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# **POSITION PAPER ON REGULATION OF ATMOSPHERIC SULFATES**

**U.S. ENVIRONMENTAL PROTECTION AGENCY**

**Office of Air and Waste Management**

**Office of Air Quality Planning and Standards**

**Research Triangle Park, North Carolina 27711**



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Strategies and Air Standards Division

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Depicted on the cover is the 16th Century symbol for vitriol, a term used to represent sulfuric acid and certain other sulfates.

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# TABLE OF CONTENTS

Page

List of Figures . . . . .	v
List of Tables . . . . .	vi
Executive Summary . . . . .	vii
Introduction . . . . .	1
1. Review of Current Knowledge . . . . .	2
1.1 Health and Welfare Effects . . . . .	3
1.1.1 Toxicological Results . . . . .	3
1.1.2 Epidemiological Results . . . . .	6
1.1.3 Welfare Effects and Acid Rain . . . . .	11
1.2 Emission and Concentration Patterns . . . . .	13
1.3 Atmospheric Chemistry and Transport . . . . .	20
1.3.1 Formation and Mechanisms for Removal . . . . .	22
1.3.2 Relationship Between Sulfur Dioxide and Sulfate Trends . . . . .	29
1.3.3 Long Range Transport . . . . .	35
1.4 Control Alternatives . . . . .	41
1.4.1 Precursor Control . . . . .	41
1.4.2 Alternatives for SO <sub>2</sub> Emission Control . . . . .	44
1.4.3 Use of Naturally Clean Fuels . . . . .	45
1.4.4 Flue Gas Desulfurization (FGD) Technology . . . . .	47
1.4.5 Fuel Pretreatment . . . . .	47
1.4.6 Alternative Coal Combustion Systems . . . . .	49
1.4.7 Intermittent Control Systems . . . . .	50
1.4.8 Employment of New Energy Sources . . . . .	51
1.4.9 Energy Conservation . . . . .	52
1.4.10 Applicability and Availability . . . . .	53
1.5 Information Gaps and Research Needs for Sulfate Regulation . . . . .	53
1.5.1 Monitoring . . . . .	56
1.5.2 Health and Welfare Effects . . . . .	56
1.5.3 Atmospheric Chemistry and Transport . . . . .	57
1.5.4 Improve Control Technologies for Sulfur Dioxide and Sulfates . . . . .	59
2. Potential Regulatory Strategies . . . . .	60
2.1 Potential Scope of the Problem . . . . .	61
2.2 Regulatory Options Under the Clean Air Act . . . . .	63
2.2.1 National Ambient Air Quality Standards . . . . .	63
2.2.2 New Source Performance Standards . . . . .	65
2.2.3 National Emission Standards for Hazardous Pollutants . . . . .	67
2.2.4 Emission Standards for Mobile Sources . . . . .	67
2.2.5 Other Clean Air Act Options . . . . .	67
2.2.6 Alternate Regulatory Options . . . . .	69
2.2.6.1 Regional Emission Control . . . . .	69
2.2.6.2 Emissions Tax as Component of Sulfates Control Strategy . . . . .	69
3. Policy Implications . . . . .	71
References . . . . .	81

## FIGURES

	<u>Page</u>
Figure 1. Rainfall Acidity 1965-1966 . . . . .	12
Figure 2. Projected Nationwide Sulfur Oxides Emission Trends, 1950-1990 . . . . .	15
Figure 3. Fossil Steam Generating Capacity, 1970 . . . . .	16
Figure 4. Nationwide Geographic Variation in Annual SO <sub>2</sub> Emission Density . . . . .	17
Figure 5. 1970 Annual Urban Sulfate Concentrations . . . . .	19
Figure 6. 1970 Annual Nonurban Average Sulfate Concentration . .	19
Figure 7. NASN Sulfate Data, 1972, 1973 . . . . .	21
Figure 8. Bimodal Distribution of Atmospheric Particles . . . . .	26
Figure 9. Velocity of Deposition for Unit Density Aerosol as a Function of Diameter . . . . .	28
Figure 10. NASN Urban and Nonurban Sulfate Trends . . . . .	31
Figure 11. NASN Urban Sulfur Dioxide Trends . . . . .	31
Figure 12. Nonurban Sulfate Trends in the Northeast (12 Sites). . .	36
Figure 13. Nonurban Sulfate Trends in the Northeast (8 Sites) . . .	36
Figure 14. Aircraft Measurements of Sulfates and SO <sub>2</sub> Concentrations . . . . .	39
Figure 15. Forty-eight-hour Back Trajectories for Air Parcels Arriving during Sampling shown in Figure 14 . . . . .	39
Figure 16. Maximum Availability of SO <sub>2</sub> Control Technologies . . . .	55
Figure 17. 24-State Region with High Sulfate Levels . . . . .	72
Figure 18. Projected SO <sub>x</sub> Emissions from 24-State Region, 1970-1985 . . . . .	79

## LIST OF TABLES

	<u>Page</u>
Table 1. $\text{SO}_x$ Compounds Toxic to Man and/or Lower Animals. . . . .	3
Table 2.    Results of Epidemiological Studies . . . . .	10
Table 3.    U. S. Manmade $\text{SO}_2$ Emissions, 1972 . . . . .	14
Table 4.    Mechanisms that Convert Sulfur Dioxide to Sulfates . . . . .	23
Table 5.    Comparison of Sulfur Dioxide and Sulfate Trends for Selected Urban and Nonurban Sites. . . . .	33
Table 6.    Environmental Inventory of Sulfate Precursor Agents: Preliminary View . . . . .	42
Table 7.    Summary of Control Alternatives for $\text{SO}_2$ . . . . .	46
Table 8.    Summary Description of Major Flue Gas Desulfuri- zation Process . . . . .	48
Table 9.    Electric Energy Conservation Savings, 1980 . . . . .	52
Table 10.   Partial Summary of Power Plant Technical Control Options - Removal and Energy Efficiency, Costs, Timing . . . . .	54
Table 11.   Diffusion Model Sensitivity of Sulfate Formation to $\text{SO}_2$ Oxidation Rate for 1500-MW Power Plant . . . . .	62
Table 12.   Summary of Assumptions Used for $\text{SO}_x$ Emissions Projections of Table 13 . . . . .	76
Table 13.   24-State Region $\text{SO}_x$ Emissions . . . . .	76

## EXECUTIVE SUMMARY

The suspected adverse health effects of atmospheric suspended sulfates\* have been of growing concern to the Environmental Protection Agency (EPA). The sulfate issue is beginning to have a significant influence on EPA policies and programs that affect levels of sulfur oxides emissions: the power plant intermittent emission control policy, the power plant tall stack emissions policy, the energy-related program for conversion of power plants from oil or gas to coal operation, the automotive emissions control program, and general policy regarding the need for additional sulfur oxides regulation. In view of the importance of these current regulatory efforts and the potential need for a general sulfate control program, EPA has conducted an extensive review of currently available information regarding sulfates. Also, the National Academy of Sciences (NAS), at the request of the Senate Public Works Committee, and the EPA Science Advisory Board (SAB), at the request of the EPA Administrator, have recently completed independent studies pertaining to sulfates. This report summarizes current scientific and technical information concerning sulfates, and identifies needs for research and development. The report also discusses the implications of our current knowledge for present and long-term regulatory control of sulfur oxides, and presents and evaluates a policy for sulfates.

### Health and Welfare Effects

Health-related research indicates that the transformation products of sulfur dioxide ( $\text{SO}_2$ ) in ambient air, principally sulfates, are more likely than sulfur dioxide alone to be responsible for many of the adverse health effects typically associated with sulfur oxides. Toxicological (animal) studies provide evidence that  $\text{SO}_2$ , in the absence of other pollutants such as ozone or particulates, is a mild respiratory

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\*"Sulfates," as used in this report, is defined by the measurement method used in health studies and the National Air Surveillance Network, i.e., material collected on a high-volume sampler filter and analyzed as water soluble sulfates. These can include acid-sulfates (e.g., sulfuric acid, ammonium bisulfate), neutral metallic sulfates, adsorbed  $\text{SO}_2$ , and sulfites.



irritant, while certain specific sulfate compounds, especially submicron-sized sulfuric acid aerosol, are more severe respiratory irritants. Epidemiological studies\* conducted in several U.S. cities suggest that high daily or annual sulfate levels are associated with increased attack frequency in asthmatics, worsened symptoms in cardio-pulmonary patients, decreased ventilatory function in school children, and symptoms of acute and chronic respiratory diseases in children and adults. The association of these health indicators with sulfates was stronger than that for  $\text{SO}_2$ . Accurate quantification of effects levels\*\* must await future research. When viewed together, the results of the toxicological and epidemiological studies suggest that specific sulfate compounds may also be responsible for the observed excess mortality associated with high  $\text{SO}_2$  concentrations. However, an association between sulfates and mortality has not yet been tested by field studies.

Considerable research is necessary before the complex relationship between sulfates and health effects can be well understood. The health effects associated with given sulfate levels can be expected to vary with the chemical form and physical size of sulfates, the presence of other pollutants, temperature, and other environmental factors. As both the NAS and SAB reports state, due to the inadequacy of present monitoring capabilities and the current incomplete understanding of the influence of other variables on the sulfates/health relationship, the preliminary sulfate health effects studies should only be considered as indicative of the potential health impact of sulfates. Until more specific information regarding the sulfates/health effects relationship is available, EPA considers total sulfate measurements to be an imperfect but useful indicator of the presence of the toxic sulfate components. Although cautioning that such measurements will contain varying proportions of toxic and relatively non-toxic sulfate compounds, the NAS reports appears to support the EPA position. However, the SAB considers

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\*Principally conducted through the EPA Community Health and Environmental Surveillance System program (CHESS).

\*\*Best judgement sulfate levels tentatively associated with adverse health effects in the preliminary epidemiological studies were as low as 6 to 10  $\mu\text{g}/\text{m}^3$  (24 hour average) and 10 to 15  $\mu\text{g}/\text{m}^3$  (annual) average.

measurements of water soluble sulfate alone to be unreliable indicators of the presence of toxic components in what they term the "sulfur oxides/particulate complex," and, instead, would view high concentrations of total suspended particulate and/or  $\text{SO}_2$  as better indicators of areas for concern with respect to the health impact of acid sulfates and other toxic sulfate components. These divergent opinions can be resolved only by further monitoring and health effects research.

Economic welfare effects associated with sulfates are ecological and agricultural damage, materials damage, and visibility degradation. Sulfates appear to be a major factor in producing acid rain in a large portion of the Eastern U.S. Research into the significance of these effects is limited and, for the most part, such effects cannot now be quantitatively related to levels of sulfates in the air.

#### Emission and Concentration Distribution

Emissions of sulfur compounds, both natural and manmade, are the principal source of atmospheric sulfates. Although manmade  $\text{SO}_2$  emissions represent only half of the total sulfur emissions in the Northern hemisphere, these manmade emissions are concentrated in the relatively small industrialized areas where they far outweigh natural production. Power plant emissions, which currently account for about 55% of manmade  $\text{SO}_2$  emissions in the U. S., have been a rapidly growing component of the  $\text{SO}_2$  emission complex. While total manmade emissions increased by 45% between 1960 and 1970, power plant emissions increased by 90%.

Currently, about 1% of sulfur oxides emissions come from automobiles, emitted predominantly as  $\text{SO}_2$ . Although the contribution to total sulfur oxides loading is not likely to change, emissions from new cars equipped with the catalytic converter are partially in the form of highly toxic submicron sulfuric acid aerosol. Since automotive emissions do not contribute significantly to current sulfate levels and originate from mobile sources, control strategies differ from those for stationary sources. The automotive sulfuric acid issue is not considered in this discussion of the more general sulfate problem.

Based on National Air Surveillance Network data, a large portion of the eastern United States has recorded sulfate concentrations significantly higher than concentrations generally observed in other sections of the country. Urban levels range from 10 to 24 micrograms per cubic meter ( $\mu\text{g}/\text{m}^3$ ) and nonurban levels range from 8 to 14  $\mu\text{g}/\text{m}^3$  (annual average) in a 24-state region east of the Mississippi, roughly bounded by Illinois and Massachusetts to the north and Tennessee and North Carolina to the south. In this 24-state region, the 1972 average of nonurban concentrations exceeded 10  $\mu\text{g}/\text{m}^3$  (annual average) with an urban concentration average of about 13.6  $\mu\text{g}/\text{m}^3$ . The high sulfate levels in the 24-state area appear to be spatially correlated with high  $\text{SO}_2$  emission density, high rainfall acidity patterns, and a high density of power plant locations. The remainder of the country does not exhibit similar sulfate concentrations on a regional scale. The 1972 urban average outside the 24-state northeastern region was 7.9  $\mu\text{g}/\text{m}^3$ ; whereas, the nonurban annual average was 4.4  $\mu\text{g}/\text{m}^3$ . There are some areas, however, such as the Southern California Coastal Basin, in which high sulfate levels are observed. The 11 stations in this area measured an annual average sulfate concentration of 11.1  $\mu\text{g}/\text{m}^3$ . Similarly, Tampa, Florida, recorded an annual sulfate level of 11.9  $\mu\text{g}/\text{m}^3$  in 1972, although nonurban stations in this region averaged 4.9  $\mu\text{g}/\text{m}^3$  for the same period. Thus, while these areas do not exhibit the regional concentration problems characteristic to the northeastern U.S., they do have high local sulfate concentrations.

#### Atmospheric Chemistry and Transport

Sulfur dioxide is oxidized to sulfuric acid and other sulfates by several mechanisms, most involving reactive agents such as photochemical smog, ammonia, catalytic metals, and fine particulates. Temperature and humidity also influence the reaction. These agents can complicate the relationship between  $\text{SO}_2$  and sulfates; for example, reductions or increases in  $\text{SO}_2$  concentrations may not result in proportional reductions or increases in sulfate levels because of the presence of other agents that affect the formation reaction. Inadequate knowledge concerning formation mechanisms currently precludes quantitative assessment of catalytic agent influences.

regional sulfate increases that, on balance, offset the local decreases. This explanation is supported by the apparent increase in manmade sulfates at a limited number of eastern nonurban sites for which data are available. This increase roughly parallels the increase in overall  $\text{SO}_2$  emissions during that time. Although, in aggregate, urban sulfate levels showed little change, variable trends were observed for different cities. Variations in both the spatial distribution of sulfur oxides emissions and in atmospheric chemistry could affect the relative magnitude of local versus imported sulfates and account for the variable trends for individual cities.

Both EPA and NAS consider the above stated hypotheses plausible; however, the SAB suggests that the impact of nonurban sources on urban areas is likely to be minor and prefers to explain the observed trends in terms of precursor-limited sulfate formation mechanisms, primary emissions of sulfates, and errors in the measurement methods. The SAB bases their conclusions regarding transport on the assumption that dilution and removal would reduce sulfates to negligible levels during transport and not on the evidence of any transport models or measurements. In the opinion of the SAB, the available evidence does not substantiate the validity of their assumptions or any of the explanations offered. These divergent views underscore the need for additional research.

Despite the uncertainties concerning the relationship between  $\text{SO}_2$  emissions and ambient sulfate concentrations, EPA believes that the available evidence suggests that further increases in  $\text{SO}_2$  emissions are likely to produce increases in regional sulfate levels. Sulfate increases are not likely to be proportional to the total  $\text{SO}_2$  emissions increase because of the spatial distribution of the important sources and the complex formation mechanisms.

### Control Alternatives

Although control of catalytic agents such as particulates and oxidants may eventually prove to be an important component of a sulfate control strategy, an examination of current emission trends and pollutant formation information indicates that control of atmospheric sulfates will probably depend primarily on control of  $\text{SO}_2$  emissions. A variety of systems and

concepts for SO<sub>2</sub> control that are applicable through 1990 have been examined to provide better definition of research and development objectives. The primary SO<sub>2</sub> control technologies for sulfate control and their applicability are discussed:

1. Naturally occurring low sulfur fuels will continue to be important in controlling SO<sub>2</sub> emissions, but are not likely to provide a total solution to the problem of SO<sub>2</sub> and sulfates control due to their limited supply, at least in the near-term.
2. Clean fuel allocation and redistribution, fuel switching, intermittent control, and tall stacks may contribute to achieving present SO<sub>2</sub> standards; however, the regional transport theory suggests that these methods would have minor impact on preventing increases in regional atmospheric sulfate levels.
3. Flue gas desulfurization will probably be the principal large stationary source control technology available for at least the next 10 years. Physical coal desulfurization can also contribute to sulfur oxides control during this same time interval.
4. Advanced fuel pretreatment technologies (liquefaction, gasification) have a significant potential for reducing SO<sub>2</sub> emissions, especially for small point and area sources, but will probably not have a major impact before 1985.
5. Ultimately, alternate energy supply systems (solar energy, thermonuclear fusion), improved combustion technologies (fluidized bed combustion), and general improvements in energy utilization efficiency should provide more effective use of energy resources with less environmental degradation.

#### Information Gaps and Research Needs for Sulfate Regulation

As evidenced by the previous discussion of health effects and transport mechanisms, considerable uncertainty exists in interpreting the limited scientific data base on sulfates. Both the SAB and the NAS place a high priority on the initiation of comprehensive research programs that are needed before major control strategy decisions can be made.

An EPA analysis of major research needs indicates that development of the data and information necessary for a sulfate regulatory program would require 3 to 5 years. In this regard, if EPA were to set a National Ambient Air Quality Standard (NAAQS) for sulfates, it could not realistically be proposed before 1980 or 1981. Important research needs are summarized below:

#### Monitoring

A critical path in the research effort is the need to develop advanced monitoring methods. Important components of the epidemiological, toxicological, and atmospheric formation studies depend on the characterization of sulfates by such monitoring methods. The development of these techniques is expected to take 1 to 3 years, thus extending the completion of some aspects of all research areas.

#### Health and Welfare Effects

Appropriate dose-response curves should be developed. The chemical composition and physical characteristics of the harmful sulfur compounds should be identified. The program to accomplish this will include toxicological work, clinical studies of human response to specific sulfates, and continued epidemiologic studies. Sulfuric acid and other specific sulfate monitors are needed for more precise quantification of exposures. Dose-response functions are needed to assess the effects of atmospheric sulfates on ecological systems and structural materials. These effects include economic losses to materials and crops caused by sulfates and/or acid rain.

#### Physical/Chemical Transformation and Transport

The oxidation rates of  $\text{SO}_2$  to sulfate in ambient air as well as in power plant and ground-level urban plumes must be determined. The role of precursors and catalysts in sulfate formation should be further assessed. Models must be developed to estimate both local concentrations and long-range transport phenomena for sulfates. Research in this area is critical to the development of control strategies.

#### Improved Control Technologies for Sulfur Dioxide and Sulfates

If stringent control of large  $\text{SO}_2$  point sources is required as part of a sulfate control strategy, current  $\text{SO}_2$  scrubbing technologies

may not have adequate removal efficiencies. Demonstration projects designed to achieve greater than 90% removal efficiency, particularly for new sources, will be evaluated. Also, further development of alternative combustion technologies, such as fluidized bed systems, is needed.

### Potential Regulatory Strategies

Although the information required to determine and support a control program for sulfates may not be available for several years, a preliminary analysis of regulatory options has been made to aid planning and to help focus research and development programs.

Regulatory approaches currently available under the Clean Air Act (CAA) were examined to assess their potential utility in a sulfate control program. Options such as hazardous standards (CAA Section 112), emergency powers (CAA Section 303), and abatement conferences (CAA Section 115) do not appear appropriate as primary approaches for regulating sulfates because of their inherently limited scope.

Sulfates exhibit those characteristics\* identified in the Clean Air Act for pollutants to be controlled through National Ambient Air Quality Standards (NAAQS). Despite the legal rationale for this approach, potential implementation problems exist. NAAQS implementation generally has required the use of a pollutant emissions/ambient air quality relationship to develop emission regulations that apply within currently established Air Quality Control Regions. Due to the complex sulfate formation mechanisms and the apparent pollutant transport through multistate regions, such a relationship may be impossible to develop. Another problem with the NAAQS approach would be the definition of a sulfate "threshold" level below which sulfate-associated health effects are insignificant.

Although New Source Performance Standards (NSPS) can be a valuable tool for ensuring minimal emissions of SO<sub>2</sub> from new major sources, this approach may not provide adequate control to deal fully with the sulfate problem.

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\*The pollutant must have "an adverse effect on health and welfare" and result from emissions from "numerous or diverse" sources.

Since long-term implementation difficulties are possible with an NAAQS and resulting state implementation plans, alternative regulatory options must be explored over the next several years. These options include regional emission limitations for  $\text{SO}_2$ , increased control of sulfate precursors, and economic incentives.

#### Policy Implications

In view of the available data, it is the judgment of EPA that an air quality standard or other major regulatory program for sulfates is not supportable at this time. Additional research is needed in order to fill the information gaps described earlier. The EPA research effort will focus on improving monitoring capability to permit identification of particle size and chemical form of toxic sulfates, developing more comprehensive health effects data, and characterizing the long range transport and transformation mechanisms. This research program will require several years to complete; consequently, it is doubtful that a comprehensive regulatory program specifically for sulfates could be initiated before the end of the decade.

Nevertheless, until further research makes a comprehensive regulatory program possible, EPA must respond to the potential sulfate problem suggested by the preliminary sulfate/health effects information cited earlier. Although considerable uncertainty exists concerning the relationship between measured ambient sulfate concentrations and adverse health effects, the preliminary health effects information can be useful in identifying areas of potential health concern.

As described previously, a large portion of the northeastern United States is experiencing relatively high annual sulfate concentrations. Nonurban concentrations have averaged in excess of  $9 \mu\text{g}/\text{m}^3$  throughout this 24-state region. The average of urban concentrations has been about  $13 \mu\text{g}/\text{m}^3$  (annual average). As mentioned earlier, this area of high sulfate concentrations correlates spatially with high  $\text{SO}_2$  emission density, high rainfall acidity patterns, and a high density of power plant locations. Furthermore, the region exhibits widespread violation of the national primary ambient air quality standard for suspended



particulate matter, a potential precursor agent in the formation of sulfates. High sulfate concentrations have also been observed in several isolated urban areas through the remainder of the country.

