illegible]	.06
32	Connecticut	171,775		.56	.06
33	Montana	163,588		.13	2.1
34	Colorado	163,586		.19	.66
35	Oregon	149,637		.19	.64
36	Arizona	136,544		.15	.69
37	Nebraska	112,378		.18	.68
38	Nevada	98,032		.11	1.8
39	North Dakota	94,477		.17	1.3
40	Utah	89,285		.13	.71
41	Maine	84,592		.34	.76
42	Wyoming	79,997		.1	2.1
43	New Hampshire	74,195		1.0	.89
44	Delaware	64,383		3.9	1.11

TABLE 14-22 (cont.)

Rank	State	State Emissions	Running % U.S. Total	Ratio Emission Densities (State/US)	Ratio Emission Capita (State/US)
45	South Dakota	54,443		.09	.71
46	Idaho	53,519		~~.08~~	~~.67~~
47	Rhode Island	51,722		6.1	.48
48	District of Columbia	51,615		104.	.61
49	Hawaii	48,745		.93	.56
50	Alaska	36,108		.008	1.1
51	Vermont	26,771		.35	.54

* Combined, the remaining states and D.C. contribute less than 9% to the total U.S. NOx emissions.

sities over 40 tons/sq. mile. (It should be noted, however, that the Pennsylvania data include a comprehensive survey of petroleum refinery emissions near Philadelphia. Since such surveys do not exist elsewhere, emissions from states other than Pennsylvania may be greater than shown here.)

A comparison of per capita emissions is given in the column of Table 14-22 labeled Ratio Emission\Capita (State/U.S.). The per capita NOx emissions range from 2.6 for Indiana to 0.4 for Alabama. Pennsylvania, Michigan, Indiana, Missouri, Montana and Wyoming had per capita emissions which were two or more times the National average of 225 pounds per person per year. Montana and Wyoming contributed less than 1 percent each to the total U.S. emissions but had a high per capita emission ratio because of their small population.

Table 14-22 also compiles the running accumulating percent contribution of state's emissions to the total U.S. NOx emissions (24.64 million tons/year). Interestingly, six states (Pennsylvania, Michigan, California, New York, Indiana and Texas) were responsible for about 50 percent of the nationwide NOx emissions. Pennsylvania, itself, contributed over 13 percent. Over 90 percent of the annual NOx emissions are accounted for in the first 27 states listed, with the remaining 23 states and the District of Columbia combining to contribute less than 10 percent.

Nationwide NOx Emission Trends

1940-1972

Nationwide NOx emission trends from 1940-1972 are displayed in Table 14-23 and Figure 14-15 (USEPA 1973a). Total nationwide emissions tripled in the three decades from 1940-1972. The stationary source fuel combustion category revealed the largest emission growth increasing from 3.5 million tons in 1940 to 12.3 million tons in 1972. In this category, emissions from electric power generation showed the greatest increase, climbing nearly 800 percent over the three decades. Transportation emissions tripled during this period.

TABLE 14-23

Nationwide NOx Emissions, 1940-1970 (EPA 1973b)

Source Category	NOx Emissions (10^6 tons/year)					
	1940	1950	1960	1968	1969	1970
Stationary Fuel Combustion	3.5	4.3	5.2	9.7	10.2	10.0
Electric Generation	0.6	1.2	2.3	4.2	4.3	4.7
Industrial	1.9	2.0	1.8	3.7	4.6	4.5
Commercial-Institutional	0.1	0.1	0.2	1.0	0.4	0.2
Residential	0.9	1.0	0.9	0.8	0.9	0.6
Industrial Process Losses	<0.1	0.1	0.1	0.2	0.2	0.2
Solid Waste Disposal	0.1	0.2	0.2	0.4	0.4	0.4
Transportation	1.7	2.9	4.3	7.5	8.3	8.8
Road Vehicles	1.4	2.2	3.5	5.5	5.8	6.2
Gasoline	1.4	2.15	3.2	4.8	5.1	5.4
Diesel	<0.1	0.05	0.3	0.7	0.7	0.8
Other	0.3	0.7	0.8	2.0	2.5	2.6
Miscellaneous	0.8	0.4	0.2	0.2	0.2	0.1
Total	6.1	7.9	10.0	18.2	19.3	19.5

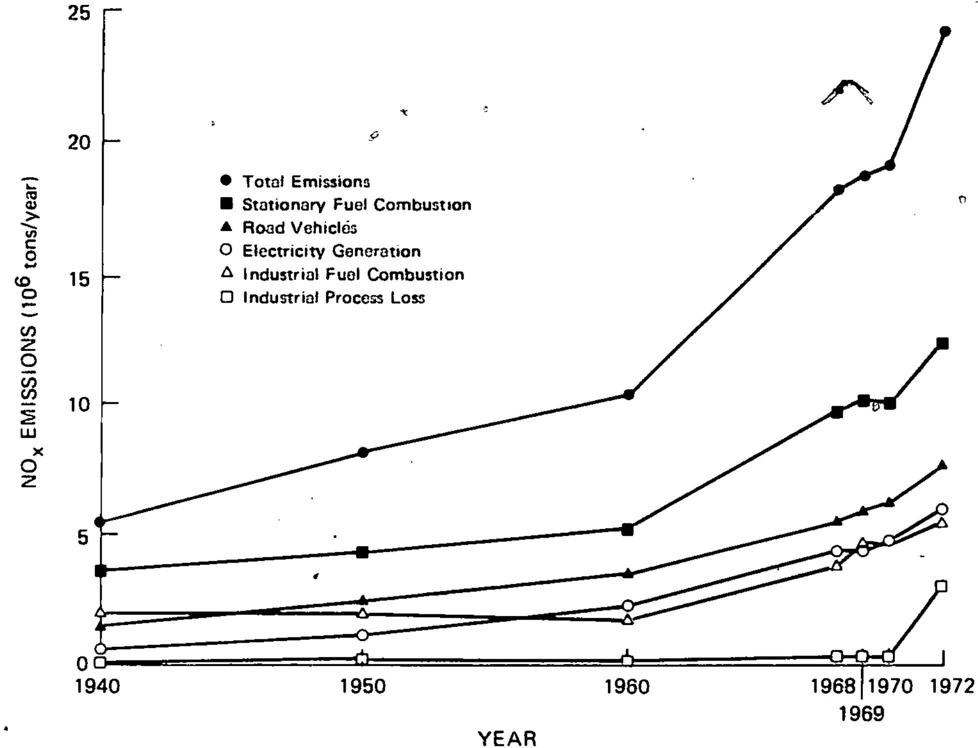


FIGURE 14-15: Nationwide NOx Emission Trends 1940-1972 (EPA 1973b).

Total Emissions: 1972-1990

In Appendix 14-C, total emissions of nitrogen oxide are projected to the year 1990 under various assumptions about the implementation of regulatory programs and about the growth rate of stationary and mobile sources.

Assuming the implementation of the present statutory program for NOx control; using emission projections for electric power generation based on forecasts by the Federal Energy Administration (USEPA 1975) and the National Academy of Engineering (1974); assuming a slowing of the growth rate of NOx emissions from industrial, commercial, and institutional fuel combustion; and using other assumptions presented in detail in Appendix 14-C, a conservative estimate yields a 36 percent increase in total NOx emissions from 1972-1990 (see Figure 14-16). The largest increases will result from industrial process losses, non-highway transportation, industrial fuel combustion, and electric power generation. Highway-related transportation emissions will decline under the assumptions of this projection as Federal controls on nitrogen oxide emissions from automobiles are implemented.

Appendix 14-C examines future NOx emissions from electric power generation and highway vehicles in some detail. Protections of power generation emissions are critically dependent upon the rate at which nuclear power generation increases. If for any reason no new nuclear power plants were built after 1975, NOx emissions from power generation facilities could more than double. Road vehicle emissions are projected under three options: (1) no control of NOx emissions, (2) implementation of a proposed five year delay of the 1977 statutory standards and (3) implementation of the present statutory program. The analysis shows a 25 percent reduction in road vehicle NOx emissions by 1990 (compared with 1972) under the present statutory program (if vehicle miles of travel increase by 4 percent per year). The five year delay leads to a 17 percent reduction in emissions by 1990. Emission reductions are somewhat smaller than might be expected from the

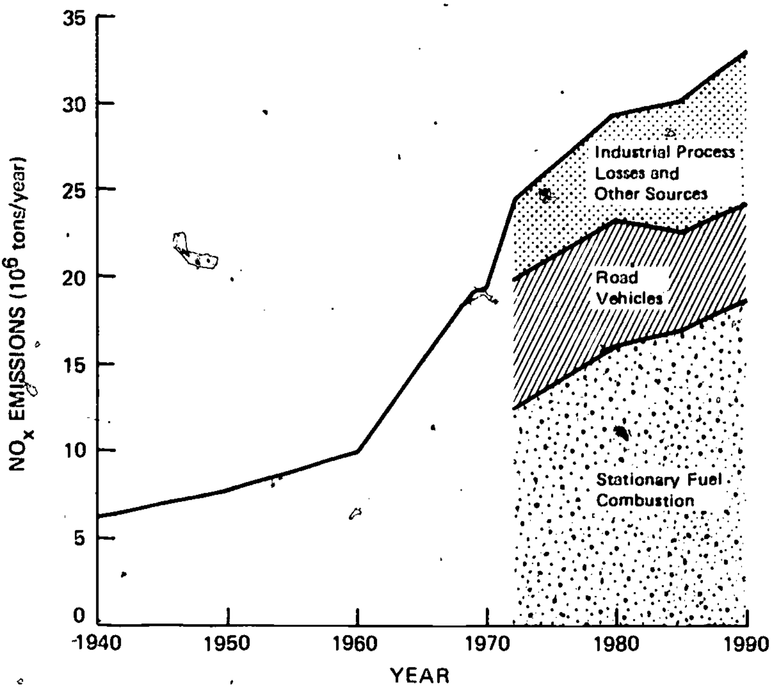


FIGURE 14-16: Nationwide NOx Emissions Projected to 1990 Assuming the Present Statutory Program.

large percentage decrease in allowable emissions per mile travelled because of the projected increase in miles travelled and because of the growth of emissions from uncontrolled heavy duty gasoline and diesel trucks.

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APPENDIX 14-A

ATMOSPHERIC REACTIONS

Nitrogen oxides play a principal role in the atmospheric reactions which produce photochemical smog. The complex reactions and the increased toxic potential associated with these secondary pollutants complicate the evaluation of control strategies significantly.

Dr. J.G. Calvert (1973) and Dr. E.R. Stephens (1973) have discussed the atmospheric reactions in detail in the Proceeding of the Conference on Health Effects of Air Pollution of the National Academy of Sciences.

PRODUCTION OF NITROGEN OXIDES

Nitrogen oxides are produced during combustion by the oxidation of organic nitrogen compounds in fossil fuels and the thermal fixation of atmospheric nitrogen gas, N_2.

Nitrogen gas (N_2) is generally inert with respect to tropospheric reactions. The equilibrium of the reaction

$$N_2 + O_2 \rightleftarrows 2NO \qquad (1)$$

is far to the left at normal atmospheric temperatures and NO concentrations are small. However at temperatures found in combustion chambers this equilibrium shifts to the right. Rapid cooling of the resultant N_2, O_2, and NO mixture quenches the reverse reaction, $2NO \quad N_2 + O_2$. The return to equilibrium proceeds very slowly and the NO persists. Although NO_2 may be an important intermediate during combustion, the amount released is small. For temperatures greater than 2000 F (1093 C), only 0.5 percent of the nitrogen oxides emitted are NO_2 (EPA 1970).

NO TO NO_2 CONVERSIONS

The levels of NO, NO_2 and O_3 as a function of time for typical diurnal smoggy conditions were described in detail in Chapter 14 in the discussion of diurnal NOx concentrations. Nitric oxide emissions increase during the morning rush hour.. However NO concentrations begin to fall rapidly as the NO_2 concentrations climb toward their maximum. Ozone levels do not begin to increase until most of the NO has disappeared, reaching a maximum at midday and falling off in the afternoon. There is no peak in NO concentrations in the afternoon corresponding to the evening rush hour.

These observations imply that NO is rapidly being converted to NO_2 in the atmosphere. Recent observations tracking power plant plumes have shown similar processes occurring (Davis et al. 1975). The time required for 50 percent conversion of the NO to NO_2 in these plumes was estimated to be between 12 and 60 minutes.

During initial dilution in the atmosphere NO_2 is produced by the reaction

$$2NO + O_2 \rightarrow 2NO_2. \qquad (2)$$

Because this is a three body reaction requiring two NO molecules, the rate of production is proportional to the square of the nitric oxide concentration. Therefore, nitrogen dioxide production by this mechanism is only important for high NO concentrations (greater than about 100 ppm) and significant O_2 concentrations. As the nitric oxide is diluted to concentrations below 1 ppm, these direct reactions with oxygen become unimportant (EPA 1970).

These conversions during the initial dilution can produce nitrogen dioxide equal to up to 25 percent of the total NOx (Calvert 1973). However, this reaction cannot account for the conversion of NO to NO_2 in the atmosphere at ambient levels of NO which are typically less than 1 ppm.

Ozone (O_3) oxidation of the NO does provide an explanation; nitric oxide is rapidly oxidized in the atmosphere by ozone,

$$O_3 + NO \rightarrow NO_2 + O_2. \qquad (3)$$

Ozone is naturally found in the atmosphere in concentrations of 10 to 50 ppb (Com et al. 1975). The 50 percent conversion time for this reaction is 0.3 minutes for nitric oxide and ozone concentrations of 100 ppb (Stephens 1973). Therefore, in the presence of excess atmospheric ozone, this reaction appears to be the primary link in NO to NO_2 conversion.

The brown gas, nitrogen dioxide, is a strong absorber of sunlight, and is rapidly photolyzed by the ultraviolet radiation to produce the reactive ground state oxygen atom, $O(^3P)$, and an NO molecule (EPA 1970):

$$NO_2 + \text{sunlight } (2900\text{-}4300 \text{ Å}) \rightarrow O(^3P) + NO \qquad (4)$$

The highly reactive $O(^3P)$ reacts with an oxygen molecule and a third molecule, M, which removes excess vibrational energy, to form ozone:

$$O(^3P) + O_2 + M \rightarrow O_3 + M. \qquad (5)$$

The net effect of these two reactions is the reverse of the equation (3) above. Thus we have a dynamic balance between two very fast (2 to 3 minutes) reactions (Stephens 1973):

$$O_3 + NO \underset{UV}{\rightleftarrows} NO_2 + O_2 \qquad (6)$$

This mechanism does not yet explain the net conversion of NO to NO_2 as observed in the atmosphere. Moreover, this reaction sequence does not explain the conversion of NO_2 to ozone.

OXIDANT PRODUCTION

The key reactions in shifting this equilibrium to convert NO to NO_2 are chains involving various transient molecules and reactive species such as carbon monoxide and hydrocarbons found in polluted air.

Carbon monoxide reacts with the hydroxyl radical HO through a chain of reactions to drive the equilibrium to the left:

$$HO + CO \rightarrow H + CO_2 \qquad (7)$$

$$H + O_2 + M \rightarrow HO_2 + M \qquad (8)$$

$$HO_2 + NO \rightarrow NO_2 + HO \qquad (9)$$

$$HO + CO \rightarrow H + CO_2, \text{ etc.} \qquad (7)$$

Each hydroxyl radical produced by this method is able to recycle back and oxidize many NO molecules to NO_2.

In the presence of ozone, the HO radical is produced by the reaction sequence (Levy 1971):

$$O_3 + \text{sunlight } (2900\text{-}3400\text{Å}) \rightarrow O(^1D) + O_2 \qquad (10)$$

$$O(^1D) + H_2O \rightarrow 2HO \qquad (11)$$

where $O(^1D)$ is the highly reactive ground state oxygen atom. Only 1 percent of the $O(^1D)$ atoms can be expected to react with water. The remainder are rapidly quenched by other molecules

$$O(^1D) + M \rightarrow O(^3P) + M. \qquad (12)$$

The HO production rate is then linearly dependent on the absolute H_2O concentration (Davis 1974).

Hydrocarbon reactions are major sources of the hydroperoxyl radical, HO_2, as in the sequence (Calvert 1973):

$$CH_3 \xrightarrow{(O_2)} CH_3O_2 \xrightarrow{(NO)} CH_3O \xrightarrow{(O_2)} CH_2O + HO_2 \qquad (13)$$

The HO_2 radical can then oxidize NO to NO_2 and also produce a hydroxyl radical:

$$HO_2 + NO \rightarrow NO_2 + HO \qquad (9)$$

The reaction chains of hydrocarbon radicals, R, are very complex, but the key reactions convert NO to NO_2 analagously to reactions (8) and (9) (Stephens 1973):

$$R + O_2 \rightarrow RO_2 \tag{14}$$
$$RO_2 + NO \rightarrow NO_2 + RO \tag{15}$$

The net effect of these radical reaction chains is to promote the conversion of NO to NO_2:

$$NO \xrightarrow{(radicals)} NO_2 \tag{16}$$

This drives the dynamic equilibrium between NO and NO_2 towards NO_2 production:

$$O_3 + NO \underset{UV}{\overset{(radicals)}{\rightleftharpoons}} NO_2 + O_2 \tag{17}$$

By scavenging NO, these reactions permit O_3 to accumulate (Stephens 1973).

REMOVAL PROCESSES

Ozone is lost by reacting with nitrogen dioxide:

$$O_3 + NO_2 \rightarrow NO_3 + O_2 \tag{18}$$

The transient NO_3 reacts with nitrogen dioxide:

$$NO_3 + NO_2 \rightarrow N_2O_5 \tag{19}$$

N_2O_5 is also a transient molecule which rapidly dissociates or reacts with water to form nitric acid, HNO_3:

$$N_2O_5 \rightarrow NO_3 + NO_2 \tag{20}$$

$$N_2O_5 + H_2O \rightarrow 2HNO_2 \tag{21}$$

In addition, nitric acid is formed by the OH removal process:

$$OH + NO_2 + M \rightarrow HNO_3 + M \tag{22}$$

Davis et al. (1974) have calculated that this reaction for power plant plumes in summer daylight conditions should have a 50 percent conversion time of 2 to 3 hours. The nitric acid formed presumably reacts with various other contaminants in the air to form nitrates. Ammonium for example, will react rapidly with nitric acid to form ammonium nitrate. In fact, composite samples in the Los Angeles basin have shown ammonium nitrate to comprise 10-15 percent of the total suspended particulates (Gordon and Bryan 1973).

The net effect of these reactions is to remove nitrogen dioxide from the air by loss reactions with the important reactive molecules to produce nitrate aerosols.

The other important loss mechanism for nitrogen dioxide is through the production of the peroxyl acyl nitrates, PAN's. The PAN's are a family of organic compounds with the general formula:

$$\mathrm{R\overset{\overset{O}{\|}}{C}OONO_2}$$

The PAN's have a temporal variation which is very similar to that of ozone. Stephens (1973) suggests that PAN formation is delayed because NO and NO_2 compete for the peroxyl acyl radicals:

$$\mathrm{R\overset{\overset{O}{\|}}{C}OO + NO \rightarrow R\overset{\overset{O}{\|}}{C}O. + NO_2} \tag{23}$$

$$\mathrm{R\overset{\overset{O}{\|}}{C}OO + NO_2 \rightarrow R\overset{\overset{O}{\|}}{C}OONO_2} \tag{24}$$

The first reaction, (23), converts NO to NO_2 and claims the major portion of the peroxyl acyl radical. As soon as the NO has been depleted however, the second reaction becomes important and PAN's are formed. This is very similar to the reaction scheme for ozone.

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APPENDIX 14-B

MEASUREMENT OF AMBIENT LEVELS OF NO_2

Ambient air concentrations of NO_2 are not well known because of problems with the Federal Reference method of measurement which was employed prior to 1972. The reference method used to determine compliance with national air quality standards was the Jacobs-Hochheiser (J-H) method as modified for the National Air Surveillance Networks (NASN). This method, along with other NO_2 measurement techniques, is summarized in Table App. 14-B1 (Saltzman 1973).

Jacobs-Hochheiser Method

Briefly, the Jacobs-Hochheiser (J-H) method involves bubbling ambient air through an aqueous solution for 24 hours. The nitrate ions formed in the reagent are treated to form an azo dye which is measured colorimetrically. The intensity of the absorbed light at a specific wave length is related to the integrated 24-hour NO_2 concentration. The collection efficiency of the reference method for NO_2 was previously determined as 35 percent, and a correction for this efficiency had been applied to all data.

In 1971-72 it became apparent that the J-H method has two inherent problems which affected its ability to accurately estimate air quality in the field. These are: (1) the collection efficiency varies with concentration of NO_2 and (2) a small portion of the nitric oxide (NO) in the ambient air shows up as NO_2. The collection efficiency of the J-H method at NO_2 concentrations in excess of 110 $\mu g/m^3$ has been known for some time and calibration curves developed. These

TABLE App. 14-B1

Manual Methods Using Alkaline Absorbing Reagents

Name	Reference	Volume and Composition of Absorbing Reagent	Absorbing Apparatus	Sampling Rate and Time	Procedure for Analysis	Remarks
Jacobs and Hochheiser (J-H)	2	30-35 ml: 0.1N NaOH, 0.2% v/v butyl alcohol	Coarse Fritted glass bubbler in sequential sampler	1.3 lpm, 40 min.	Add 1 drop 1% H_2O_2, 10 ml of 2% sulfanilamide in 5% v/v H_3PO_4; 1 ml of 0.1% N-(1-naphthyl)-ethylene diamine de-hydrochloride	No validation or stoichiometry studies. Data shows inverse relationship between SO_2 and NO_2 analyses, suggesting interference. 90% collected in first of two bubblers.
Perry and Tabor (NASN)	15, 16	20 ml: Same as J-H	Polypropylene test tube containing glass inlet tube with orifice 0.36 ± 0.05 mm. THERMOSTATED AT 35°C.	0.15-0.20 lpm, 24 hrs.	Autoanalyzer, using J-H reagents.	Absorption in first of 4 bubblers varied from 30-69%. Collected samples stable for 3 weeks. Assumed 50% overall conversion. Large mass of data available using this method.
Meadows and Stalker (Alabama Study)	14	35 ml: 0.1N NaOH (in each of 2 bubblers)	Two NASN bubblers in series.	0.35-0.50 lpm. 24 hrs.	Same as NASN.	Absorption in 1st of 6 bubblers was 19%, in 1st 2 was 30%. Overall conversion of 36% used, with assumption of 100% stoichiometry.
Federal Reference Method	1	50 ml: 0.1N NaOH	Membrane filter leading to polypropylene test tube with fritted glass inlet of porosity B (70-100 μm max. pore diameter).	0.20 lpm, 24 hrs.	Similar to J-H.	Overall conversion of 35% is used.

TABLE App. 14-B1 (cont.)

Name	Reference	Volume and Composition of Absorbing Reagent	Absorbing Apparatus	Sampling Rate and Time	Procedure for Analysis	Remarks
Christie, et al.	17	5 ml: 0.025 N NaOH, 0.1% $NaAsO_2$, 0.75% sulfanilic acid	Glass bubbler with open tube inlet.	0.12 lpm, 1 min.	Add 3 ml of 0.02% N-(1 naphthyl)-ethylene diamine dihydrochloride, 6% oxalic acid.	Overall conversion 94% at 6 ppm NO_2. For industrial hygiene use.
Nash	18	4 ml: 0.1N NaOH, 0.05% guaiacol (2-methoxy phenol)	Glass Arnold Tube (with open tube inlet)	0.6 lpm, 2hrs.	Add 4 ml of 0.2N HCl 0.5% sulfanilic acid, 1.5% glycine, 0.002% N-(1-naphthyl)-ethylene diamine dihydrochloride	Limited amount of H_2O_2 may be added to reduce SO_2 interference. Stoichiometric factor 0.73, absorption efficiency 99%.
Huygen and Steerman	19	15 ml: 0.1N NaOH, 0.02% R-salt (2-naphthol-3, 6-disulfonic acid disodium salt), 0.1% triethanolamine (in each of two bubblers).	Two bubblers in series, each with coarse glass frit (90-150 µm) of 6.5 cm^2 area.	1 lpm, 24 hrs.	Add to each absorber 15 ml of 0.4% sulfanilamide, 0.03% Cleves acid (8-aminonaphthl-2-sulfonic acid), 0.12N HCl. A drop of H_2O_2 may be added first to eliminate SO_2 interference.	Stoichiometric factor varied from 95% to 80% in range 40-2200 µg/m^3 NO_2. Absorption efficiency averaged 96%.
EPA Tentative Candidate Arsenite Method	6	50 ml: 0.1N NaOH, 0.1% sodium arsenite	Polypropylene tube containing glass inlet tube with orifice 0.6 ± 0.2 mm i.d., 6 mm from bottom of tube.	0.2 lpm, 24 hrs.	To 10 ml of absorbing solution, add 1 ml 0.024% H_2O_2; 10 ml of 2% sulfanilamide, 5% v/v H_3PO_4; and 1.4 ml of N-(1-naphthyl) -ethylene diamine dihydrochloride.	Overall conversion is 85% over range 50-750 µg/m^3 NO_2 concentration.
Levaggi, Siu, and Feldstein (triethanolamine)	20	50 ml: 1.5% triethanolamine, 0.3% v/v n-butanol.	Polypropylene tube with fritted glass inlet of 70-100 µm max. pore diameter.	0.15-0.20 lpm, 24 hrs.	Similar to J-H.	Overall conversion efficiency 76-92% over range 56-750 µg/m^3 NO_2, collection efficiency 95-99%. Constant 0.85 factor recommended.

show an approximate 35 percent collection efficiency at NO_2 concentrations of 110 to 180 $\mu g/m^3$. This means that about one-third of the NO_2 in the air that bubbles through the sampler is trapped and bubbles through the sampler is trapped and analyzed. Late in 1971 lab methods became available to generate reliable low level NO_2 concentrations and it was possible to extend the calibration curve for NO_2 concentrations below 100 $\mu g/m^3$. The extended curve showed that collection efficiencies greatly increased as NO_2 concentrations decreased below 100 $\mu g/m^3$. For example, at 100 $\mu g/m^3$ the collection efficiency is just below 40 percent, whereas at 30 $\mu g/m^3$ the collection efficiency is 60 percent. This variable collection efficiency may have been a major source of error in the observations used for classifying regions for NO_2.

The J-H method as published in the Federal Register uses a constant average correction factor on a collection efficiency of 35 percent based on the early efficiency curves. It is clear (see Figure App. 14-B1) that a 35 percent collection efficiency is too low for concentrations in the 30 to 60 $\mu g/m^3$ range. Therefore, at lower concentrations where collection efficiencies are much higher than 35 percent, the calculated and reported concentrations of NO_2 obtained by the J-H method are higher than actual ambient NO_2 concentrations. Since the NO_2 concentration in the air varies throughout the day, the collection efficiency may vary significantly throughout any one 24-hour sample, making accurate calculation of NO_2 concentrations impossible.

The NO interference previously mentioned is a second factor leading to an additive effect and is particularly important in areas of low NO_2 concentrations. Table B2, The Effect of NO on the Reference Method for NO_2, displays this additive interference of NO (Hauser and Shy 1972).

Because of the questionable data base, EPA reviewed all 47 AQCR's in 29 states that were classified as Priority I for NO_2 control and thus required stationary source controls. In July 1973, 43 of the original Priority I AQCR's were reclassified as Priority III for NO_2. The Los Angeles and Chicago AQCR's are the only ones

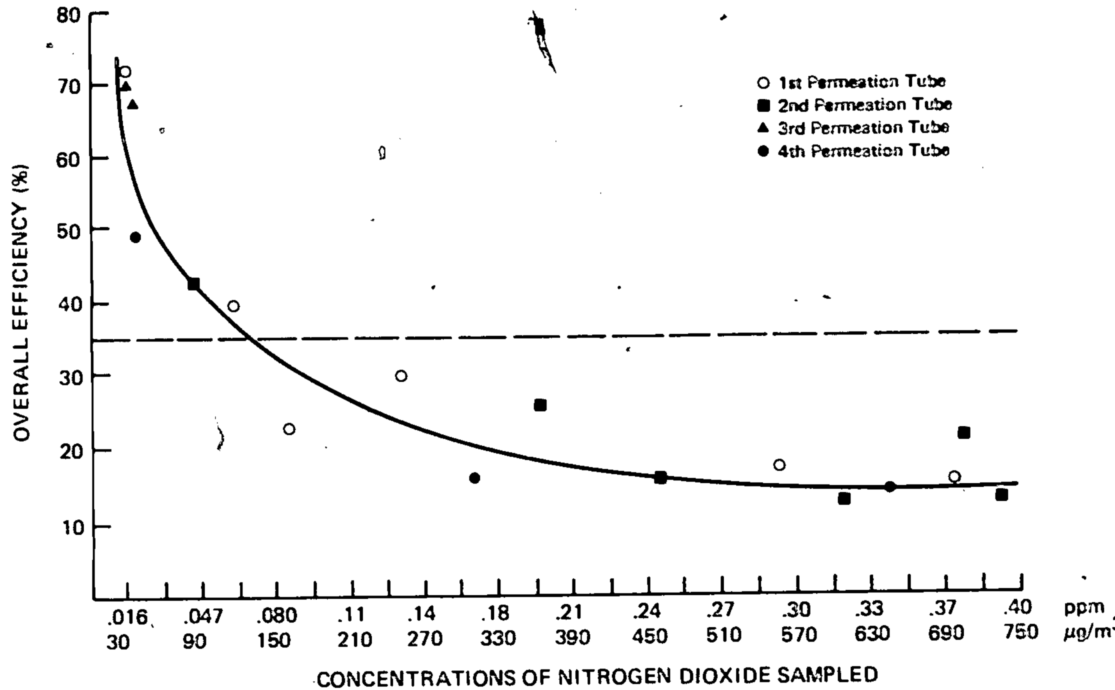

FIGURE App. 14-B1: Response of the NO_2 Reference Method

TABLE App. 14-B2

Effect of NO on the Reference Method for NO_2

$\mu g/m^3$		Ratio	Expected NO: recovered,	Apparent NO: recovered,
NO_2	NO	NO/NO:	%	%
100	0	0.0	39	38
102	63	0.6	39	38
105	127	1.2	38	52
122	627	5.1	36	57
189	0	0.0	29	29
244	1205	4.9	24	45
248	1279	5.2	23	55
215	1242	5.8	26	50
311	0	0.0	20	17
316	111	0.4	20	30
318	332	1.1	20	33
356	1060	3.0	18	44

where the data indicated NO_2 concentrations exceeding 110 $\mu g/m^3$ annual average. Both of these AQCR's remain Priority I. In the New York-New Jersey-Connecticut, Wasatch Front (Salt Lake City) and Denver AQCR's (originally Priority III), arsenite data shows concentrations below the cutoff point for a Priority III classification, but chemiluminescence and/or Saltzman data show concentrations above it.

For more complete details of comparative sampling data performed in the original 47 Priority I AQCR's, refer to Federal Register 38 FR 15181 (Federal Register 1973).

Proposed Reference Methods

Three methods of ambient NO_2 monitoring have been proposed as the Federal Reference method: the modified sodium arsenite method, the continuous Saltzman method and the chemiluminescence detection method.

Each method has its own inherent limitations of either interferences, stability, complexity, toxicity, or cost. Each of the methods is briefly described.

Arsenite Method

Figure App. 14-B2 (Federal Register 1973) displays the apparatus used to determine NO_2 (24-hour sample) by the Arsenite Method. Air is bubbled through a reagent of sodium hydroxide-sodium arsenite solution to form a stable solution of sodium nitrite (Christie et al. 1970). The nitrite ion produced during sampling is reacted with phosphoric acid, sulfanilamide, and N-1-naphthylethylenediamine dihydrochloride to form an azo dye. NO_2 concentrations are infered by measuring the azo dye colorimetrically.

The method has an overall conversion efficiency of 85 percent over the range of 50-750 $\mu g/m^3$ NO_2. However, it has been reported that there is a 12 percent positive interference for NO which can be deducted (Saltzman 1973). The toxicity of the absorbing reagent is an obvious disadvantage of this method.

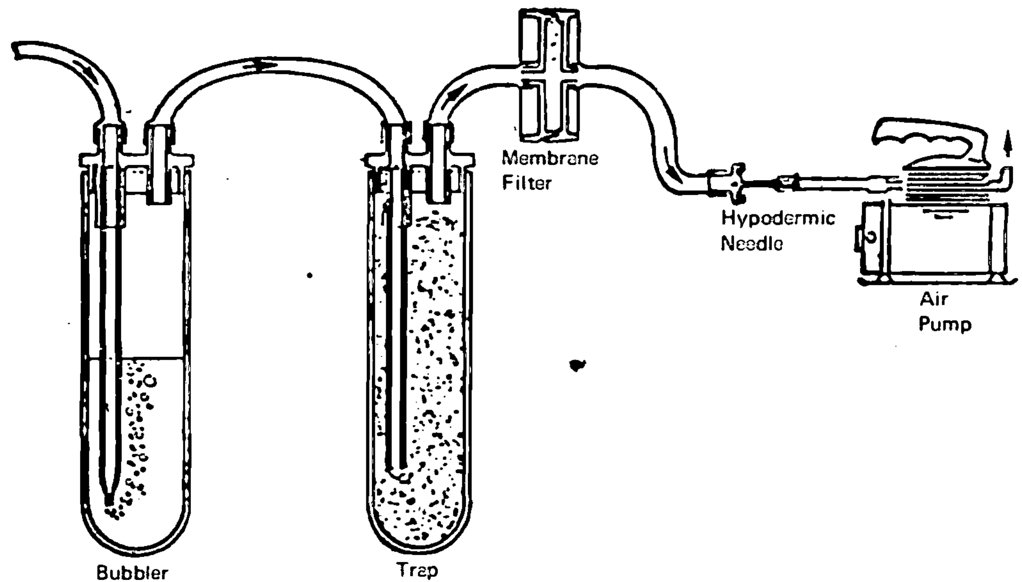

FIGURE 14-B2: Sampling Train for EPA Tentative Candidate Arsenite Method (USEPA 1972b).

Continuous Saltzman Method

The principles of this method have been used for ambient air sampling for over 14 years (Saltzman 1960, Lyshkow 1965). By contacting an air flow with a liquid diazotizing-coupling reagent, the NO_2^- (nitrite ions) form a deeply colored azo dye which is measured colorimetrically. The absorbance of the azo dye is directly proportional to the number of NO_2 molecules in the air stream that are converted to NO_2^-.

This continuous method is applicable for concentrations from 18.8 $\mu g/m^3$ to 1880 $\mu g/m^3$ (.01-1 ppm). Apparently there is some interference from ozone. This is a negative interference reported as 5.5 percent for $NO_3/NO_2 = 1$, 19 percent for $O_3/NO_2 = 2$ and 32 percent for $O_3/NO_2 = 3$ (Federal Register 1973).

This method has the drawback of requiring considerable skill and effort for calibration and maintenance, especially replacement of the liquid reagent.

Chemiluminescence Method

The chemiluminescence instrument represents a major break with absorbing wet chemistry methods of ambient air sampling. Figure App. 14-B3 schematically illustrates the NOx chemiluminescent analyzer (Federal Register 1973). In general, the analyzer draws an air sample into a light-tight reaction chamber and mixed with ozonated air. The chemiluminescence produced by the reaction of nitric oxide is measured by a photomultiplier tube, and the data is stored in a peak-holding amplifier.

The first generation chemiluminescence instruments alternately sampled NO and total NOx. The total NOx was determined by converting all NO_2 to NO and measuring as described above. The difference between total NOx and NO was taken as NO_2. This 1-minute cycling of samples often gave negative NO_2 values when NO concentrations were high and rapidly varying, such as near traffic. The latest analyzers operate

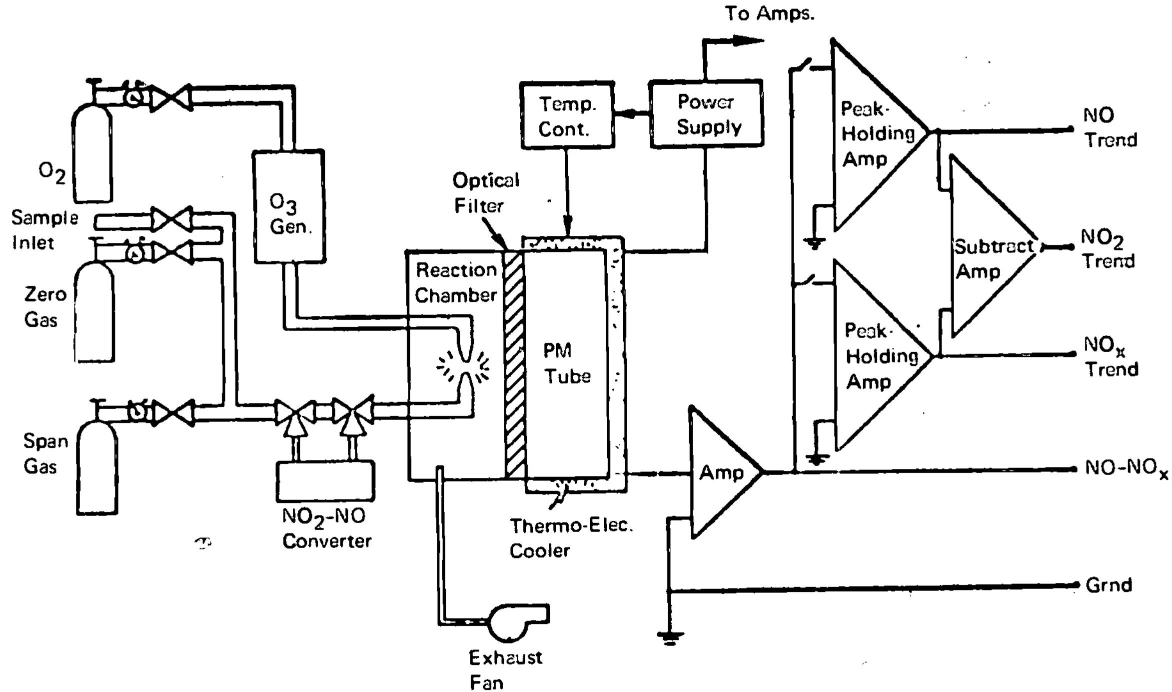

FIGURE App. 14-B3: Schematic diagram of automated chemiluminescent analyzer for NO and NO_2.

with dual chambers or a rapidly cycling single chamber. This improvement has apparently removed spurious negative values.

Since the instrument measures the light emissions of individual molecules, its lower detectable values are dependent on photomultiplier sensitivity. The lower limit of this method is approximately 9.4 $\mu g/m^3$ (0.005 ppm).

The direct measurement of NO has no interferences. However, the conversion of NOx to NO by the converter can decompose unstable nitrogen compounds to give a falsely high NOx reading. Fortunately interference is small (Federal Register 1973).

Selection of a New Federal Reference Method

An extensive field study is now being conducted by EPA to determine the proper priority classifications of numerous air quality control regions. The arithmetic average results for 42 regions by simultaneous analysis with the Tentative Candidate Chemiluminescent Method and the Tentative Candidate Arsenite Method have been tabulated in 38 FR 15174. Figure App. 14-B4 shows a plot derived from these data by Saltzman. Even though the methods were selected as best, after much testing, there is a disturbing scatter in the comparison plot. The straight line fit by the method of least squares has the equation:

$$Y = 0.51\ X + 25.2$$

The diffuculty and expense of validating analytical methodology has been grossly underestimated in the past.

Calibration Techniques

Standardized calibration techniques are essential to all ambient air quality monitoring. Precise procedures for calibration of each of the tentative NO_2 methods are given in 38 FR 15174. There are two techniques described:

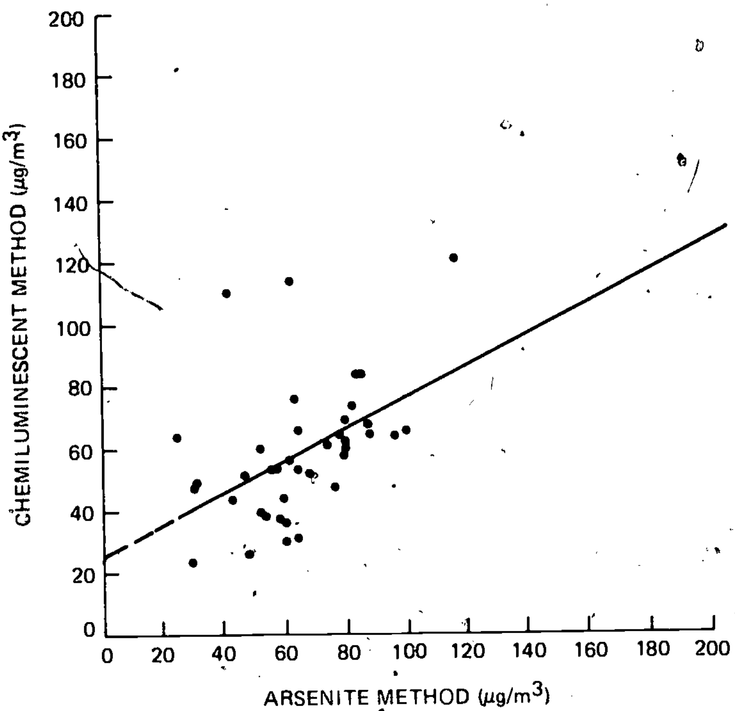

FIGURE App. 14-B4: Comparisons of average NO_2 Concentrations by Two EPA Tentative Candidate Methods in 42 Air Quality Control Regions (Junge 1963).

NO_2 permeation tube and dilution, and NO_2 gas-phase titration.

The latter, gas-phase titration, is diagrammed in Figure App. 14-B5 (Federal Register 1973). This apparatus can calibrate chemiluminescent NO, NO_2, and O_3 instruments.

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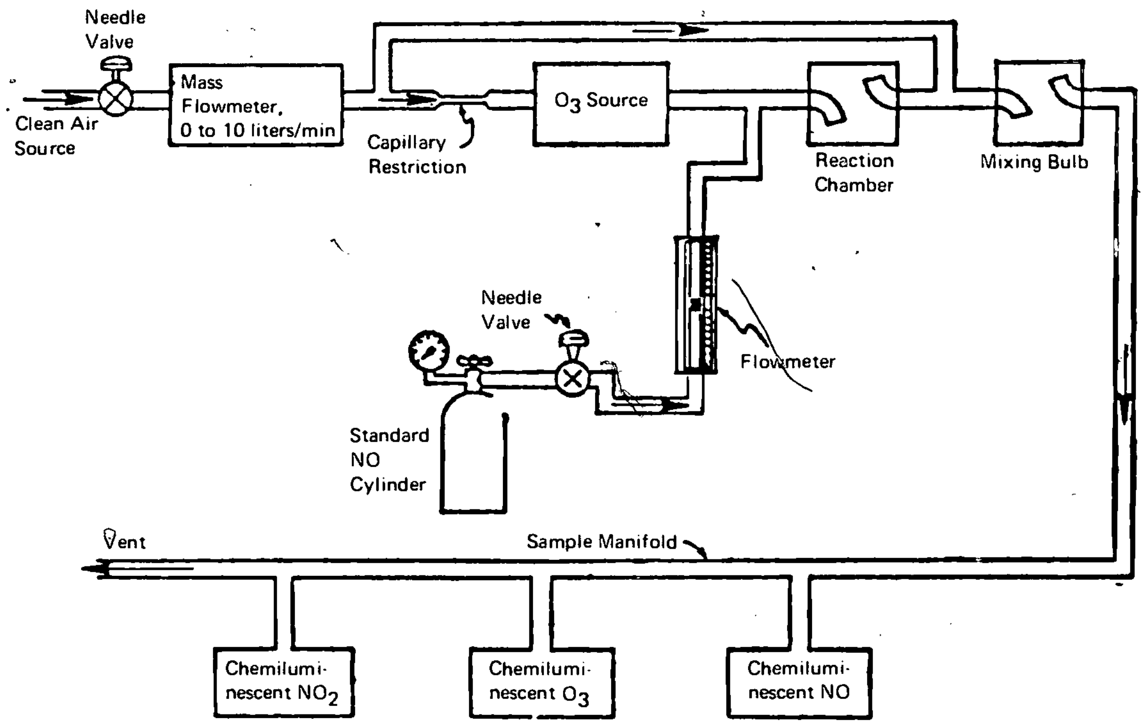

FIGURE APP. 14-B5: Flow scheme for calibration of NO, NO_2, NO_x, and O_2 monitors by gas-phase titration.

APPENDIX 14-C

PROJECTIONS OF FUTURE NOx EMISSIONS

TOTAL EMISSIONS: 1972-1990

Total emissions of nitrogen oxides projected to 1990 assuming implementation of the present statutory program for NOx control are displayed in Table App. 14-C1 and Figure 14-16 in Chapter 14. This estimate projects a 36 percent increase in total emissions from 1972-1990. The largest increases in emissions will be produced by industrial process losses (98 percent), non-highway transportation (72 percent), industrial fuel combustion (54 percent) and electric power generation (49 percent). Highway-related transportation emissions will decline by 24 percent during this period assuming the statutory emission standards for automobiles are achieved. The estimate is considered to be conservative since it assumes that (1) most new electric power generation will be produced by nuclear reactors, (2) the statutory automotive emission standards will remain in effect and be achieved and (3) the 1940-1970 annual growth rate of NOx emissions from industrial, commercial and institutional fuel combustion will slow during the next two decades.

Emission projections for electric power generation are based on recent electric generation forecasts by the Federal Energy Administration (USEPA 1975) and the National Academy of Engineering (NAE 1974). These projections are discussed in greater detail below.

Industrial fuel combustion emissions of nitrogen oxides grew at the rate of 9.5 percent per year during the 1960-1972 period due to a

TABLE App. 14-C1

Nationwide NOx Emissions Projected to 1990 Assuming the Present Statutory Program

Source Category	NOx Emissions (10^6 tons/year) 1972	1980	1985	1990
Stationary Fuel Combustion	12.27	15.96	16.82	18.46
Electric Generation	5.94	8.16	8.20	8.88
Industrial	5.39	6.73	7.46	8.31
Commercial-Institutional	0.65	0.76	0.84	0.93
Residential	0.29	0.31	0.32	0.34
Industrial Process Losses	2.88	3.91	4.72	5.71
Solid Waste Disposal	0.18	0.22	0.25	0.28
Transportation[a]	8.45	8.47	7.49	7.60
Road Vehicles	7.48	7.14	5.89	5.68
Gasoline	6.59	5.97	4.30	3.95
Diesel	0.89	1.17	1.59	1.73
Other	0.97	1.33	1.60	1.92
Miscellaneous[b]	0.59	0.74	0.87	1.02
TOTAL	24.37[c]	29.30	30.15	33.07

[a] Assumes a 4% annual VMT growth rate

[b] Includes New York City Point sources assumed to grow at 4% per year

[c] 1972 total emissions are 270,000 tons lower than those summarized in Table 2.2 due to the use of new road vehicle emission factors

large increase in natural gas and industrial process gas combustion. In this projection it is assumed that the growth rate will slow to approximately 2.5 percent per year through 1990 due (1) to a gradual shift to purchase of electricity from utility companies, and (2) to an assumed 3 percent annual growth rate in use of natural and industrial process gas, and in the use of coal. A 3 percent growth rate for industrial process gas use follows the growth rate for steel and petroleum production. No increase in oil consumption is assumed after 1975. Since there are no federal new source performance standards and few state regulations for NOx emissions from medium-sized and small boilers, all emissions are uncontrolled. Because historical trends show a much larger growth rate than assumed here, these projections are considered conservative.

NOx emissions from commercial and institutional fuel combustion have grown at the rate of 4.5 percent per year over the last three decades. In this projection it is assumed that the growth rate will slow to approximately 2 percent per year through 1990 due to (1) a gradual shift to the purchase of power for space heating and electricity from utility companies, and (2) energy conservation measures. No controls were assumed for industrial fuel combustion sources. These projections are also quite conservative since they depart significantly from historical trends.

Residential fuel combustion emissions of NOx have declined at the rate of 2 percent per year over the last three decades. This is primarily due to a shift in fuel usage from coal to distillate oil and natural gas and to the purchase of power for space heating from electric utilities. In this projection it is assumed that residential emissions will grow 1 percent per year through 1990 since: (1) the benefits of a coal to oil or natural gas shift will all have been realized by 1975; (2) the number of housing units increase at a rate of 2 percent per year; and (3) approximately half of the new housing units will be heated by electricity.