Given the significant potential for sulfate-related health risk due to the multiple influence of high sulfate concentrations, high precursor concentrations, and high SO<sub>2</sub> emission density, prudence dictates that EPA adopt a policy of avoiding aggravation of existing conditions by minimizing further increases in the relatively high sulfate levels in the northeastern United States and other more localized problem areas. In addition, close attention must be paid to sulfate trends in areas of lower sulfate concentrations. Although this goal of avoiding sulfate increases will primarily be achieved by minimizing SO<sub>2</sub> emission increases, existing programs for control of pollutants such as oxidants and particulates may provide some measure of sulfate control by limiting sulfate formation processes.

Opinions differ over where, and to what extent, SO<sub>2</sub> emissions should be limited to adequately address the potential sulfate problem. There is general agreement that SO<sub>2</sub> emissions increases should be avoided in or near urban areas where ambient concentrations of sulfates, SO<sub>2</sub>, or total suspended particulates are high. The NAS placed high priority on abating SO<sub>2</sub> emissions from sources located in or near urban areas with high concentrations of sulfur dioxide and sulfates. In addition, the NAS is concerned about the effects of area-wide increases in SO<sub>2</sub> emissions on regional sulfate levels. The SAB, however, does not share the same degree of concern for the impact of area-wide SO<sub>2</sub> emissions. Rather, the SAB suggests that increased SO<sub>2</sub> and other sulfate precursor emissions may have primarily a local impact on sulfate formation, and "that increases in exposure to sulfur oxides or particulates in localities where the sulfur dioxide and/or total suspended particulates exceed primary standards should be viewed with grave concern."

EPA considers the points addressed in both reports as important and essentially compatible with the Agency's assessments. Current efforts to attain the primary standards for the criteria pollutants are responsive to the SAB's concerns. In addition, EPA believes that the area-wide

concern must be addressed by minimizing increases in SO<sub>2</sub> in the areas of maximum sulfate impact. A strategy of minimizing regional and local increases in SO<sub>2</sub> emissions can be implemented through existing regulatory options such as State Implementation Plans (SIPs) and New Source Performance Standards (NSPS).

A policy of minimizing SO<sub>2</sub> emission increases is generally consistent with other Agency policies previously announced. These policies include the Clean Fuels Policy, the limited application of intermittent control systems (ICS), and the significant risk aspect of oil-to-coal conversions.

The EPA Clean Fuels Policy is intended to make it unnecessary for plants to switch to lower sulfur fuels to comply with state regulations where such compliance is not needed for attainment and maintenance of the national health-related standards for SO<sub>2</sub>. By revising SIPs appropriately, plants currently in areas meeting primary air quality standards could continue to burn currently available fuels; no switch to higher sulfur fuel is intended. Therefore, sulfur emissions from these sources should not increase.

With respect to EPA policy on intermittent control systems (ICS), a limited number of isolated power plants may be permitted to use intermittent emission control to meet air quality standards, temporarily deferring expenditures for costly continuous emission controls. Eligible plants are already burning coal and, under ICS, will continue to burn existing fuel except during adverse meteorological conditions, at which point they will reduce emissions by switching to a lower sulfur fuel or shifting generation load. Again, total sulfur emissions from these sources should not increase and may actually be slightly reduced.

The Energy Supply and Environmental Coordination Act of 1974 (ESECA) provides the Federal Energy Administration with the authority to prohibit a power plant from burning oil or natural gas subject to certification by EPA of the plant's ability to burn coal in compliance with certain environmental requirements. Two requirements relate to a plant's ability to burn coal without contributing to a violation of primary standards for total suspended particulate or sulfur dioxide. In this regard, if certain legal criteria are satisfied (regional limitation),

EPA may specify alternate emission requirements (primary standard conditions) to be met temporarily by converting plants. An additional requirement of ESECA states that conversions cannot result in an increase in the emission of unregulated pollutants or pollutant precursors to levels that may result in a significant risk to public health. Based on currently available health effects information, EPA has decided to apply this "significant risk" provision only with respect to sulfates. Under the significant risk provision, EPA plans to restrict emissions of sulfate precursor pollutants--sulfur dioxide or particulate matter -- if a converting plant is located in an area with high sulfate concentrations and with concentrations of particulate or sulfur dioxide in excess of primary standards.

Though currently available information does not now permit the establishment of a comprehensive sulfate regulatory program, the information does suggest a need to minimize increases in regional sulfur oxides emissions as a means of preventing increased levels of atmospheric sulfates. In the interim, prior to the initiation of any comprehensive control program, existing regulatory options can be effective in limiting increases in sulfate concentrations. The "significant risk" policy for converting power plants, the vigorous enforcement of state implementation plans for the control of sulfur dioxide and particulates, and the increasing application of new source performance standards to power generating facilities are vital components of an overall strategy that should limit growth of ambient sulfate levels. EPA analysis indicates that these regulations and policies should prevent major SO<sub>2</sub> emission increases through 1980 in the regions of maximum sulfate impact. Until information is available to support the enforcement of a more rigorous sulfate regulatory program, the use of these currently applicable regulatory measures should provide reasonable protection against increased health risk from sulfates.

# POSITION PAPER ON REGULATION OF ATMOSPHERIC SULFATES

## INTRODUCTION

Atmospheric suspended sulfates have been of growing concern to the Environmental Protection Agency (EPA) in recent months for a number of reasons. CHESS\* epidemiological results of 1970-71 gave preliminary indications that health effects previously associated with sulfur dioxide ( $\text{SO}_2$ ) and particulates were related more strongly to sulfate concentrations. Other studies, including toxicological work, have also indicated that sulfuric acid aerosol ( $\text{H}_2\text{SO}_4$ ) and certain other sulfates are stronger irritants than sulfur dioxide. Recently published precipitation studies suggest that the acidity of United States rainfall (which may be related to acid sulfate formation) is at levels that may produce serious ecological consequences. Although current control strategies have been successful in reducing ambient concentrations of  $\text{SO}_2$  and particulates in urban areas, sulfate levels in some nonurban areas appear to have increased between 1962 and 1972, and if sulfur dioxide emissions were to increase due to the projected rise in fossil fuel consumption, there is concern that sulfate levels in urban areas could increase. Thus, EPA concerns about permanent use of tall stacks and Intermittent Control Systems (ICS) have been predicated, in part, on the conviction that widespread use of these measures probably would not prevent increases in sulfate levels. This concern also influences EPA policy regarding

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\*EPA's Community Health and Environmental Surveillance System.

the energy-related program for conversion of power plants from oil or gas to coal combustion. In addition, information concerning the emission of sulfuric acid aerosols from automotive catalysts has prompted alterations in the automotive emissions control program.

Although the scientific understanding of health and welfare effects, atmospheric chemistry, and transport of sulfates is incomplete, a preliminary assessment of possible control strategies is needed so that a coherent Agency approach to the sulfate problem can be developed. Recognizing the importance of these issues, the EPA Administrator requested an external review by the Science Advisory Board (SAB)<sup>1</sup> of the scientific and technical issues relating to sulfates. In addition, the National Academy of Sciences (NAS),<sup>2</sup> has prepared a comprehensive review largely devoted to the sulfate question. This report will provide a brief review of the scientific information developed to date, discuss technical and regulatory alternatives for sulfate control, and present and evaluate a policy for sulfates.

Unless otherwise specified, "sulfates", as used in this report, are defined by the measurement method used in the preliminary health studies and the National Air Surveillance Network. The method measures material that is collected on glass fiber filters of high-volume samplers and analyzed as water-soluble sulfate. This can include sulfuric acid, soluble sulfate salts, adsorbed sulfur dioxide, sulfite salts, and sulfates formed due to sampling and analytical artifacts.

## 1. REVIEW OF CURRENT KNOWLEDGE

It is not within the scope of this report to present a complete review of the scientific and technical information regarding health effects, atmospheric chemistry and transport, monitoring, and control of sulfur oxides. Several comprehensive reviews of these subjects have been published recently.<sup>1-4</sup> This section summarizes the major areas relevant to development of control strategies and policy. An earlier draft has been reviewed by members of the Science Advisory Board, and their comments have been considered in development of this version. There seems to be general agreement among EPA, SAB, and the NAS report with the interpretations presented here, although some exceptions are noted in the body of the report.

## 1.1 Health and Welfare Effects

The current Air Quality Standard for  $\text{SO}_2$  is based upon studies summarized in the sulfur oxides criteria document.<sup>5</sup> Recent reviews<sup>1,3,6</sup> of the scientific literature indicate that studies conducted since the criteria document was published have tended to confirm the association of adverse human health effects with ambient  $\text{SO}_2$  concentrations at or near the primary standard ( $80 \mu\text{g}/\text{m}^3$  annual average,  $365 \mu\text{g}/\text{m}^3$  24-hr average). Although  $\text{SO}_2$  may cause health effects in combination with ozone and/or respirable particles, it has long been suspected that the transformation products of  $\text{SO}_2$  may have more significant health effects than  $\text{SO}_2$  alone. Studies of polluted air have demonstrated that sulfur dioxide undergoes atmospheric oxidation leading to the formation of sulfuric acid aerosol and other fine-particulate sulfates.<sup>3</sup> Recent evidence from both toxicological and epidemiological studies suggests that these compounds are more likely than sulfur dioxide alone to be responsible for observed adverse human health effects.

### 1.1.1 Toxicological Results

A variety of toxicologic studies of sulfur oxide inhalation have been conducted on lower animals and, to a limited extent, on humans. Most of the studies have dealt with the effects of  $\text{SO}_2$ . Only a few investigations of sulfuric acid ( $\text{H}_2\text{SO}_4$ ) and inorganic particulate sulfates have been conducted. The sulfur oxides that have been investigated are shown in Table 1. Most studies of sulfur oxides have used acute respiratory

Table 1.  $\text{SO}_x$  COMPOUNDS TOXIC TO MAN AND/OR LOWER ANIMALS

- 
- |    |  |
|----|--|
| 1. | Sulfur dioxide   |
| 2. | Specific sulfate aerosols (decreasing irritant toxicity) <sup>8</sup>  |
|    | sulfuric acid, zinc ammonium sulfate, ferric sulfate, zinc sulfate, ammonium sulfate, ammonium bisulfate, cupric sulfate, ferrous sulfate, manganese sulfate |
| 3. | Pollutant mixtures   |
|    | e.g. sulfur dioxide/ozone, sulfur dioxide/sodium chloride aerosol, sulfur dioxide/vanadium aerosol   |
-

physiologic and pathologic effects as indices of response. A few biochemical studies and one investigation of the cocarcinogenic potential of sulfur dioxide have been conducted. These studies have been extensively reviewed by Amdur et al.<sup>6</sup> and Goldstein.<sup>7</sup> The results of the toxicologic studies indicate that sulfur dioxide, by itself, is a mild respiratory irritant; whereas, sulfuric acid aerosol and certain specific inorganic sulfates are greater respiratory irritants.

The extent and type of biological response to exposure to sulfur oxides are complex due to this interaction of many parameters. The particle size, physico-chemical composition, and presence of interacting factors as well as mass concentration, are of paramount importance as factors that affect both the site of deposition of sulfur oxides in the respiratory tract and the magnitude of the response. Sulfur dioxide is thought to be a mild irritant because, as a gas, most of it is absorbed in the nose or mouth and very little penetrates into the lung. By contrast, the reason that  $H_2SO_4$  and several other particulate sulfur oxides are greater respiratory irritants is thought to be because they are intrinsically more toxic and/or because, as small particles, they are deposited deeper in the respiratory tract than gaseous  $SO_2$ . By a similar mechanism, the response to  $SO_2$  is enhanced when it can be adsorbed onto such small particles. The effective size is apparently important even for particles generally considered to be in the respirable range. Sulfuric acid aerosol of  $0.7\ \mu m$  size produces a four-fold greater irritant response than  $2.5\ \mu m$  particles of the same compound.<sup>8</sup> A similar size/response relationship exists for fine particulate zinc ammonium sulfate aerosols.<sup>6</sup>

The specific chemical composition of particulate sulfur/oxide may have an important bearing on its toxicity. One line of investigation examined respiratory physiologic responses to a variety of sulfates of similar aerosol size and mass concentrations.  $H_2SO_4$  was found to be the greatest respiratory irritant, followed by metallic and ammonium-containing sulfates in the order listed in Table 1. Although copper, ferrous, and manganese sulfates were the least toxic of the compounds tested, they were found to enhance irritant response when mixed with  $SO_2$ .<sup>8</sup> The differences in the inhalation response of some of these sulfates was

small. Although these data do not constitute an adequate basis for a determination of the comparative toxicity for specific inorganic sulfates, the data do suggest that the toxicological evaluation of particulate sulfur oxides must consider the cation as well as the anion of the molecule, and that aerosol acidity is of great importance.

These studies were based upon a sensitive respiratory physiologic response, primarily increased pulmonary airflow resistance in guinea pigs. This response results from narrowing of the airways within the respiratory system. A similar response has been observed in men exposed to sulfur dioxide and  $\text{H}_2\text{SO}_4$  aerosol. This physiological response is a generally accepted, sensitive measure of airway irritation.

A crucial question arises as to the pathophysiologic significance of subtle increases in pulmonary airflow resistance. A recent investigation in guinea pigs<sup>9</sup> demonstrated that the total respiratory deposition rate of inhaled particles and the pattern of regional respiratory deposition of these particles was altered by  $\text{H}_2\text{SO}_4$  inhalation. These effects were noted at  $\text{H}_2\text{SO}_4$  concentrations as low as  $30 \mu\text{g}/\text{m}^3$ , particle size  $< 1 \mu\text{m}$ , for 1 hour. This response was probably associated with increased pulmonary airflow resistance. Increased pulmonary airflow resistance is the principal physiologic response in uncomplicated asthma. It has been hypothesized, therefore, that  $\text{H}_2\text{SO}_4$  inhalation may act to increase the incidence of asthma attacks through increased deposition of inhaled particles and/or a shift in the principal site of deposition of inhaled particles to airway regions where asthma can be triggered. On a qualitative basis, the toxicologic data support the epidemiologic evidence of a better correlation of health responses with particulate sulfur oxides than with gaseous sulfur dioxide.

Experimental investigations have not yet dealt adequately with the contribution of covariate factors on the health response to sulfur oxides. McDilton et al.<sup>10</sup> have recently shown that high relative humidity is important in potentiating the physiologic response of guinea pigs to  $\text{SO}_2$ -NaCl (sodium chloride) mixtures. High relative humidity ( $\sim 80\%$ ) served to deliquesce the NaCl particles, resulting in particle growth. An acid aerosol developed, apparently through  $\text{SO}_2$  adsorption on the droplets.



Analysis indicated that the aerosol may have contained acid sulfite. Under these circumstances, a physiologic response was elicited which did not occur at low (~40%) relative humidity. Mixtures of SO<sub>2</sub> and ozone also produce greater response than either pollutant alone. It is thought that the reaction of those gases in the respiratory tract to produce sulfuric acid may be responsible for this synergism.<sup>7</sup> These examples demonstrate the need not only for further investigation of the effects of specific sulfates, but also for studies of the biologic response to gas-aerosol mixtures under conditions that can better simulate the complex physico-chemical environment to which mankind is exposed.

In summary, much of the limited toxicological work supports the widely held contention that the neutralized sulfate ion is not innately toxic, and that the observed associations between health effects and sulfates and SO<sub>2</sub> are more likely to be caused by small particulate acid sulfates and possibly sulfites. They also highlight the need for physical and chemical characterization of the material currently measured as particulate sulfates.

#### 1.1.2 Epidemiological Results

Epidemiological studies of air pollution give evidence of associations that must be replicated under a variety of conditions to be accepted as cause effect relationships. The goal of such studies is to provide data indicative of dose-response relationships. However, since these are field studies, it is often very difficult to distinguish the effects of a specific variable from the multitude of other environmental factors. Unfortunately, dose measurements in the majority of epidemiological studies involving sulfur oxides have been of total suspended particles and sulfur dioxide. No studies have included measurements of respirable-sized sulfuric acid or other specific sulfate particles. In addition, assessment of response, whether acute (resulting from daily variations in pollutants) or chronic (resulting from long term exposures), is difficult.

Although epidemiological work to date is not yet sufficient to provide accurate dose-response relationships, recent studies (primarily CHESS) have been conducted using water soluble sulfate measurements as an indicator of acid sulfates and other potentially toxic sulfur oxides.

The studies were preliminary in nature and interpretation of the results is hampered by a number of factors that are discussed by the authors<sup>9</sup> as well as reviewers.<sup>1,3,6</sup> Shortcomings existed in determination of pollutant exposures which, in long term studies, had to be based on historical monitoring data of uncertain quality; in some cases, past sulfate levels were imputed from available SO<sub>2</sub> and particulate data. Furthermore, as noted above, water soluble sulfate measurements had to be used as a proxy for fine particulate acid sulfates. The studies differed in locale and response rate of study subjects, and the methods of ascertainment of illness used by CHESS differ from those used by other investigators. These difficulties are being corrected in current and future replicative studies. Reviewers such as the NAS<sup>2</sup> and the SAB<sup>1</sup> consider the CHESS studies to be of significant value and agree that, taken in total, they point out the potentially important role of certain sulfates in air-pollution-related health effects. The preliminary studies suggest that human respiratory disease is more closely associated with relatively low levels of sulfates than with sulfur dioxide or total suspended particulate concentrations. Adverse effects observed included an increase in attack frequency in asthmatics, worsening of symptoms in cardio-pulmonary patients, and decrements in ventilatory function in school children. In addition, studies, in some cases using reconstructed past pollutant trends, indicate increased incidence of acute and chronic respiratory diseases in children and adults.

Daily reports from two panels of asthmatics, one in New York and one in the Salt Lake Basin, and from a panel of cardiopulmonary subjects in New York<sup>11</sup> were used in studies of daily exposures of sulfates. For asthmatics, the frequency of attacks was analyzed; the response for the cardiopulmonary panel was aggravation of one or more specific symptoms. In the New York asthma study, there was a 9 percent increase in risk of attack associated with 24-hour sulfate levels of 8.1 to 10.0 µg/m<sup>3</sup> when the minimum temperature ranged between 30 and 50°F, and a 10 percent increase in risk with 24-hour levels greater than 10 µg/m<sup>3</sup> when the minimum temperature exceeded 50°F. In the Salt Lake study, a slight decrease in risk was noted with increased sulfate levels between

6 and 10  $\mu\text{g}/\text{m}^3$  when temperatures ranged between 30 and 50°F. However, when temperatures exceeded 50°F, a 17 percent increase in attack risk was associated with 24-hour average sulfate levels between 6.1 and 8.0  $\mu\text{g}/\text{m}^3$ . The New York cardiopulmonary study indicated a worsening of symptoms, such as shortness of breath, cough, and increased production of phlegm, was associated with sulfates in the 24-hour average concentration range of 6 to 10  $\mu\text{g}/\text{m}^3$ . Total suspended particulates were also correlated with aggravation of symptoms.

Studies of school children<sup>11</sup> in a moderately polluted industrial valley of Cincinnati indicated that ventilatory function was significantly lower than that of children living in a clean area of the metropolitan region. Differences in sulfate levels were found to be closely associated with differences in ventilatory response. A similar study in New York indicated that older children (9 to 13 years) who had been exposed to elevated levels of sulfur oxides and particulates in their early years had decreased ventilatory function relative to unexposed children. No consistent effect was noted for younger children (5 to 8 years).

Other studies<sup>11</sup> observing effects of long-term exposures involved reconstructing some past sulfate exposures from emission and/or other pollutant data. These studies are suggestive of a relationship between sulfates and (1) excess acute respiratory disease in children and families, and (2) excess risk for chronic bronchitis in adults. The authors suggest that these effects may occur as a result of long-term (up to 10 years) exposures to sulfates at annual average levels of 10 to 15  $\mu\text{g}/\text{m}^3$ .

An earlier study by Dohan<sup>12</sup> found a significant association between mean annual ambient sulfate concentration and numbers of absences caused by illness for female employees of a large, multicity U.S. corporation. Sulfate concentrations in the four areas with the highest illness absence rates were 13.2 - 19.8  $\mu\text{g}/\text{m}^3$  annual average.

Although no studies of pollutant relationships with excess mortality have involved direct sulfate measurements, a significant association between ambient sulfur dioxide levels in the presence of particulates and mortality has been noted in several studies.<sup>5,7,13</sup> Since some evidence suggests that effects linked with sulfur dioxide are more

closely associated with sulfates, Finklea et al.<sup>14</sup> have suggested that increased mortality may be associated with high sulfate levels. Studies relating mortality to actual sulfate measurements are needed to confirm this suggestion.

The results of the preliminary CHESS epidemiological studies are summarized in Table 2. Concentration ranges at or above which adverse effects were noted are listed. The difference between 24-hour and annual average times should be noted. A location that experiences several days a year in which 24-hour sulfate levels exceed  $10 \mu\text{g}/\text{m}^3$  is likely to have an annual average considerably below  $10 \mu\text{g}/\text{m}^3$ . This may be of great importance in development of possible future control strategies, which must be directed toward the lowest level at which health effects are observed.

Because health effects can be expected to vary depending upon the chemical form and particle size of sulfates, and upon synergisms with other pollutants such as acid nitrates, levels of total sulfates at which effects are observed might be expected to vary for different cities, regions, or changing atmospheric conditions within given regions. Quantification of the effects of these variables is not now possible. Therefore, the numbers presented in Table 2 are only indicative of the relative magnitude of the concentrations that are likely to be associated with adverse effects. As pointed out by NAS<sup>2</sup>, these first approximations are subject to considerable uncertainty.