Lack of knowledge of NOx emission factors from industrial process losses in the past make historical trends for these sources difficult to interpret. It is assumed that refinery emissions which amount to 86 percent of all industrial process loss emissions, will increase at the rate of 4 percent per year, which is the growth rate for petroleum usage. Since there are no federal new source performance standards for NOx from refineries it is assumed that these sources will be uncontrolled. Nitric acid manufacturing emissions of NOx are assumed to grow at 2.6 percent per year which reflects the mix of old and new sources and the 90 percent control of emissions from new sources required by federal new source performance standards. These emission estimates are conservative since baseline 1972 petroleum refinery emissions are probably underestimated.

NOx emissions from solid waste disposal grew at a rate of 4 percent per year from 1940-1970 due to open burning and incineration. State implementation plans required by the Clean Air Act will eliminate most of the open burning emissions so that it is more realistic to assume only a 1 percent annual growth rate of such emissions through 1990. The scarcity of land for sanitary landfill, and the increase in per capita solid waste production will probably result in at least a 5 percent annual growth rate in incineration emissions. Overall, it is assumed that solid waste emissions will grow at a rate of 2.6 percent per year.

Non-highway transportation sources grew at the rate of 5 percent per year from 1940-1960 and 12.5 percent per year from 1960-1970. As reliance on petroleum products decreases with energy conservation measures, this projection assumes that the growth rate will slow to 4 percent per year through 1980 and 3.8 percent per year from 1980-1990. The slower increase in the 1980-1990 period is due to the effect of new aircraft emission standards effective on 1979 and later model year aircraft engines. Again, it is likely that the estimates are conservative since they depart significantly from historical trends.

ELECTRIC POWER GENERATION EMISSIONS: 1972-1990

Emission projections for electric power generation were based on recent electric generation forecasts by the Federal Energy Administration (USEPA 1975) and the National Academy of Engineering (NAE 1974). These forecasts include generating capacity and actual power output for 1980 and generating capacity projections for 1985. Actual power output for 1985 and both generating capacity and actual power output for 1990 were projected from 1972-1980 data which indicates a 6.5 percent annual increase in power usage. The ratio of actual power output to generating capacity, i.e., load factor, was assumed to remain constant from 1980 to 1990 for each power source. The projections of generating capacity, actual power output and emissions are summarized in Table App. 14-C2.

As seen in Table App. 14-C2, nuclear power generation will increase significantly from 1972-1990 under the assumptions of Project Independence. Coal generation will also increase and natural gas generation will decrease by 10 percent during that same period. All new coal, oil and natural gas generation is assumed to meet federal new source performance standards for NOx emissions. One-quarter of the existing plants are assumed to be retired and replaced by new facilities every 5 years. Resulting emissions from electric power generation will increase 49 percent from 1972-1990 due to the increase in emissions from coal fired plants.

These emission estimates reflect the projected predominance of nuclear power in electric generation. If for some reason a decision should be made not to build any new nuclear power plants after 1975, quite a different picture would emerge. As can be seen in Table App. 14-C3 and Figure App. 14-Cl, such a policy decision would result in nearly a doubling of NOx emissions in 1990 if coal fired plants provided the capacity that was projected for nuclear power. If the projected growth of nuclear generation were slowed rather than halted then the resulting NOx emissions would fall somewhere between the projected extremes displayed in Table App. 14-C3 and Figure App. 14-Cl.

TABLE App. 14-C2

NOx Emission Projections to 1990 from Electric Power Generation Assuming Project Independence

Energy Source	Gen. Cap. (MW)	Power Output (10^9 kwh)	Emissions (10^6 tons)	Gen. Cap. (MW)	Power Output (10^9 kwh)	Emissions (10^6 tons)	Gen. Cap. (MW)	Power Output (10^9 kwh)	Emissions (10^6 tons)
		1980			1985			1990	
Coal	295,000	1608	7.21	330,000	1734	7.21	402,000	1972	7.89
Oil	59,000	165	0.52	52,000	146	0.52	52,000	146	0.52
Natural gas	86,000	145	0.48	78,000	178	0.44	78,000	178	0.44
Gas Turbine	61,000	53	0.03	61,000	53	0.03	61,000	53	0.03
Hydro-electric	88,000	325	-	75,000	276	-	75,000	276	-
Nuclear	73,000	394	-	250,000	1314	-	500,000	2,452	-
Other	-	-	-	61,000	53	-	75,000	66	-
Total	662,000	2,740	8.16	907,000	3,754	8.20	1,243,000	5,143	8.88

TABLE App. 14-C3

Nationwide Emissions of NOx from Electric Power Generation Projected to 1990 for Two Policy Options

YEAR	NOx Emissions (10^6 tons/year)							
	Project Independence				No New Nuclear Plants Built After 1975			
	Total[a]	Coal	Oil	Natural Gas	Total[a]	Coal	Oil	Natural Gas
1972	5.94	3.95	0.85	1.14	5.94	3.95	0.85	1.14
1980	8.24	7.21	0.52	0.48	9.32	8.29	0.52	0.48
1985	8.20	7.21	0.52	0.44	12.81	11.82	0.52	0.44
1990	8.88	7.89	0.52	0.44	17.56	16.57	0.52	0.44

[a]Total contains o.o3 x 10^6 tons/year from gas turbines

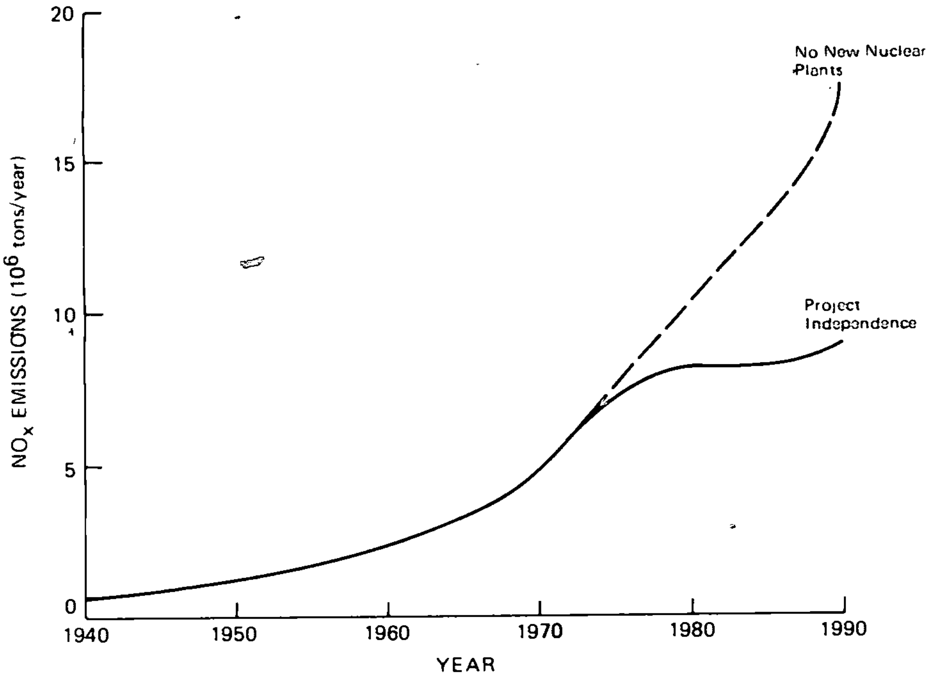

FIGURE App. 14-C1: Nationwide Emissions from Electric Power Generation Projected to 1990 for Two Policy Options.

ROAD VEHICLE PROJECTIONS

Past highway statistics (FHA 1972) on vehicle-miles-of-travel (VMT) are used to project NOx emissions to 1990 for road vehicles including light-duty passenger vehicles, light and heavy duty gasoline trucks and heavy duty diesel trucks and buses. Three options for control are considered: (1) no controls on NOx emissions with the statutory program for CO and HC in effect; (2) a 5 year delay of the 1977 statutory standards, i.e., maintenance of the interim standard for NOx emissions of 2 g/mile until 1983; and (3) the present statutory program (NOx emissions of 0.4 g/mile by 1977). Two different VMT growth rates are considered: (1) 2 percent annual increase and (2) 4 percent annual increase. The former assumes stringent gas conservation measures while the latter assumes no restraint. The resulting projections are displayed in Table App. 14-C4 and Figure App. 14-C2.

The present statutory program will result in a 25 percent reduction in 1972 road vehicle emissions by 1990 for a VMT growth rate of 4 percent. A 5 year delay of the 1977 automotive standards will result in only a 17 percent reduction in 1972 emissions by 1990 for the same VMT growth rate. In addition to growth in total VMT, growth in uncontrolled heavy duty gasoline and diesel truck usage will be responsible for the fact that emission reductions predicted in 1990 are considerably smaller than the percentate reduction in emissions per mile required by the standards for light-duty automobiles. These heavy duty sources become major contributors to total transportation emissions during the 1980's.

TABLE App. 14-C4

Nationwide NOx Emissions From Road Vehicles Projected to 1990 for Three Policy Options

	NOx Emissions (10^6 tons)					
	2% Annual Growth in VMT			4% Annual Growth in VMT		
YEAR	No Control	Present Statutory Program	Proposed 5-year Standards Delay[a]	No Control	Present Statutory Program	Proposed 5-year Standards Delay[a]
1972	7.62	7.48	7.48	7.62	7.48	7.48
1975	8.55	7.82	7.82	9.08	8.31	8.31
1980	9.49	61.4	7.37	11.06	7.14	8.60
1985	10.35	4.44	5.60	13.48	5.89	7.38
1990	11.22	4.03	4.38	15.86	5.68	6.18

[a]1977 auto emission standards for NOx postponed until the 1982 model year. California interim standard of 2.0 g/mi applies to 1977-1982 model years.

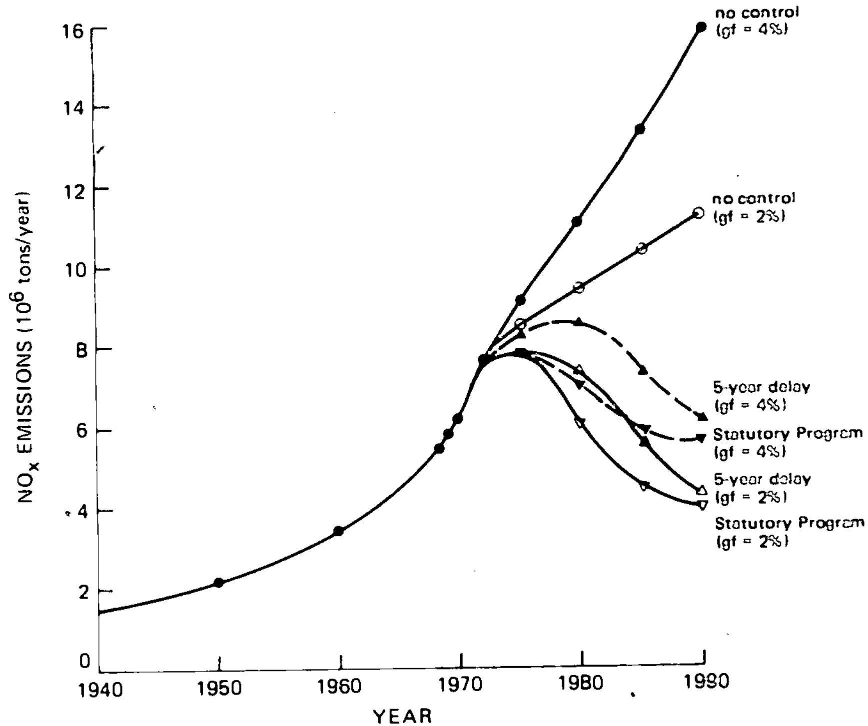

FIGURE App. 14-C2: Nationwide NOx Emissions from Road Vehicles Projected to 1990 for Three Policy Options.

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CHAPTER 15

NITROGEN OXIDE CONTROL TECHNIQUES

OVERVIEW

This chapter makes substantial use of reports and information made available by a large number of government and private institutions. Special thanks are due to the Combustion Research Section of the EPA Control System Division.

Throughout this section, emissions have been converted to lbs/10^6 BTU expressed as NO_2 in order to facilitate comparison of emissions from different sources. A table of conversion factors appears in Appendix 15-A.

Typically, nitrogen oxides are formed in localized, high-temperature regions in combustors by the oxidation of both atmospheric nitrogen (thermal NOx) and nitrogen that may be contained in the fuel (fuel NOx). The formation of NOx in combustion systems can be suppressed, with varying degrees of success, by reducing the oxygen content and the temperature in the localized regions in the combustor contributing to emissions, usually in the vicinity of the flame. Reductions in the oxygen content in the flame zone reduce the emissions of both fuel and thermal NOx; reductions in temperature, however, produce significant reductions in only the thermal NOx.

Injection of cooled combustion products, steam, or water into the flame volume; reduction of temperature to which combustion air is preheated; and extraction of heat from the flame volume are methods that have been used to reduce the temperature in the combustor. The injection of cooled combustion products into the flame volume can be achieved externally by ducting relatively cool combustion products to the burner area (referred to as flue gas recycle for furnaces or exhaust gas recycle for engines) or internally by modifying the burner design so as to induce the entrainment of colder combustion products by the hot gases leaving a burner.

Methods for reducing the oxygen content in the flame zone involve lowering the volume of air supplied to the burners by reducing the overall air/fuel ratio to the combustor (referred to as low-excess-air firing) or by reducing the air/fuel ratio for some burners without reducing the overall air/fuel ratio (referred to as staged combustion).

Low excess-air firing can achieve modest reductions in NOx emissions. However, its application is limited by the ability to reduce the air/fuel ratio to burners without producing excessive emissions of carbon monoxide and particulates.

There are several variations on staged combustion, referred to as biased firing, off-stoichiometric firing, overfire air and two-stage combustion. In general, the air/fuel ratio to some or all of the burners is reduced, to as low as seventy percent of the value required for combustion. The combustion is then completed by the addition of the balance of the air requirement after an interval in which the combustion product is allowed to partially cool. In engines a form of staged combustion is achieved by stratifying the fuel charge to provide local regions of relatively high fuel concentration, facilitating combustion at overall air/fuel ratios considerably higher than those practical for a uniform charge.

Many of the results reported in this chapter are from tests performed in laboratories or on small-scale units. Some of the results point to promising techniques or methods for nitrogen oxide emission control. However, it should be borne in mind that the application of test results to large scale, operational systems often introduces significant problems, many of which are indicated throughout the chapter. The success of a test does not necessarily imply easy transfer to a large, commercially practicable system and considerable effort is being, and will continue to be, directed toward the resolution of the problems which arise when such a transfer is attempted.

Utility boilers and gas turbine engines have received priority in the development of nitrogen oxide emission control technology. Other major sources, particularly stationary reciprocating engines, have received relatively little attention.

Utility Boilers

As noted in Chapter 14, utility boilers fired by coal , gas, and oil accounted for nearly half of the stationary emissions of NOx in 1972. Based upon a field test performed by Esso Research and Engineering and other data, EPA has developed emission factors which indicate that NOx emission rates for existing boilers range from 0.6 to 1.2 lbs/10^6 BTU for coal-fired units, from .27 to .7 lbs/10^6 BTU for oil-fired units, and from .12 to .7 lbs/10^6 BTU for tangentially fired boilers using gas. The lowest emissions were from tangentially-fired units and the highest from units with intense combustion. Emissions decreased substantially with decreased furnace load.

Low-excess-air firing, staged combustion, flue-gas recirculation, water injection and reduced air preheat are control techniques that have been successfully demonstrated on field units. The latter two methods, however, have an associated and usually unacceptable penalty in thermal efficiency of the boiler. Flue gas recirculation is most effective for gas-fired units but is relatively uneffective for oil-fired and coal-fired units in which the fuel NOx contribution is significant. By the utilization of a combination of control techniques an average reduction in emissions of 60 percent has been achieved for gas-fired units, 48 percent for oil, and 37 percent for coal. In the case of coal, the locally fuel rich conditions may contribute to slagging or corrosion problems. Accelerated corrosion testing over 300 hours showed no adverse effects but longer range trials for a wide range of coals are needed before the control techniques can be commercially accepted.

The applicability of combustion process modification to existing furnaces must be evaluated on a case-by-case basis. Boilers can generally be adapted for low-excess-air firing and staged combustion without major modification. Flue-gas recirculation, however, may be impractical for retrofitting except on units which have had the ducting already installed for steam temperature control. The capital costs vary widely with specific installation size and design; they range from under $0.50/KW for staged combustion to $6.0/KW for flue gas recirculation on existing units and from negligible costs for staged combustion to $4.0/KW for flue gas recirculation on new units.

Equipment manufacturers can meet existing emission standards on new units but usually specify an upper limit on the nitrogen content of the fuel oils that can be burned without exceeding the standards. EPA has proposed emission levels that might be achieved in the future: 0.12 $lb/10^6$ BTU(100 ppm) for gas, 0.20 $lb/10^6$ BTU(150 ppm) for oil, and 0.28 $lb/10^6$ BTU (200 ppm) for coal by 1980; and 0.06 $lb/10^6$ BTU(50 ppm) for gas, 0.12 $lb/10^6$ BTU(90 ppm) for oil, and 0.14 $lb/10^6$ BTU(100 ppm) for coal by 1985. These are technological goals that can probably be met on individual units if adequate research is carried out on new control techniques, pilot-plant development and field-demonstration. It should be recognized that high-intensity combustion burners may be required to burn certain types of coals, and these will yield higher emissions than the projected goals. Further, there is a significant lag time between the development of new designs and installation of field-units by the utilities.

Industrial Boilers

Most boilers with 10 to 500 million BTU/hr capacity can meet the new source performance standards for units greater than 250 million BTU/hr capacity with only minor modifications in operating conditions. Still lower emission levels are possible, but redesign of the boilers would be required to allow off-stoichiometric or staged-combustion in units burning heavy fuel oils or coal since very few existing units possess the necessary flexibility. Problems which must be considered in the design of new units, and particularly in the modification of existing units, include corrosion and deposits on boiler tubes, flame instability, and combustion noise. The level of control that is achievable on industrial boilers is close to but not as great as that attainable with utility boilers.

Commercial and Residential Space Heating

Small space heating equipment (less than 10 million BTU/hr) fueled with natural gas or distillate oil generally has the lowest specific NOx emissions of any class of combustion equipment (i.e., 0.05 to 0.20 lb $NO_2/10^6$ BTU) and produces about seven percent of the stationary source NOx. Existing units have little flexibility for NOx control, and the emissions are unaffected by normal maintenance operations. Combustion improving devices generally increase NOx emissions by producing a more intense flame region, but optimized burners, based either on conventional designs or on new combustion concepts such as catalytic combustion, which emit from 0.015 to 0.05 lb $NO_2/10^6$ BTU have been demonstrated in tests.

Stationary Engines

About 18.8 percent of the stationary source NOx is produced by reciprocating engines. Spark-ignition gas engines produce about 63 percent of the engine NOx, primarily in applications associated with the gas industry, i.e., pipelines and natural gas production and processing.

Moderate reduction in NOx emissions from these engines (20 to 40%) can be achieved, while reducing fuel consumption, by cooling the fuel/air mixture. Further reductions in NOx emissions from existing engines are possible, but have generally been accompanied by a significant increase in fuel consumption. New engine designs, such as those using stratified charge concepts, may produce substantial reductions in NOx without increasing fuel consumption; however, further development work is required.

Diesel engines produce about 34 percent of the engine NOx in agricultural and industrial applications. As in spark ignition engines, cooling the intake air, can reduce NOx emissions while decreasing the fuel consumption. Water injection can also reduce NOx emissions with little change in fuel consumption; however, corrosion may be a problem, particularly if the engine is used on a standby basis. NOx emissions have been reduced by as much as 50 percent with little penalty in fuel consumption, but lower emission levels may require substantial engine redesign, e.g., prechamber engines; or exhaust gas treatment using catalytic reduction of NO to N_2; or may result in significant increases in fuel consumption. EPA projects that emission levels of 0.14 lb $NO_2/10^6$ BTU for spark ignition gas engines and 0.16 lb $NO_2/10^6$ BTU for diesel engines may be reasonably achieved by 1980, but there may be some increase in fuel consumption.

Stationary gas turbine NOx emissions can be reduced to the levels of currently proposed emission standards through the use of steam or water injection into the combustion chamber. Further reductions are possible through combuster design modifications including use of out-of-line combustion chambers; improved fuel atomization and mixing; prevaporized, premixed combustion; internal recirculation of combustion products; and surface combustion. The use of heavy fuel oils, rather than natural gas or distillate oils, will probably result in higher emission levels due to fuel nitrogen and may require new measures to control NOx.

The efficiency of gas turbines varies from 24 percent for simple cycle gas turbines to more than 40 percent for combined gas turbine - steam turbine cycles. Emission standards based upon thermal input (e.g., lb.$NOx/10^6$ BTU or ppm NOx at 15 percent oxygen) provide no credit for higher efficiencies and, in fact, may inhibit the development of more efficient systems.

Fluidized Bed Combustion

Fluidized bed combustion of coal provides a potential alternative to current utility boiler design that is competitive when a low-sulfur fuel or flue-gas treatment is required to meet the emission standards for sulfur oxides. Atmospheric pressure and pressurized combustors with dolomite injection for control of sulfur oxide emissions are at a pilot-plant stage. A demonstration of a 30 MW atmospheric pressure fluidized-bed combustor is scheduled for 1975 and it is projected that 200 MW and 800 MW units may be built as early as 1977 and 1980, respectively.

At the low temperatures of operation of fluidized bed combustors, typically 1500F to 1800F, most of the NOx is contributed by the oxidation of fuel nitrogen. Tests on laboratory and pilot-scale fluidized-bed combustors have yielded emissions that meet the current standards for new coal-fired units. Emissions as low as 0.11 lbs/10^6 BTU have been obtained without the benefit of staging. Tests on larger-scale units are needed to establish practical emission levels for commercial units.

Tall Stacks and Intermittent Control Strategies

The only intermittent control strategy that appears practical for NOx emission reduction is load switching of electric power generation. Load switching has limited applicability because of the variability in the contribution of electric power generation to local emissions. The advantages of tall stack release of sulfur dioxide to reduce ground level concentrations do not apply for NO. Tall stacks potentially reduce ground level NO concentrations; however, NO converts to nitric acid and nitrates faster than does SO_2 to sulfuric acid and sulfates and since the reaction products precipate, there is a greater potential for local impact. There is considerable uncertainty about the effects of NOx release from tall stacks on the formation of photochemical oxidants and the ground level concentrations of oxidants and nitrogen dioxide.

Future Trends

A major shift from gas to coal is expected in fuel utilization for utility boilers, and to a lesser extent smaller combustors. Emissions would be expected to increase unless new levels of control are achievable for the combustion of coal. Shale and synthetic fuels from coal may have higher nitrogen contents than existing fuels and may yield high NOx emissions.

Although significant success has been achieved in reducing NOx emissions from stationary sources, the reductions that have been achieved, typically 50 percent, are much smaller than existing potential. New combustion systems, utilizing surface or catalytic combustion, have the potential to eliminate thermal NOx. Significant reduction in the emissions from coal-fired units have been achieved by burner modification on a pilot scale, and the potential exists of designing burners with emission levels for coal below 0.2 $lb/10^6$ BTU.

Flue gas treatment is more expensive than combustion process modification but at present can attain much higher reduction in NOx emissions. A number of flue gas treatment methods are at a pilot plant demonstration stage, primarily in Japan.

Institutional Constraints

The reduction of NOx from stationary sources is subject to a number of economic and logistic constraints. Capital availability may limit the rate at which new, more expensive equipment with reduced NOx emissions can be brought on line and may severely constrain the rate at which existing equipment is brought into compliance with emission standards by retrofit. Increased operating costs due to new maintenance requirements, increased fuel consumption, decreased capacity, or increased labor to provide the closer control of combustion conditions required for NOx control must also be factored into the decision making process.

Equipment outages for modification of existing facilities present an additional constraint, particularly in the electric utilities. Some modifications may be prepared in advance and installed during scheduled outages; however, prolonged outages, which may be required for the modification of some units, may strain the system generating capacity and will, therefore, require careful scheduling in order to permit utilities to meet demand. The logistics of modifying a very large number of units of diverse types may inhibit the retrofit of smaller sources.