In summary, the CHESS studies show that sulfates may be a better indicator of sulfur oxide pollution than  $\text{SO}_2$  and point out that sulfates may play an important role in the causation of air-pollution-related health effects. Until improved monitoring capabilities and further health research permit more accurate dose-response functions, EPA believes that the consistency of the preliminary studies justifies consideration of total sulfate measurements as an imperfect but useful indicator of the presence of acid sulfates and other toxic sulfur-containing components. It is recognized that in some cases, the bulk of specific sulfates collected may not be of concern to health. The NAS report<sup>2</sup> appears to support use of sulfates as an indicator, cautioning that such measurements will contain changing

Table 2. RESULTS OF EPIDEMIOLOGICAL STUDIES<sup>11,12,14</sup>

Adverse health effect	Concentration <sup>a</sup> at which effect was observed		Averaging time
	SO <sub>2</sub> , μg/m <sup>3</sup> (ppm)	Sulfates, μg/m <sup>3</sup>	
Increased mortality	300-400 (0.11-0.15)	NA <sup>b</sup>	24 hr
Aggravation of symptoms in elderly	365 (0.14)	8-10	24 hr
Aggravation of asthma	180-250 (0.07-0.09)	6-10	24 hr
Decreased lung function in children	220 (0.075)	11	Annual mean
Increased acute lower respiratory disease in families	90-100 (0.034-0.037)	9	Annual mean
Increased prevalence of chronic bronchitis	95 (0.035)	14	Annual mean
Increased acute respiratory disease in families	106 (0.039)	15	Annual mean
Increased respiratory disease related illness absences in female workers	NA <sup>b</sup>	13	Annual mean
Primary standard	365	-	24 hr
Primary standard	80	-	Annual mean

<sup>a</sup>Effects levels are best judgment estimates based on a synthesis of several studies.

<sup>b</sup>NA = not available.

amounts of components with variable toxicities. The likelihood of variable composition has caused the SAB<sup>1</sup> to consider such measurements as unreliable indicators of the toxic components in what they term the "sulfur oxides/particulate complex." The SAB considers elevated levels of total suspended particulates and/or SO<sub>2</sub> indicative of areas of concern about acid sulfates and the other toxic components. Further monitoring and health effects research is urgently needed to clear up these uncertainties. Important areas for further research are discussed below in Section 1.5

### 1.1.3 Welfare Effects and Acid Rain

Sulfate-related welfare effects include ecological damage, materials damage, visibility deterioration, and possible climatological alterations. Of principal concern is the suspected role of sulfur oxides in causing acid rain. Acid rain, observed in regions of high sulfate concentrations in both Europe and the United States, can lower the pH of soils and natural waters, cause mineral leaching, and damage vegetation.<sup>15-18</sup> Although the effects are not always deleterious, the potential exists for serious ecological disruption in some areas of the country. A recent study<sup>15</sup> suggests that acid precipitation and dry deposition of acid aerosols and gases may be causing depletion of fish populations in lakes in the Adirondack Mountains of New York.<sup>18</sup> Figure 1 illustrates the pattern of acid rainfall over the Eastern U.S. in 1965-66. The pH of normal rainfall is about 5.7. Rainfall of pH 4.7 is ten times more acidic because the scale is logarithmic. Values as low as pH 2.1 have been reported in individual storms.<sup>15</sup>

In a comprehensive review of the literature on sulfates and acid rain, Nisbet<sup>19</sup> indicates that both the acidity of precipitation and the rate of deposition of sulfates in precipitation has increased in the northeastern U.S. in recent years. The distribution of acid rainfall is closely related to observed ambient levels of suspended sulfates. Acid rain, and dry deposition of acid-sulfate aerosols, as well as SO<sub>2</sub> in combination with particulates can cause materials damage. The role of sulfates in forming acid rain must be considered in relation to other possible acid formers such as nitrates. Nisbet<sup>19</sup> estimates that nitrates account for about 24% of observed acidity.

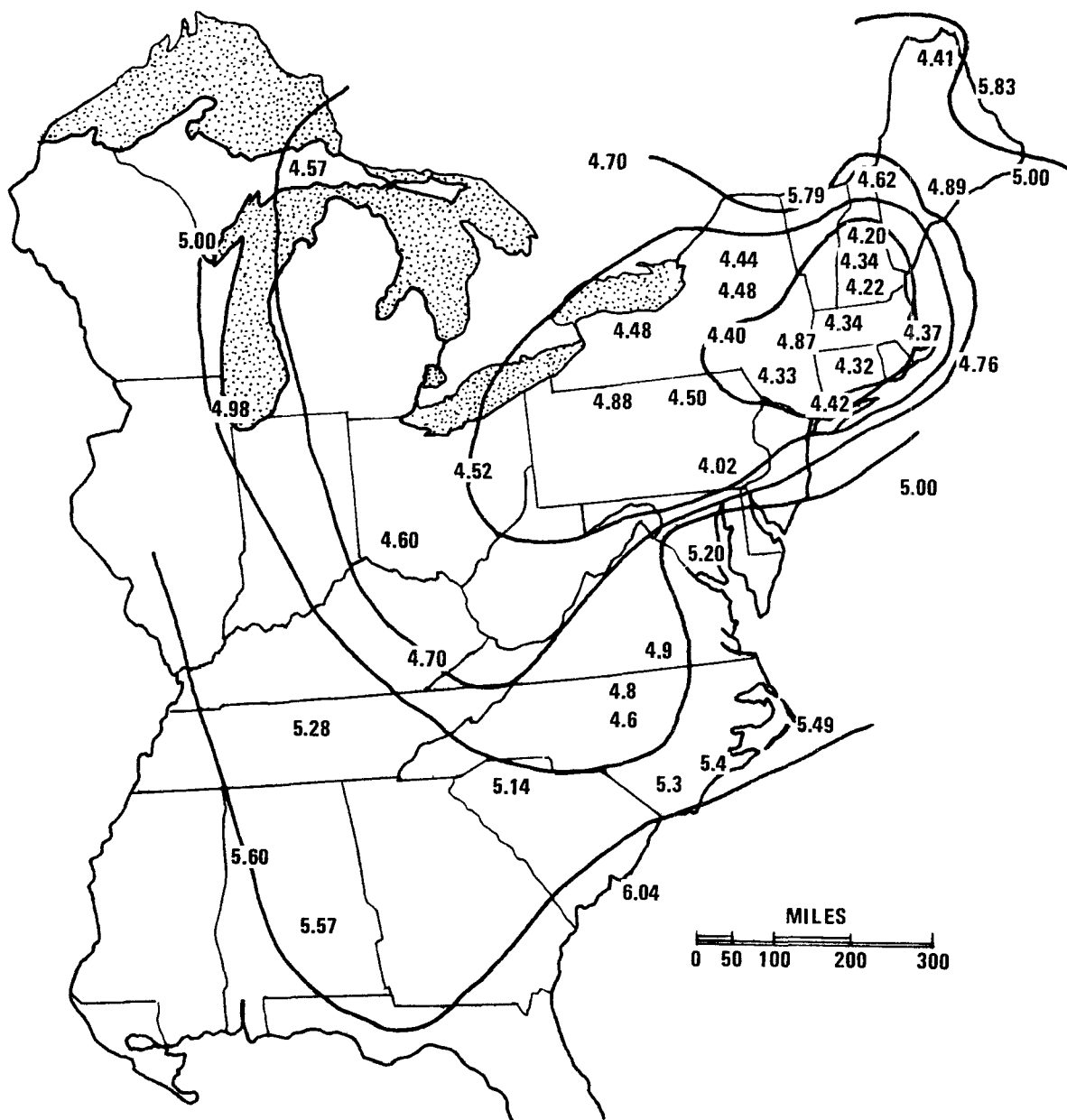


Figure 1. Rainfall acidity, 1965-1966 (pH units). 16

Sulfates, as a major component of atmospheric fine particulate material, have a significant role in visibility reduction.<sup>20</sup> Because some sulfates have a water absorbing capacity, their impact on visibility is greatly increased at high humidities. Widespread atmospheric hazes observed throughout the eastern U.S. are apparently increasing with SO<sub>2</sub> emissions.<sup>21</sup> Sulfates may comprise two-thirds of all fine particles in this section of the country.<sup>22</sup> Existing information, however, does not permit adequate quantification of these effects, and considerable work is needed to provide damage estimates.

## 1.2 Emission and Concentration Distributions

Most sulfates are not emitted directly, but are secondarily formed from other sulfur compounds. In the Northern Hemisphere approximately half of the total annual sulfur emissions are from widely dispersed natural sources, principally biogenic decomposition processes and sea salt spray.<sup>23</sup> Anthropogenic contributions, however, are concentrated in nations of high industrial density such as the United States, where they can far outweigh the natural impact.<sup>23,24</sup> Approximately 95 percent of the total pollution related sulfur emissions is in the form of sulfur dioxide, most of the remainder being hydrogen sulfide, sulfur trioxide, and a small amount of directly emitted sulfates. Hydrogen sulfide oxidizes to sulfur dioxide, ultimately forming sulfate, and sulfur trioxide rapidly reacts with water vapor to form sulfuric acid. When describing source distributions for sulfates, it is necessary to present emissions patterns for sulfur dioxide.

A breakdown of 1972 United States manmade sulfur oxide emissions expressed as SO<sub>2</sub> is presented in Table 3. Significantly, 55 percent of the total emissions in 1972 were from electrical production in coal-fired and oil-fired power plants and occurred largely in the eastern half of the country. Figure 2 shows past and projected trends in total and power plant SO<sub>2</sub> emissions, and it can be seen that most of the 45% increase in total SO<sub>2</sub> emissions shown in Figure 2 for 1960 to 1970 was accounted for by the increase in power plant emissions. Figures 3 and 4 illustrate the geographical distribution of power plant generating



Table 3. U. S. MANMADE SO<sub>2</sub> EMISSIONS, 1972<sup>a</sup>

Source	Emissions, 10 <sup>6</sup> tons	% of total emissions
Stationary fuel combustion	25.1	77
Electric utilities <sup>b</sup>	18.0	55
Coal	16.5	
Oil	1.5	
Industrial/commercial (point sources)	3.6	11
Coal	2.5	
Oil	1.0	
Other	0.1	
Area sources	3.5	11
Industrial processes	6.8	21
Transportation	0.6	1.8
Automotive	0.2	0.6
Other	0.4	1.2
Solid waste	0.1	0.3
Miscellaneous	0.1	0.3
Total, all sources	32.7	100

<sup>a</sup>NEDS data.<sup>25</sup>

<sup>b</sup>SASD data file.

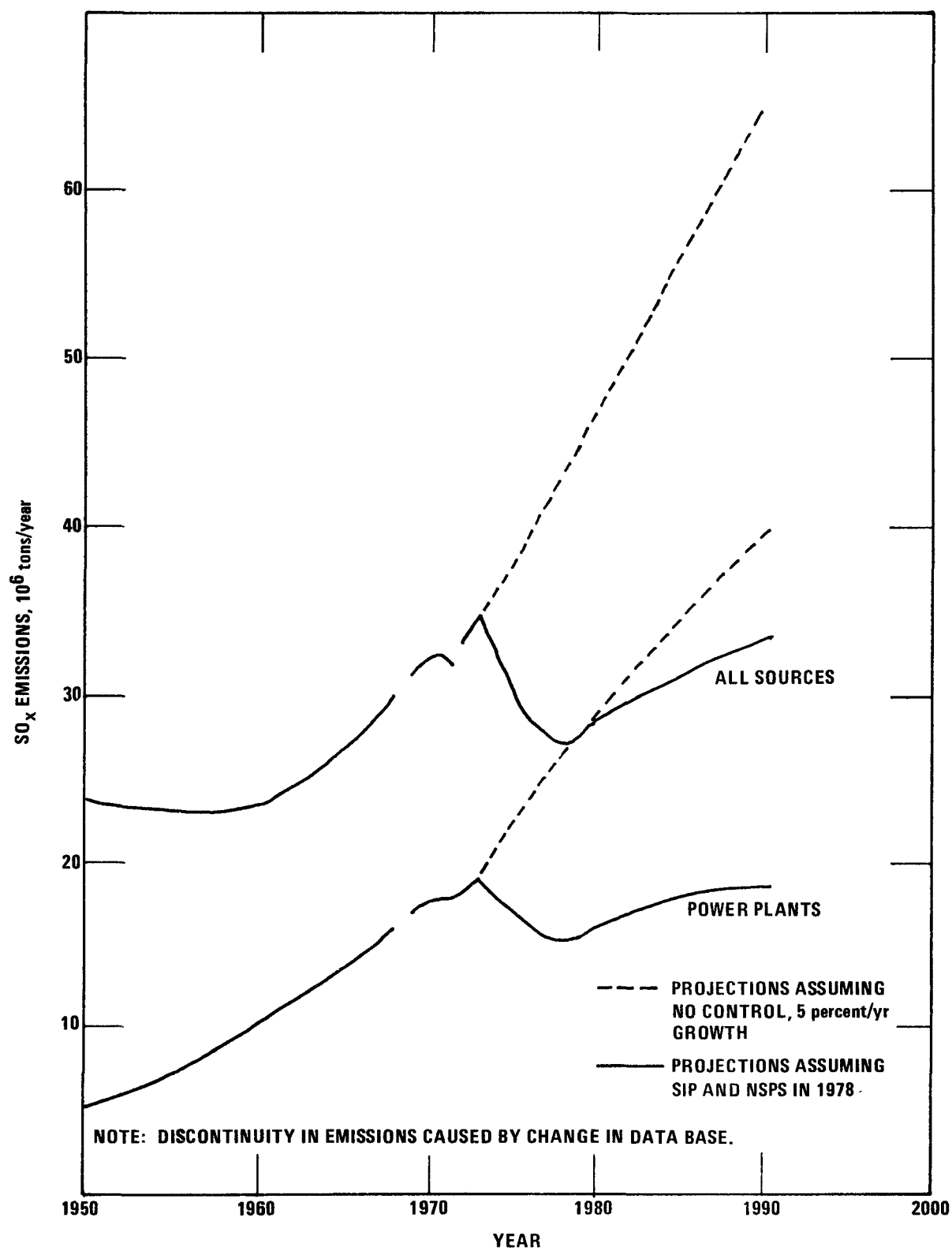


Figure 2. Projected nationwide sulfur oxides emission trends, 1950-1990<sup>14,25</sup>

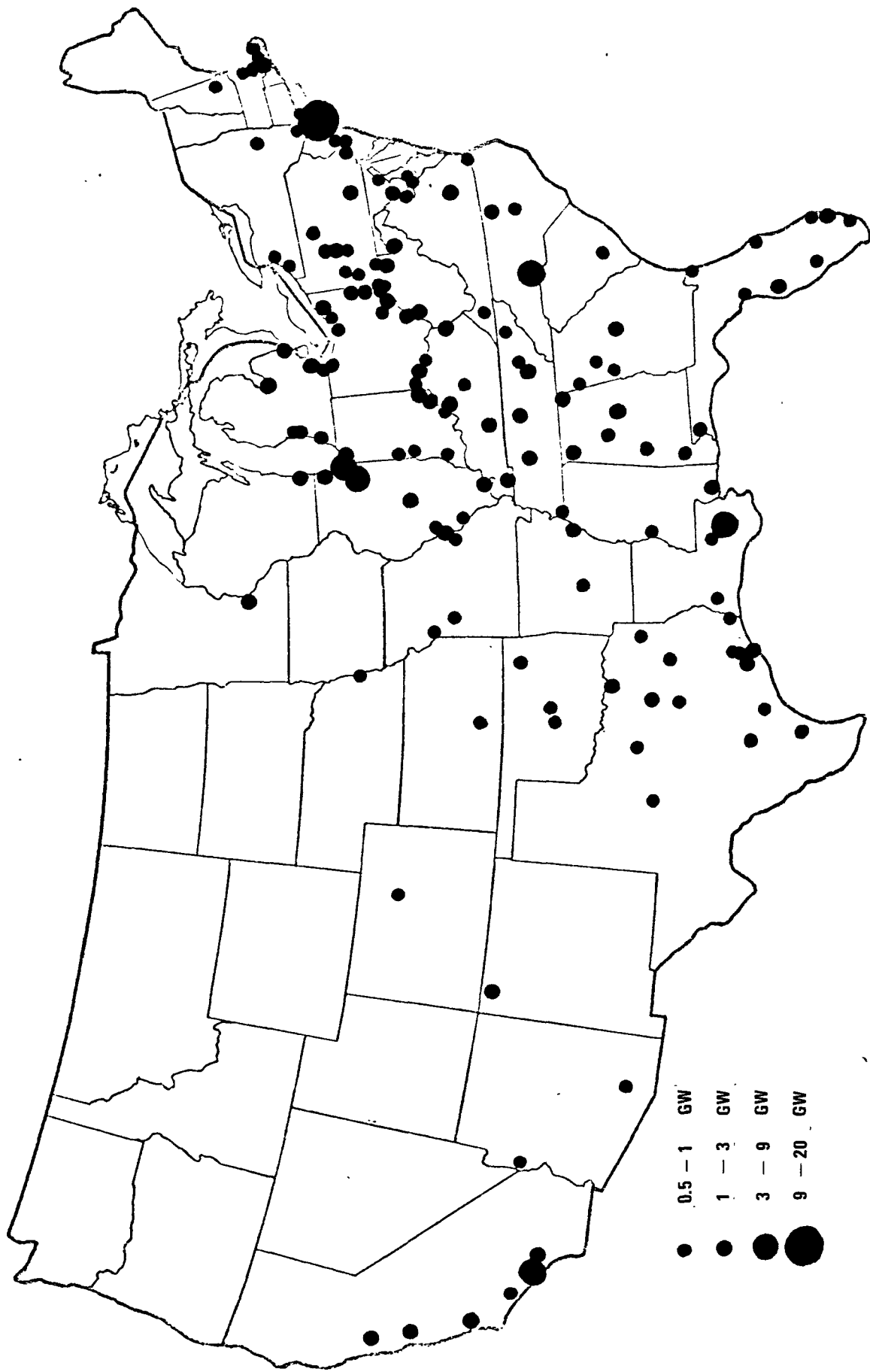


Figure 3. Fossil steam generating capacity, 1970.26

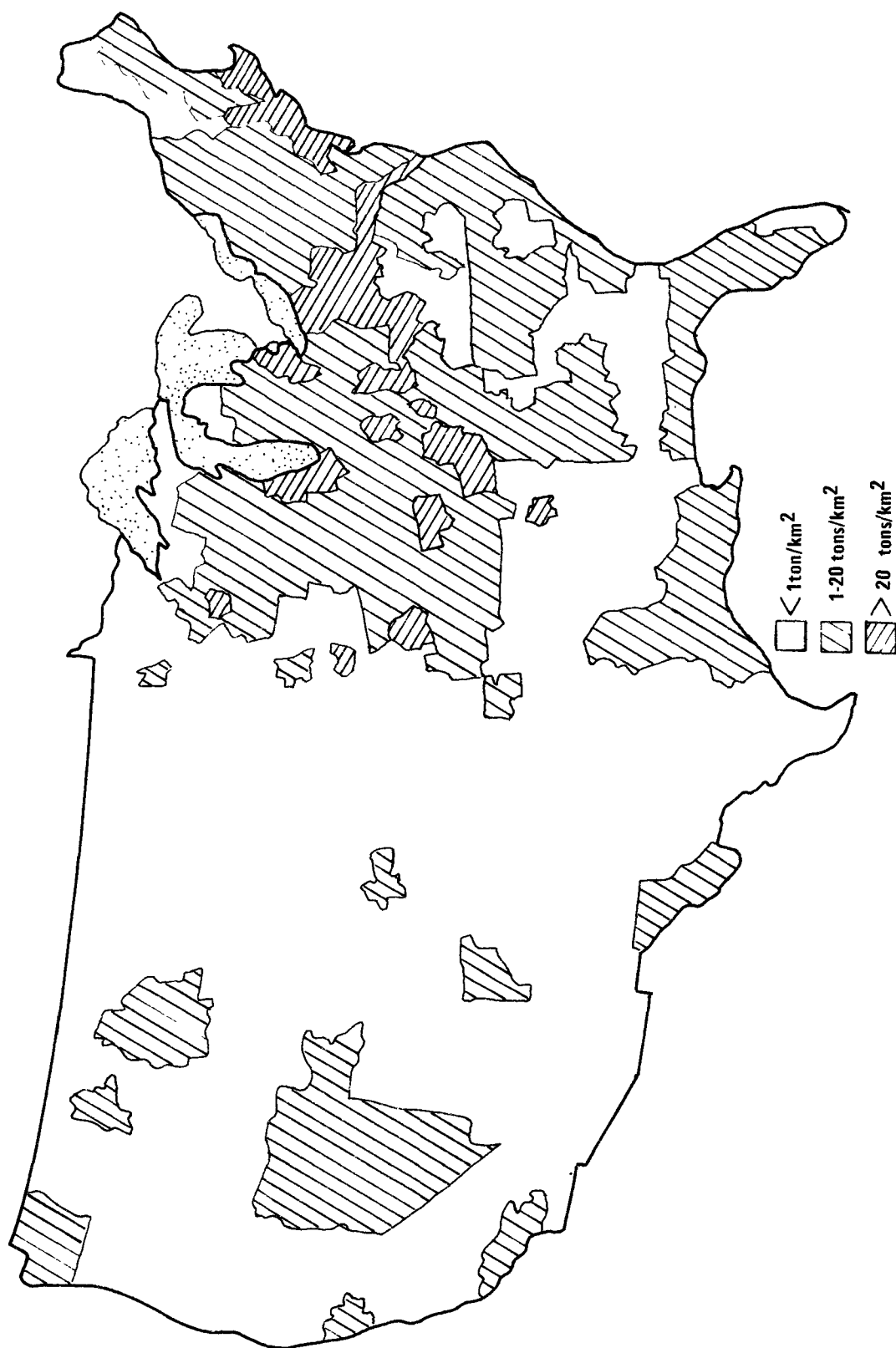


Figure 4. Nationwide geographic variation in annual SO<sub>2</sub> emission density.<sup>27</sup>

capacity and of total  $\text{SO}_2$  emission density, respectively. It is apparent that power plants account for a large share of  $\text{SO}_2$  emissions and are concentrated in the northeast quadrant. Many of the industrial process emissions of  $\text{SO}_2$  result from the smelting of sulfur-bearing ores at plants located in the western portion of the nation.

Although automotive sources are responsible for only about 1 percent of total sulfur emissions, they may become increasingly important with the introduction of catalytic converters. These devices will cause some of the sulfur in gasoline that was formerly emitted as  $\text{SO}_2$  to be directly emitted as fine particulate sulfuric acid.<sup>28</sup> Because automotive emissions are not currently an important component of urban sulfate levels and because both technical and regulatory control strategies for vehicles must differ from those for stationary sources, the automotive sulfate issue will not be examined in any detail in this report.

The best available data base on national ambient concentrations of water-soluble sulfates in the United States is provided by the National Air Surveillance Network (NASN). Approximately 250 monitoring sites are distributed in both urban and nonurban locations across the country. Urban sites were placed in center city areas; nonurban sites were chosen to be representative of general background areas in their respective regions. These sites monitor total suspended particulates once every 2 weeks. The samples are later analyzed for water soluble sulfates. This is the major available data base from which to examine the extent of spatial/temporal distributions of suspended sulfate levels and to determine the degree of correlation between measured ambient sulfate concentrations and its major precursor,  $\text{SO}_2$ . Because urban sites are usually located in center city areas, little information is obtainable for micro scale analysis of metropolitan areas.

Figures 5 and 6 illustrate the urban and nonurban annual average sulfate concentrations for 1970 as measured by NASN. The available data are limited and represent approximately 25 samples taken at each of 164 urban and 25 nonurban sites. As such, the figures can be considered as being only indicative of the actual spatial concentration distributions. Throughout the eastern portion of the country, nonurban concentrations

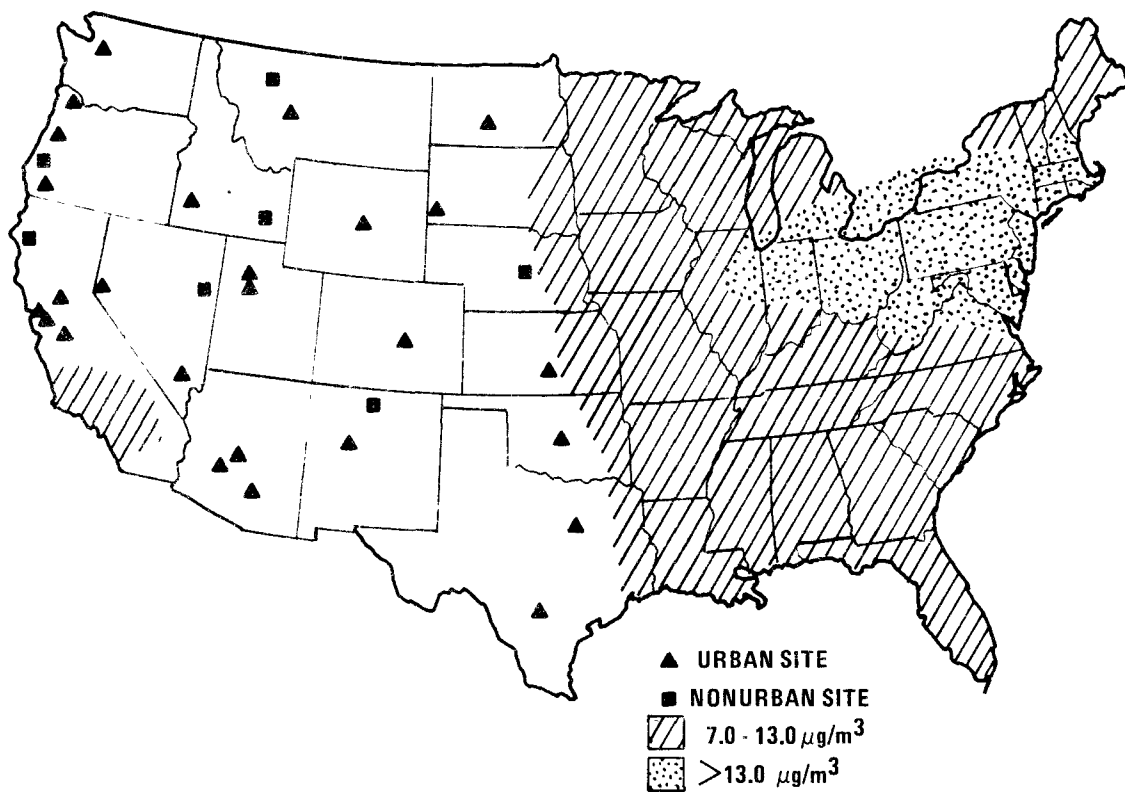


Figure 5. 1970 annual urban sulfate concentrations.<sup>14</sup>

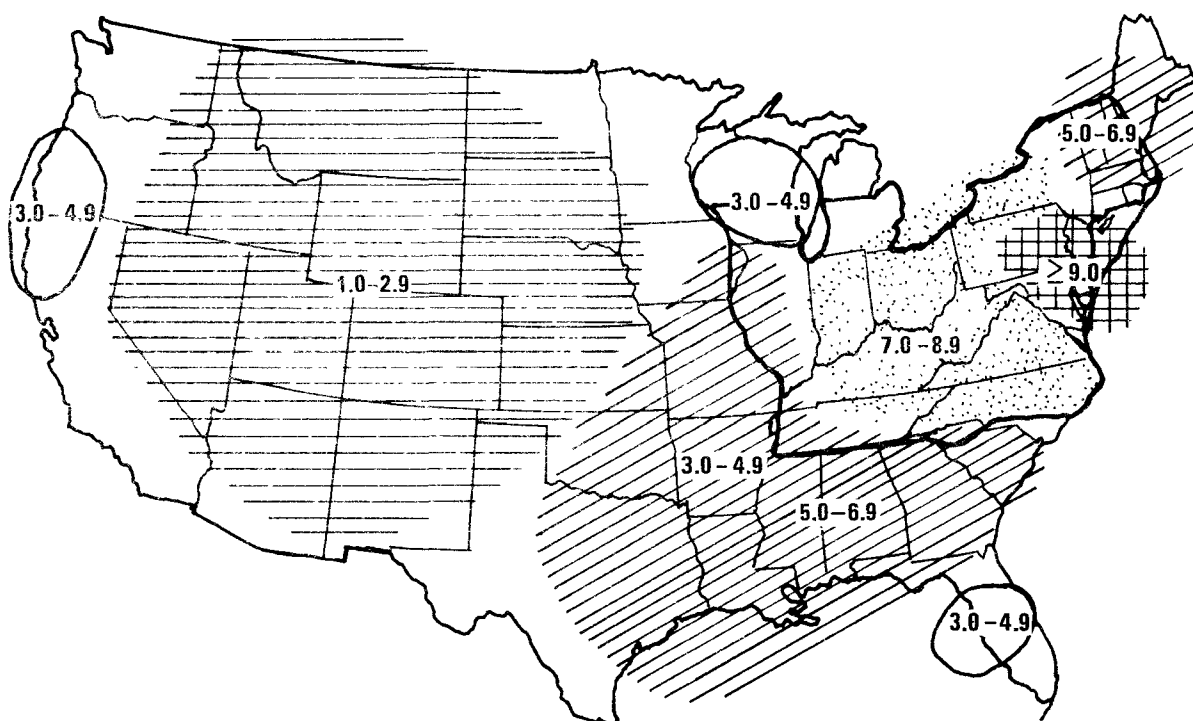


Figure 6. 1970 annual nonurban average sulfate concentrations.<sup>14</sup>

reached high levels and only in portions the west were they so low as to approximate the global background of 1 to 2  $\mu\text{g}/\text{m}^3$  (annual average). Both figures point out the distinct differences between the sulfate levels in various regions of the country, with a 24-state region (outlined in Figure 6) east of the Mississippi and north of South Carolina having significantly higher concentrations at both urban and nonurban sites. In this 24-state region, urban levels ranged from 10 to 27  $\mu\text{g}/\text{m}^3$  and at nonurban sites levels generally exceeded 7  $\mu\text{g}/\text{m}^3$ , averaging 9  $\mu\text{g}/\text{m}^3$  (annual average). In the remainder of the country, annual concentrations averaged about 6  $\mu\text{g}/\text{m}^3$  for urban stations and about 4  $\mu\text{g}/\text{m}^3$  for non-urban locations.

The distribution of sulfate levels in 1972 indicates that the general patterns have not changed greatly since 1970. In 1972, concentrations at the urban sites in the 24-state region averaged 13.6  $\mu\text{g}/\text{m}^3$ , and the average at the nonurban sites was 10.2  $\mu\text{g}/\text{m}^3$ . Outside of this region, urban levels averaged 7.9  $\mu\text{g}/\text{m}^3$  and nonurban levels 4.4  $\mu\text{g}/\text{m}^3$  in 1972. Representative sulfate data for 1972 as well as available 1973 data are presented in Figure 7. As shown in the figure, although concentrations are highest in the eastern section of the nation, some locations outside this region, such as Southern California, also exhibit high concentrations.

Comparison of Figures 5 and 6 with  $\text{SO}_2$  emission density patterns (Figure 4), and power plant locations (Figure 3), indicates that observed distribution of high sulfate concentrations are geographically related to  $\text{SO}_2$  emissions distribution. These patterns as well as complementary evidence for acid rain (Figure 1) and incidence of atmospheric hazes suggest that sulfate levels are higher than natural background throughout the northeast quadrant of the nation.

### 1.3 Atmospheric Chemistry, Trends, and Transport

Once emitted, sulfur compounds are ultimately removed from the atmosphere, primarily by precipitation and dry deposition on the ground and on vegetation. Most of the sulfur dioxide is converted to sulfate, either before or during the removal process.<sup>4</sup> Scavenging rates for  $\text{SO}_2$  and sulfates are not well quantified, but global mass balance estimates of

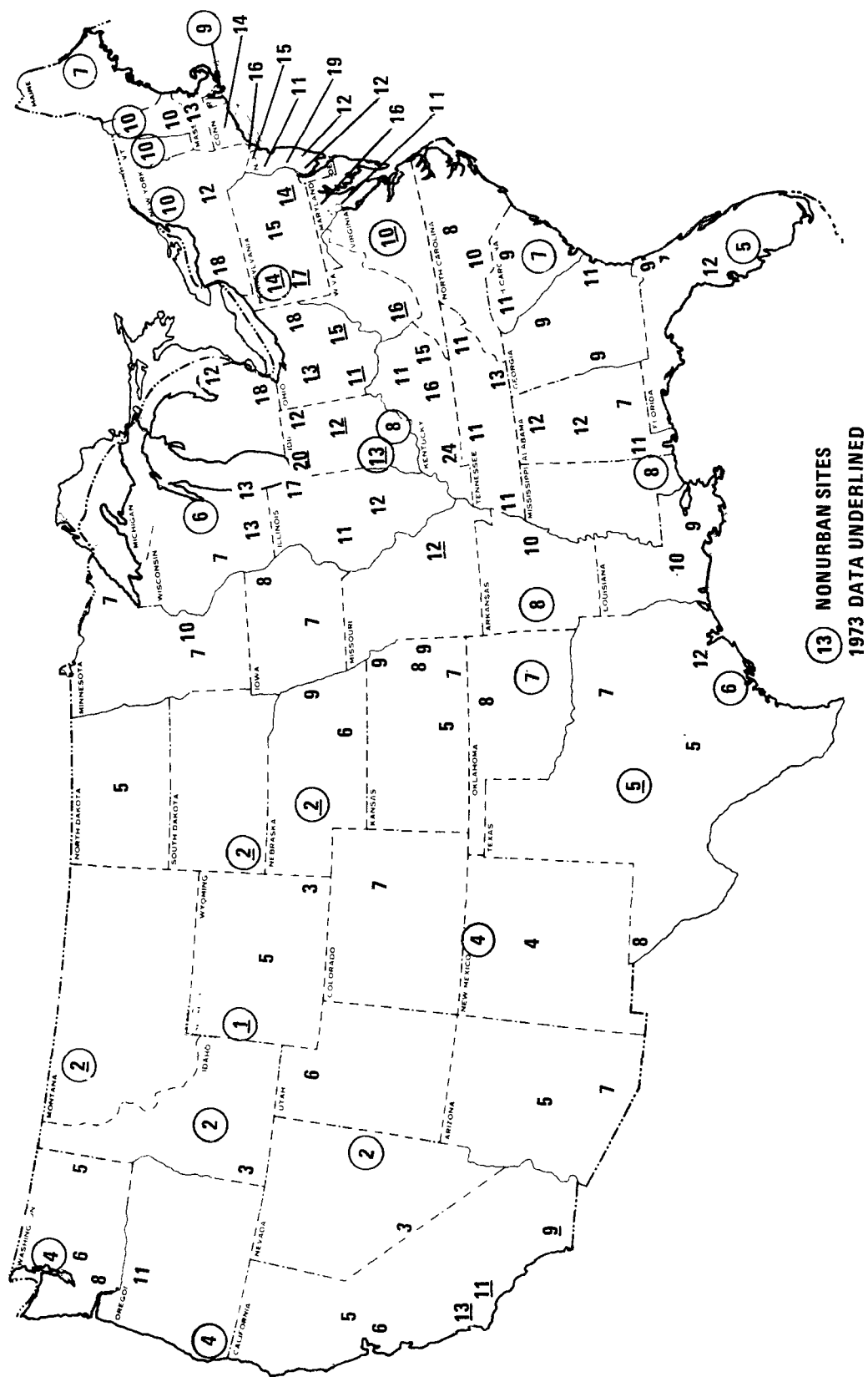


Figure 7. NASN sulfate data, 1972, 1973 (annual average,  $\mu\text{g}/\text{m}^3$ ).



atmospheric residence times for sulfur compounds range between 1 and 8 days,<sup>23,24</sup> suggesting the possibility of long-range transport.<sup>17</sup> These processes, which must be understood for development of a relationship between sulfur dioxide emissions and sulfates in the ambient air, are discussed more fully below.

### 1.3.1 Formation and Removal Mechanisms

The mechanisms by which sulfur dioxide is oxidized to sulfates are not well understood but are important because they determine the formation rate and, to some extent, the final form of sulfate. Atmospheric sulfur dioxide may be oxidized to  $\text{SO}_3$  and converted to sulfuric acid aerosol, or it may form sulfite ions that are then oxidized to sulfate. Subsequent to the oxidation, sulfuric acid or sulfate may interact with other materials to form other sulfate compounds. The most important sulfate formation mechanisms identified to date are summarized in Table 4.

The rates of oxidation associated with these mechanisms can vary with humidity, temperature, and concentrations of the reactants.<sup>4</sup> At ambient concentrations, Mechanism 1 is slow and probably important only for very long range transport of low pollutant levels on a global scale. Rates for Mechanisms 2 through 5 are thought to range between 1% and 20% of the sulfur dioxide oxidized per hour, which could make them of significance in urban or other areas near important emissions sources. Sulfate formation rates are usually enhanced by increases in humidity. Some idea of the pseudo first order reaction rates for the oxidation of  $\text{SO}_2$  in the ambient air is available for Los Angeles and St. Louis, two cities with different atmospheric characteristics. For Los Angeles, with photochemical smog and carbonaceous aerosol, the observed rate ranged between 1 and 13% per hour.<sup>30</sup> For St. Louis, which has less photochemical smog and perhaps more metal-containing aerosol, the rate was from 1 to 2% per hour.<sup>31</sup> Conversion rates for sulfur dioxide formation in plumes are even less well defined. Rates between 1 and 50% per hour have been reported. A review of the reports published before 1975, however, indicates that all of these studies of plume conversion rates are likely to be subject to substantial errors due to inadequate sampling capability.<sup>31</sup> Preliminary

Table 4. MECHANISMS THAT CONVERT SULFUR DIOXIDE TO SULFATES<sup>4,29</sup>

Mechanism	Overall reaction	Factors on which sulfate formation primarily depends
1. Direct photo-oxidation	$\text{SO}_2 \xrightarrow[\text{water}]{\text{light, oxygen}} \text{H}_2\text{SO}_4$	Sulfur dioxide concentration, sunlight intensity.
2. Indirect photo-oxidation	$\text{SO}_2 \xrightarrow[\text{hydroxyl radical (OH}\cdot\text{)}]{\text{smog, water, NO}_x, \text{ organic oxidants,}} \text{H}_2\text{SO}_4$	Sulfur dioxide concentration, organic oxidant concentration, $\text{OH}^\cdot$ ; $\text{NO}_x$
3. Air oxidation in liquid droplets	$\text{SO}_2 \xrightarrow{\text{liquid water}} \text{H}_2\text{SO}_3$	Ammonia concentration
4. Catalyzed oxidation in liquid droplets	$\text{NH}_3 + \text{H}_2\text{SO}_3 \xrightarrow{\text{Oxygen}} \text{NH}_4^+ + \text{SO}_4^-$	Concentration of heavy metal (Fe, Mn) ions
5. Catalyzed oxidation on dry surfaces	$\text{SO}_2 \xrightarrow[\text{carbon, water}]{\text{oxygen, liquid water, heavy metal ions}} \text{SO}_4^-$ $\text{SO}_2 \xrightarrow{\text{oxygen, particulate}} \text{H}_2\text{SO}_4$	Carbon particle concentration (surface area)

results from recent studies indicate that for distances of up to 30 miles downwind of the source, power plant plume  $\text{SO}_2$  oxidation rates were 1 to 2% per hour for coal combustion<sup>32</sup> and 10 to 20% per hour for oil combustion.<sup>33</sup> The more rapid rate in oil-fired plumes is thought to be due to the lack of particulate control, which permits vanadium or other catalytic particulates to accelerate  $\text{SO}_2$  oxidation in the plume. Further studies must be completed before firm conclusions can be drawn. However, coal-fired plant plumes contain higher concentrations of  $\text{SO}_2$ , which can convert to more sulfate over wide areas downwind than plumes from low-sulfur-oil-fired plants.

Current EPA studies suggest that  $\text{SO}_2$  oxidation rates in coal fired power plant plumes are initially inhibited by high concentrations of nitric oxide ( $\text{NO}$ ) with conversion rates increasing miles downwind as the plume encounters ambient oxidants and perhaps ammonia from rural background or urban plumes.<sup>31</sup> The observation by Davis<sup>34</sup> of conditions favorable to photochemical oxidant formation in a power plant plume 25 miles downwind of the source supports this conclusion. Near the source,  $\text{NO}$  levels were sufficient to depress preexisting oxidant concentrations; whereas, farther downwind the  $\text{NO}$  was depleted and increases in oxidant levels were observed. A consideration of possible reaction mechanisms suggests that  $\text{SO}_2$  conversion will be slow during the early stages of plume dispersion, but will increase as more ambient air mixes with the plume. Background oxidants that are entrained into the plume will preferentially oxidize  $\text{NO}$  to  $\text{NO}_2$ . However, after some  $\text{NO}$  is converted to  $\text{NO}_2$ ,  $\text{HNO}_2$  can form and be dissociated by sunlight to yield  $\text{OH}$ , which proceeds to oxidize  $\text{SO}_2$  by Mechanism 2. Entrained oxidants can react directly with  $\text{SO}_2$  dissolved in liquid droplets or wet aerosol particles after  $\text{NO}$  is converted to  $\text{NO}_2$ . The amount of  $\text{SO}_2$  dissolving in the liquid (droplet or film) is governed by the pH of the solution. As the liquid becomes acidic,  $\text{SO}_2$  dissolution is inhibited. Ammonia, present in the ambient air, probably controls this process (Mechanism 3). Thus, the EPA observation of a slow initial rate of conversion in plumes, increasing as the plume disperses, is mechanistically reasonable.

Because the extent of oxidation for Mechanisms 2 to 5 may be more dependent on concentrations of precursors other than  $\text{SO}_2$ , a reduction in ambient  $\text{SO}_2$  concentration may not produce a corresponding decrease in sulfate production if these precursors are limiting factors. The actual

oxidation rates and controlling mechanisms under realistic environmental conditions are poorly understood, thus precluding a quantitative assessment of the role of such precursors.

The ultimate chemical and physical forms of sulfate are influenced by the formation mechanisms and associated precursors. These properties are of paramount importance in determining the effects associated with sulfates. Although most sulfate measurements available to date do not provide species information, chemical forms of sulfate in the atmosphere are thought to be (roughly in order of suspected abundance) ammonium and ammonium acid sulfates, sulfuric acid aerosol, various metallic sulfates, and organic sulfates.<sup>4,24</sup> Electron Spectroscopy for Chemical Analysis has identified seven distinct species of particulate-related sulfur in California aerosols.<sup>35</sup> This and complementary evidence<sup>36</sup> suggest that some forms of sulfites may exist in the atmosphere. The relative abundance of these particulate sulfur oxide species can be expected to vary with both location and time. It is therefore important to be able to monitor specific compounds of greatest toxicity.