NOx control techniques have been developed for those applications for which emission regulations have been proposed (e.g., utility boilers and stationary gas turbines) providing an economic incentive for equipment manufacturers to develop NOx controls. Where funding has been available for research on NOx control, significant progress has been made toward developing control techniques. On the other hand, there is little incentive to develop burners or small package boilers with low-NOx emissions, so progress in development is slow for these systems. Reduction of NOx emissions from systems for which development incentives do not exist, (e.g., industrial process furnaces) has received little attention.

INTRODUCTION

Stationary Sources of Nitrogen Oxides

Stationary sources currently contribute more than half of the man-made emissions of NOx in the U.S. The NOx emissions are dominated by combustion sources burning coal, gas, and oil (Mason and Shimzu 1974). The major sources are utility boilers and stationary engines. Nitric acid and nitrogen fertilizer plants contribute a small fraction of the national total, although their emissions may be locally significant in some areas.

The nitrogen oxides emitted by combustion sources are predominantly in the form of nitric oxide (NO) with the residual, usually less than 5 percent, in the form of nitrogen dioxide (NO_2). The oxides are formed either by the oxidation of atmospheric nitrogen at high temperatures (thermal NOx) or by the oxidation of nitrogen compounds in the fuel (fuel NOx). The relative contributions of thermal and fuel NOx depend on combustor design and operating conditions, as well as on the nitrogen content of the fuel. As much as half of the total stationary emissions may be contributed by the oxidation of the nitrogen in the fuel, primarily in units burning heavy-oils and coals.

Control Options

The technology for controlling the emission of nitrogen oxides from combustion sources is based on either the modification of the combustion process to prevent formation of the oxides or the treatment of the product gases to destroy or remove the oxides. The latter option is made difficult by the relative inertness and insolubility of NO; wherever combustion process modification has yielded the needed reduction in NOx emissions, it has proven to be the most economical method for doing so. In some cases, where stringent emission controls have been imposed on combustion sources, a combination of combustion process modification and product gas treatment has been proposed (Tohata 1974). For NOx emissions from chemical plants, a variety of control techniques are available, including catalytic destruction, afterburning, absorption and adsorption.

MECHANISMS FOR NITRIC OXIDE FORMATION

Thermal Fixation of Atmospheric Nitrogen

The formation of thermal NOx is determined by highly temperature dependent chemical reactions, the so-called Zeldovich (1946) reactions. The rate of formation is significant only at high temperatures (greater than 3300F) and doubles for every increase in flame temperature of about 70F. The rate of formation increases with increasing oxygen concentration (rate proportional to the square root of the oxygen concentration) except in a small region near the flame zone in which superequilibrium concentrations of oxygen atoms are found (Thompson et al. 1972, Sarofim and Pohl 1973, **Livesey** et al. 1971). Although the NOx emitted by most practical combustors is predominantly in the form of NO, there is evidence from laboratory studies that a significant fraction of the NOx may be present as NO_2 in localized regions of a flame (Merryman and Levy 1974).

Formation of Fuel NOx

An upper bound for the potential contribution of fuel-nitrogen to NO_x formation may be gauged from the nitrogen content of fuels. Natural gas has negligible amounts of organically bound nitrogen, U.S. crude oils have nitrogen concentrations that average about 0.15 percent by weight (Martin and Berkau, Ball et al. 1951) and U.S. coals have nitrogen concentrations that average about 1.4 percent by weight. The nitrogen content in the crude oil varies between oil fields (Table 15-1), and is high for California oils. During the refining of oils, the nitrogen

TABLE 15-1

Nitrogen Content in U.S. Crude Oils in Million Barrels
(Ball et al. 1951, Ball and Wenger 1958, Vandaveer 1965, Aga et al. 1973)

	Proved Reserves (1972)		Production (1972)		Average Weight % Nitrogen
	MM bbl	%	MM bbl	%	
Texas	12,144	33.4	1,258	38.3	0.074
Alaska	10,096	27.8	73	2.2	----
Louisiana	5,028	13.8	780	23.8	0.056
California	3,554	9.8	346	10.5	0.49
Oklahoma	1.303	3.6	198	6.0	0.151
Wyoming	950	2.6	139	4.2	0.183
New Mexico	582	1.6	106	3.2	0.082
U.S. Total	36,339	92.6	3,231	88.2	----

concentrates in the heavy fractions so that, residual oils from a California crude may have as much as 1.1 percent nitrogen whereas light distillate oils generally have nitrogen concentrations under 0.1 percent. The spread in nitrogen concentrations for coals is shown in Figure 15-1. The concentration is reported as lbs-NO_2 per million BTU assuming 100 percent conversion of fuel nitrogen to NO_2. Also shown on the plot are the sulfur concentrations, reported as lbs-$SO_2/10^6$ BTU, and the percentage of U.S. coal reserves that have nitrogen and sulfur concentrations below certain levels. There is no apparent correlation between coal nitrogen content and coal sulfur content.

A fraction of the fuel nitrogen is converted to nitric oxide in practical combustors. Tests on carefully controlled laboratory-scale units have typically shown that 15 to 100 percent of the fuel nitrogen is converted to NOx (Figure 15-2), with the higher conversion efficiencies obtained when the nitrogen content is low or when the combustor is operated lean.

Because it is not easy to separate the contribution of fuel NOx from thermal NOx, the conversion efficiency of fuel nitrogen to NOx in large scale units is not known with certainty, but estimates obtained from field tests show trends similar to those obtained in the laboratory. A plot of the total uncontrolled NOx emissions from large and small scale units, Figure 15-3, suggests that the emissions increase with increases in fuel nitrogen content for units ranging in size from laboratory units to utility boilers. Emissions from large gas-fired units account for the high emission data at zero nitrogen content.

The mechanism by which the fuel nitrogen is converted to nitrogen oxides is imperfectly understood. The conversion efficiency of fuel nitrogen to NOx is found to increase markedly with increased oxidizing conditions in the flame but is insensitive to changes in temperature (Fenimore 1972, Martin and Berkau).

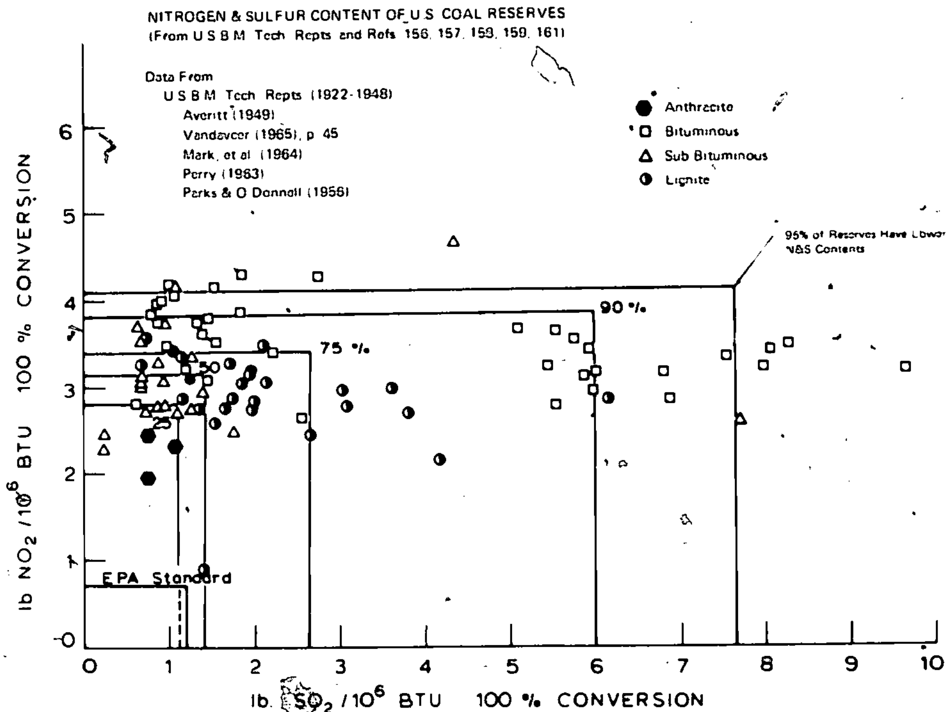

FIGURE 15-1: Nitrogen & Sulfur Content of U.S. Coal Reserves (From U.S.B.M. Tech. Repts. and Parks and O'Donnell 1956, Mark et al. 1964, Perry 1963, Vandaveer 1965, Avexitt 1949).

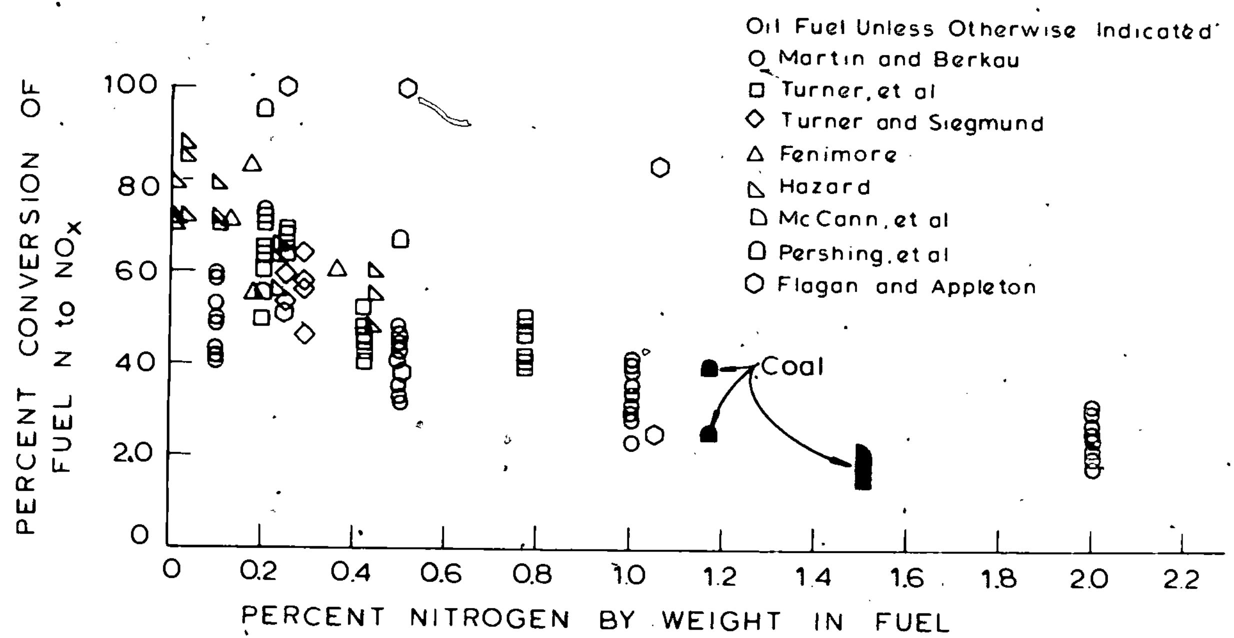

FIGURE 15-2: Percent Conversion of Fuel N to NOx in Laboratory Scale Combustors.

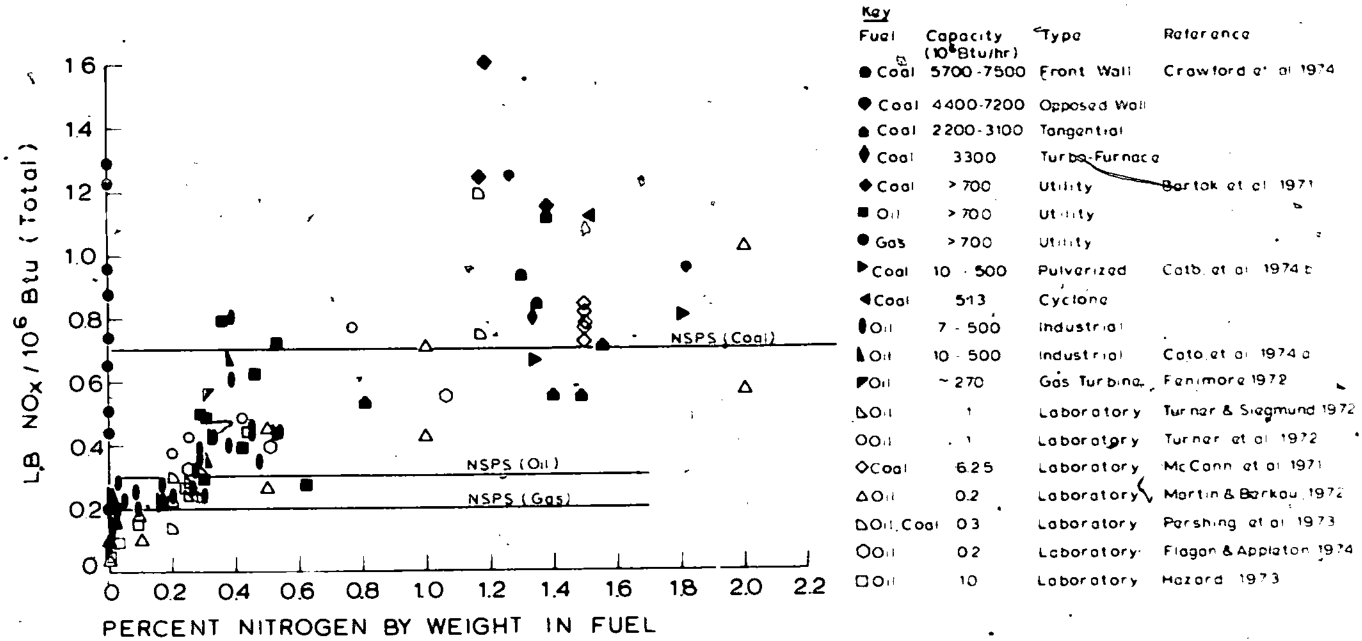

FIGURE 15-3: NOx Emissions from Uncontrolled Combustors as a Function of Fuel Nitrogen Content.

FACTORS INFLUENCING NOx EMISSIONS

The rate of formation of nitric oxide in flames by the two mechanisms discussed above is a sensitive function of oxygen content and, for thermal NOx, temperature. Any factor which influences temperature and oxygen concentration profiles in combustors may therefore have an influence on NOx emission. This makes generalizations for practical systems difficult since fuel/air ratios, fuel/air mixing patterns, fuel type, interaction of different burners, and placement of heat transfer surfaces all influence emissions. Additional complications are introduced by the fact that NO may be reduced by reactions with hydrocarbons in flames and that hydrocarbons can react with atmospheric nitrogen to form nitrogen-containing compounds that are in turn oxidized to nitric oxide. The following sections indicate some of the factors that influence emissions.

Air Preheat

Increased air preheat increases flame temperature and the contribution of thermal NOx. The increases observed in practice for premixed flames are in agreement with predictions from theory (Lange 1972). Reducing air preheat has been shown to decrease emissions on full-scale utility boilers (Blakeslee and Burbach 1972) but it carries with it an associated penalty in thermal efficiency.

On the other hand, in reciprocating engines reducing the inlet air temperature increases efficiency and is thus a very attractive NOx control technique for these systems.

Water or Steam Injection

Flame temperatures can be reduced through the use of steam or water injection. A 90 percent reduction in NOx emissions has been achieved by the use of 15 percent (by weight) water in the fuel air mixture in a gas-fired system (Halstead et al. 1972). The water may be introduced by forming a water-in-oil emulsion which has the additional benefit of providing better fuel atomization and lower soot and particulate emissions (Toussaint and Heap 1974). Water injection, as expected, has little effect in controlling the emission of fuel NOx since fuel NOx formation is insensitive to temperature changes. Although the effectiveness of water injection for reducing NOx emission has been demonstrated on a full-scale utility boiler (Blakeslee and Burbach 1972), the associated loss in thermal efficiency was high. The major application of water injection for NOx control is in engines where it provides substantial NOx reductions without incurring large losses in fuel economy.

Flue Gas Recirculation

The recirculation of flue gases reduces temperature and dilutes the oxygen in the air. It reduces thermal NOx by an amount equal to that of an equivalent thermal load of water injection, and has little influence on fuel NOx. To be effective the flue gas has to be injected into the flame zone. A recirculation of 10 percent (by weight) of the flue gases has achieved a 60 percent lowering of thermal NOx and 20 percent flue gas recycle a 75 percent reduction on a small scale furnace (Halstead et al. 1972). Flue gas recycle is sometimes used in furnaces for temperature control. In such cases, the cost of implementing NOx control is small; otherwise the cost of installation of ducting makes flue gas recycle impractical in retrofitting units. Flue gas recycle, unlike water injection, does not have an adverse effect on the thermal efficiency of a furnace, but does on an engine.

Furnace Load and Size

As the size of a furnace of a given type increases the emissions per unit weight of fuel increase slightly (Woolrich 1961). This is consistent with the postulate that flames radiate less effectively to the walls in larger furnaces and, therefore, are somewhat hotter. The level of emissions per unit weight of fuel varies much more dramatically with furnace load, being a little more than proportional to load for gas-fired field units and a little less than proportional to load for coal and oil-fired units (Bartok et al. 1971). This difference between fuels can be explained by the influence of fuel NOx which is expected to be relatively load insensitive. Reduction of NOx emissions by use of reduced loads or the oversizing of combustion chambers carries with it a capital cost associated with underutilized equipment.

Excess Air

As the amount of excess air is increased in a combustor, the oxygen content in the flame zone generally increases and the temperature decreases. As a consequence of these two opposing effects, the emissions of thermal NOx pass through a maximum(Armento 1974) (Wasser et al. 1968). Most, but not all, furnaces operate in a range where NOx emissions decrease with a reduction in excess air. Fuel NOx ,being temperature insensitive, increases monotonically with increased excess air (Fenimore 1972, Martin and Berkau, Turner et al. 1972). Low excess air firing can reduce NOx emissions, but care must be taken not to replace those emissions with CO and soot emissions. When operating a multi-burner system near stoichiometric, care must be taken to regulate the fuel/air ratio of each burner so as to prevent some burners from running fuel-rich. Low excess air firing of furnaces, although necessitating more control equipment and instrumentation to monitor emissions, provides fuel savings through increased thermal efficiency. In contrast, fuel lean combustion with high excess air can reduce the NOx emissions from engines, but improved carburation, fuel injection, or stratification may be necessary for efficient operation.

Staged Combustion

Staged combustion takes advantage of both low temperatures and low oxygen concentration by operating some burners fuel rich, allowing the partially combusted products to cool between stages, and completing the combustion by addition of the residual air. Several variations of staged combustion can be used. In a multi-burner system the lower burners may be run fuel rich and the upper burners lean; this is known as biased or off-stoichiometric combustion. Alternatively, all of the fuel can be injected through the lower burners and the air introduced through the upper burners or higher in the furnace through 'NO' ports; this is referred to as overfire-air (Winship and Brodeur 1973) or two-stage combustion (Barnhart and Diehl 1959, Barnhart and Diehl 1960). Staged combustion reduces the emission of both thermal and fuel NOx. By operating the first stage with as little as 70 percent of stoichiometric air, the emission from a 0.25 percent nitrogen oil could be reduced to under 100 ppm (Siegmund and Turner 1974). As much as a 90 percent reduction in the fuel nitrogen has been observed by use of fuel rich operation in laboratory burners (Fenimore 1972).

The potential for NOx control in practical systems depends on many factors. In retrofitting units the number of burners, the ability to change fuel rates to individual burners, and the availability of 'NO' ports for air addition above the burners are physical constraints. In addition, in changing operation or design conditions to reduce NOx, care must be taken that the other pollutants are not substituted for the NOx. Low excess air operation increases the potential for emissions of unburned hydrocarbons, carbon monoxide and soot. Staged combustion may result in the formation of hydrogen cyanide or ammonia in the first stage, which will be partially oxidized to NO in the second stage (Yamayishi et al. 1974). Changes of fuel/air ratio on burners may create problems of fuel stability and noise (Lockling et al. 1974). These complicating factors and the very wide variety of combustion systems in use make it difficult to state any generalization on the potential reduction in emissions that may be achieved by combustion process modification. Staged combustion, however, provides the most successful method currently in use for the control of NOx emission from combustion sources.

Burner Design

Variation in the momentum of the fuel and air streams, in the swirl or rotation of the air stream, in the shape of a burner, in the positioning of the fuel nozzle in a burner, in the atomization of liquid fuels, in the amount of air that is premixed with the fuel, or in the positioning of the flame stabilizer, if any, can make large differences in mixing and combustion patterns and hence in NOx emissions from different burners. The potential for major reductions in NOx emission by modification of burner design has been established on experimental burners (Heap et al. 1973). Although a complete characterization of the processes is not possible, some general rules seem to apply.

High intensity combustion favors the formation of thermal NOx. Evidence for this is provided by an inverse correlation of NOx emission with residence time in the combustion zone in a laboratory burner as the swirl was varied (Wasser and Berkau 1972). Other, less quantitative, evidence is provided by the observation that burners which entrain air gradually and produce relatively long flames, such as in a tangential boiler, produce low thermal NOx whereas high intensity burners for example cyclones, yield high emissions. Low emissions of thermal NOx also result when a burner produces entrainment of relatively cold combustion products into the flame, a form of internal flue gas recirculation (Calvert 1973, Hemsath et al. 1972).

High emissions of fuel NOx are found when air is premixed into a burner or when a flame is lifted so that air is entrained before ignition (Heap et al. 1973). For oil and coals containing fuel nitrogen it is preferable that the fuel nitrogen be released into an oxygen deficient ambient atmosphere (Heap et al. 1973). Long diffusion flames therefore favor low fuel NOx in addition to low thermal NOx.

Through the use of multiple concentric fuel and air ports, apparently providing delayed mixing or a form of staging, emissions as low as 150 ppm have been reported for an experimental coal-fired burner (Heap et al. 1973).

These results suggest that with developmental effort, significant reduction in NOx emissions may be derived from changes in burner design.

Fuel Atomization

Under normal operating conditions for practical systems, the technique of fuel atomization and atomized pressure has little effect on either thermal NOx or fuel NOx. Normal atomization procedures produce fuel droplets which are sufficiently large and which have a velocity relative to the air flow sufficiently low that the droplets burn with an attached flame. This diffusion type flame results in near stoichiometric combustion of much of the fuel vapor and results in both thermal and fuel NOx levels which are relatively insensitive to the overall fuel/air ratio of the combustor (Flagan and Appleton 1974, Pompei and Heywood 1972). However, by using more efficient atomizers which produce much smaller droplets with a high velocity relative to the air flow, distillate fuels may evaporate and partially premix with air prior to burning. Thus, in a burner operating fuel lean, thermal NOx may be significantly reduced by increasing the atomizer efficiency (Pompei and Heywood

1972). Fuel NOx, on the other hand, tends to increase with increasing atomizer efficiency (Flagan and Appleton 1974).

Burner Interaction

Interaction between burners either on the same wall (Lowes et al. 1974) or on opposed walls (Bartok et al. 1971) leads to higher emissions, probably as a consequence of the reduced heat transfer from the central flames to the walls.

Reduction of NO by Reaction with Hydrocarbons

Laboratory experiments in which hydrocarbons and ammonia where injected into the post-flame zone have shown significant reduction in NOx levels (Wendt et al. 1973), and may suggest new methods for NOx control. Related experiments demonstrate that NO was destroyed with 30 to 95 percent efficiency by injection into the combustion air of a burner (Turner and Siegmund 1972), into a fluidized bed coal combustor (Hammons and Skopp 1971), and into a diffusion flame (Sarofim et al. 1973). In practical turbulent combustors, some NO may be destroyed by these processes as the turbulent eddies bring fuel rich pockets together with NO.

The factors that influence NO formation are many and are imperfectly understood. The results reported here suggest some directions, but what is practically achievable must be determined from field studies for the various classes of stationary sources, as described in subsequent sections. The large variation in peak temperatures, pressures, sizes and residence times in different combustion systems explains the wide range of emissions that will be reported, with residential heating units having the lowest emissions per unit fuel consumption and reciprocating engines the highest. These same differences necessitate different control methods for the different classes of units and may limit the degree of control which can be achieved by combustion modification alone.

UTILITY BOILERS

Background

Utility boilers are a major source of NOx emissions. The NOx from utility boilers is discharged from a relatively small number of tall stacks. The boilers are fired by coal (54.3 percent of the energy supplied in 1972) (Mason and Shimzu 1974), gas (27 percent) and oil (18.6 percent), sometimes in combination. Environmental constraints on sulfur emissions had resulted in a substitution of low-sulfur oils and gas for coals, with a short term decrease in percentage use of coal, but the trend has been reversed and projections (NERC 1974) indicate a continued reliance on coal, a decrease in the rate of installation of oil-fired units, and a gradual phasing out of gas. Emissions are contributed by over 3000 boilers in use for steam-electricity generation, varying widely in size, design, and age. The most common boiler types, designated by the location of the burners in the combustion chamber, are tangential (T), horizontally opposed (HO), front wall (FW), cyclone (Cyc), vertical (V), turbo-fired (turbo) and all wall (AW) (Bartok et al. 1969).

Control Methods for Gas-Fired Units

The emissions from gas fired units are due entirely to thermal NOx and fall typically in the range of 0.30 to 1.31 lbs/10^6 BTU for uncontrolled units at full-load (Table 15-2 taken from an EPA-sponsored systematic field study) (Bartok et al. 1971). The lowest emissions are expected for units with delayed mixing of fuel and air and in which the flames can radiate effectively to the walls; the highest emissions

TABLE 15-2

Uncontrolled Emissions from Gas, Oil, and Coal-fired Utility Boilers Operated at Full, Intermediate, and Low Load (Bartok et al. 1971)

Fuel	Size (MW)	Type of firing	Full Load (MW)	PPM @ 3%	lbs NO_2 / 10^6 Btu
Gas	180	FW	180	390	0.51
Gas	80	FW	82	497	0.65
Gas	315	FW	315	992	1.29
Gas	350	HO	350	946	1.23
Gas	480	HO	480	736	0.96
Gas	600	HO	559	570	0.74
Gas	220	AW	220	675	0.88
Gas	320	T	320	340	0.44
Gas	66	V	66	155	0.20
Oil	180	FW	180	367	0.50
Oil	80	FW	80	580	0.79
Oil	250	FW	250	360	0.49
Oil	350	HO	350	457	0.62
Oil	450	HO	455	246	0.33
Oil	220	AW	220	291	0.39
Oil	320	T	320	215	0.29
Oil	66	T	66	203	0.27
Oil	400	CY	415	530	0.72
Coal	175	FW	-	-	-
Coal	315	FW	275	1490	2.04
Coal	600	HO	563	838	1.15
Coal	800	HO	778	905	1.24
Coal	575	T	-	-	-
Coal	300	T	300	568	0.78
Coal	700	CY	665	1170	1.60

TABLE 15-2 (cont.)

Intermediate Load (MW)	PPM @ 3% O_2	lbs NO_2 / 10^6 Btu	Low Load (MW)	PPM @ 3% O_2	lbs NO_2 / 10^6 Btu	% N in Fuel
120	230	0.30	70	116	0.15	-
50	240	0.31	20	90	0.12	-
223	768	1.00	186	515	0.67	-
-	-	-	150	341	0.44	-
360	610	0.79	250	363	0.47	-
410	335	0.44	325	253	0.33	-
190	550	0.72	125	313	0.41	-
240	230	0.30	-	-	-	-
-	-	-	-	-	-	-
120	322	0.44	80	266	0.36	0.29
50	361	0.49	21	258	0.35	0.36
172	306	0.41	-	-	-	0.31
-	-	-	150	264	0.36	0.46
365	219	0.30	228	186	0.25	-
170	267	0.36	120	324	0.44	0.42
220	220	0.30	-	-	-	0.30
-	-	-	-	-	-	0.62
258	205	0.28	-	-	-	0.53
140	660	0.90	-	-	-	1.36
190	1280	1.75	160	1200	1.64	1.36
462	781	1.07	363	643	0.88	1.38
580	741	1.01	-	-	-	1.17
470	405	0.55	310	264	0.36	-
240	418	0.57	-	-	-	-
545	882	1.21	-	-	-	1.19

are expected for large units with high combustion intensities. Generally, tangentially-fired boilers yield the lowest emissions. Reduction in load has a major influence on emissions (Table 15-2); to a first approximation there is a proportionality between emissions and furnace load.

The control techniques that have been successfully demonstrated on field units are low-excess-air firing, staged combustion, flue-gas recirculation, water injection, and reduced air preheat. The concept of staged combustion was pioneered on gas units in the late 1950's and 1960's by Babcock and Wilcox and Southern California Edison (Bagwell et al. 1970, Barnhart and Diehl 1959, Barnhart and Diehl 1960, Teixera and Breen 1973). A 33 to 75 percent reduction in the emissions level has been attained on 12 units of 175 ~~MW~~ to 480 MW by the use of staged combustion (two-stage and/or off-stoichiometric combustion). The off-stoichiometric combustion, in which the fuel from up to 25 percent of the burners was diverted to the remaining burners, yielded better results than the two-stage combustion, in which part of the air to all of the burners was diverted to 'NO' ports higher in the furnace.

Southern California Edison also tested the potential for control of NOx by flue-gas recirculation on tangentially-fired boilers designed with flue-gas recirculation to the windbox (the compartment confining the air to the burners). Recirculation of flue gases at a rate of 15 percent by weight of the sum of the fuel and air reduced the emissions by 30 to 60 percent; recirculation of 30 percent yielded emission levels below 100 ppm (0.131 lb $NO_2/10^6$ BTU) (Teixera and Breen 1973).

The EPA funded systematic field study of NOx emission control methods determined the control achievable on representative front wall, horizontally opposed, and tangentially fired boilers at three load levels, by use of various combinations of low-excess-air firing, staging, and flue gas recirculation (see Table 15-3).

TABLE 15-3

Percentage Reduction in NOx Emissions Achieved by Staging, Low Excess Air Firing, and Flue Gas Recirculation (Bartok et al. 1971)

Combustion Operating Modification and Furnace Load(1)

Fuel Fired	Type(2) of Firing	% Reduction in NO_x Emission: Low Exc. Air Full	Low Exc. Air Int.	Low Exc. Air Low	Staging Full	Staging Int.	Staging Low	LEA + Staging Full	LEA + Staging Int.	LEA + Staging Low	Flue Gas Rec. Full	Flue Gas Rec. Int.	Flue Gas Rec. Low	"Full"(3) Full	"Full"(3) Int.	"Full"(3) Low
GAS	FW	13	24	7	37	30	30	48	42	36	--	--		48	42	36
	HO	17	15	32	54	35	59	61	48	68	--	--	20	73	52	72
	T	--	--	--	--	--	--	--	--	--	--	60	--	66	65	--
	ALL (Average)	16	19	26	45	31	52	54	44	52	--	60	20	64	51	60
OIL	FW	27	20	28	29	20	20	39	32	21	46	31	--	50	41	21
	HO	10	16	12	34	34	47	35	44	42	--	--	--	38	35	55
	T	28	22	--	--	17	--	--	45	--	10	13	--	--	59	--
	ALL (Average)	19	19	18	30	22	34	38	37	32	28	23	--	47	42	38
COAL	FW	--	14	--	--	40	--	--	55	--	--	--	--	--	60	--
	HO	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
	T	27	18	--	--	39	--	--	50	42	--	--	--	--	50	42
	ALL (Average)	27	17	--	--	39	--	52	52	42	--	--	--	--	55	42

(1) Furnace load: "Full" = 85%-105%, "Intermediate" = 60%-85%, Low = 50%-60% of rating.
(2) Type of firing: FW = Front Wall, HO = Horizontally Opposed, T = Tangential.
(3) "Full control": combination of techniques achievable on boilers tested.

Use of low excess air achieved modest reductions in emissions (13 to 32 percent). The minimum value of the excess air that could be used was determined by the problem of CO emissions which increased dramatically below a critical excess air level. The minimum practical level of excess air is a function of burner and furnace design; low-excess-air firing therefore requires careful control of the fuel and air rate to individual burners and careful monitoring of products of incomplete combustion in the stack. These disadvantages, however, are partially offset by the increase in the thermal efficiency of a boiler which results from a reduction in the excess air level. Staging and flue gas recirculation produced improvements in the systematic field study similar to those observed in Southern California. A combination of control techniques, as expected, yields a smaller effect than that computed from the gains observed when each technique is applied individually. The final three columns in Table 15-3 show that a 50 to 60 percent reduction in emissions was achievable when combinations of all the techniques available on each of the boilers in the field study were utilized to their fullest.

Injection of water, use of water-in-oil emulsions, and reduction of air preheat are all less desirable methods of NOx emissions control because they impose a penalty in thermal performance. Water injection tests carried out by Combustion Engineering on a 150 MW tangentially fired unit showed a maximum reduction of 50 percent in NOx emission at a water injection rate of 45 pounds per million BTU fired and an associated 5 percent decrease in furnace efficiency (Blakeslee and Burbach 1972). Significant reductions in NOx emission and furnace efficiency were also observed when the air preheat was reduced from 490F to 81F at three-quarter and half loads (Blakeslee and Burbach 1972).

In summary, the techniques most readily adapted to reducing emission from existing gas-fired units are low excess air firing and staged combustion, particularly in units where the number of burners permits the changes to be made without any equipment modification. Flue gas recirculation provides a viable option only for those existing units already equipped with a recirculation system. Water injection or reduction in air preheat are less desirable control techniques in view of the penalty of increased fuel consumption entailed in their use. For gas units, a reduction in emissions of 50 to 60 percent can be expected with the use of low excess air and staged combustion alone, and greater reductions are possible when these can be combined with flue-gas recirculation. It should be noted, however, that units differ significantly in design and individual units may show much lower potential for emission control.

Control Methods for Oil-Fired Units

Emissions from oil-fired units are due to a combination of thermal and fuel NOx. Control methods which are based on a temperature reduction, such as flue gas recirculation, water injection, and load reduction, are effective in controlling the thermal NOx contribution but have little effect on the fuel NOx. Methods which reduce the oxygen availability in the primary conbustion zone, such as low excess air firing, staged combustion, and long flames with delayed oxygen-uptake are effective in controlling both sources of NOx. The emissions from nine oil fired utility boilers included in the systematic field survey (Bartok et al. 1971) range from 0.27 to 0.79 lbs/10^6 BTU at full load (Table 15-2). Tangential-fired units were found to yield the lowest NOx emissions for a given size unit, as had been previously found for gas fired boilers. It was estimated from results obtained using oils with different nitrogen contents that 30 percent of the fuel nitrogen in the nitrogen concentration range of 0.3 to 0.6 percent by weight was converted to NOx, corresponding approximately to an incremental emission of 0.06 lb/10^6 BTU per 0.1 percent increment in fuel nitrogen. Independent tests on tangentially-fired boilers indicate a range of conversion of fuel nitrogen to NOx from 43 percent for 0.2 percent nitrogen oils to 30 percent for 1 percent nitrogen oils when the boilers where operated with 3 percent O_2.

Reduction in load for oil-fired boilers resulted in a reduction in emissions but the dependence of emissions on load is less marked than for the case of gas-fired boilers (Table 15-2).

The control methods tested in the field study included low-excess-air firing, staging, flue gas recirculation and combinations thereof (Table 15-3). The trends are similar to those obtained for gas-fired units but the fractional reduction in NOx emissions is lower. The results show an average reduction in NOx for the best combination of control techniques ranging from 48 percent at full load to 38 percent at low load. Although the uncontrolled emissions of gas fired units were higher than the emissions from oil units of the same design and size, the emissions with the best control achieveable were generally lower for the gas-fired units. Part of the difference may be due to the fuel-nitrogen contribution and part due to the difficulty of controlling the very complicated processes that define the atomization and combustion of liquid fuels.

Control Methods for Coal-Fired Utility Boilers

Uncontrolled emissions from coal-fired boilers are higher than those for gas and oil (Table 15-2 and 15-4), ranging from 0.53 to 2.04 lbs/10^6 BTU at full load. Tangential-fired units again show the lowest emissions. Emissions from coal-fired units show a less than proportional dependence on load (Table 15-2). From the dependence of the emissions on load it is inferred that about 20 percent of the fuel nitrogen content, typically 1.3 percent by weight, is converted to NOx and that the fuel nitrogen contributes 50 percent of the emission at full-load (Crawford et al. 1974). The relative proportions of fuel NOx and thermal NOx depend

TABLE 15-4

Uncontrolled Emissions from Coal-Fired Utility Boilers and Percent Reduction in Emission Achievable by Staged Combustion (Crawford et al. 1974).

Fuel	Size (MW)	Type of Firing	Load (MW)	PPM @ 3% O_2	$\frac{\text{lb } NO_2}{10^6 \text{ Btu}}$	%N in Fuel	% Reduction at Full Load
Coal	105	FW	101	454	0.60	——	53
Coal	125	FW	125	634	0.84	1.35	40
Coal	256	FW	253	703	0.93	1.31	48
Coal	320	FW	350	832	1.11	1.38	34
Coal	218	Ho	219	569	0.76	-----	34
Coal	480	Ho	490	711	0.95	1.81	35
Coal	800	Ho	800	935	1.24	1.27	48
Coal	250	T	250	410	0.55	1.49	25
Coal	330	T	334	531	0.71	1.56	59
Coal	350	T	350	415	0.55	1.4	35
Coal	348	T	306	434	0.53	0.80	4
Coal	850	Turbo	370	600	0.80	1.34	34

upon many variables including coal moisture content, nitrogen content, burner design, furnace design and operating conditions.

More conclusive evidence on the contribution of fuel nitrogen in coal is provided by laboratory studies in which pulverized coal was burned in argon-oxygen mixtures (Pershing et al. 1973). In these tests 25 to 40 percent of the 1.2 percent nitrogen content of the coal was converted to NOx.

Based on the importance of both thermal NOx and fuel NOx it is expected that techniques that reduce temperature in the flame zone will be less effective in controlling NOx emission than those that reduce the oxygen content. Low-excess-air firing and staged-combustion have proven effective in reducing NOx emissions from coal-fired units by an average of 37 percent for the 12 units included in the EPA funded field tests (Crawford et al. 1974). The carbon content of the ash and the carbon monixide in the stack gas increased slightly during the tests (Crawford et al. 1974). Accelerated corrosion studies (Crawford et al. 1974) for the 300 hour duration of the trials with staged combustion showed no adverse effects but additional long range trials will be needed to confirm these preliminary findings.

Design Modifications and Costs

The success of low-excess-air and staged combustion has enabled all manufacturers to develop boilers that will meet the national NOx emission standards of 0.2 lbs/10^6 BTU for gas, 0.3 lbs/10^6 BTU for oil, and 0.7 lb/10^6 BTU for coal. Some manufacturers specify a maximum nitrogen content on the oil to be fired in new oil fired units. Further reduction in emissions can be achieved at considerably increased costs by use of flue gas recycle. Water injection may be used but with a penalty in thermal efficiency.

Severe constraints may be imposed on the ability to make modifications on existing units by the design of the particular unit and by the space available for adding ducting. The modifications easiest to apply are low-excess-air firing and off-stoichiometric combustion. Addition of NO ports may be feasible in many units but the addition of flue-gas recycle would require a major effort.

Costs for modifying new and existing coal-fired tangentially-fired units have been estimated by Combustion Engineering (Blakeslee and Seler 1973) and are presented in Figures 15-4 and 15-5. (Additional data supplied by a utility and by a boiler manufacturer are consistent with the figures presented here). The costs were calculated for the introduction of 20 percent of the total combustion air over the fuel firing zone as overfire air, for the recirculation of 30 percent of the flue gas to the secondary air ducts and windbox, for a combination of overfire air and flue-gas recirculation, for the recirculation of 17 percent of the flue-gas through the coal pulverizers (mills), and for water injection into the fuel-firing zone at 5 percent of the steam rate for the boiler. The practical limit for single cell furnaces is about 600 MW; larger units use divided furnaces with two times as many burners which results in an increased cost for installation of ductwork for the various control methods (Blakeslee and Seler 1973). Cost figures are expected to vary considerably between units particularly for the case of retrofit. It should be noted that the costs in Figures 15-4 and 15-5 are only capital costs; in addition to capital costs, operating (Bartok et al. 1969, NAE 1972) and testing costs will be incurred which will be totally dependent on the specific situation.

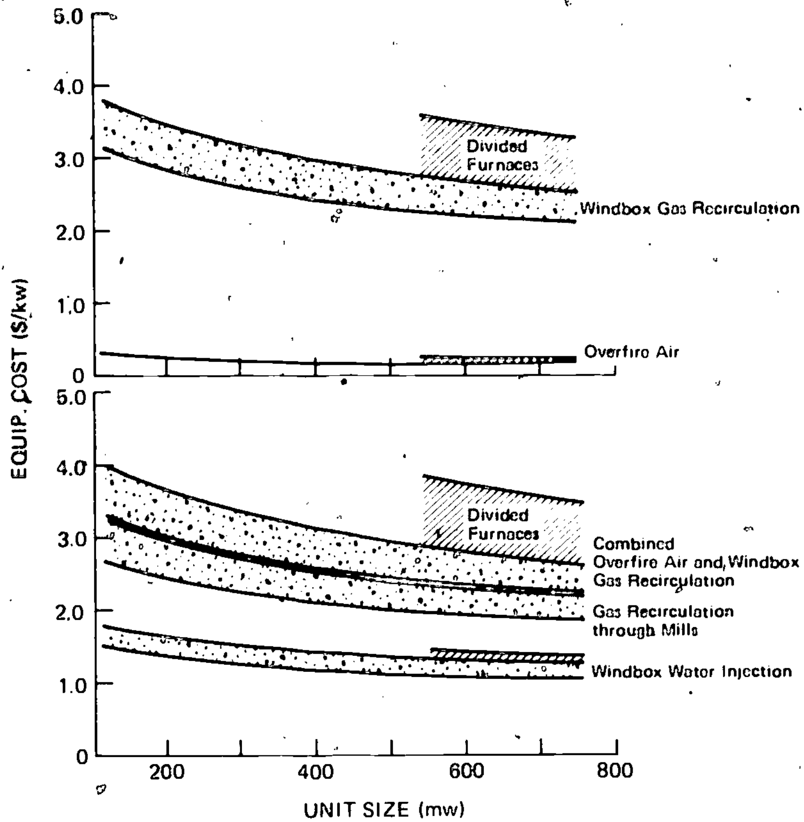

FIGURE 15-4: Costs of NOx Control Methods for New Coal-fired Units (included in Initial Design). (Blakesler and Selker 1973b).

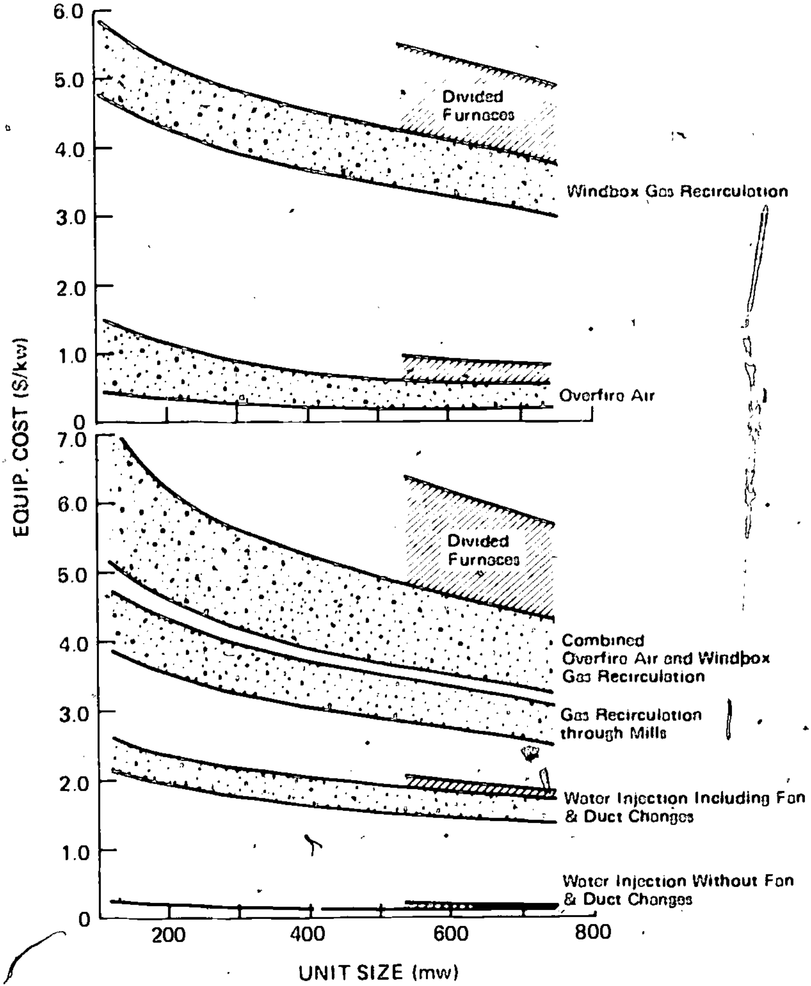

FIGURE 15-5: Costs of NOx Control Methods for Existing Coal-fired Units (Heating Surface Changes Not Included). (Blakesler and Selker 1973b).

INDUSTRIAL BOILERS

Background

Boilers with capacities from 10 to 500 million BTU per hour emit an estimated 18.1 percent of the U.S. stationary source nitrogen oxides. The industrial boiler population is made up of about 75000 units of various types, ages, and applications which burn gas (57 percent of the energy supplied in 1968), coal, (23 percent) and oil (20 percent) (Lockling et al. 1974). Residual oils and a small quantity of distillate oils are used. As a result of regulations on the emissions of sulfur oxides, recent trends have been toward the use of natural gas and low sulfur oils in all but the largest units. However, due to increased prices and reduced availability of these fuels, the industrial sector may be forced to rely increasingly upon coal and residual oil.

About a third of the industrial boilers, accounting for 10 percent of the industrial capacity, are small units (10 to 16 million BTU per hour capacity) (Lockling et al. 1974). These are primarily packaged firetube boilers which burn oil or gas. The 17 to 100 million BTU per hour units, which are primarily packaged watertube boilers, account for 53 percent of the units and 45 percent of the capacity. The larger units are watertube boilers, mostly field erected. Currently only 1 percent of the units and 9 percent of the capacity are greater than 250 million BTU per hour capacity, the size range for which new source performance standards have been established.

The data on emission levels and combustion modification effectiveness for NOx control are limited, mostly derived from a field study performed by KVB, Engineering ((Cato et al.) in which about 80 units were tested for baseline emission levels and minimum NOx achievable without hardware modifications. Due to the limited flexibility in combustion operating conditions for these units, only moderate reductions were obtained in most cases.. The results are presented in Table 15-5.

When combustion modification strategies are considered, it will be necessary to consider several problems which may arise such as corrosion and deposits on boiler tubes, flame instability, blow-off, flashback, combustion driven oscillations, and combustion noise or roar. The need for tuning individual units to minimize these problems will place constraints on retrofit programs due to the large number of units in existence and their diverse characteristics (Lockling et al. 1974).

Gas-Fired Boiler Emissions

Gas-fired units without air preheat, firetube boilers, and some of the watertube boilers had NOx emission levels less than 0.14 lb NO_2 /10^6 BTU. (Cato et al.) The watertube units with air preheat had generally higher emissions, from 0.08 to 0.45 lb $NO_2/10^6$ BTU. By comparing units with different degrees of air preheat, it was estimated that the preheat effects vary from 0.02 lb $NO_2/10^6$ BTU per 100F for units of less than 30 million BTU per hour capacity to 0.15 lb $NO_2/10^6$ BTU per 100F for much larger units. (Cato et al.) This appeared to be a function of the burner firing rate (BTU/hr/burner). Reducing the oxygen level had only a small effect on emissions from units without air preheat, but did decrease the NOx levels from preheated boilers. Off-stoichiometric firing, achieved by taking one burner out of service, resulted in NOx reductions of 16 to 42 percent. By combustion modification, the percentage of units with emissions below 0.2 lb $NO_2/10^6$ BTU was increased from 75 percent to 82 percent, (Cato et al.)

Oil-Fired Burner Emissions

The baseline emission levels for those units fired with No. 2 fuel oil were 0.13 to 0.26 lb

TABLE 15-5

Uncontrolled NOx and Low NOx Emissions from Industrial Boilers (Cato et al.)