Studies<sup>37</sup> over the past 5 years of the size distribution of both sulfate aerosol and the general atmospheric aerosol have led to important changes in our understanding of the behavior of particles or aerosols such as sulfates in the ambient atmosphere. A schematic diagram of a typical atmospheric aerosol size distribution is shown in Figure 8. The plot is constructed so that the area under any section of a curve is proportional to the concentration in that size range. The number concentration is dominated by particles in the 0.002- to 0.02-  $\mu\text{m}$  size range, and the surface area is characterized by particles in the 0.05- to 0.5-  $\mu\text{m}$  size range. The volume and mass, however, show a bimodal distribution. Within the past 2 years, this bimodal distribution has been confirmed by a variety of studies in many locations that determined particle number and particle mass as a function of size.<sup>37</sup> The size range between 0.1 and 1.0  $\mu\text{m}$  is called the "accumulation mode." Particles in the accumulation mode are formed by physical and chemical processes that convert gases to particles, including metal fumes from high temperature sources, sulfates, nitrates, organic materials, etc. On the other hand, coarse

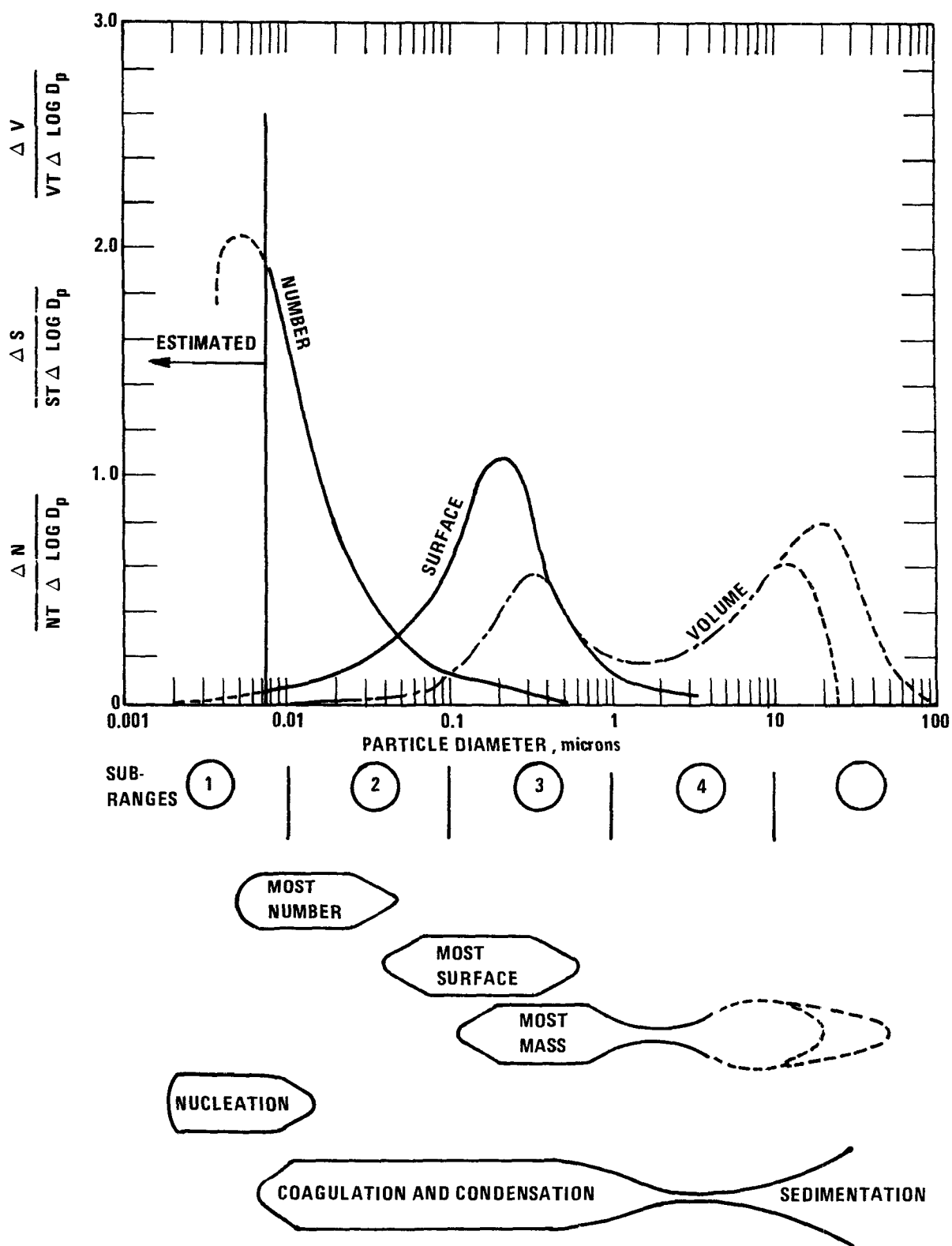


Figure 8. Bimodal distribution of atmospheric particles.<sup>37</sup>

particles are formed by mechanical processes such as grinding or rubbing-- for example soil, street dust, and rubber tire wear. Sulfates formed by the conversion of  $\text{SO}_2$  are found in the accumulation mode; whereas,  $\text{MgSO}_4$  from sea salt, and  $\text{CaSO}_4$  from gypsum are found in the "coarse particle mode". Typically, 80% of atmospheric sulfate is found in the accumulation mode.<sup>31</sup> These small sulfate particles, formed by homogeneous nucleation, grow by condensation of vapors into the accumulation mode. However, they do not grow into the coarse mode because there are so many more very small particles that dominate the condensation growth process.

It is important to understand that most sulfate aerosol is secondary, i.e., formed by gas-to-particle conversion, and occurs in the accumulation mode. Particles in this size range have very long life-times, the only efficient removal mechanism being cloud formation processes; they are also most effective in scattering light and therefore control visibility reduction. Most importantly, these particles are within the respirable size range and can pass into the lower respiratory tract. Sulfate aerosols can absorb water and increase in size;  $\text{H}_2\text{SO}_4$  and  $\text{NH}_4\text{HSO}_4$  do so at all humidities, and  $(\text{NH}_4)_2\text{SO}_4$  does so at relative humidities above about 65%.<sup>20</sup> As a result, visibility reduction caused by aerosols can be much greater than might be expected on a mass basis, and the lung deposition cannot be predicted on the basis of existing models for non-hygroscopic, non-deliquescent substances.

Some confusion has existed regarding the relative removal of  $\text{SO}_2$  and sulfate aerosol by ground and other surface dry deposition. As shown in Figure 9, the deposition velocity of aerosols depends on the particle size and reaches a minimum at the accumulation mode size range between 0.1 and 1.0  $\mu\text{m}$ . The deposition velocity of sulfate aerosol is theoretically, therefore, 100 to 1000 times smaller than for gaseous  $\text{SO}_2$ .<sup>31</sup> It should be clear then that, although  $\text{SO}_2$  is removed fairly rapidly by dry deposition on soil, vegetation, and other surfaces, once the  $\text{SO}_2$  has been converted to sulfate, the removal processes become very slow. Therefore, sulfates, or any secondary accumulation-mode aerosol (fine particulate matter) can travel long distances before removal by fallout or rainout.

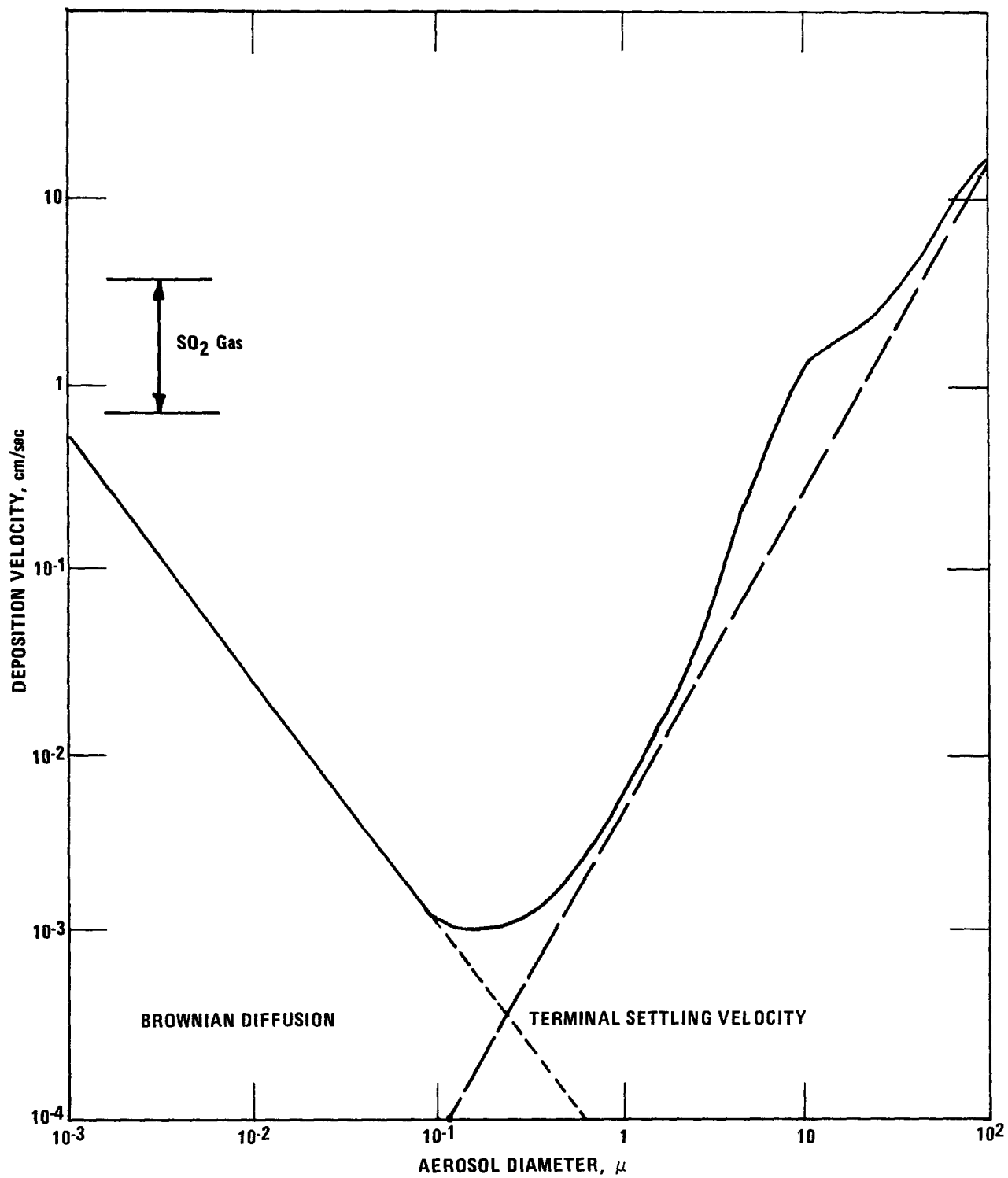


Figure 9. Velocity of deposition for unit density aerosol as a function of diameter.

These aspects of reaction mechanisms, aerosol size, and deposition rates are critical to an understanding of the possible effects of tall stacks. When  $\text{SO}_2$  is emitted near the ground, as from home heating units, the  $\text{SO}_2$  can be removed by surface removal mechanisms (dry deposition). When  $\text{SO}_2$  is emitted higher in the air, as from the tall stacks of fossil fuel-fired electric power plants, the  $\text{SO}_2$  is diluted before it reaches the ground, and the surface removal rates are reduced. Emissions may be trapped above the inversion layer and remain trapped for hours. Thus, elevated stacks theoretically permit a longer residence time in the ambient atmosphere for  $\text{SO}_2$  and promote fine particulate sulfate formation by the mechanisms discussed previously. However, they also provide increased dilution of the sulfate and  $\text{SO}_2$  and reduce impacts in the vicinity of the source.

### 1.3.2 Relationship Between Sulfur Dioxide and Sulfate Trends

The existence of a number of mechanisms by which  $\text{SO}_2$  is transformed into sulfates, and the possibility of transport of these fine particulate compounds into areas with low  $\text{SO}_2$  concentrations complicates the relationship between ambient  $\text{SO}_2$  concentrations and ambient sulfate levels. However, a good association between these two pollutants would be expected in source areas relatively isolated from the influence of other major emission sources and not subject to high levels of substances that influence sulfate formation. Monitoring in such areas has shown a reasonably good correlation between  $\text{SO}_2$  and sulfates. Four CHESS sites monitored  $\text{SO}_2$  and sulfates in the vicinity of a large, relatively isolated smelter in the Salt Lake City, Utah, area. The sites were located about 5, 10, 15, and 40 miles from the smelter. Ambient  $\text{SO}_2$  and sulfate levels were significantly associated at all sites.<sup>38</sup> During July 1971, a strike closed down the smelter. Sulfate levels, which had averaged from 13 to 6  $\mu\text{g}/\text{m}^3$  at the four sites (decreasing with distance from the source), dropped to about 4  $\mu\text{g}/\text{m}^3$  at all sites during the strike. The effect of the  $\text{SO}_2$  emissions on sulfates apparently extended at least 40 miles from the smelter. A similar study<sup>39</sup> was conducted during a nationwide steel strike in 1956. Although ambient  $\text{SO}_2$  measurements were not made, sulfate levels in four cities with large steel



mills were markedly lower during the 2-week strike period than they were afterwards. In two cities in which the steel industry was the dominant air pollution source, sulfates were about twice as high after the plants were put back into operation than during the strike. In this strike, emissions of both  $\text{SO}_2$  and catalytic metals were affected. These studies illustrate that (1) ambient  $\text{SO}_2$  concentrations and sulfate levels can be significantly associated when complicating factors such as high levels of background sulfates and substantial amounts of sulfate precursors are not present and (2) a clear relationship exists between  $\text{SO}_2$  emissions and ambient sulfate levels in the vicinity of large point sources in the absence of such complicating factors.

The relationship between  $\text{SO}_2$  and sulfate measurements from the NASN sites is not as apparent as that in the above examples. Seasonal and long term trends for national average rural and urban sulfates and sulfur dioxide for 1960 to 1970 are shown in Figures 10 and 11. Any actual trends are somewhat obscured by the fact that the number of NASN sites with valid data changes from year to year. Before 1965, sulfates show the same high-winter, low-summer peaks that characterize  $\text{SO}_2$  emissions. In contrast, after this time, nonurban, and to some extent urban, average sulfate levels begin to reach maximum values in the summer months. This is the opposite result that would be expected if sulfates were principally derived from area and other low elevation  $\text{SO}_2$  emission sources located near the monitors, because these sources are generally responsible for winter peaks in  $\text{SO}_2$ . Although correlations between ambient concentrations of  $\text{SO}_2$  and sulfates at NASN sites were not high, they were significantly better before 1967 than during the period 1967 to 1970.<sup>14</sup> Figures 10 and 11 show that, as  $\text{SO}_2$  concentrations trended downward, no long-range trends were observed for sulfates. A comprehensive analysis of urban  $\text{SO}_2$  and sulfate trends by Frank<sup>27</sup> concluded that if the data can be taken as representative of actual trends, no simple relationship exists between ambient  $\text{SO}_2$  levels and sulfate concentrations.

Although trends analyses for post 1970 NASN data have not been completed, the data available for 1970 to 1973 suggest that mixed urban sulfate trends continued during a period of continued decreases in  $\text{SO}_2$

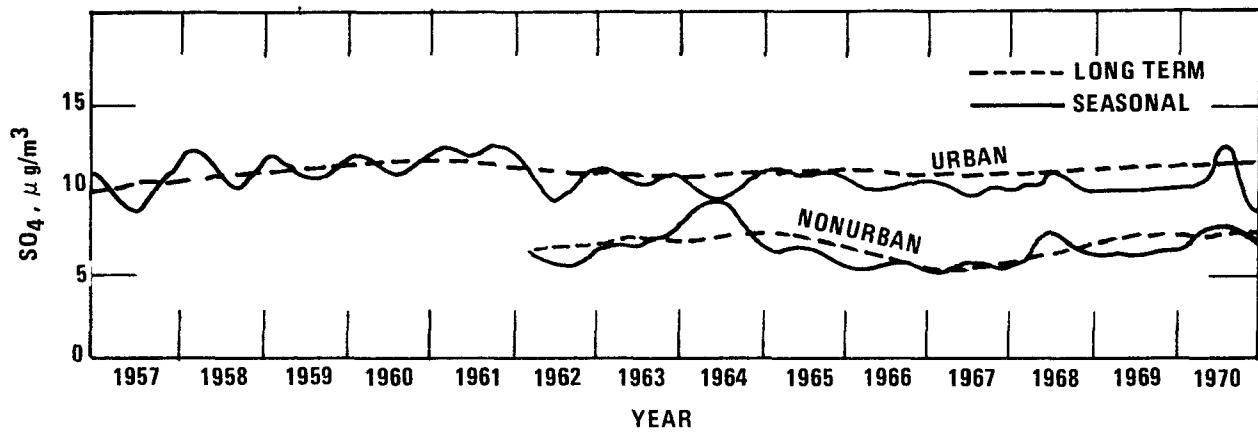


Figure 10. NASN urban and nonurban sulfate trends.<sup>4</sup>

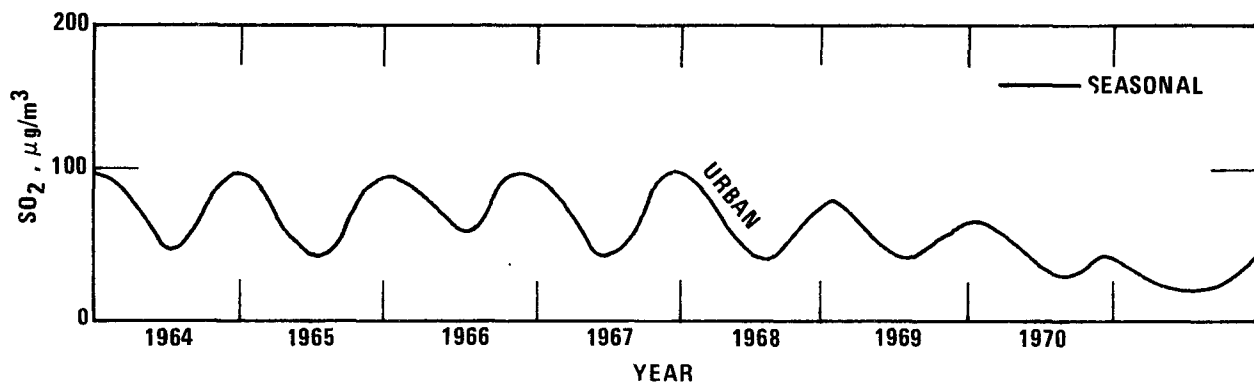


Figure 11. NASN urban sulfur dioxide trends.<sup>4</sup>

concentrations in urban areas. Table 5 indicates concentration trends for sulfates and  $\text{SO}_2$  for several urban and nonurban NASN sites. Although  $\text{SO}_2$  concentrations in most of these cities have decreased since the mid-1960's, sulfate levels have increased in some areas and decreased in others. When both trends are downward, the decrease in  $\text{SO}_2$  is often greater than the decrease in sulfates.

Although the lack of proportional response in long term and seasonal sulfate concentrations is not completely understood, several tentative explanations have been offered to account for the data. One interpretation suggests that if the potential error in sulfate sampling that results from  $\text{SO}_2$  being captured on the filter could be subtracted from the ambient sulfate levels, changes in  $\text{SO}_2$  levels would be better correlated with sulfates.<sup>2</sup> This is unlikely to be a satisfactory explanation because it does not account for elevated nonurban averages where  $\text{SO}_2$  levels are insufficient to produce such an error, or areas where sulfates increased when  $\text{SO}_2$  levels dropped. Unfortunately, the effect may not be consistent and has not been adequately quantified.

Other explanations offered suggest that local sulfate levels have been increased by switching from high sulfur fuels to vanadium containing, low-sulfur oil<sup>40</sup> and/or increased production of "natural" sulfates from biogenically reduced sulfur<sup>41</sup> compounds produced in polluted waters and marshy areas without increasing  $\text{SO}_2$  levels. Although these interpretations may have merit in specific locations, they are probably insufficient to explain the observed trends throughout the entire eastern U.S. As noted above, a more plausible explanation for at least part of the lack of trends is that some of the atmospheric mechanisms described previously for converting  $\text{SO}_2$  to sulfate are more dependent on the presence of precursor agents such as photochemical smog than on  $\text{SO}_2$  concentrations. A drop in sulfur dioxide concentrations would, therefore, not produce a corresponding drop in sulfates if unusually high levels of such precursor agents existed. This complicating effect may well cause part of the observed lack of correlations. For example, the greater levels of photochemical oxidants present in the summer may partially explain the observation that peak sulfate concentrations also occur at

Table 5. COMPARISON OF SULFUR DIOXIDE AND SULFATE TRENDS FOR SELECTED URBAN AND NONURBAN SITES

Site locations	SO <sub>2</sub> , µg/m <sup>3</sup>				Sulfate, µg/m <sup>3</sup>		% change
	Average, 1964-1966	Average, a 1970-1973	Trends <sup>a</sup>		Average, 1964-1966	Average, b 1971-1973	
			1964-1971	1968-1971			
Urban sites							
New York, New York	411 <sup>c</sup>	45 <sup>c</sup>	NA <sup>c</sup>	Down	26.9	18.5 <sup>c</sup>	-31.2
Glassboro, New Jersey	NA	14 <sup>c</sup>	Down	Low	9.3	11.0 <sup>c</sup>	+18.3
Jersey City, New Jersey	NA	29 <sup>e</sup>	NA	Up	22.2 <sup>c</sup>	15.8 <sup>c</sup>	-28.8
Newark, New Jersey	174	8 <sup>e</sup>	Down	Down	16.4	13.4 <sup>c</sup>	-18.3
Philadelphia, Pennsylvania	NA	64 <sup>c</sup>	NA	Down	23.3	14.9	-36.1
Charleston, West Virginia	21 <sup>c</sup>	15 <sup>e</sup>	NA	Down	25.6	17.9	-30.1
Pittsburgh, Pennsylvania	90	56	Down	Down	14.3	19.4	+35.4
Youngstown, Ohio	62	28	Down	Down	13.3	15.4	+15.8
Cincinnati, Ohio	NA	26	NA	Down	13.7	11.7	-14.6
Toledo, Ohio	NA	13	NA	Down	12.0	13.5	+12.5
Detroit, Michigan	13 <sup>c</sup>	30	Up	Down	15.3	14.7	- 3.9
Indianapolis, Indiana	57 <sup>c</sup>	20 <sup>c</sup>	Down	Down	14.0	12.1	-13.6
East Chicago, Indiana	105 <sup>e</sup>	58 <sup>c</sup>	Down	Down	17.7	19.1	+ 7.9
Chicago, Illinois	NA <sup>e</sup>	79	NA	Down	15.5	15.4	- 0.6
St. Louis, Missouri	86 <sup>e</sup>	32 <sup>c</sup>	Down	Down	15.6	13.5 <sup>e</sup>	-13.5
Denver, Colorado	19 <sup>c</sup>	10 <sup>c</sup>	NA	Down	4.3	6.7 <sup>c</sup>	+55.8
Salt Lake City, Utah	17	9 <sup>c</sup>	NA	Down	7.4	7.2 <sup>c</sup>	- 2.7
Los Angeles, California	NA	23 <sup>e</sup>	NA	NA	11.2 <sup>c</sup>	11.4	+ 1.8
Nonurban sites							
Shenandoah Nat. Pk., Va.	NA	NA	NA	NA	5.9 <sup>c</sup>	9.2	+55.9
Clarion Co., Pennsylvania	NA	NA	NA	NA	8.3 <sup>c</sup>	11.0	+32.5
Parke Co., Indiana	NA	NA	NA	NA	8.0 <sup>c</sup>	13.6	+70.0

<sup>a</sup>Sulfur dioxide averages are computed for 3 years within the 4-year time span. Qualitative "trends are defined over several sub-intervals and are based on statistically significant changes in geometric mean concentration."

<sup>b</sup>Some 1-year averages do not include enough samples to meet usual criteria for a valid sampling year. Their use for calculating 3-year means is not a significant problem.