Fuel	Size MBH	Burners Type	Burners Number	Test Load MBH	Baseline PPM @ 3% O_2	Baseline lbs NO_2/10^6 Btu	Low NO_x PPM @ 3% O_2	Low NO_x lbs NO_2/10^6 Btu	% reduction
Gas	25	Ring	1	20	72	.086	65	.077	10
Gas	29	Ring	1	22	70	.083	-	-	-
Gas	29	Ring	1	11	97	.115	68	081	29.6
Gas	59	Ring	6	48	116	.138	-	-	-
Gas	60	Ring	2	48	101	.124	86	102	17.?
Gas	60	Ring	4	46	242	.288	138	.164	43.1
Gas	169	Ring	1	136	374	.445	355	.423	4.9
Gas	158	Ring	4	125	299	.343	110	131	61.8
Gas	300	Ring	4	259	199	.237	169	.201	15.2
Gas	10	Ring	1	8	55	065	38	045	30.8
Gas	20	Ring	1	14	107	.127	75	089	29.9
Gas	11	Ring	1	10	87	104	83	099	4.8
Gas	?	Ring	1	6	71	.085	67	.080	5.8
Gas	10	Ring	1	?	91	108	90	107	9
Gas	13	Ring	1	12	57	068	-	067	1.5
Gas	18	Ring	1	17	56	067	53	063	6.0
Gas	20	Ring	1	13	79	.094	67	080	14.9
Gas	8	Ring	1	6	70	083	45	.054	35.0
Gas	20	Ring	1	10	101	.120	-	-	-
Gas	33	Ring	1	24	90	107	81	096	10.3
Gas	65	Ring	6	53	98	.117	-	-	-
Gas	115	Ring	1	85	94	.112	-	-	
Gas	225	[illegible]	8	190	212	.252	147	175	30.6
Gas	325	[illegible]	8	260	320	.381	230	.274	28.1

TABLE 15-5 (cont.)

Fuel	Size MBH	Burners Type	Burners Number	Test load MBH	Baseline PPM @ 3% O_2	Baseline lb NO_2 / 10^6 Btu	Low NO_x PPM @ 3% O_2	Low NO_x lb NO_2 / 10^6 Btu	% reduction
#2 Oil	110	Steam	2	88	177	.231	-	-	-
#2 Oil	10	Air	1	7	169	.220	149	.194	11.8
#2 Oil	18	Steam	1	14	65	.085	63	.082	3.5
#2 Oil	18	Air	1	14	97	.127	86	.112	11.8
#2 Oil	18	Mech	1	12	80	.104	80	.104	-
#2 Oil	11	Air	1	11	128	.167	-	-	-
#2 Oil	18	Air	1	16	116	.151	-	-	-
#2 Oil	18	Steam	1	16	118	.154	-	-	-
#2 Oil	20	Air	1	16	193	.252	141	.184	27.0
#2 Oil	29	Steam	1	10	103	.134	-	-	-
#2 Oil	7	Air	1	7	127	.166	-	-	-
#2 Oil	158	Steam	4	115	181	.236	120	.157	33.5
#2 Oil	33	Steam	1	23	123	.160	104	.136	15.0
#2 Oil	13	Air	1	11	84	.110	-	-	-
#5 Oil	19	Cup	1	12	200	.261	139	.181	30.[illegible]
#5 Oil	85	Steam	4	59	329	.429	243	.317	26.1
#5 Oil	11	Air	1	12	183	.239	162	.211	11.7
#5 Oil	18	Air	1	18	177	.231	154	.201	13.0
#5 Oil	18	Steam	1	17	161	.210	152	.198	5.7
#5 Oil	13	Air	1	11	181	.236	171	.223	5.5
#5 Oil	7	Air	1	7	275	.359	-	-	-
#5 Oil	59	Steam	6	46	619	.807	516	.673	16.6
#5 Oil	65	Steam	6	50	466	.608	431	.562	7.6
#5 Oil	[illegible]	Steam	1	100	337	.440	322	.420	4.5
#5 Oil + LG	110	Steam	4	88	172	.224	166	.217	3.1
#5 Oil + RG	110	Steam	4	90	215	.280	211	.275	1.8

TABLE 15-5 (cont.)

Fuel	Size MBH	Burners		Test Load MBH	Baseline		Low NO_x		% reduction
		Type	Number		PPM @ 3% O_2	$\frac{\text{lb } NO_2}{10^6 \text{ Btu}}$	PPM @ 3% O_2	$\frac{\text{lb } NO_2}{10^6 \text{ Btu}}$	
NSF oil	17	Cup	1	15	184	.240	-	-	-
#6 oil	18	Steam	1	14	350	.457	331	.432	5.5
#6 oil	18	Air	1	15	334	.436	277	.361	17.2
#6 oil	80	Steam	1	51	305	.398	268	.350	12.1
#6 oil	90	Steam	3	71	246	.321	175	.228	29.0
#6 oil	65	Steam	2	54	186	.243	174	.227	6.6
#6 oil	105	Steam	4	80	251	.327	222	.290	11.3
#6 oil	260	Steam	4	130	240	.313	201	.262	16.3
#6 oil	500	Steam	9	400	267	.348	180	.235	32.5
#6 oil	7	Air	1	7	298	.389	249	.325	16.5
O/C	513	Cyc	2	320	716	.934	-	-	-
O/C	513	Cyc	2	320	710	.926	-	-	-
Coal	60	UFS	7	48	266	.327	188	.263	29.3
Coal	60	UFS	7	46	224	.314	198	.277	11.8
Coal	135	Sprd	2	110	370	.518	335	.469	9.5
Coal	50	Sprd	1	40	465	.651	330	.462	29.0
Coal	75	Sprd	1	63	465	.651	387	.542	16.7
Coal	225	Pulv.	8	181	378	.529	360	.504	4.7
Coal	210	Sprd	5	120	553	.774	471	.659	14.9
Coal	230	Sprd	6	162	547	.766	360	.504	34.2
Coal	500	Pulv	6	400	580	.812	-	-	-
Coal	513	Cyc	2	320	800	1.120	742	1.039	7.2
Coal	10	UFS	1	8	273	.382	-	-	-
Coal	10	UFS	1	8	346	.484	-	-	-
Coal	325	Pulv	8	260	484	.678	-	-	-

TABLE 15-5 (concluded)

Abbreviations

NSF	=	Navy Standard Fuel
Cup	=	Rotary cup fuel atomizer
Air	=	Air assist fuel atomizer
Steam	=	Steam-fuel atomizer
Cyc	=	Cyclone furnace coal combustor
UFS	=	Underfed stoker coal burning equipment
Sprd	=	Spreader stoker coal burning equipment
Pulv	=	Pulverized coal burning equipment
MBH	=	Million BTU per hour

$NO_2/10^6$ BTU (Cato et al.). Increasing combustion intensity resulted in increased NOx, but other changes in operating parameters had little effect.

The heavy fuel oil emissions were higher, 0.20 to 0.81 lb $NO_2/10^6$ BTU, and more sensitive to the operating conditions. The nitrogen content of the fuel had a particularly strong effect since an estimated 44 percent of the fuel nitrogen was converted to NOx. NOx emissions decreased with decreasing oxygen levels, burner heat release rate (BTU/hr/burner), and combustion intensity. Emission reductions of 6 to 29 percent were obtained by using stoichiometric or staged combustion. The technique of fuel atomization did not strongly influence the emissions as long as good atomization was achieved. The two highest emission levels for No. 5 oil were the result of preheating the oil to only 130F, rather than 160 to 180F as in most other tests. Increasing the oil temperature improved the atomizer effectiveness and reduced the NOx emission levels.

Coal-Fired Boiler Emissions

Due largely to the high nitrogen content of coals (1.29 to 1.80 percent by weight in the KVB study), the emissions from coal-fired units were generally higher than those from oil or gas fired units, i.e., 0.31 to 1.12 lb $NO_2/10^6$ BTU. (Cato et al.) The lowest emissions from coal-fired units were from underfed stoker coal burning equipment. In these units the air fed up through the grating is insufficient for complete combustion, so additional air must be introduced above the grating through overfire air ports. The combustion is, therefore, effectively staged, and the NOx emissions were quite low, .31 to .48 lb $NO_2/10^6$ BTU.

Spreader stokers, in which the fuel is introduced with the air flow above the grate, had intermediate emission characteristics, 0.52 to 0.77 lb $NO_2/10^6$ BTU. Some of the fuel is burned in the fuel spray; the remaining fuel is burned on the grate as in the underfed stoker. The resultant combustion is only partially staged. The combustion intensities are also higher than for underfed stokers, possibly increasing thermal NO formation.

Pulverized coal units, in which all of the fuel is burned in suspension, had higher emissions, 0.53 to 0.81 lb $NO_2/10^6$ BTU. A unit equipped with two cyclone-type coal combustors produced 1.12 lb $NO_2/10^6$ BTU, the highest emissions of all the units tested due to the very high combustion intensity. (Cato et al.)

Reductions of about 0.07 lb $NO_2/10^6$ BTU percent reduction in oxygen were obtained by reducing the amount of excess air. A decrease in underfire air-rate with a compensating introduction of air through openings higher in the furnace for a spreader stoker-fired boiler resulted in about a 46 percent NOx reduction, but the grate temperature increased beyond its allowable limits. The most acceptable operating conditions reduced NOx by 20 to 25 percent. Controlled NOx emissions were below 0.68 lb $NO_2/10^6$ BTU for all but the cyclone fired boiler. (Cato et al.)

RESIDENTIAL AND COMMERCIAL SPACE HEATING

Background

Space heating equipment of less than 10 million BTU hr capacity emits about 7.1 percent of the U.S. stationary source NOx. The emissions are largely near ground level in areas of high population densities. (Hall et al. 1974). The problem of NOx emission from these sources may, in fact, be more severe than this number would indicate since the emissions are confined to the heating season.

Commercial units, which generally fall in the range of 0.3 to 10 million BTU hr capacity, include a mix of packaged firetube, cast iron, and watertube boilers. (Barrett et al. 1973). These units are equipped to burn natural gas (56 percent), number 2 oil (38 percent), number 4 and 5 oils (8 percent), and number 6 oil (4 percent). About 6 percent of the units have dual fuel capabilities. Commercial boilers are designed to operate with a minimum of manual control and maintenance for a life of from 10 to 45 years depending upon the type of unit.

Residential units are much smaller; about 70 percent have capacities of less than 0.2 million BTU/hr, and 96 percent have capacities of less than 0.42 million BTU/hr. (Barrett et al. 1973). The units include warm air furnaces, hot-water and steam boilers, and water heaters, and are fueled primarily by distillate oil (No. 2) or natural gas. Operation is generally controlled by a thermostat and maintenance is on an annual schedule, at most.

Emissions from Residential Space Heating Equipment

Specific NOx emission levels from residential units average about 0.14 lb $NO_2/10^6$ BTU, (Barrett et al. 1973) which is low, primarily as a consequence of low combustion intensities in these units. About 90 percent of the residential units tested in a recent field survey (Barrett et al. 1973) of oil-fired space heating equipment had emissions below 0.2 lb $NO_2/10^6$ BTU; the highest emission level measured in that survey was about 0.29 lb $NO_2/10^6$ BTU. Normal service and adjustment practice had very little effect on the emission level of NOx or any pollutant other than smoke.

A recent EPA study (Hall et al. 1974) (USEPA 19) provides more detailed information on the effects of operating conditions and burner design on residential heating equipment emissions. Combustion improving devices, which utilize flame retention to produce a more intense flame region, tend to increase efficiency and to increase NOx emissions. A spark ignition system was found to produce between 7 and 10 percent of the NOx due to continuous operation of the ignition arc during the burning cycle. Turning the ignition system off after startup, or using low power output ignition could eliminate this source. Other modifications to existing equipment designs were found to have very little effect in this study. However, significantly lower levels of NOx emissions have been demonstrated without increases in other pollutant emissions, or loss of efficiency with both optimized operation of conventional systems (Dickerson and Okuda 1973), and new combustor concepts (Dickerson and Okuda 1974, Hall et al. 1974, Thompson et al. 1973). In a recent EPA-sponsored program, Rocketdyne has developed low emission burners based on conventional design practices. (Dickerson and Okuda 1974). One unit tested for 128 hours was a 20000 BTU/hr, residential size burner which produced about 0.048 lb $NO_2/10^6$ BTU. A commercial size, 180,000 BTU/hr, unit was also tested for 112 hours and produced only 0.025 lb $NO_2/10^6$ BTU. Burners which use hot combustion products to prevaporize the liquid fuel can achieve very low levels of all pollutants with 0.02 to 0.07 lb $NO_2/10^6$ BTU during steady state operation. During startup, however, the fuel vaporization is not effective in some systems and the CO and smoke emissions are high. Internal recirculation of combustion products is also very effective in reducing thermal NOx formation by reducing flame temperatures. NOx emissions as low as 0.015 lb $NO_2/10^6$ BTU have been reported. The lowest emission levels reported, less than .005 lb $NO_2/10^6$ BTU, have been achieved using catalytic combustion; (Dickerson and Okuda 1974, Thompson et al. 1973), however, the associated hydrocarbon emissions were very high. These studies have demonstrated that very low NOx emission levels are achievable for residential and commercial heating units. However, further development will be required before these systems become practical for wide scale application.

Emissions from Commercial Boilers

The specific NOx emissions from commercial boilers fired with natural gas or No. 2 oil are comparable with the emissions from residential units, i.e., 0.07 to 0.20 lb $NO_2/10^6$ BTU, (Barrett et al. 1973). Combustion of residual oils, No. 4, 5, or 6 oil, resulted in emissions as high as 0.47 lb $NO_2/10^6$ BTU in a Battelle field study, (Barrett et al. 1973) primarily due to the nitrogen content of these fuels, about 60 percent of which was converted to NOx.

For heavy oil combustion, reduced excess air resulted in reduced NOx emissions, but also high smoke and carbon monoxide emission levels. Techniques such as staged and low excess air combustion which have been applied for fuel NOx control in utility boilers might be adaptable to large commercial boilers, but have not been applied to date. Switching to low nitrogen content fuels, such as natural gas and No. 2 oil, if they are available, would be one means of NOx control for commercial units for which modification of the combustion process is not feasible. New combustion concepts such as pre-vaporization of the fuel or surface combustion also have potential for commercial units, but have not been demonstrated.

INTERNAL COMBUSTION ENGINES

Types and Locations

Internal combustion engines produce about 21.3 percent of the total stationary source NOx emissions, second only to utility boilers (Martin 1974). The installed capacity of stationary engines is distributed about equally between reciprocating engines (34.7 million horsepower) and gas turbines (35.5 million horsepower) (McGowin 1973). The applications and NOx emissions of these two classes of engines are quite different. Estimates of power generation by engines indicate that reciprocating engines, generate about 71.8 percent of the power while gas turbines generate the remaining power (McGowin 1973) corresponding to utilization of 58 and 22 percent, respectively, of their installed capacities. Emission estimates for these two sources are somewhat uncertain, particularly for gas turbines whose total emissions have been estimated at 131 (Martin 1974) to 291 (McGowin 1973) thousand tons of NOx per year. Nonetheless, it is clear that reciprocating engines produce the vast majority of the stationary engine emissions.

Recent survey estimates (McGowin 1973) of the installed power, fuel usage, emissions, and power generation for stationary engine applications are tabulated in Table 15-6. In terms of installed horsepower, the major uses of engines are electric power generation (54.8 percent), oil and gas pipelines (22.4 percent), agriculture (10.7 percent), natural gas processing (5.6 percent) and production (4.6 percent). The major sources of stationary-engine NOx emissions are oil and gas pipelines (41.6 percent), natural gas processing (19.2 percent), and natural gas production (13.8 percent) which use primarily spark ignition gas engines, and agriculture (14.2 percent) which uses primarily diesel engines. Electric power generation, which accounts for most of the installed horsepower, produces only about 5 to 10 percent of the stationary engine NOx due to two factors: (1) the overall load factor for engines used in electric power production is only 12 percent since the primary application of such engines is for peak and stand-by power generation; and (2) most of the installed horsepower is in the form of gas turbines which produce much lower NOx emissions than reciprocating engines. Reciprocating engines are used for peak power generation in older installations and continue to find application for electricity generation in municipalities, hospitals, schools and shopping centers which are too small to use gas turbine units. In recent years, both large electric utilities and natural gas pipelines have been favoring large gas turbines.

Recent projections from the electric utility industry (NERC 1974) for the period 1974-1983 indicate a 75 percent increase in the gas turbine generating capacity and a four-fold increase in electric power generated by gas turbines for continuous power generation in addition to peak power generation. This will be accompanied by slightly reduced usage of natural gas and increased usage of both

TABLE 15-6

Stationary Engines in the United States (McGowin 1973)

	Installed Horsepower - 10^3 Bhp						Fuel Consumption[a] 10^{12} Btu	
	Load Factor %	Diesel	Fuel	Gas Engine	Gas Turbine	Total	Natural Gas	No. 2 Oil
Electric Power	12	1,570	3,710	90	30,440	35,810	127.24	280.14
Oil & Gas Pipeline	69	830	390	10,990	3,520	15,730	802.06	40.77
Natural Gas Processing	-	-	-	2,410	1,530	3,940	432.60	-
Oil & Gas Exploration	15	1,500	-	500	-	2,000	5.09	14.67
Crude Oil Production	-	-	-	852	-	852	42.66	-
Natural Gas Production	-	-	-	3,237	-	3,237	190.04	-
Agricultural	40	7,500	-	-	-	7,500	-	198.94
Industrial Process	-	O	-	230	-	230	11.80	-
Municipal Water & Sewage	75	465	-	465	-	930	24.08	23.14
TOTAL	-	11,865	4,100	18,774	35,490	70,229	1635.75	557.66

(a) Calculated assuming Higher heating values of 5.8×10^6 Btu/Bbl for oil and 1070 Btu/SCF for natural gas.

TABLE F5-6 (cont.)

Power Generation 10^6 bhp hr		Annual NO_x Emissions Tons			Emission Factors gm NO_x/bhp hr	
Recip	Gas Turbine	Recip	Gas Turbine	Total	Recip	Gas Turbine
5,900	33,240	62,440	62,920	125,360	9.61	1.72
73,700	21,260	39,000	39,800	970,000	11.46	1.70
31,200	15,010	429,690	28,130	457,820	12.47	1.70
2,580	-	31,720	-	31,720	11.16	-
5,410	-	62,370	-	62,370	10.47	-
24,100	-	308,200	-	308,200	11.61	-
26,280	-	318,700	-	318,700	11.01	-
1,510	-	19,300	-	19,300	11.61	-
6,110	-	76,100	-	76,100	11.31	-
176,870	69,510	2,238,720	130,850	2,369,570	-	-

Per cent Capacity
58.12% 23.36

distillate and residual oils as gas turbine fuels. (NERC 1974).

Large concentrations of stationary engine horsepower, e.g., 10,000 to 60,000 Bhp, reciprocating engines at some pipeline compressor stations, may represent significant local pollution sources. Most pipeline compressor stations are remote from population centers; however, there is some human exposure at most compressor stations and the NOx levels may be high (McGowin 1973).

Stationary reciprocating engines can be classified in several categories. The fuel/air mixture may be ignited in reciprocating engines by either an electrical spark discharge (spark ignition engines) or by compression heating (diesel engines). Natural gas engines are almost always spark ignited. In diesel engines combustion is controlled by injecting fuel into the cylinder through a spray nozzle at the proper time during the compression stroke. Dual fuel engines can operate on 100 percent fuel oil or natural gas to which fuel oil has been added (about 5 percent on the basis of heating value) to serve as a pilot for ignition.

Stationary gas turbines have evolved from aircraft jet engines. In its simplest form, the gas turbine engine (simple cycle) consists of a compressor in which the air is compressed from 100 to 200 psig, followed by a combustor in which fuel (liquid or gaseous) is added. Combustion occurs at mixtures of fuel and air which are overall very fuel lean. The combustion gases are then expanded through a turbine or turbines to near atmospheric pressure, converting their energy to power. The simple cycle gas turbine thermal efficiency is 24 to 31 percent. The efficiency of the gas turbine can be increased by further extraction of heat from the exhaust gases which normally leave the turbine at 800 F to 1100F. In a regenerative cycle this is done by passing the air leaving the compressor through a heat exchanger in which some of the heat of the exhaust gases leaving the turbine is transferred to the pressurized air. In such a cycle less fuel is required for fixed turbine inlet temperatures increasing the thermal efficiency to about 34 to 38 percent. Another way of utilizing the exhaust waste heat is in a combined cycle. In this case the exhaust gases are used to produce steam which can then be used to drive a steam turbine to produce additional power. Due to its high thermal efficiency, currently 40 to 42 percent, the combined cycle gas turbine is being favored by electric utilities in new installations.

Emission Levels

Table 15-7 summarizes the emission factors used in the estimates of exhaust emissions from stationary engines described above. While there is considerable variation in the emissions, indicated by the numbers in parentheses, these figures provide a useful basis for comparison of the uncontrolled emission levels of the various engines.

Except for gas turbines and prechamber diesel engines, the NOx levels, in the range 2.8 - 4.2 lb NOx/10^6 BTU, exhibit only minor differences among the different engine types (McGowin 1973). Turbocharging results in slight increases in NOx emissions over naturally aspirated engines due to increased temperatures. Prechamber diesel engines, in which combustion is initiated in a fuel rich environment, emit about half as much NOx as direct injection diesels.

Gas turbine NOx emissions are one-tenth the emissions of reciprocating engines, primarily as a result of lower peak temperatures and shorter residence times in the combustion chamber. Emissions from distillate-oil fired gas turbines are about a factor of two higher than emissions from gas turbines fueled with natural gas (Johnson and Schiefer, Roessler et al. 1974). Since the electric utility projections indicate increasing reliance on oil as gas turbine fuel, including residual oils as a substantial fraction of total fuel by the mid-1980's (NERC 1974), the uncontrolled specific

TABLE 15-7

Stationary Engine Emission Factors (McGowin 1973)

	Engine Type	Cycle	Charging Air	Charging Fuel	BSFC Btu/Bhp-hr	NO_x (lb/10^6 Btu)	CO (lb/10^6 Btu)	HC (lb/10^6 Btu)
13 Mode Test	Diesel	4	TC	DI	7252	42 (3.5-4.0)	1.2	0.43
			NA	DI	0420	2.9 (1.9-5.6)	1.4	1.4
			TC	PC	7252	1.6 (1.1-2.2)	0.49	0.10
			NA	PC	0420	1.5	0.65	0.00
		2	SC	DI	7840	4.1	1.7	0.22
Constant Conditions		4	TC	DI	7252	3.3	1.2	0.04
			TC	PC	7252	2.3	0.20	0.03
			NA	PC	0420	1.6	0.25	0.05
	Dual Fuel	4	TC	DI	5970	(2.9-3.1)	0.74	1.1
	Natural Gas	4	TC	DI	6030	3.9 (3.5-4.9)	0.32	0.65
			NA	DI	7150	3.6 (2.4-4.9)	0.43	0.62
			TC	High Speed	7000	4.0 (3.5-4.9)	1.0	0.66
		2	TC	DI	6635	3.5 (2.3-6.7)	0.90	1.5
			NA	DI	7100	3.3 (1.4-5.1)	0.09	3.3
	Gas Turbine (natural gas)				11105	0.34 (0.17-0.51)		

TC = Turbocharged: NA = Naturally Aspirated; SC = Supercharged; DI = Direct Injection; PC = Prechamber Engine: Oil: HHV = 19600 Btu/lb.

emissions for gas turbines may be expected to increase. Fuel-nitrogen may play an important role in determining the achievable emission levels, since the limited data available on fuel-NOx formation in gas turbines suggests a high conversion efficiency of fuel-N to NOx (Fenimore 1972, Dilmore and Rohrer 1974).

NOx Emission Control Technology

Many emission control techniques which have been developed for mobile sources are potentially applicable to stationary engines; however, different design constraints may make techniques either more or less desirable for stationary engines than for mobile engines. Size and weight constraints are severe for mobile engines, and durability is an important characteristic of an engine used in stationary applications. Techniques to reduce NOx include engine derating, fuel injection and ignition timing retard, exhaust gas recirculation, catalytic converters, water or steam injection, and engine component and operating condition modifications. Methods developed or evaluated for mobile-source NOx-control may be applicable to stationary engines, but the emission control achievable and the operating parameters in stationary applications cannot be directly inferred from data obtained on smaller mobile engines. Since the carbon monoxide and unburned hydrocarbon emissions of large, stationary, reciprocating engines tend to be quite low, moderate increases in these emissions due to NOx control may be acceptable although this must be determined on a case-by-case basis.

A detailed discussion of control techniques for various engine types appears in Appendix 15-A but the major conclusions are summarized here. In diesel engines, adjustment of the fuel injection timing can reduce NOx emissions by about 40 percent without major hardware alteration. The NOx reduction is accompanied by increases in fuel consumption and in carbon monoxide emissions. Cooling the intake air can reduce NOx emissions and improve fuel economy. Optimization of the fuel injection system may produce modest reductions (20 percent for one engine) in NOx without increasing fuel consumption.

Diesel NOx emissions can also be reduced by as much as 50 percent by water injection into the intake system with very little change in fuel consumption but corrosion problems must be resolved before water injection techniques become useful for stationary engines.

Exhaust gas recirculation (EGR) has been shown to be a very effective NOx abatement technique for diesel engines. Fifty percent reductions in NOx emissions have been achieved using EGR with only a small penalty in fuel consumption. However, the NOx emission reductions are accompanied by a 100 to 150 percent increase in carbon monoxide and smoke.

NOx can be removed from diesel exhaust gases by catalytic reduction. Very high removal efficiencies have been observed but the costs are high and significant development work will be required before wide scale application will be practical.

In spark ignition engines, a fuel rich mixture results in low NOx emissions at the expense of increased carbon monoxide and hydrocarbon emissions and of increased fuel consumption. Stratified charge engines offer the possibility of substantially reduced emissions as well as reduced fuel consumption.

Reducing the temperature of the fuel/air mixture in a spark ignition engine results in a moderate reduction in NOx emissions while reducing fuel consumption. Water injection can reduce the NOx emissions by as much as 80 percent, but this may be accompanied by a significant increase in fuel consumption. Exhaust gas recirculation can reduce NOx emissions by as much as 80 percent, but the fuel economy loss is considerable. Recirculating only five to ten percent of the exhaust gas and advancing the spark timing can minimize the loss, making 40 percent reductions in NOx emissions possible.

For all techniques designed to reduce NOx emissions from spark ignition engines, the effects on engine durability must be determined before long term use on stationary engines is practical.A meaningful assessment of the cost of emission control for large stationary gas engines has not yet been performed.

Water or steam injection has been shown to be a very effective technique for stationary gas turbine NOx emissions. For small gas turbines (less than 2 Mw) the costs of this technique may be prohibitive. NOx reductions of 50 to 75 percent have been achieved with water injection with only slight increases in carbon monoxide emissions. Water injection is currantly being used by some utilities without observed deleterious effects as long as the water purity is maintained at a high level.

INDUSTRIAL PROCESSES

Industrial Process Heating

It is estimated that industrial process heating contributed 3.3 percent of total stationary source emissions in 1972. The sources include open-hearth furnaces with high air-preheat used for glass-melting and to a decreasing extent in steel making; rotary, vertical, and tunnel kilns used in cement, lime, and ceramic industries; pyrolysis process furnaces for ethylene and propylene production; petroleum heaters, cat-cracking regenerations, and CO boilers in the petroleum industry; billet-reheating and ingot-soaking furnaces, coke-ovens and blast furnaces in the steel industry; and a large variety of heat-treatment furnaces in the metallurgical industry. The emissions from these sources have not been well documented. In view of the small contribution to the total national emissions of NOx and the diversity of sources, the control of emissions from process heating furnaces has received comparatively little attention other than through the development of low-emission burners by the equipment manufacturers that supply the combustion equipment. The applicability of combustion modification techniques to process-heaters depends upon the specific heater design and no generalizations can be made at present about the potential for control of emissions from these sources.

Non-Combustion Sources

The major non-combustion sources of NOx are the manufacture and use of nitric acid which were estimated to contribute 1.3 percent of the stationary source emissions in 1972. The techniques for controlling NOx emission from non combustion-sources are based on removal of the NOx from the tail gas by selective or non-selective reduction, adsorption, and absorption. Uncontrolled emissions from nitric acid plants are in the range of 2000 to 3000 ppm (approximately 28 to 41 lbs per ton of acid) and are characterized by approximately equal concentrations of NO and NO_2. As a consequence of the cost of installation of tail-gas treatment processes to meet emission standards of 3.0 lbs/ton of acid for new plants, nitric acid plants are being designed to yield lower concentration effluents.

The most commonly used emission-control technique at present is the catalytic combustion of the NOx using a fuel, generally natural gas, and a platinum or palladium catalyst (Bartok et al. 1969, Gillespie et al. 1971). In this process sufficient fuel must be supplied to consume the oxygen present in the tail-gas and part of the energy released is recovered in a waste heat boiler and/or a power recovery turbine. A selective reduction technique has been developed using ammonia to reduce the NOx without consuming the oxygen. For this technique to work, the catalyst bed temperature must be carefully controlled and maintained in the range of 410F to 520F. Several selective abatement techniques have been installed on commercial units (Bartok et al. 1969).

A commercial adsorption process has been developed utilizing a molecular sieve bed to catalytically convert NO to NO_2 and to adsorb the NO_2 product. Continuous operation is achieved through the use of two beds in an adsorption-desorption cycle. The NO_2 is recycled to the nitric acid plant. High recovery efficiencies are claimed for this process at costs competitive with catalytic reduction (Fornoff 1972). Two units have been installed on commercial plants.

Absorption techniques for NOx are hindered by the relatively low solubility of NO. A process enhancing the absorption of NOx by catalyzing the oxidation of NO to NO_2 has been commercialized (Mayland and Heinze 1973).

For typical NOx concentrations in a tail gas of about 3000 ppm, the methods described above can achieve over 90 percent removal. The best abatement technique for a given unit depends on the specific conditions at a plant. Investment and operating costs are of the order of \$1500/ton-per-day capacity and \$1.50 per ton of acid produced, but vary greatly with location, size, and design of a unit.

FLUIDIZED BED COMBUSTORS

State of Development

The combustion of coal in fluidized beds operated in the temperature range of 1400F to 2000F appears to have a number of advantages over conventional utility pulverized-coal boilers with stack-gas clean-up. These advantages include:

(a) high heat release rates per unit volume permitting the construction of relatively compact boilers without penalizing the over-all combustion efficiency;

(b) high rates of heat transfer to tubes immersed in the bed and, therefore, a relatively small tube surface area;

(c) sulfur dioxide removal capabilities of 80 to 95 percent by injection of limestone or dolomite in the bed;

(d) operation below the ash softening temperatures and, hence, elimination of slagging problems and a reduction in emissions of hazardous trace metals [operation at temperatures above 2050F may be used, however, to promote self-agglomeration of the ash (Ehrlick et al. 1974)];

(e) use of coarsely pulverized coal in contrast to under 200 mesh for a pulverized-coal boiler;

(f) low nitrogen oxide emissions;

(g) potential for increased cycle efficiency by use of pressurized boilers in a combined steam/gas-turbine cycle; and

(h) ability to handle low-grade solid fuels

Studies of fluidized bed combustion for coal burning were initiated in 1961 by the National Coal Board in England on small-scale units operated at pressures from atmospheric up to 6 atmospheres (Hoy and Roberts 1969, Broadbent 1970). In 1965, work was initiated in the U.S. at Pope, Evans, and Robbins under contract to the Office of Coal Research (Bishop et al. 1966). With the recognition of the potential of fluidized bed combustion for reducing the emissions of sulfur and nitrogen oxides, the U.S. program has expanded to include a number of organizations working under the sponsorship of the Office of Coal Research and the Environmental Protection Agency.

A number of system design studies for the use of pulverized combustion have been conducted by Pope, Evans, and Robbins (Bishop et al. 1966, Bishop et al. 1968, Bishop et al. 1972, Ehrlich et al. 1974, Ehrlich and Chronowski 1974, Ehrlich and McCurdy, Ehrlich et al. 1971, Ehrlich 1970), Westinghouse (Archer et al. 1971), and Foster-Wheeler (Gamble 1974). These studies lead to the conclusion that fluidized bed combustion offers its greatest potential in competition with conventional fossil-fuel utility boilers using low-sulfur fuel or stack-gas clean-up. The economic analyses indicate that the pressurized units used in combined cycles have greater potential in the long-range than the atmospheric pressure units. The development of atmospheric pressure combustors is, however, more advanced and a 30 MW unit is scheduled for completion in 1975 in Rivesville, Virginia. Depending on the successful operation of this cell, it is expected that multicellular 200 MWe units will be installed before 1980 and 800 MWe units shortly thereafter. It is anticipated that the installation of pressurized units will follow the atmospheric-pressure units. It is clear that the acceptance by the utility industries of the fluidized bed combustors will depend greatly on whether or not these units will be able to match the relatively trouble-free operation of conventional pulverized-coal units.

NOx Emissions from Fluidized-Bed Combustors

Fluidized-bed combustors have the potential of meeting the existing standards for NOx emissions from coal-fired steam-generating plants by a considerable margin. However, the factors that control these emissions are imperfectly understood. At the temperatures of the fluidized bed negligible amounts of thermal NOx are expected and the major source of NOx emission is the fuel nitrogen. This has been proven conclusively in tests at the Argonne National Laboratories which showed no change in NOx concentrations from a fluidized-bed coal combustor when the nitrogen in the combustion air was replaced by argon (Jonke et al. 1969). The emissions from fluidized bed combustors are therefore a function of both the fuel nitrogen content and the fraction of fuel nitrogen converted to NOx during combustion. Many factors influence the conversion of fuel nitrogen to NOx. Tests on a batch laboratory reactor have shown that part of the NOx is contributed by oxidation of the volatiles emitted by the coal and the remainder by oxidation of the char, with the fractional contribution of the volatiles increasing with increasing temperature (Pereira et al. 1974). The emissions from laboratory and pilot units have been found to increase with increasing temperature and increasing excess air (Hammons and Skopp 1971, Jonke et al. 1969, Jonke et al. 1970, Jonke et al. 1971). Increased pressure in some cases yields lower emissions (Vogel et al. 1974). The emissions of NOx from the beds are also found to be a function of the composition of the solids in the bed. Calcium sulfate catalyzes the reduction of nitrogen oxide with carbon monoxide and the emissions from a bed using calcium sulfate as a bed material are lower than the corresponding emissions with an alundum bed material (Hammons and Skopp 1971). In addition a partially sulfated lime is found to reduce nitric oxide (Hammons and Skopp 1971). Some additives, such as cobaltic oxide, however, cause the emissions to increase (Jonke et al. 1970, Jarry et al. 1970), possibly as a consequence of catalysis of the oxidation of the fuel nitrogen. Representative values of emissions for a range of conditions are shown in Table 15-8.

Emission levels of 0.10 to 0.17 lbs $NO_2/10^6$ BTU have been attained in both atmospheric and pressurized combustors operated slightly fuel lean (15 percent excess air or 3 percent oxygen) (Demski et al. 1973, Vogel et al. 1974). Other atmospheric pressure tests have yielded higher emissions than these although the emissions were still lower than the standard for coal units (Hammons and Skopp 1971). If needed, significant additional reduction in NOx emissions may be attained in fluidized bed combustors by the use of staged combustion (Hammons and Skopp 1971).

TALL STACKS AND INTERMITTENT CONTROLS FOR NITROGEN OXIDES

Until recently the U.S. Environmental Protection Agency has consistently argued against intermittent controls as a means of achieving compliance with air quality standards. As several states have reviewed the cost-effectiveness of their clean fuel regulations, intermittent controls have gained more support. The use of tall stacks and of intermittent controls (such as fuel switching and load switching) has been directed primarily toward the control of sulfur dioxide emissions and their effects and, to a lesser extent, toward particulate emission control. This section reviews the applicability of tall stacks and intermittent controls to achieve compliance with NOx standards or to reduce photochemical smog formation.

TABLE 15-8

NOx Emissions from Fluidized Bed Combustors (Jonke et al. 1974, Skopp et al. 1971)

T(°F)	O_2(%)	Pressure (atm)	NOx lb NO_2 / 10^6 Btu	% N in Fuel	Comments
1500	6-7	1	0.11		
1500	4	1	0.11-0.17		
1400	4	1	0.45	1.4	$CaSO_4$ bed material
1500	4	1	0.00	1.4	" " "
1000	4	1	0.99	1.4	" " "
1600	1	1	0.01	1.4	" " "
1600	4	1	0.95	1.4	" " "
1600	6	1	1.05	1.4	" " "
1600	0	1	1.12	1.4	" " "
1600	1	1	1.05	1.4	Alundum bed material
1600	4	1	1.12	1.4	" " "
1600	6	1	1.19	1.4	" " "
1600	0	1	1.26	1.4	" " "
1445	2.7	0	0.27	1.1	
1565	3.0	0	0.29	1.1	
1575	3.0	0	0.20	1.1	
1650	3.0	0	0.25	1.1	
1460	3.0	0	0.21	1.1	
1565	3.0	0	0.27	1.1	
1550	3.0	0	0.22	1.1	
1460	2.7	0	0.21	1.1	
1630	3.0	0	0.30	1.1	
1665	2.9	0	0.17	1.1	

Tall Stacks: The Effects of Elevated Release on Nitrogen Oxides

Tall stacks (greater than 500 feet) have been proposed as possible control methods for sulfur dioxide but their use may have an effect on nitrogen oxides. For pollutants with long half-lives (several hours to several days) predicted ground level concentrations of pollutants are reduced significantly by the use of taller stacks. As seen in Figure 15-6, increasing the effective stack height by a factor of 10 decreases the predicted maximum downwind concentration by two orders of magnitude. Of course, there are many simplifying assumptions in these calculations, for example the existence of flat terrain, which restrict their direct application.

Moreover, air pollutants are not chemically inert. Nitric oxide is oxidized to nitrogen dioxide which can dissolve in water droplets to form nitric acid. The acid is neutralized to nitrates, and the nitrates are eventually deposited on the surface. The conversion times for nitric acid and nitrate formation are a factor of three to four times faster than those for sulfuric acid and sulfates, the result of similar processes which occur with sulfur dioxide emissions. Thus secondary nitrogen oxide products such as nitric acid can potentially have a much more serious impact in the vicinity of the source than sulfur oxide decay products.

The effects of tall stacks on the formation of secondary pollutants is somewhat speculative. Assume that these stacks will service large, high efficiency combustion processes, for example power plants. Hydrocarbon and carbon monoxide emissions should therefore be extremely small. The NO emissions at higher levels will be separated from the ground level hydrocarbon and carbon monoxide automobile emissions. Hydrocarbons and carbon monoxide are important in the production and recycling of the transient radicals which drive the net dynamic balance equation to the conversion of NO to NO_2 as shown in Chapter 14, Appendix 14-A. Calvert (1973), for example, has shown in numerical simulations, that if hydrocarbons are absent, decreased CO concentra-

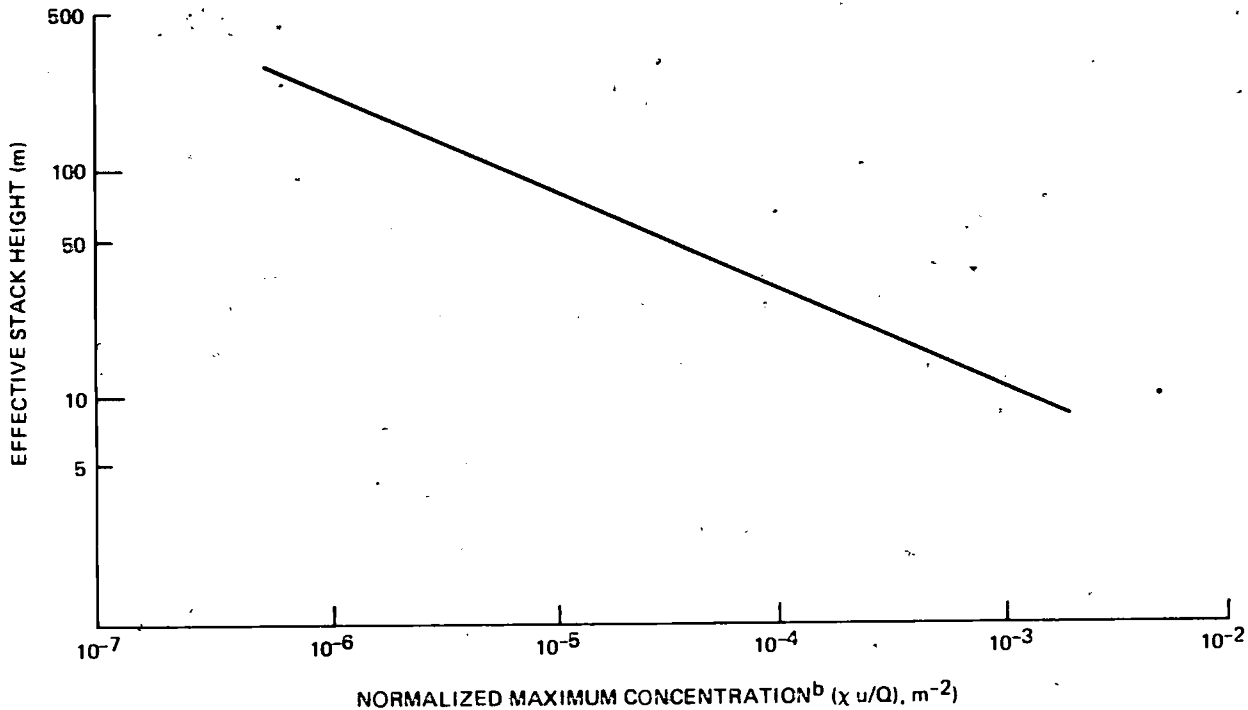

FIGURE 15-6: Normalized Maximum Concentrations as a Function of Effective Stack Height[a]

tions lower the maximum levels of NO_2 and O_3.

On the other hand, tall stacks may release NO into regions rich in ozone. Coffey and Stasiuk (1975) have recently suggested that urban peak ozone concentrations are primarily a result of high background levels of ozone rather than local photochemical production. They argue that since urban areas are major sources of nitric oxide, they act as sinks for ozone. Maximum surface ozone levels are associated with maximum downward mixing. Unpublished night ozone measurements by the authors show uniform ozone levels from just above the ground to 5,000 feet and peak values occurring in a haze layer between 5,000 and 5,500 feet. (See Figure 15-7.)

Davis et al. (1974) have tracked a power plant plume from two 200 meter stacks. Ambient ozone concentrations at this level had been consistently 60 to 80 ppb for several days. These high ozone levels produced rapid NO to NO_2 conversion. Ozone was completely depleted in the plume out to 24 km downwind. However beyond this point ozone concentrations in the plume began to rise, eventually exceeding ambient levels out of the plume by 20 ppb. In the absence of hydrocarbons and carbon monoxide, the authors have suggested a photochemical reaction scheme with the sulfur oxide emissions to account for the long distance O_3 generation. Altshuller (1974) has suggested that the entrainment of ambient air containing hydrocarbons and carbon monoxide allows the classical photochemical reactions to occur after sufficient dilution of the plume.

At present, there are not enough field measurements to determine the relative effects of near ground level and high level nitrogen oxide emissions. There is evidence that high level sources (power plant plumes) may lead to increased ozone many kilometers downwind but field studies are needed to assess rural and urban oxidant levels as well as their vertical distribution.

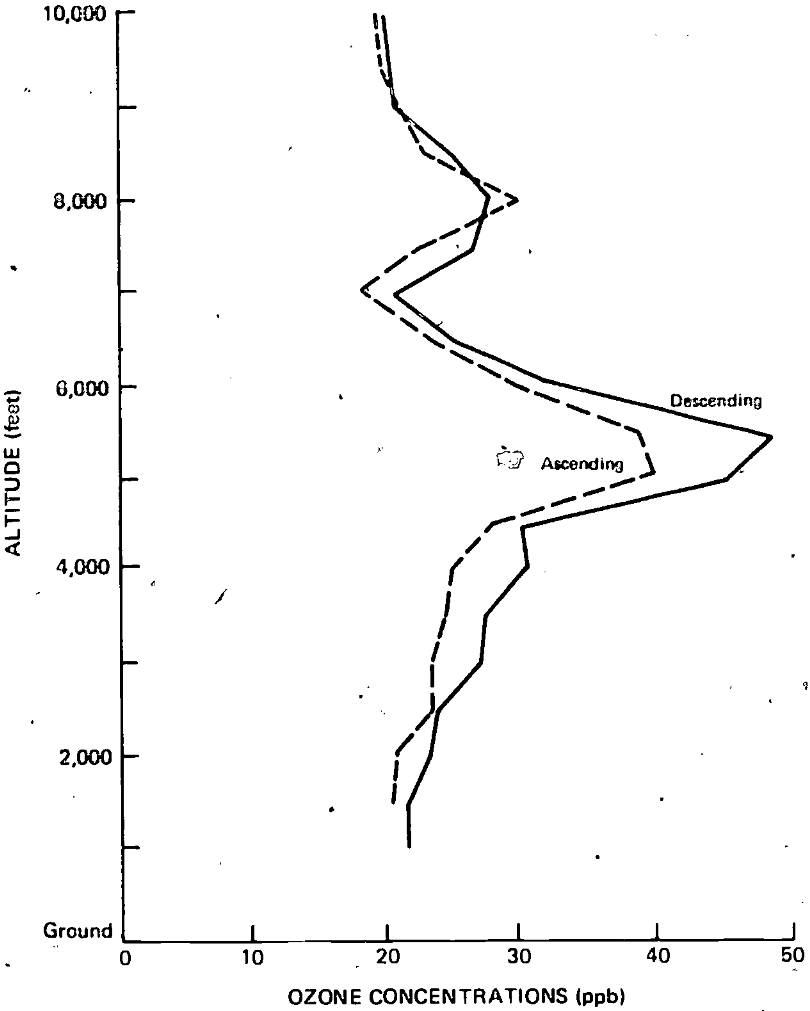

FIGURE 15-7: Vertical Ozone Profile Over Schenectady, N.Y., October 22, 1974. 2200-2300 Hours.

Intermittent Control Strategies

The use of intermittent methods and procedures for NOx controls is limited for the following reasons:

(1) although fuel switching is successfully being employed to control ambient sulfur dioxide levels, the organic nitrogen component of fossil fuels varies only from 0.1 percent for distillate oils to greater than 2 percent for Colorado shale residual oil and bituminous coal: this range is far smaller than the natural variation in the amount of sulfur in fossil fuels;

(2) there are at present no processes available which can readily and economically remove nitrogen from the fuel;

(3) unlike sulfur dioxide, which is formed entirely from oxidation of sulfur in the fuel, some nitrogen oxides originate from thermal fixation of air N_2 during high temperature combustion so that nitrogen oxides can be emitted even in the absence of fuel nitrogen;

(4) while fuel switching based on meteorological parameters to meet one-hour, three-hour and 24 hour sulfur dioxide standards (depending on the state) can be economical for large fuel consumers with dual fuel storage capacity, there are no short term Federal ambient air quality standards for NOx. The need for fuel switching would have to be determined on the basis of predicted or measured photochemical oxidant levels. Episodes of high oxidant concentrations can last several days, but the annual frequency of such occurrences is quite low (less than 0.01) for most of the country. It is unlikely that large storage capacity could be devoted to low nitrogen-content fuels when the use of such fuels would be rare;

(5) because of complicated reactions between NOx and other pollutants, the prediction of ground level NOx or

photochemical oxidant concentrations is very uncertain.

In spite of these limitations, there are some situations in which NOx may be dynamically controlled. Further, through allocation of low nitrogen content fuels some reduction of NOx emissions may be realized. Intermittent or supplementary control techniques applicable to NOx emission reductions are summarized in this section.

Fuel Type Substitution: Oil for Coal

The nitrogen content of distillate oil averages 0.1 percent, residual oil 0.4 percent and coal 1.4 percent. On this basis it would appear that fuel conversion may lower NOx emissions (Archer et al. 1971, Armento and Sage 1973a, Armento and Sage 1973b, Armento 1974, Axworthy and Shuman 1973, Bagwell et al. 1970, Barnhart and Diehl 1959). However, this is not necessarily the case. On a BTU rating, the emissions from existing coal and residual oil fired units of comparable size are sometimes nearly equivalent. (See Table 15-9, which summarizes EPA emission factors based on contractors' fuel tests.) There is an emission factor difference between new utility coal and oil fired units; in new plants, for coals NOx emission limitations are 0.7 lbs. per million BTU's, and for oil the NOx emission limitations are 0.3 lbs. per million BTU's.

However, this 0.4 lbs. per million BTU advantage (57 percent) would be difficult to achieve simply by changing fuels. The lower emissions factor for new utility boilers is a result of new design characteristics. This design advantage may be inappropriate for maintaining lower emission rates if fuel is switched.