<sup>c</sup>Two-year average. <sup>d</sup>NA = No data available. <sup>e</sup>One-year average.

this time. This suggestion is supported by a study<sup>44</sup> that found an association between peak 24-hr sulfate concentrations and air masses that contained high oxidant levels.

According to EPA scientists as well as others,<sup>3,20</sup> much of the observed lack of correlation between long term ambient  $\text{SO}_2$  and sulfate trends is explainable by long-range transport of fine particulate sulfates coupled with a change in the spatial distribution of emissions. According to this hypothesis, urban sulfates are thought to consist of two components: (1) locally produced sulfates that are related to  $\text{SO}_2$  emissions and concentrations in the immediate vicinity of the city, and (2) imported sulfates, which are formed from  $\text{SO}_2$  emissions upwind of the area and transported into the city. From the mid-1960's to the present, urban emissions of  $\text{SO}_2$  have decreased from area and industrial sources. This is reflected in the observed decline in  $\text{SO}_2$  concentrations. During that same time period, a change in the spatial distribution of  $\text{SO}_2$  emissions was taking place because much of the growth in the electrical utility industry was occurring in nonurban areas upwind of many major urban centers. As previously shown in Figure 2, the growth in emissions of power plants through 1970 more than counter-balanced the urban emissions decreases and caused the total sulfur dioxide emissions to rise. It is hypothesized that these emissions in nonurban areas were transported over long distances forming fine particulate sulfates that increased regional and urban concentrations of sulfates. Although it is reasonable to suggest that locally produced urban sulfate levels may have declined due to decreased urban sulfur dioxide emissions, the increase in regional backgrounds obscured this effect in many cities, resulting in either no observable trends or actual increases.

Finklea et al.<sup>14</sup> tested this explanation of trends for a number of U.S. cities. The procedure involved comparing trends in sulfate levels with trends in (1)  $\text{SO}_2$  emissions from sources in the city (assumed proportional to ambient  $\text{SO}_2$  trend) and (2) trends in power plant emissions in large areas surrounding the city. Contributions of the two source areas to sulfate levels were assumed to be equal. Usually,  $\text{SO}_2$  emissions in cities declined and contiguous area emissions increased. The net

trends in  $\text{SO}_2$  emissions predicted the sulfate trends with a reasonable degree of consistency particularly in larger eastern cities. Predictions were less good in areas with high photochemical oxidants or catalytic metal concentrations. The net trends in regional emissions predicted increases in sulfate levels in the few southeastern cities for which data was available and increased levels were observed. This analysis tends to confirm the transport explanation of obscured  $\text{SO}_2$  and sulfate trends during the 1960's. It also suggests that as regional  $\text{SO}_2$  emissions increase, increases in urban sulfate levels can be expected.

### 1.3.3 Long Range Transport

The hypothesis that long-range transport of sulfates from power plants is influencing urban sulfate levels is supported by the limited data base on emission and concentration trends. Comparisons of  $\text{SO}_2$  emission density (Figure 4), power plant locations (Figure 3), ambient sulfate concentrations (Figures 5 and 6), acid rainfall patterns (Figure 1), and incidence of atmospheric hazes<sup>20</sup> all show a somewhat similar distribution pattern concentrated in the northeastern and north central United States. According to the hypothesis, because  $\text{SO}_2$  emissions have been increasing, regional sulfate levels should also be increasing. This is not apparent from a cursory look at national nonurban trends as presented in Figure 10. The national trends, however, are based on data from changing numbers and locations of sites from year to year and include areas in the west where sulfates are not regionally elevated. Unfortunately, only a limited number of nonurban sites exists from which to determine long-term trends. Furthermore, because "nonurban" is a relative term in some sections of the densely populated east, any regional sulfate increases due to transport over great distances may be partially obscured by the influence of nearer source areas. Figure 12 shows the trends in nonurban sulfates from 12 sites in the northeastern area outlined in Figure 6. Not all of these sites reported valid data every year. Figure 13 presents trends for those 8 sites for which continuous data from 1965 to 1970 are available. Data from 1972, available for only 7 of these sites is also included. Three-year moving means are also

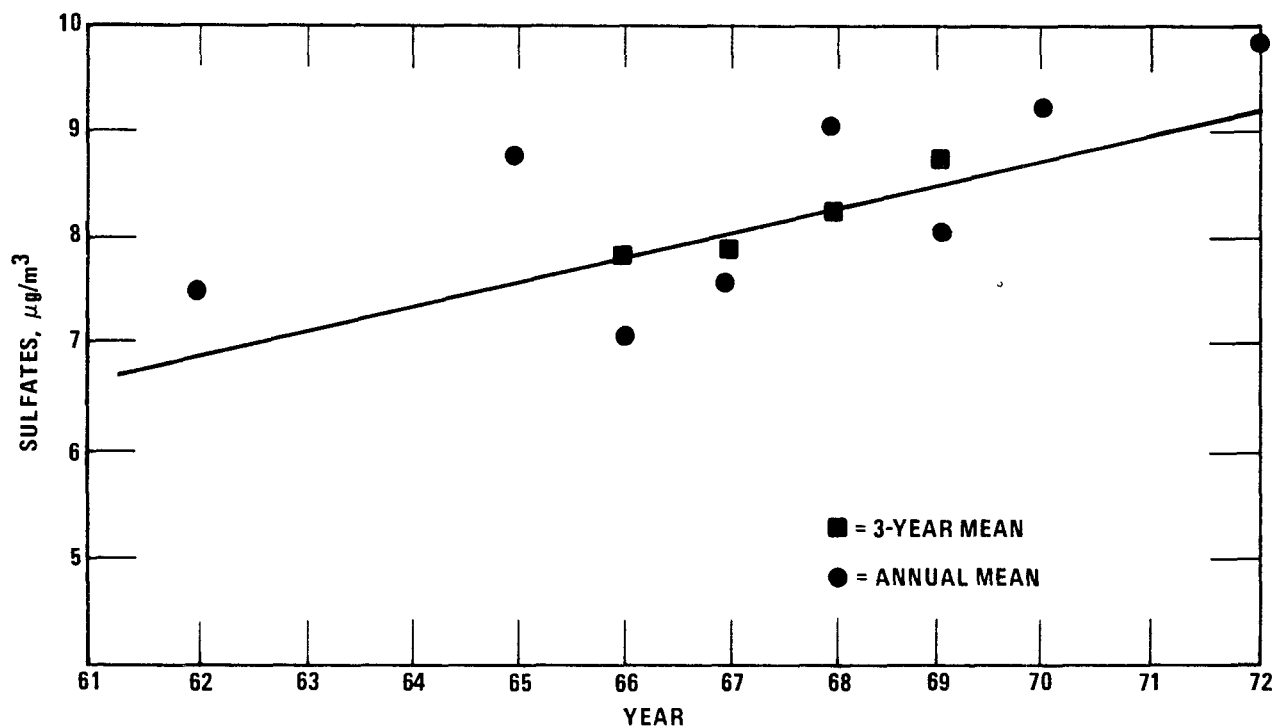


Figure 12. Nonurban sulfate trends in the northeast (12 sites).

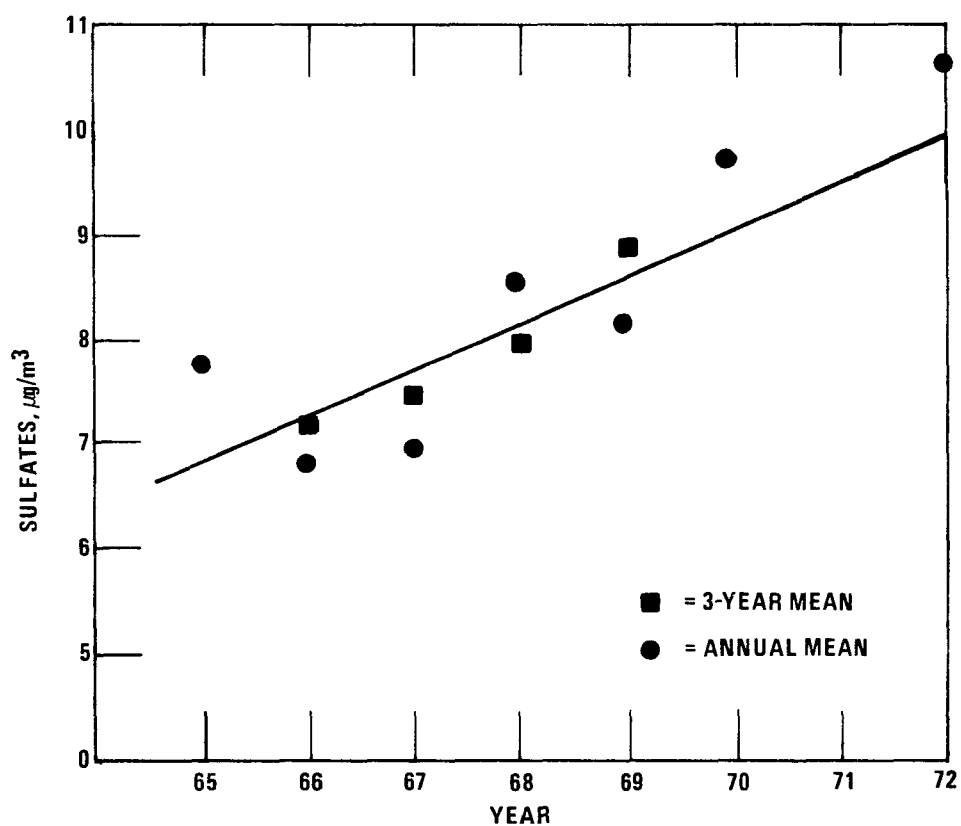


Figure 13. Nonurban sulfate trends in the northeast (8 sites).

plotted to smooth out variations due to fluctuations in weather patterns and the limited number of samples taken each year (20 to 26). The sites used in these figures are the same as those used by Trijonis<sup>44</sup> in a similar analysis, with the addition of the Cape Hatteras, N.C., site. Both figures suggest a general increase in nonurban sulfate concentrations in the 24-state northeast quadrant outlined in Figure 6.

The apparent increase in average nonurban sulfate levels is not evenly distributed throughout this region. Eight monitoring sites that had generally higher levels in 1970 and 1972 than in prior years are located in the northern New York-New Hampshire-Vermont, midwest, and southern portions of the 24-state region. Four sites at which concentrations appear to show no increases are located near the east coast from Maryland to Maine. Any increases in sulfates at these sites due to long-range transport may have been obscured by the decreases in urban SO<sub>2</sub> emissions in this heavily populated region. It is of note that none of the NASN nonurban sites located in the 24-state region of Figure 6 reported sulfate concentrations below 8 µg/m<sup>3</sup>, and 10 had levels in excess of 9 µg/m<sup>3</sup> in 1972 or the latest year of record.

Due to the fluctuations in annual averages and to the limited number and distribution of existing monitoring sites, these trends do not provide definitive verification or quantification of transport. However, Trijonis<sup>44</sup> points out that if the average trend lines were accepted as valid, subtraction of 1 to 4 µg/m<sup>3</sup> due to natural background indicates that anthropogenic sulfates may have increased by 40 to 100% in eastern nonurban areas during the 1960's. The increase in suspended sulfates is paralleled by the apparent rise in the sulfate ion content in precipitation noted earlier. These results are qualitatively in agreement with the increases in SO<sub>2</sub> emissions shown in Figure 2. The spatial distribution of the sulfate increases is not in conflict with the siting of many new power plants in the midwest and Ohio river valley area and the inception of stringent sulfur emission regulations in the urbanized east coast area. Based on the increases in SO<sub>2</sub> emissions past 1970 indicated in Figure 2, nonurban sulfate levels would not be expected to have increased markedly through 1975. Nonurban data for 1973 are only available at 3 sites in this region. Concentrations at these sites remained in excess of 10 µg/m<sup>3</sup>.



Evidence from particle characterization and plume chemistry studies presented previously is also supportive of regional transport. Although very little field work on this subject has been done in the United States, European studies have indicated SO<sub>2</sub> and sulfate transport cover distances of hundreds of kilometers.<sup>45,46</sup> Illustrative of this work are the results of an aircraft sampling run over the North Sea conducted as part of a transport model validation effort.<sup>47</sup> Observed sulfate and SO<sub>2</sub> concentrations are shown in Figure 14 and back trajectories (850 millibar) are plotted in Figure 15. Air from large emission sources in central Europe was transported towards England across the Netherlands, and the southern North Sea. Air from relatively clean areas in central Scandinavia was transported towards the southern parts of Norway. As the aircraft moved southward, a gradual increase of sulfates and SO<sub>2</sub> was measured. High sulfate levels (20 µg/m<sup>3</sup>) were noted at a spot some 100 miles from the nearest land mass. The peak concentrations were noted west of the Netherlands, as expected from the trajectories and source distribution.

The results of a summer 1974 study<sup>44</sup> of sulfates and oxidants in New York strongly suggest that high sulfate levels are associated with certain air masses. Sulfate levels at 11 to 13 urban and one nonurban sites located throughout the state were noted to have high or low concentrations on the same days. The authors concluded that "The high concentrations which occasionally existed simultaneously over the entire state suggests that there is a sulfate level associated with certain air masses probably due to long range transport."<sup>44</sup> A separate study<sup>16</sup> found that acid precipitation events in central New York were strongly associated with air masses that had passed through areas of high SO<sub>2</sub> and NO<sub>2</sub> emission densities 1 to 2 days earlier. Rainfall associated with air parcels from lower emission density areas was less acidic.

Both EPA scientists and the NAS consider long range transport to be an important factor that may explain much of the differences in trends between ambient sulfate and SO<sub>2</sub> concentrations observed during the 1960's. Using various assumptions based on current understanding of formation and transport, the NAS<sup>2</sup> presents estimates of the impact of large emission sources on downwind sulfate concentrations. Their analysis

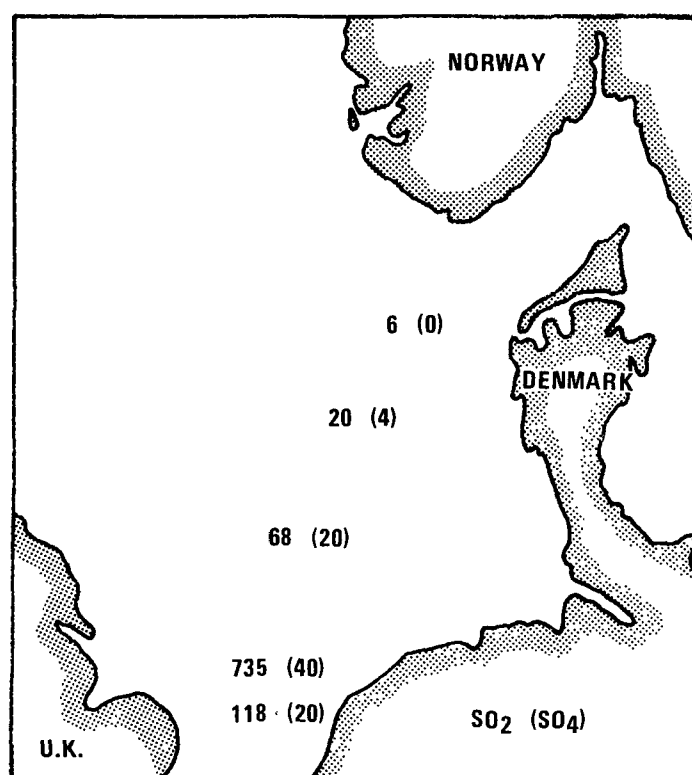


Figure 14. Aircraft measurements of sulfates and sulfur dioxide concentrations ( $\mu\text{g}/\text{m}^3$ ).<sup>47</sup>

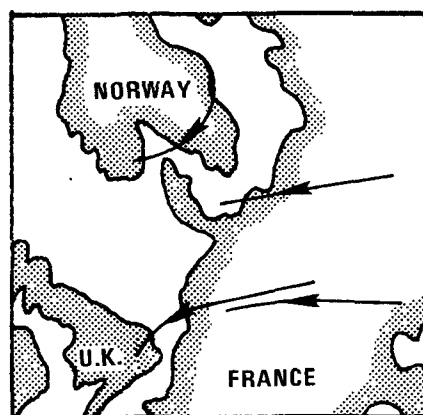


Figure 15. Forty-eight-hour back trajectories for air parcels arriving during sampling shown in Figure 14.

suggests appreciable impacts on sulfate levels at distances of 300 miles downwind. The NAS report<sup>2</sup> states that "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 emissions." As the report points out, the relative impact of distant versus local SO<sub>2</sub> emissions on sulfate levels will depend on regional characteristics such as spatial distribution of emissions and the presence of precursors involved in sulfate formation mechanisms. Accordingly, regional sulfate levels would not be expected to increase as rapidly as regional SO<sub>2</sub> emissions.

Although significant long range transport is considered plausible by both EPA and NAS<sup>2</sup>, the SAB<sup>1</sup> suggests that the impact of nonurban sources on urban areas is likely to be minor. The SAB prefers to explain the observed disparity in ambient SO<sub>2</sub> and sulfate trends discussed in the previous section wholly in terms of some of the alternate explanations noted above. In particular, the SAB believes that the sulfate levels extant in an urban area are almost completely the result of local sulfur emissions. The lack of relation between SO<sub>2</sub> and sulfates is then explained by the lack of proportionality expected from complex formation mechanisms, consistently inflated monitoring data, and significant direct emissions of sulfur trioxide and sulfates. The SAB based its assessment of the impact of transport on the assumption that dilution and removal would reduce sulfates to negligible levels during transport and did not analyze transport models or measurements. The SAB believes that the available evidence is insufficient to confirm the validity of their assumptions or any of the explanations offered to date for the observed urban sulfate trends.<sup>1</sup> This only highlights the need for additional research in these areas. EPA recognizes the uncertainties involved and the fact that a quantifiable relationship between SO<sub>2</sub> emissions and sulfate air quality must await further study of formation mechanisms, oxidation rates, and long range transport. Even so, once applicable emission limits have been met by all sources in urban areas thus reducing locally produced sulfates, EPA believes that, based on the available evidence concerning transport, further increases in regional

and urban sulfates can be expected if nonurban SO<sub>2</sub> emissions from power plants and other sources continue to rise. Given the general levels of sulfates, other fine particles, and sulfur oxides in the northeast, the Agency's assessment of the preliminary health data suggests that such increases should be viewed with concern.

#### 1.4 Control Alternatives

As the previous section has indicated, sulfate formation is a complex phenomenon that involves many factors. Some parameters such as humidity, temperature, and natural production of hydrogen sulfide and ammonia are obviously not controllable. This limits the available technical control options to control of anthropogenic emissions of precursors that accelerate the formation of sulfate from SO<sub>2</sub> or control of the major precursor SO<sub>2</sub>. These alternatives are discussed below.

##### 1.4.1 Precursor\* Control

Considering sulfate formation mechanisms and the lack of a perceptible relationship between SO<sub>2</sub> concentrations and sulfate trends, control of non-SO<sub>2</sub> precursors, such as fine particulates and nitrogen oxides, might be necessary. Such a control strategy is currently being applied to oxidants. Table 6 lists the principal agents involved in the conversion mechanisms shown in Table 4 that affect sulfate formation and presents expected trends and control feasibility.

As pointed out in the table, control of those precursors already included in other control strategies should be dictated in large part by considerations other than sulfate formation. For example, if the critical hydrocarbon/nitrogen oxides ratio necessary to reduce sulfate formation differs from that for oxidants, the oxidant strategy should prevail. Similarly, fine particulates should be controlled to prevent intrinsic health and welfare effects as well as the synergistic effects possible when they combine with sulfur oxides.

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\*Although SO<sub>2</sub> is the most essential sulfate "precursor," the term as used here refers to those chemical agents (e.g. oxidants) that are essential in the previously outlined formation mechanisms.

Table 6. ENVIRONMENTAL INVENTORY OF SULFATE PRECURSOR AGENTS: PRELIMINARY VIEW 29

Precursor agent	Ambient concentration trends	Emission trends	Control feasibility
Sulfur dioxide, H <sub>2</sub> S	Rapidly declining in urban areas, but possible reversal in future.	Moderate increase with possible acceleration in future.	Extensive control possible; should result in SO <sub>4</sub> reduction.
Hydrocarbons/NO <sub>x</sub> oxidant formation	HC, oxidants: limited data. Possible decline with more controls proposed; NO <sub>x</sub> : small increase.	HC: minor, steady increase; NO <sub>x</sub> : increase; oxidant: uncertain.	Control strategy based on oxidant standard has priority.
Particulate matter: catalytic metals and carbon	Moderate, steady decline in TSP, but possible increase in fine particle emissions.	TSP: decrease slowly and variable. Fine particles: uncertain.	Fine particles should be controlled for intrinsic effects; sulfate formation potential unknown.
Ammonia	Increasing over time; little monitoring and control.	Growing, with very limited control technology alternatives; high regional natural emissions.	Some control of point sources possible; background uncontrollable.
Humidity, sunlight, other meteorology	NA	NA	No control possible.

The effectiveness of controlling sulfate levels by the control of pollutant precursors is impossible to predict with currently available information. Ideally, elimination of precursors would slow the sulfate formation rate, reducing sulfate concentrations in the vicinity of SO<sub>2</sub> emissions and increasing the percentage of SO<sub>2</sub> that will be removed from the atmosphere by rainout or dry deposition before forming sulfates. However, if the relatively low oxidation rate tentatively measured in St. Louis is typical of eastern cities, ambient precursor control in urban areas might do little to reduce local urban sulfate concentrations. In addition, favorable meteorological conditions and natural backgrounds of humidity, oxidant-related hydrocarbons, and ammonia may be sufficient under some conditions to allow significant regional formation of sulfates from urban and rural SO<sub>2</sub> emissions even in the absence of urban precursors. Thus, reduction of precursors involved in a given mechanism may only result in a shift to another formation mechanism without greatly affecting sulfate levels. Control of precursors such as catalytic vanadium particulates, which are released simultaneously with SO<sub>2</sub>, may slow oxidation in plumes and be a more successful strategy than control of ambient precursors for reducing local sulfates if plume oxidation rates without such controls are rapid. Even if control of catalytic particulates would reduce local sulfate formation in the plume, rural ambient conditions may still produce regional sulfate formation and transport. Thus, given current information although catalytic particulates and photochemical oxidant control may be of importance in a sulfate strategy, control of such precursors without also restricting sulfur dioxide emissions might be of limited value.