Fuel Type Substitution: Gas for Oil, Gas for Coal

The new source NOx emission limitation for

TABLE 15-9

NOx Emission Rates (Pounds/Million BTU's)[a]

Boiler Types	Coal	Oil	Gas
New Utility Boilers	0.7	0.3	0.2
Existing Boilers (>100 million BTUs/hr)	0.7-1.2	0.7	0.3[b]-0.7
Intermediate Boilers (10-100 million BRU/hr)	0.6	0.27[b]-0.53	0.12[b]-0.23
Commercial/Domestic Boilers	0.27	0.28[b]-0.54	0.08[b]-0.12

[a]Converted from EPA, Emission Document AP-42, and assumes
coal - 12,500 BTU per pound,
residual oil - 150,000 BTU per gal.,
and natural gas - 1,000 BTU per cu. ft.

[b]Tangentially fired boiler.

large utility boilers using gaseous fossil fuel is 0.2 lbs. per million BTU. Provided, as stated above, if there were no loss in boiler efficiency, there would be a 71 percent reduction in NOx emissions when substituting gas for coal. For an oil to gas substitution the maximum reduction would only be 33 percent.

Provided emission limitations are met, substitution of natural gas thus may be an attractive method of reducing NOx emissions from new utility boilers during times of elevated oxidant levels.

Because the NOx emissions per million BTU can be comparable for coal, oil, and gas in existing utilities and large industrial fired boilers (greater than $100x10^6$ BTU/hr), there is no apparent advantage in substituting fuels for intermittent controls in all existing facilities. The situation is different for smaller boilers. There, using different fuels results in a sizable difference in NOx emissions for the same heat equivalence. This difference can be translated to a reduction of NOx emission when substituting cleaner fuels. (See Table 15-9)

It should be noted that fuel substitution is only feasible when dual fuel capabilities are built in. The cost and time necessary to convert to either a new fuel or a dual fuel system make fuel substitution impractical as a short-term intermittent control technique.

Fuel Switching: Low Nitrogen Oils for High Nitrogen Oils

There is some variation in the fuel nitrogen content of oils. U.S. crude oil nitrogen content ranges from 0.056 percent nitrogen by weight for Louisiana crude to 0.49 percent for California crude. (See Table 15-1) In the refining of crude the heavier nitrogen molecules tend to concentrate in the higher boiling point distillates and residual oils.

It would not be practical to implement a fuel switching supplemental control program to reduce ambient levels of NO_2. In most urban areas large utility and industrial point sources represent 25 percent to 35 percent of the NO_x emissions. The urban point sources with fuel switching capabilities probably contribute less than half of these emissions or approximately 10 to 15 percent of the total urban NOx emissions. Assuming a stationary source fuel combustion process which produced NO in the flue gas of which 50 percent is derived from nitrogen organically bound in the fuel and 50 percent from nitrogen thermally fixed from air, a fuel switching program could reduce total urban emissions by no more than about 5 percent. On balance, there is extremely limited value in fuel switching to control NOx on a short-term local basis.

This does not imply that there are no environmental concerns about the use of high nitrogen fuels. These fuels sould be allocated to areas that do not have high NOx or oxidant levels.

Load Shifting of Electric Power Generation

With large regionally integrated electric power exchanges, the potential exists to shift power generation loads to reduce pollutant emissions during adverse meteorological conditions. Depending on the flexibility of individual power exchanges, load switching could reduce NOx significantly in certain areas. For example, steam and power generation contributes 30 percent of the NOx emissions in the 30-town Boston area. The effect of fulfilling electrical demand by generating electricity elsewhere in the New England Power Exchange or in plants with lower NOx emissions could be sufficient to prevent violation of ambient photochemical oxidant and NO_2 air quality standards during adverse conditions.

Water Injection to Lower Combustion Temperature

As has been noted, since NO formation from the reaction between atmospheric nitrogen and oxygen is highly temperature-dependent, reducing peak flame temperature can be effective in reducing NOx emissions. Steam or water injections to lower peak flame temperature could be used as an intermittent control technique. During elevated oxidant conditions reduction of NOx emissions could be rapidly effected by this procedure. However, even assuming a 50 percent trimming of the NOx emissions, only 5 to 20 percent of an urban area's NOx would be reduced at most. It is not known whether even the most optimistic reduction of 20 percent would significantly influence photo-oxidant formation during adverse conditions, for the reduction in NO concentrations, in general, only increases the reaction time for oxidant formation (Calvert 1973).

FUTURE TRENDS

Fuel Usage

It is projected that in the near future the U.S. will need to rely increasingly on fuels derived from its major domestic reserves of coal and possible shale. As the nitrogen content of both coal and shale are high, increased utilization of fuels from these sources will result in increased emissions of nitrogen oxides unless the nitrogen content is reduced prior to combustion or a capability for burning high-nitrogen content fuels with acceptable NOx emission levels is developed.

The forecast for changes in fossil fuel requirements by the utility industry (NERC 1974) for the period 1974 to 1983 is that coal usage will increase from 417 to 742 million tons/year (approximately 10.8 to 19.3 x 10^{15} BTU/year), oil from 685 to 968 million barrels/year (approximately 4.0 to 5.6 x 10^{15} BTU/year), and natural gas from 2.8 to 1.9 x 10^{12} cubic feet/year (approximately 2.9 to 2.03 x 10^{15} BTU/year). These projections lead to the conclusion that if all utility boilers by 1983 emitted at levels equal to the standards for new sources, the total emissions would increase from the estimated 5.9 x 10^6 tons/year in 1972 to about 8 x 10^6 tons/year in 1983. (More detailed projections of future emissions under various assumptions can be found in Appendix 14-C.)

It is expected that synthetic gaseous and liquid fuels derived from coal will be used to meet the deficit between the U.S. production and demands for these fuels. Little is known on the NOx emission potential of new fuels.

High BTU gas is expected to follow the behavior of natural gas. Low BTU and intermediate BTU gas will generally have relatively low flame temperatures and therefore low thermal NOx emissions. EPA sponsored tests with a 180 BTU gas yielded NOx levels less than a tenth the value obtained with a natural gas fired under the same conditions (Martin 1974). Ammonia, pyridine, and hydrogen cyanide and other nitrogen containing compounds are produced in the coal gasification process. The fuel NOx emissions will depend to a large extent on the gas clean-up process; they will be low when conventional low-temperature high efficiency scrubbing systems are used but may be significant if a high temperature desulfurization step is used. The nitrogen content is more likely to be high in synthetic liquid fuels since the sulfur is more readily removed than the nitrogen by catalytic hydrogenation. Potential problems with fuel NOx should be anticipated in the combustion of synthetic fuels derived from coal.

Alcohol fuels have been proposed as alternatives to liquid natural gas and also as a possible product from conversion processes for coal or solid wastes. A probable composition of alcohol fuels is 90 percent methanol with the balance containing higher alcohols (Martin 1974). Methanol can be considered the equivalent (from an energy viewpoint) of a mixture of approximately equal mass of a CH_2 fuel and water. Studies by the EPA on the combustion of methanol with 5 percent excess air in a hot refractory wall furnace (wall temperatures in excess of 2500F) found NOx emission levels a factor of 4 lower than those from a distillate oil and a factor of 3 lower than those from propane (Martin 1974). Blending higher alcohols in the methanol increased the emissions but even with a 50-50 blend of methanol and isopropanel the emissions were below 150 ppm (approximately 0.2 lbs/10^6 BTU) (Martin 1974).

Surface Combustion

In surface combustion very low thermal NOx emissions are achieved by reducing the peak flame temperatures below the levels attainable in conventional combustors. The low temperatures are achieved either by very lean fuel combustion on a catalyst bed or by heat transfer from a flame layer to an adjacent cooled solid surface (Roessler et al. 1973).

Emission levels as low as 0.005 lb $NO_2/10^6$ BTU have been reported for a catalytic combustor designed to simulate gas turbine operating conditions and achieving very high combustion efficiencies at an energy release 3.5 x 10^6 BTU/(atm)(hr)(ft)(Blazowski and Bresowar 1974). Catalytic surface combustion has been proposed for stationary gas turbines, electric utility boilers, and small space heating equipment; however, several problems must be solved before catalytic systems become accepted commercially.

Emissions from non-catalytic surface combustion systems are quite low, 0.005 to 0.10 lb $NO_2/10^6$ BTU (Roessler et al. 1973). Porous plate combustors have been proposed for utility boiler and stationary gas turbine applications, but several problems must be resolved before the technique is practical.

Flue Gas Treatment

The costs for removal of NOx from tail-gases are generally higher than those associated with reducing NOx formation in the combustion process. Major emphasis in the U.S. has been on combustion process modification since it permits the construction of new equipment that can meet existing standards. A considerably smaller effort has been devoted to flue-gas treatment. Faced with more stringent emission standards, Japanese industry has been much more interested in flue-gas treatment and has several major pilot plants in operation and full-scale plants under construction (Tohata 1974).

A number of methods have been proposed for reducing NO concentrations in stack gases. The processes that have proceeded farthest towards full-scale demonstration are the catalytic reduction of NO with ammonia or hydrocarbons; the oxidation of NO to NO_2 using ClO_2 followed by the reduction of NO_2 to N_2 by Na_2SO_3; and the addition of NO_2 to produce an equimolal mixture of NO and NO_2 followed by absorption in $Mg(OH)_2$, a process that has the advantage of also removing SOx. A number of problems remain to be resolved, however, before these methods can be considered to be practical.

Selective reduction of NO to N_2 using NH_3 and a noble metal catalyst is being explored on a pilot scale in the U.S. For a stack gas with a concentration of 225 ppm NOx, ninety percent removal of the NOx has been achieved but, in the process, some unreacted NH_3 (approximately 25 ppm) escaped. The selective reduction of NO by NH_3 can also be achieved without a catalyst by carrying out the reaction in the temperature range of 1600 to 2000F (Lyon 1974). In laboratory tests, efficiencies of NOx removal as high as 99 percent have been attained with negligible emissions of NH_3.

Although flue-gas treatment cannot compete economically with combustion process modification at present it provides an option for additional control when the need for such control is dictated by severe local environmental problems.

INSTITUTIONAL CONSTRAINTS

Due to the nature of the industries involved, and because of the number, and diversity of types and modes of operation of the equipment, there are a number of constraints on the achievement of low nitrogen oxide emissions from stationary sources, particularly in retrofit operation. The costs of retrofit to meet NOx emission standards may be small compared to the value of the equipment to be modified; however, the total capital available may not be adequate to finance the unbudgeted retrofit of a network of equipment.

The scheduling of equipment outages for modifications may pose a severe constraint on the rate of retrofit in electric utilities.

Some modifications may be prepared in advance and installed during scheduled outages. Major modifications which require longer times for installation may require special outages thus putting extra demands on other equipment. In order to maintain adequate system generating capacity, these modifications would require careful scheduling, particularly if neighboring utilities from whom the required power would usually be purchased are undergoing similar outages for system modification.

The number, lifetime, and diversity of smaller stationary sources may limit the rate of changeover to low NOx operation. Units operated by a small number of users, such as stationary gas turbines for electric power generation, stationary engines for pipelines, and large industrial boilers, might be modified under similar constraints as utility boilers. But other engines and smaller boilers and furnaces are distributed among a large number of small installations. Retrofit of all units of a particular type may be difficult even if a straightforward modification is developed, since it may be impossible to locate and assure compliance of all units.

Fuel availability poses one of the most severe constraints on achieving low NOx emissions. The fuels for which the NOx emissions are generally lowest, i.e., natural gas and distillate oils, are now, and are likely to remain, in short supply. This will probably result in increased reliance on heavy, nitrogen containing oils for large commercial boilers, some industrial boilers, and stationary gas turbines with a corresponding increase in NOx emissions in the absence of controls. The large sources, utility boilers and industrial boilers, will probably use more coal, also with an increase in the uncontrolled NOx emissions. Synthetic fuels, both liquid and gas, produced from coal, which is the largest fossil fuel reserve of the U.S., and shale oil contain very high proportion of fuel nitrogen unless they undergo expensive denitrogenation (although some, but not all, of the fuel nitrogen is removed during the desulfurization of these fuels). Therefore these fuels, too, may significantly increase NOx emissions from the stationary sources which use them unless control techniques are developed and applied.

APPENDIX 15-A

CONVERSION FACTORS

NOx emissions from combustion sources are commonly reported on several bases including:

1. Lbs/10^6 BTU, where the lbs are pounds of NOx expressed as the equivalent weight of NO_2 and where BTU refers to the heating value in the fuel fired. The metric equivalent of grm/10^6 calories can be obtained by multiplying the figures for lbs/10^6 BTU by 1.8.
2. Parts per million (ppm) for a dry gas at 3 percent O_2. The conversion from ppm to lbs/10^6 BTU depends on fuel composition and heating value. For the three major fossil fuels in use to-day lbs/10^6 BTU may be converted to ppm at 3 percent O_2 using the following multiplication factors:

gas	840
oil	767
coal	714

3. Parts per million (ppm) for a dry gas at 15 percent O_2. This emission factor is favored by EPA for gas turbines. To convert from ppm at 15 percent O_2 to ppm at 3 percent O_2, multiply by 3.
4. Lbs/ton of acid for nitric acid plants. To convert to ppm at 3 percent O_2, multiply by a factor of approximately 75.

For the most part, emissions in the text were reported in lbs/10^6 BTU because the Federal emission standards for new large steam-generating plants are stated in these units. It should be noted that this method of describing emissions provides no credit for either increased thermal efficiency of a steam generating plant or increased cycle efficiency of a steam-electric cycle. It would seem appropriate that as advanced cycles are considered with potential increases in efficiency of conversion of thermal to electrical energy, emissions should be compared on the basis of electrical power output (or useful energy output) rather than on the basis of thermal input.

APPENDIX 15-B

CONTROL TECHNIQUES FOR INTERNAL COMBUSTION ENGINES

DIESEL ENGINES

The effect of fuel injection timing retard on diesel engine emissions has been determined experimentally by many investigators, and the available test data are in reasonable agreement for all diesel classes. The first few crank angle degrees retard are particularly effective in reducing NOx (McGowin 1973, Roessler et al. 1974, Schaub and Beightol 1971). For most engines, injection retard is probably limited to about 6 degrees or less because fuel consumption increases rapidly beyond that point. Typically, 6 degrees retard reduces NOx emissions by about 40 percent while increasing specific fuel consumption and carbon monoxide emissions by about 4 and 50 percent, respectively. This technique requires only minor hardware changes, or none at all, and can be incorporated in both new and existing engines. Potential problems include effects upon exhaust valve and turbocharger durability which might be adversely affected by increased exhaust gas temperatures.

Optimization of the fuel injection system may produce modest reductions (20 percent for one engine) in NOx without increasing fuel consumption. These techniques might be applied to both new engines and retrofit installations, but little information is available to make cost effectiveness evaluations.

Water injection into the intake system (water induction) is an effective method for reducing NOx emissions (McGowin 1973, Roessler et al. 1974, Schaub and Beightol 1971). Typically a water induction rate approximately equal to the fuel injection rate reduces NOx emissions by about 50 percent with very little change in fuel consumption. Water induction could be applied fairly easily to both new and existing engines, but problems of corrosion and wear of the intake system, intake valves, and water injection nozzles, and of degradation of the lubricaiton oil would have to be resolved before the technique could be applied to stationary engines. These problems might be alleviated by using distilled or demineralized water, which would increase cost, or by water injection in the form of emulsified fuel, which could cause increased fuel system corrosion.

Exhaust gas recirculation (EGR) has been shown to be a very effective NOx abatement technique, particularly where the exhaust gas is cooled before admission into the intake manifold (McGowin 1973, Roessler et al. 1974). Incorporation of 10 percent cooled EGR typically results in about 50 percent reduction in NOx emissions, 1.5 percent increase in fuel consumption, and 100 to 150 percent increase in CO and smoke (Roessler et al. 1974). Corrosion and deposit build-up in the EGR circuit, particularly in the EGR cooler; excessive engine wear; and fuel oil contamination are potential problem areas.

Reduction of the air intake temperature reduces NOx emissions by about 20 percent per 100F temperature drop, with a slight reduction in fuel consumption (Roessler et al. 1974). In turbocharged diesels, intercooling (cooling the compressed intake air) is a proven technique to increase the power output capability of the engine and to lower its specific fuel consumption. Additional improvements in engine performance and NOx emissions might be achieved by further cooling the compressed intake air. By combining turbocharging (which generally increases NOx emissions), retarded injection timing, and intercooling, NOx reductions of up to 35 percent have been demonstrated without any loss in fuel consumption compared to the equivalent naturally aspirated engine (Roessler et al. 1974).

Derating an engine by reducing engine load at rated speed may reduce NOx slightly for turbocharged, direct injection diesels, while increasing the investment cost per horsepower (McGowin 1973, Roessler et al. 1974). Combustion chamber modifications, other than divided chamber, also provide small benefits. Prechamber diesels have inherently lower emissions than comparable direct injection diesels and warrant further development.

NOx can be revoved from the exhaust gases by catalytic reduction by CO or hydrogen in the presence of low oxygen concentrations or by an added reducing agent such as natural gas or ammonia (McGowin,1973). Very high removal efficiencies have been observed. Significant development work will be required before wide scale application will be practical and the cost is likely to be relatively high (McGowin, 1973). (One estimate for a 1000 Bhp engine suggests a price of $3/Bhp).

It is difficult to assess the initial and maintenance costs for the various control techniques accurately in view of the very limited emission control work conducted to date. Incorporation of a turbocharger on a medium size diesel has been estimated to increase the cost by about 10 percent, or $2.50 to $3.00 per horsepower. The percentage cost increase in large stationary diesels is somewhat lower (Roessler et al. 1974). Addition of an intercooler would add about $0.30 to $0.50/hp while increasing the operating efficiency. It is estimated that the initial cost of a water injection system would be comparable to that of a fuel injection system, with possible additional costs for water purification or distillation equipment. The cost of a typical emission control system has been estimated at $1.25 to $3.00/hp with a related maintenance cost increase of about 10 to 15 percent.

Estimates of the operating cost of NOx reduction equipment and techniques in terms of fuel cost alone indicate that timing retard is the least cost-effective technique (Roessler et al. 1974). Water induction produces only small changes in the operating cost. Intake cooling, either alone or combined with 10 percent EGR, reduces the specific fuel consumption. Exhaust gas recirculation or increased fuel injection rate combined with timing retard may achieve moderate NOx reductions with small fuel consumption increases.

Of the techniques discussed, only fuel injection timing retard requires no hardware changes for NOx control, but it is the least cost effective technique considered. Injection timing retard combined with an optimized fuel injector improves the fuel consumption and is applicable for both new and retrofit application. Intake cooling combined with EGR, water injection, or timing retard appears to be a very attractive technique. Water injection and prechamber design diesels are other techniques which show promise for near term control.

Over the longer term, prechamber diesel engines utilizing a combination of techniques such as intake cooling, EGR, or water injection and catalytic reduction of NOx in the exhaust may yield lower NOx emissions with low specific fuel consumption (McGowin 1973, Roessler et al. 1974).

SPARK IGNITION ENGINES

Due to the efforts over recent years to develop emission control techniques for automobile engines, many NOx control strategies are well defined. A fuel rich mixture results in low NOx emissions at the expense of increased CO and hydrocarbon emissions (which can be reduced with catalytic converters or thermal reactors) and increased fuel consumption (Roessler et al. 1974). Very fuel lean combustion which would result in the lowest emissions of all three pollutants appears feasible only by means of fuel stratification or improved carburation or fuel injection. Stratified charge offers the possibility of substantially reduced emissions and of reduced fuel consumption (Jonke et al. 1971, McGowin 1973). Since application of this technique would require redesign of the engine, further development work is required.

Reducing the temperature of the fuel/air mixture results in a decrease in NOx emissions and generally, in the specific fuel consumption (McGowin et al.). Temperature reduction can be accomplished by eliminating inlet manifold heating in gasoline engines and by passing the air through an evaporative cooler in gas engines (Roessler et al. 1974). In the latter case NOx emissions would be even further reduced because of the increased moisture content of the air. Reducing the coolant temperature also results in moderate NOx reductions, e.g., 20 percent by decreasing the coolant temperature from 370 to 212F.

Water injection can reduce the NOx emissions by as much as 80 percent, but this may be accompanied by more than a 10 percent increase in fuel consumption based on tests on a larger gas engine. Results obtained on a single cylinder test engine have shown comparable reductions in NOx with a 2 percent reduction in fuel consumption suggesting that the fuel consumption of the larger engine might be improved (Roessler et al. 1974).

Exhaust gas recirculation can reduce NOx emissions by as much as much as 80 percent, but the fuel economy loss is considerable (McGowin 1973, Roessler et al. 1974). However, if only five to ten percent of the exhaust gas is recycled, advancing the spark timing or alternating the mixture fuel-air ratio can be used to minimize the loss, making 40 percent NOx reductions possible. However, as with all techniques for NOx control for spark ignition engines, the effects of the control technique on engine durability must be established before it can be applied to stationary engines, which may have a useful life of 30 years.

Increasing the engine speed at constant power results in a substantial reduction in NOx, but the effects on engine life might be significant (McGowin et al., McGowin 1973, Roessler et al. 1974). Valve timing can be modified to increase the quantity of combustion products retained in the cylinder from cycle to cycle (in effect, internal EGR) reducing NOx with little or no penalty in fuel consumption (Roessler et al. 1974).

Ignition timing retard can produce moderate reductions in NOx accompanied by a loss in engine power and fuel economy. Furthermore, at high loads, overheating or burn-out of exhaust valves may occur (McGowin et al., McGowin 1973, Roessler et al. 1974).

A meaningful assessment of the cost of emission control for large stationary gas engines has not yet been performed.

GAS TURBINE ENGINES

Emission control technology for gas turbines has been developed primarily in studies of automotive and aircraft gas turbine engines (Roessler et al. 1974) While the methods developed for mobile sources may be applicable to stationary engines, strategies for stationary gas turbine NOx control should not be limited to these techniques since stationary engines are not subject to the severe size and weight limitations of mobile engines. In addition, the problems of emission control may be complicated by the combustion of residual oils which will, according to a recent electric utility projection (NERC 1974), account for 40 percent of the fuel burned in gas turbine generating sets by 1983. The heavier fuels require increased residence times of the hot gases for complete combustion, resulting in increased thermal NO formation (Johnson and Scheifer). Further, the heavier fuels may contain significant fuel nitrogen. These problems have not been resolved in the studies to date. It is clear that fuel nitrogen may substantially increase the NOx emissions (Johnson and Scheifer), making limitations on the nitrogen content of the fuel necessary if the proposed EPA emission standards for oil combustion (75 ppm NOx at 15 percent oxygen) are to be met.

Several combustion modifications, which are applicable to present combustor designs, have achieved modest NOx reductions. These include air-blast or air assist fuel atomization (which can reduce the fuel droplet size by more than 50 percent), leaving the primary zone, quenching the flame early in the combustor, and internal flow recirculation (Roessler et al. 1974). NOx reductions achievable using these techniques are small, but the costs are low.

Water or steam injection has been shown to be a very effective technique for NOx control, capable of meeting current standards for stationary gas turbine NOx emissions (Dibelius et al. 1970, Hilt and Johnson 1971, Klapatch and Koblish, Roessler et al. 1974, Shaw 1974b, Singh et al. 1971, Winship and Brodeur 1972). Siting flexibility is somewhat reduced by the requirements for water in quantities comparable the fuel supply. Furthermore, to avoid detrimental effects on turbine durability the water has to be purified to a maximum of 2 to 5 ppm of dissolved solids. The added costs of the water injection system (6 to 10 percent of the baseline cost of a large gas turbine), water treatment, and the water (5 to 7 percent of the baseline operating cost) must be considered in the application of this technique (Roessler et al, 1974). For small gas turbines (less than 2 Mw) the costs may be prohibitive. A decrease in the cycle efficiency is observed with water injection since the heat of vaporization is not recovered in the cycle. Steam injection increases the mass flow without additional turbine work, increasing the thermal efficiency. NOx reductions of 50 to 75 percent have been achieved with water injection with only slight increases in the CO emissions (Roessler et al. 1974). Water injection is currently being used by some utilities without observed deleterious effects as long as the water purity is maintained at a high level.

Several advanced combustor designs have been proposed which may meet the NOx standards with "dry" (no water injection) operation (Roessler et al. 1974). Internal or external recirculation combined with fuel prevaporization and lean primary zone operation has demonstrated this potential (Roessler et al. 1974). Surface combustion, in which combustion occurs in close proximity to a cooled solid surface, i.e., either a porous bed through which the prevaporization fuel and air are injected or a catalyst bed, looks promising for long term emission control. These advanced methods will require considerable research and development before they are applied to stationary gas turbines.

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