Reliance on tall stacks and precursor control to reduce local sulfates could result in increased rainfall acidity. As discussed previously, SO<sub>2</sub> emissions from tall stacks can travel long distances before deposition. In addition to allowing more time for sulfate formation, the lengthy residence time increases the dependence on precipitation scavenging for removal.<sup>48</sup> Recent evidence suggests that this would only aggravate the already serious acid rain problems described in Section 1.1.3.

Furthermore, for air masses that travel large distances with no rain or that are stagnant for several days, sulfate formation could be extensive, even with precursor control at source areas and with slow oxidation.

Based on the current understanding of atmospheric chemistry, transport, and health effects, the most feasible approach to sulfate control is to control atmospheric emissions of sulfur compounds and pursue precursors primarily through existing programs aimed at their control. However, the role of precursors, including those which are uncontrollable, should be considered before implementing any sulfate-related strategy; for example, more stringent  $\text{SO}_2$  controls might be necessary in areas with high smog levels, high emissions of catalytic metals, or high humidity to prevent excessive levels of acid-sulfates caused by rapid conversion. Due to the combination of precursor-limited conversion mechanisms and to the influx of sulfates from distant sources, reduction of  $\text{SO}_2$  emissions and concentrations in a given urban area may not produce proportional reductions in sulfates. Furthermore, depending on the type and concentration of precursors present, reductions in total sulfate concentrations may not produce comparable reductions in the specific acid-sulfates or other sulfur oxides thought most responsible for adverse health effects. Current knowledge does suggest that reduction in  $\text{SO}_2$  emission density over a large region would reduce regional sulfate backgrounds; under these conditions, decreases in locally produced urban sulfates would be expected to become more directly related to reductions in urban  $\text{SO}_2$  emissions.

#### 1.4.2 Alternatives for $\text{SO}_2$ Emission Control

The principal approach to sulfate control is likely to be sulfur dioxide control. Primary sulfate emission control for stationary sources involves widely utilized conventional particulate control devices. Control of sulfuric acid emissions from catalyst-equipped motor vehicles may include desulfurization of gasoline, as yet undeveloped exhaust particulate control, or switching to alternative exhaust hydrocarbon and carbon monoxide control systems or alternative engines. As discussed in Section 1.4.1, control of sulfate precursor agents such as catalytic

metals is unlikely as a principal control strategy. Control of  $\text{SO}_2$  precursors such as  $\text{H}_2\text{S}$ ,  $\text{CS}_2$ , and  $\text{COS}$  is commonly practiced in the petroleum industry and involves a variety of standard and developing technologies that will not be discussed here. Application of these technologies will become more important with the increase of precombustion removal of sulfur from fossil fuels.

Consideration of the magnitude of total emissions and the relative removal rates of  $\text{SO}_2$  released from low versus high level sources and resultant sulfate formation potential indicates that permanent control of large industrial and utility emitters will probably be an important element of any sulfate control program.

Selection of  $\text{SO}_2$  control technology is dependent upon the characteristics of the emission sources to be controlled. In the evaluation of a  $\text{SO}_2$  control alternatives for use in a sulfate control strategy, the following criteria should be used:

1. Sulfur reduction potential.
2. Relative energy efficiency or penalty.
3. Cost of sulfur removal or emission reduction.
4. Overall environmental impact.
5. Commercial availability of technology.
6. Applicability of approach to scope and/or numbers of emission sources.

Table 7 presents several major alternative  $\text{SO}_2$  control technologies together with a partial listing of their characteristics according to the above criteria. A summary of the major control alternatives that might be included in a sulfate strategy follows.

#### 1.4.3 Use of Naturally Clean Fuels<sup>49</sup>

This approach is constrained by the availability of "clean fuels": natural gas, and low-sulfur coal and oil. Due to a growing shortage, natural gas is already being allocated to high-priority uses. Therefore, until 1990 the principal natural clean fuel options involve use of low-sulfur coal and, secondarily, low sulfur oil.

Options include redistribution of clean fossil fuels to large coal consumers in those regions in which the  $\text{SO}_2$  ambient air quality standard



Table 7. SUMMARY OF CONTROL ALTERNATIVES FOR SO<sub>2</sub> 49, 59

Control technique	Sulfur removal efficiency (3% S base), %	Energy penalty, %	Incremental cost, mills/kWh	Timing	Annual capacity, 10 <sup>6</sup> tons coal	Likely applicability	Other environmental effects	Comments
Nonregenerable (lime/limestone) scrubbing (new plants)	90	3-7	3.0	1975-77	230 (1980)	All new utilities and large industrial boilers	Waste disposal problems	Operating load factor 80%, 1000-Mw power plant
Nonregenerable scrubbing (existing plants)	90	3-7	4.4-6.1	1975-77	60 (1980)	Boilers - 100 MW - 20 yr age	Waste disposal problems	Operating load factor 60%, 300-Mw power plant
Regenerable scrubbing (new plants)	80-95	3-7	3.4-7.0	1980	--	New utilities and large industrial boilers	Reduced waste disposal problems	Limited data on utilization and costs
Physical coal cleaning	30-50	3-10	0.6-1.6	1974-80	--	All coal combustors	Land, air, waste disposal at cleaning site	Cleaning costs \$1.50/ton
Coal liquefaction (solvent refined cleaning)	75-85	20	10	1985	110	Existing and mine-mouth power plants, industrial boilers, small point sources	Sulfur and other emissions partially concentrated at central plants; increased mining necessary; unidentified air, water, and solid waste problems	Economics of scale possible
Low-Btu gas	80-98	10-40	6-16	1980-85	13 200	New and large existing power plants		Energy penalty reduced by combined cycle application. Capital intensive
Pipeline quality	98+	40	NA	1980 1985	29 50-100	Small and area sources		Minimal energy penalty if used to replace electric heat
Fluidized bed	90+	Minimal	0-3	1985	150	New power plants and industrial boilers	Substantial reductions in NO <sub>x</sub> emissions possible	Energy efficiency can be greater than conventional power plants
Refuse combustion	20	Minimal	Minimal	1985-90	50	Coal mix, 10-20 refuse	Benefits due to solid waste reduction	
Low sulfur western coals	75-85	Minimal	0-5.3	1980	250	Western and fringe eastern boilers	Increased particulate control necessary	Energy penalty for transportation to eastern areas
SIP compliance coal (Eastern)	33-70	Minimal	1.0-3.0	1975	95	Eastern existing boilers		Cost for 1 S or less compared to 3 S coal
Desulfurized residual oil 0.3%	85	--	2.0	1980-85	NA	Existing oil burners	Also removes V, other trace elements	

is being exceeded. This may be accomplished by relaxing SIP requirements in certain areas, and by expanding and developing clean fuel extraction and production techniques. Apart from the uncertainty associated with attempts to regulate the distribution of fuels, this approach may merely transfer, spread out, or actually increase the sulfur emission burden and may aggravate long-term sulfate or acid rain problems.

#### 1.4.4 Flue Gas Desulfurization (FGD) Technology<sup>49</sup>

Stack gas desulfurization systems can be classified into two general categories: (1) nonregenerable processes in which the sulfur materials are disposed of as wastes, and (2) regenerable processes from which by-products such as sulfuric acid and elemental sulfur can be marketed. Many of these engineering control systems have been studied at the pilot plant level, some have been tested at the intermediate prototype scale (10 to 15 MW), and a growing number have been incorporated into full scale commercial power systems (100 MW or greater). In the United States, the majority of utilities planning full-scale system installation through 1980 will likely choose the nonregenerable FGD systems due to their advanced state of development compared to regenerable systems. In the 1980's, however, this trend may be reversed as the regenerable systems, which produce saleable by-products, are proven to be operationally and economically competitive.

It should be noted that scrubbers are a possible source of localized primary sulfate emissions, but such emissions are thought to be of small potential import due to the relatively small amounts emitted from controlled sources as opposed to uncontrolled sources, and the non-acidic chemical form of the sulfates from controlled sources. In addition, primary emissions of  $\text{SO}_3$  may be actually reduced by scrubbers and, where necessary, very effective mist eliminators are available and are being used on flue gas desulfurization units.

A summary description of the major flue gas desulfurization processes is presented in Table 8.

#### 1.4.5 Fuel Pretreatment<sup>49,50</sup>

Major coal pretreatment methods are (1) mechanical coal cleaning, (2) coal liquefaction, and (3) coal gasification.

Table 8. SUMMARY DESCRIPTION OF MAJOR FLUE GAS DESULFURIZATION PROCESSES

Process	Classification/ operating principles	SO <sub>2</sub> particulate efficiency	Development status	Application	Implementation <sup>a</sup>	Advantages	Disadvantages
Lime/limestone scrubbing	Nonregenerable process/ wet absorption in scrubber by slurry; insoluble sulfites and sulfates disposed of as waste.	Up to 90 percent SO <sub>2</sub> removal/99 percent fly ash removal by most scrubbers.	32 full-scale units (9,829 MW) in operation or plan- ned for start-up by 1977.	Old or new power plant; coal- or oil-fired.	An additional 21 units (10,349 MW) planned for installation by 1980; 15 units (8,099 MW) considered for unspecified date; 4-5 years lead time needed for new plants; 3 years for retrofit of old plants.	Cheapest of ex- isting pro- cesses; elimination of particulate control requirement.	Waste and water pollution prob- lems; reheating of scrubber exit gases needed; supply and handling of large vol- umes of react- ant may be problems.
Double alkali process	Nonregenerable process/ wet absorption in scrubber; reactants and soluble reaction products precipitated and removed from recycled reactant solution outside of scrubber; most common reactant sodium sulfite.	High efficiency >90 percent SO <sub>2</sub> removal/high particulate removal as above.	Active area but no fullscale demon- stration as yet; 2 units operating; G.M. installed a unit on a coal- fired boiler in February 1974; several sulfate removal schemes under study.	As above with potentially lower cost and greater ease of operation favoring some inroads into smaller plants.	Research-Cottrell esti- mates \$600 million a year market by 1979; a second generation lime/limestone system; lead times as above for power plants.	Potentially cheaper and more reliable than lime/lime- stone system.	Similar to above and all throw- away systems.
Magnesium oxide scrubbing	Regenerable process/wet absorption by magnesium oxide slurry; fly ash removed prior to or after scrubbing; magne- sium oxide regenerated by calcining with car- bon; SO <sub>2</sub> by-product can be converted to sul- furic acid or sulfur.	90 percent SO <sub>2</sub> removal/par- ticulates removed as required by prescrubber.	One full-scale unit on test at Boston Edison 150-MW oil- fired unit; Potomac power unit started in 1973 (coal-fired 100 MW).	Similar to lime/lime- stone but oil-fired boilers will not require particulate control up- stream of scrubber.	4 additional units (850 MW) planned or under consideration; lead times as for lime/ limestone systems.	May be more reliable than lime/limestone process; no known waste disposal prob- lems; regenera- tion facility need not be located at utility.	Cost of regeneration; marketing of sulfur products; reheat.
Wellman-Lord	Regenerable process/ sodium base scrubbing with sulfite to pro- duce bisulfite; regen- eration in an evapora- tive crystallizer. Sul- fate formed either purged or removed by selective crystalliza- tion.	>90 percent SO <sub>2</sub> removal; particulate removal as above by pre- scrubber.	Reliably operated (~9000 hours) in Japan; full-scale demonstration scheduled at Northern Indiana Public Service coal-fired 115-MW boiler to start December 1975.	As above.	5 additional units (1800 MW) planned by 1980; lead times same as for limestone systems.	More reliable than lime/lime- stone system based on Japa- nese experience, simplicity of unit operations in regenerator; waste disposal problems reduced.	Some bleed of solution to remove unde- sirable reac- tion products, a source of water pollution, otherwise as above.

<sup>a</sup> Twenty-six units (13,772 MW) under consideration have not yet selected a specific FGD process. Seven units (655 MW) are operating or planned using processes not summarized here.

1. Mechanical coal cleaning: Mechanical coal cleaning is a process in which coal is crushed, screened for size, and subjected to techniques based upon specific gravity and surface characteristics to separate coal from its heavier impurities including pyritic sulfur. Because only about 30 percent (10 to 47%) sulfur removal is routinely possible, mechanical cleaning cannot generally be used to attain very low sulfur coal from high-sulfur coals.

2. Coal liquefaction processes: The most promising liquefaction technology for short-term commercial application through 1990 is solvent refining under hydrogen pressure to dissolve the organic components of the coal, producing a solvent refined coal (SRC). The solution is filtered to remove the ash and insoluble organic materials, and fractionated to recover the solvent. The refined coal remaining is solid at room temperature. Final fuel output contains 0.6 to 0.7 % sulfur.

3. Coal gasification: Almost all processes for gaseous fuel from coal fall into one of three groups: (1) production of low-Btu gas, typically around 100-200 Btu/ft<sup>3</sup>, (2) production of medium-Btu gas, 300-500 Btu/ft<sup>3</sup>, and (3) production of pipeline quality synthetic natural gas (SNG) around 1000 Btu/ft<sup>3</sup> (high-Btu gas). Greater than 95% sulfur removal is possible, although the gasification plant can become a significant sulfur emission source.

#### 1.4.6 Alternative Coal Combustion Systems<sup>49</sup>

The pulverized-coal boiler and steam turbine systems currently dominate the power generation field. There are alternate approaches available, however, that have proponents within the energy engineering field. Two such systems are fluidized bed combustion and magneto-hydrodynamics; both systems decrease coal usage through increased overall conversion efficiency.

1. Fluidized-bed coal combustion: In fluidized-bed operations, coal less than 1 centimeter in size is suspended in a vertically rising flow of combustion air, which gives the appearance of a boiling fluid. The various processes have a number of engineering advantages, among them smaller size and cost, the ability to handle even high-ash coals without

particulate controls (the ash agglomerates in the fluidized bed and is collected in a hopper), and sulfur removal by inclusion of lime or limestone in the combustion bed. Because lower operating temperatures are possible, this system also reduces  $\text{NO}_x$  emissions. In addition, the systems can be engineered to provide hot gases to drive a gas turbine and steam generator, raising the overall thermal efficiency of the plant.

It would appear that fluidized-bed combustion with  $\text{SO}_x$  cleaning should be considered, especially with respect to low-sulfur coals with high ash resistivity. It does not, however, seem promising except in new plant application.

2. Magnetohydrodynamics (MHD): These power systems, currently in the research and pilot stage, operate on different physical principles than a conventional steam-electric plant, promising thermal efficiencies of 50 to 60 percent.

Though energy efficiency is a major promise, MHD also offers some direct  $\text{SO}_2$  emissions reduction. To aid the ionization of the combustion gases, potassium or other alkaline metals are added; these react with the sulfur and are then collected for recycling. Near-term availability does not appear feasible.

#### 1.4.7 Intermittent Control Systems (ICS)

Intermittent or supplementary control systems are a category of sulfur oxide management techniques that attempts to avoid large localized ground level  $\text{SO}_x$  concentrations by means of very tall stacks, fuel switching, load shifting, and curtailment of operation when short-term or seasonal meteorology forecasts indicate poor dispersion. As discussed in Section 1.3.1, tall stacks may actually increase potential sulfate formation by extending the time available for conversion before ground removal of  $\text{SO}_2$  is possible. This possibility has not yet been tested by comparing plume studies of very tall stacks. Extensive use of ICS would likely maintain or increase current high levels of sulfates and acid rain, because growth in  $\text{SO}_2$  emissions from new sources would be comparatively greater and this growth would not be balanced by corresponding emissions decreases

from existing sources. In two separately prepared reviews<sup>2,60</sup> of control options, the NAS points out that intermittent control is likely to be only a temporary short-term measure in the next 5 to 10 years because it does not reduce regional atmospheric loading of sulfur oxides nor does it curtail secondary sulfate pollutant formation and transport.<sup>2</sup> The strong possibility that increases in sulfates and acid rain might cause significant adverse health and welfare effects precludes permanent use of such techniques as currently applied in a sulfur oxides and fine-particle control strategy. Based on current knowledge, intermittent control will probably be an acceptable technique in the long term only in special cases where constant emission reduction measures are unavailable and/or if later information indicates that certain sources are located in such a manner that  $\text{SO}_2$  and  $\text{SO}_4^{=}$  are removed by natural mechanisms without adversely impacting on health or welfare.

#### 1.4.8 Employment of New Energy Sources<sup>49,54</sup>

Today, the major portion of our electric energy requirements is supplied by coal combustion. Therefore, the substitution of alternative basic energy sources represents an equivalent reduction of coal combustion  $\text{SO}_x$  emissions. The most readily available alternate energy sources are solar energy, nuclear fission, geothermal energy, and solid waste combustion.

Solar energy is unlikely to be of significant impact through 1990, but is potentially an attractive major future energy source. Nuclear fission is already scheduled to have a large impact on energy generation through 1990 and beyond. Further fission development as a sulfur emission reduction alternative must be balanced by environmental and economic concerns associated with planned development. Geothermal energy development is of limited capability through 1990, especially in the northeast U.S. where sulfate reduction is most needed. In addition, sulfur control must be applied to this energy source to prevent natural  $\text{H}_2\text{S}$  emissions. Combustion of solid waste could displace as much as 5% of utility coal in the 1980's with a sulfur content equivalent of about 0.3%. Consideration should be given to the relative energy and environmental impact of this method of solid waste disposal compared with recycling of the material.

#### 1.4.9 Energy Conservation

Any reduction in demand for electric power, whether through economic or conservation incentives, can result in a decrease in  $SO_x$  emissions from power plants. Detailed discussions of measures such as efficient pricing, modification of demand, and conservation are presented by Kahn<sup>61</sup> and Chapman et al.<sup>62</sup> Kahn concludes that more efficient pricing reflecting the higher costs of supplying electricity during peak hours could make a significant contribution to demand reduction. He also notes that reductions in the use of oil and natural gas due to the energy crisis may lead to increased use of electricity. This could partially offset reduced demands due to costs.

Conservation measures could have a significant impact on electrical demand. Table 9 shows that potential savings in 1980 amount to about 400 billion kilowatt-hours. Projected demand in 1980 is expected to range between 2,200 and 3,200 billion kWh. Estimated reductions in  $SO_2$  emissions from this savings are as large as 1.8 to nearly 4.0 million tons/yr of sulfur oxides nationwide.

Table 9. ELECTRIC ENERGY CONSERVATION SAVINGS, 1980<sup>62</sup>  
( $10^9$  kWh)

Type	Savings
Substitution of natural gas for electricity in selected household uses	94
Lighting levels	183
Appliance efficiency, general	23
Air conditioning efficiency	51
Aluminum production and use in cars	20
Home insulation	<u>21</u>
	392

#### 1.4.10 Applicability and Availability

It is important to note that the impact of the above technologies must be considered in reference to the total energy production system in which they are utilized. For example, utilization of synthetic natural gas in power plants entails severe energy and cost penalties, but is competitive with electricity when used for space heating. Table 10 gives a total system evaluation of several SO<sub>2</sub> control technology alternatives potentially available to the utility sector. The table clearly shows the difference in SO<sub>2</sub> emissions, energy efficiency, and costs when various control technologies are used.

The maximum potential availability of the major SO<sub>2</sub> control technologies for use in a national sulfate control strategy through 1990 is presented in Figure 16. It should be pointed out that the estimates of Figure 16 do not indicate actual application of technologies, but the maximum amount of capacity possible, given economic and technological constraints. Applicability of the systems would depend on the other factors outlined previously and included in Table 7.

It is important to note that flue gas desulfurization is the chief technology available for stationary source control through 1985. Given the cost and energy penalties of gasification and liquefaction, FGD is likely to have continued application through 1990.

#### 1.5 Information Gaps and Research Needs for Sulfate Regulation

Decisions to regulate a pollutant require that adequate information be available to facilitate assessment of health and welfare effects and design of a workable control strategy. As evidenced by the previous discussion of health effects and transport, considerable uncertainty exists in interpreting the limited scientific data base on sulfates. Both the SAB and the NAS place high priority on initiation of comprehensive research programs that are needed before a major regulatory control program is possible. This section outlines some of the more significant information gaps relevant to control strategy development and indicates important research needs. A detailed analysis of these needs conducted by the Agency<sup>64</sup> indicates that development of data and information



Table 10. PARTIAL SUMMARY OF POWER PLANT TECHNICAL CONTROL OPTIONS -  
REMOVAL AND ENERGY EFFICIENCY, COSTS, AND TIMING<sup>56</sup>

Extraction	System				SO <sub>2</sub> emissions, lb/10 <sup>6</sup> Btu	Overall system efficiency	Overall cost, mills/kW-hr	Estimated year of application
	Transportation	Processing	Transportation	Utilization				
East coal	Rail	Physical coal clean	None	Conv. boiler	2.02	0.324	10.0	Current
East coal	Rail	Physical coal clean	None	Conv. boiler lime scrub	0.2	0.307	12.4	1975
East coal, S.M.	Rail	None	None	Conv. boiler MgO scrub	0.50	0.349	11.5	1980
West coal, S.M.	Rail	None	None	Conv. boiler	1.65	0.369	11.5	Current
West coal, S.M.	Rail	None	None	Conv. boiler lime scrub	0.16	0.349	13.3	1975
East coal, S.M.	Rail	Chemical coal clean	None	Conv. boiler	1.93	0.350	10.5	1980
East coal, S.M.	Rail	Coal liquefaction	None	Conv. boiler	0.7	0.276	13.2	1985
East coal, S.M.	Rail	Gasify, low Btu	None	Conv. boiler	0.017	0.329		1985
East coal, S.M.	Rail	Fluidized bed combustion	None	Combined cycle	0.07	0.378	10.8	1988
Oil well, onshore	Oil pipeline	Refinery domestic	Oil tanker	Conv. boiler	1.83	0.321	11.2	Current

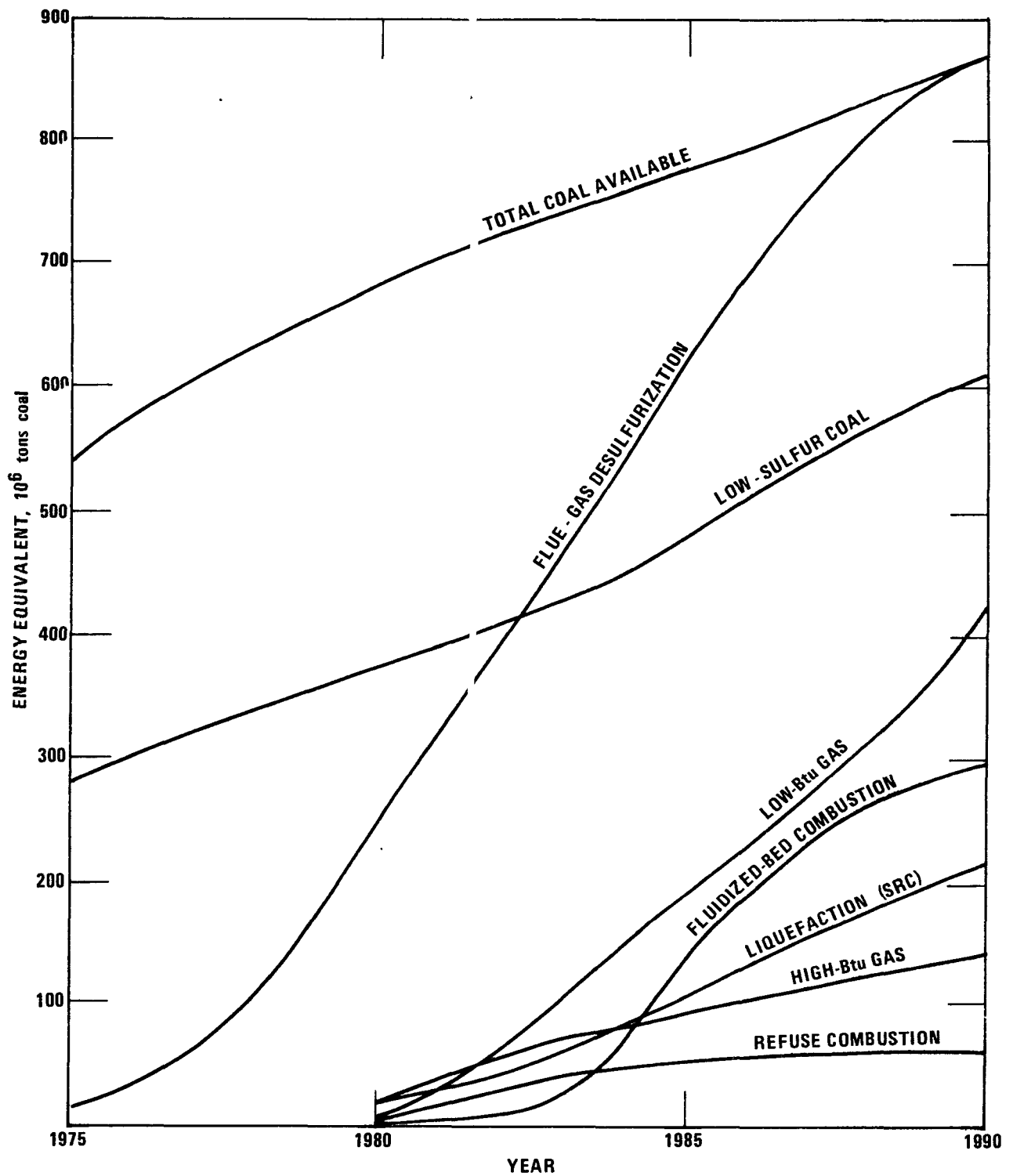


Figure 16. Maximum availability of SO<sub>2</sub> control technologies.<sup>49,53,54,63</sup>

necessary for a sulfate regulatory program would require 3 to 5 years. In this regard, if EPA were to set a National Ambient Air Quality Standard for sulfates it could not realistically be proposed before 1980 to 1981. Important research needs are summarized below.

#### 1.5.1 Monitoring

One of the reasons that such a lengthy period is needed for the research program is that many of the projects cannot begin until after successful completion of other program elements. The most prominent example of this interdependency is the development of reliable monitoring methods for total sulfates, sulfuric acid, and specific sulfates. Such methods may take 1 to 3 years to develop. Important components of epidemiological, toxicological, clinical, and characterization studies are dependent on the use of such monitors. This extends the completion date of the entire effort.

#### 1.5.2 Health and Welfare Effects

Although toxicological data have provided qualitative information regarding the relative effects of sulfur dioxide and specific sulfates, this work must be expanded to provide more quantitative data using additional response indicators. Additional human studies are especially necessary. Animal and human experiments have been performed in static gaseous or particulate atmospheres with no great potential for interaction. Toxicological and clinical studies of the biologic response to gas-aerosol mixtures, which can better simulate the complex physico-chemical environment to which mankind is exposed, are greatly needed.

Despite the accumulation of epidemiological evidence supporting the role of sulfates, there is a paucity of information concerning the role of sulfates in the production of, or exacerbation of, chronic pulmonary disease. Although preliminary epidemiological data support the occurrence of such effects, more comprehensive population studies are needed to substantiate preliminary findings. The etiology of such disease is undoubtedly complex, probably being involved with primary and secondary factors including sulfur oxides, respirable particles, oxidants, smoking,

temperature or humidity variation, and the presence of biological agents such as bacteria and viruses. The specific sulfates responsible for observed health effects should be identified. Characterization should include information on particle size. No studies of sufficient sophistication to fully resolve these complex issues have yet been undertaken.

Studies using available exposure and response assessment techniques should continue, but development of more definitive dose-response information must await the production of instrumentation required to adequately measure ambient pollution both qualitatively and quantitatively.

Although sulfates are suspected to cause significant welfare effects, damage or effect functions have not yet been developed. The role of sulfates in forming acid rain must be determined. The effects of acid rain and dry deposition of sulfate aerosols (particularly sulfuric acid) on crops, terrestrial and aquatic ecosystems, and materials are incompletely understood and remain largely unquantified. Although such information is not needed for health related regulations, the potential magnitude of the effects is such that a significant impact on the outcome of cost-benefit analyses is possible.

### 1.5.3 Atmospheric Chemistry and Transport

More work is needed to develop sulfate-precursor relations to the extent that they can be used to determine the need for control of precursors other than  $\text{SO}_2$ . This kind of work should be pursued as soon as possible because the conclusions that may be drawn will govern the directions of other control technology and strategy development programs that may be in progress.

As indicated above, although part of the work that has been done to date on the physics, chemistry, and transport of sulfates has been in the area of sulfate- $\text{SO}_2$  conversion rates for power plant plumes and urban areas, the current status of knowledge is deficient in several respects. Some of the open questions are as follows:

1. It is possible that the plume from a nonurban coal-fired power plant crossing an urban area mixes with precursor agents that significantly accelerate the rate of conversion of sulfur dioxide to sulfate.

Assessment of this possibility is difficult because plume breakup is more likely to occur in the city, and the sulfur dioxide from the plume cannot be distinguished from the sulfur dioxide from low-level combustion sources. However, certain experimental approaches may allow an estimate of the relative contribution of plumes to the overall urban sulfate levels on a day-by-day basis. Several study sites are needed to evaluate photochemical effects as well as effects of particulate composition on rates of conversion of sulfur dioxide to sulfate.

2. Limited experimental results indicate that within some types of plumes from oil-fired power plants, sulfur dioxide can be rapidly converted to sulfates. Plumes from several additional oil-fired plants need to be studied, and the influence of particulate control should be investigated.

3. Little experimental work is available on the low level urban plume. Because power plant plume contributions from upwind break up in the city, downwind of the city the urban plume containing the sulfur dioxide and sulfate from all emission sources may be a problem. We cannot fully assess the relative rates of conversion of sulfur dioxide to sulfate compared to the rate of removal of the sulfur dioxide by dry deposition or precipitation. Possible sinks for sulfates in urban and nonurban areas need to be identified.

4. The build-up of regional sulfates from numerous rural and urban sources under stagnating conditions, although possible, has not been investigated. Estimates of the impact natural sources of hydrogen sulfide and organic sulfides exact need to be quantified because under certain conditions these might also be rapidly converted to sulfur dioxide and then to sulfate. Also unassessed is the influence of natural hydrocarbons and nitrogen oxides in sulfate formation during periods of high photochemical activities.

These areas must be addressed in order to develop models that can be used to estimate both local concentrations and long-range transport phenomena for those components of sulfates which are important in producing adverse effects. Such models are essential in the evaluation of control strategies.

#### 1.5.4 Improve Control Technologies for Sulfur Dioxide and Sulfates

If, as has been implied, a high degree of control for large point sources of sulfur dioxide is needed to adequately control sulfates in some parts of the country, the efficiency required of currently demonstrated scrubbing technologies might be greater than 90% removal. Modification of current technologies to demonstrate greater than 95% removal efficiency could be accomplished by 1978. Alternative combustion technologies showing promise of high efficiency, such as fluidized bed systems, should be evaluated and encouraged. Advanced precombustion technologies and control for small point and area sources of  $\text{SO}_2$  may be necessary and should be developed. Sulfur traps or alternate hydrocarbon and carbon monoxide controls may be needed for post-1978 automobiles to control emissions of sulfuric acid mist.

## 2. POTENTIAL REGULATORY STRATEGIES

The review of current scientific and technical knowledge indicates that although a major regulatory initiative for sulfates may not be possible for several years, the strategies that are ultimately developed probably must take into account health and welfare effects at low concentration levels, emissions/concentration relationships complicated by complex formation mechanisms, and long-range transport. Although scientists and engineers proceed with research directed toward filling significant information gaps, strategy analysts must investigate the regulatory options that, based on current information, appear likely to be needed. This section outlines the considerations that must be examined in the context of available and possible regulatory alternatives. It is not to be construed as an Agency regulatory policy for sulfates. Policy implications are discussed later in the report.

### 2.1 Potential Scope of the Problem

The goal of any regulatory approach to sulfate control should be the protection of health and welfare from significant harm. Because the acute and chronic effects levels from the health studies listed in Table 2 differ considerably with regard to the degree of control necessary for achievement, strategy decisions may have to consider the benefits derived from attainments of various levels. In the long term, no segment of society should suffer the effects of illness unnecessarily. However, 24-hr maximum sulfate concentrations at western sites (where annual levels approximate the global background of 1 to 3  $\mu\text{g}/\text{m}^3$ ) can on occasion be in the range tentatively associated with sensitive health indicators. Thus, protecting the most sensitive portion of the population could ultimately involve  $\text{SO}_2$  control in excess of that required to meet current  $\text{SO}_2$  standards. Any attempt to achieve such levels of control in the short term (3 to 5 years after initiation of sulfate regulatory control) could result in failure or unacceptable social and economic consequences.

A recent analysis of strategies<sup>65</sup> assumed proportionality between  $\text{SO}_2$  emissions and regional sulfate levels to determine the maximum degree of control that might be needed to meet various presumed effects

levels. The study concluded that continuous control of all major SO<sub>2</sub> sources might be insufficient to achieve sulfate levels presumed needed to protect the most sensitive portions of the population in some areas. In addition, controls somewhat in excess of those anticipated for SO<sub>2</sub> standards could be needed to prevent excursions possibly related to mortality and to protect the general population from increased respiratory ailments. The cost of the incremental control technology assumed necessary to achieve this level of control in the northeastern U. S. totaled nearly \$30 billion over a 10-year period from 1981 to 1990.

If economic and social factors are to be considered in development of a sulfate control strategy, a phased reduction of sulfate concentrations over an extended period of time is likely to be necessary. The degree of SO<sub>2</sub> control applied within a region at a given time depends upon the following factors: (1) efficiency, availability, and cost of SO<sub>2</sub> control technologies for existing and new sources, (2) SO<sub>2</sub> emission density characteristics, (3) chemical and physical characteristics of existing sulfate levels, (4) influence of primary sulfuric acid emissions by catalyst equipped vehicles, (5) levels of other precursors, (6) population at risk from existing levels, (7) meteorology, and (8) significant welfare effects.

Because current knowledge precludes adequate assessment of the above factors, evaluation of preliminary sulfate control strategy development must include a sensitivity analysis of the significant unknowns. Studies done to date indicate that among the most sensitive parameters relative to strategy development is the uncertainty regarding the oxidation rate of SO<sub>2</sub> in plumes and under varying ambient conditions. Estimates of oxidation rates for power plant plumes range from negligible to 20% or more per hour. The sensitivity of diffusion model estimates for sulfate increments from a typical power plant to the plume oxidation rates is shown in Table 11. The difference between a 1% and a 5% oxidation rate produces significant differences in incremental sulfates at distances of 5 to over 50 km downwind. Because a low oxidation rate increases the removal potential of SO<sub>2</sub> before it can form sulfates, estimates of the degree of long range transport can be affected. North<sup>65</sup> analyzed the sensitivity of an oxidation rate of 0.5 to 2%/hour. The differences between the



rates resulted in a tripling in predicted sulfate increments over a wide front nearly 500 km downwind. Sound decisions as to the degree of control required for such a power plant might depend upon whether the plant was located in a rural or urban situation and could not be made without further information on oxidation rates and long-range transport even if health effects information were well developed.

Table 11. DIFFUSION MODEL SENSITIVITY OF SULFATE  
FORMATION TO SO<sub>2</sub> OXIDATION RATE  
FOR 1500-MW POWER PLANT<sup>a</sup>  
(24-hr sulfate concentration, µg/m<sup>3</sup>)

Distance, km	Conversion rate, %/hr		
	1	5	40
5	1	2	21
10	1	4	38
20	1	6	50
30	1	6	54
40	1	7	54
50	1	7	54

<sup>a</sup>Gaussian dispersion model applied to a typical large power plant under conditions of poor dispersion.

Only qualitative conclusions can be drawn concerning the degree of SO<sub>2</sub> control required for a possible sulfate strategy. Based on extrapolations of current knowledge, it is possible that sulfate control would require greater SO<sub>2</sub> reductions from all sources than is required to achieve SO<sub>2</sub> standards. In addition, distinct differences exist between various regions with respect to the eight criteria referred to above as necessary assessments of degree of control. The western United States clearly requires less sulfate control than does the east. Regions, subregions, and even urban areas within the east also differ in their need for control. Sulfate control strategies might recognize these differences by developing sulfate control regions based on emission, concentration, and transport patterns, and by prioritizing allocation of

limited control resources. The NAS<sup>2</sup> recognized these considerations in presenting a preliminary methodology for prioritizing major sources according to their need for control. This technique was useful in illustrating the crucial scientific information gaps and providing the general conclusion that power plants located in or near urban areas with elevated sulfates or SO<sub>2</sub> should be given high priority with regard to control. Beyond these general considerations however, the technique is of limited utility for decision-making until significant new research is completed.

## 2.2 Regulatory Options Under the Clean Air Act

The previous section presented a preliminary assessment of some important considerations associated with development of technically feasible control strategies for sulfates. In this section, the implication of these considerations with respect to available regulatory approaches is examined.

### 2.2.1 National Ambient Air Quality Standards

Under the Clean Air Act of 1970 the Administrator of EPA may establish a National Ambient Air Quality Standards (NAAQS) for "any air pollutant which in his judgment has an adverse effect on public health and welfare," and results from emissions of "numerous or diverse mobile or stationary sources." Simultaneous with the publication of health and welfare criteria, primary and secondary NAAQS are set, respectively, to protect public health with an adequate margin of safety and to prevent known or anticipated adverse welfare effects. The Clean Air Act does not allow consideration of costs of achieving the standard or benefits derived from the standard to influence the determination of the maximum permissible exposure level for a pollutant. As indicated in Section 1.5, if an air quality standard for sulfate was determined to be necessary, it could not be set before 1980-81.

Once standards are set, a time phased sequence begins. The standards are promulgated 90 days after publication in the Federal Register. Within 9 months, each state would be required to submit a state implementation plan (SIP) that would provide for implementation, maintenance,

and enforcement of the NAAQS for each air quality control region (AQCR) within that State. Guidelines issued at the time of promulgation would specify emission reductions required to achieve the prescribed ambient pollutant level. Each state implementation plan must be approved or disapproved by EPA within 4 months of submission. Achievement of the primary standard must occur within 3 to 5 years after SIP approval.

Although sulfates exhibit those characteristics described above for pollutants to be controlled through NAAQS, some implementation difficulties associated with this approach are anticipated. Because an air quality standard requires that implementation plans include measures that will ensure attainment of the standards within each AQCR, NAAQS implementation generally has required the use of a pollutant air quality/emission control relationship within that AQCR's boundary. Complex sulfate precursor formation mechanisms and transport across multistate regions, however, are likely to make such a relationship impossible within current AQCR boundaries. AQCRs with essentially no SO<sub>2</sub> emissions could exceed the standard because of sulfates imported from other regions. Areas with high SO<sub>2</sub> emissions and low imported sulfate levels might apply limited control to meet local air quality goals at the expense of downwind areas. The present AQCR boundaries were fixed by the Clean Air Act and are, in general, far too small to permit implementation of a NAAQS for sulfates in areas where long-range transport is significant.

Another possible problem is the definition of a "threshold" level for sulfates below which no serious health or welfare effects occur. Preliminary health studies have identified possible effects at near background levels, suggesting that the threshold concept may be inappropriate. In addition, achievement of very stringent control requirements might require compliance extensions beyond the 3- to 5-year period required by the act.

The effectiveness of a sulfate NAAQS for protecting health would depend in part upon the degree of discrimination possible in the health effects criteria and precursor mechanisms. As more information about specific sulfate effects becomes available, alternative standards for sulfuric acid emissions, or combined standards that take into account

synergism between sulfates and other pollutants, such as nitrates, might be set. A better understanding of complex precursor relationships is needed before emission-ambient guidelines similar in approach to those for oxidants can be established. If the above considerations cannot be resolved, evenhanded implementation of a sulfate standard in all areas of the country would produce uneven levels of protection from adverse effects, and in some cases unnecessarily stringent controls.

#### 2.2.2 New Source Performance Standards

The Clean Air Act requires that EPA set emission standards for new or modified sources that may "contribute significantly to air pollution which causes or contributes to the endangerment of public health or welfare." The emission standard for a given pollutant must reflect the best degree of available control technology, considering cost, that the Administrator believes has been adequately demonstrated. If a NAAQS has been set for the pollutant, only new sources are affected. For a non-NAAQS pollutant, existing sources of the same type may be required to meet emission standards through a SIP process similar to that described for NAAQS. If the state plan is approved, existing sources must be in compliance within 3 to 5 years. If the state plan is unacceptable, EPA must enforce the New Source Performance Standards (NSPS).

Although sulfate is a non-criteria pollutant, NSPS for the principal precursor,  $\text{SO}_2$ , already exist for steam generating plants and sulfuric acid production plants. Strengthening existing NSPS for large, fossil-fuel-fired boilers would affect only new plants and could be used as part of an interim approach until more comprehensive action is possible. Conceptually, a NSPS for total sulfur emissions as measured by a technique that would collect all  $\text{SO}_x$ , might allow retrofit of existing sources. If this approach were possible for sulfates, retrofit of existing sources would be implemented by the states, subject to EPA approval. The current NSPS for  $\text{SO}_2$  for steam generators is 1.2 lb/million Btu heat input and reflects the 70 to 80% removal efficiencies currently demonstrated.

Given the less stringent need for health criteria, an emissions standard under the NSPS section of the Act might be set 1 to 3 years

earlier than a NAAQS, or about 1978. According to the sulfate research program outlined previously, greater than 95% control efficiency with flue gas desulfurization probably could be demonstrated by that time. A revised NSPS for  $\text{SO}_2$  at this time could be used as an interim approach to limit growth in emissions past 1981. Although it is unlikely that the NSPS section of the Act could be used as the primary regulatory mechanism for sulfate control, use of this approach could avoid some of the implementation difficulties associated with a NAAQS and preclude the necessity for major statutory changes. Because costs are considered in setting a NSPS, the consequences of plant shut downs and excessive expenses associated with installing devices with control efficiencies greater than 95% could be considered. The timing associated with controlling existing sources may still present some problems, but the degree of control required can be adjusted to existing technological capabilities rather than to meeting specified ambient levels. The size of AQCR's and effect of precursors or imported sulfates would not cause SIP development difficulties.

More serious troubles with the use of the NSPS approach as the primary regulatory control method occur in the effectiveness with which health and welfare is protected. A national NSPS for given sources would disregard the regionality of the sulfate problem and result in application of stringent control measures in areas where sulfates were not a problem. The standard for new sources would have to be lenient to the extent that the best demonstrated control techniques must be generally adaptable everywhere. For example, if 95% sulfur reduction is the maximum control possible for generally available coals, a new electric plant that could economically scrub and use medium sulfur coal for 98% control would not be required to do so, because this would not be generally feasible at all new plants. Although cost-benefit considerations would suggest tighter standards in regions with more serious sulfate problems, a uniform standard would ignore this. Conversely, areas with minimal sulfate problems would be overcontrolling both new and existing plants, diverting needed control resources that might better be applied in other regions. Because use of a "total sulfur" NSPS approach for sulfates

control could preclude major statutory changes that otherwise might be needed, however, mechanisms for implementing this alternative should be investigated.

#### 2.2.3 National Emission Standards for Hazardous Pollutants

A non-criteria pollutant that in EPA's judgment "may cause, or contribute to, an increase in mortality or an increase in serious irreversible, or incapacitating reversible, illness," may be controlled by National Emission Standards for Hazardous Pollutants (NESHAP).

This option is best used for relatively rapid control of hazardous pollutants without regard to cost or available technology. This is an unlikely regulatory path for sulfates.

#### 2.2.4 Emission Standards for Mobile Sources

The Act requires the Administrator to set emission standards for any air pollutant coming from motor vehicles if the pollutant is harmful to public health and welfare. This can be done by requiring installation of emission control devices or regulation of fuels content.

The Administrator has determined that the health risk from predicted incremental concentrations of fine particulate sulfuric acid from catalytic converters represents a serious health risk and has indicated his intent to propose an Emission Standards for Mobile Sources (ESMS) for sulfuric acid. Desulfurization of gasoline, use of alternative hydrocarbons and nitrogen oxide controls, and application of exhaust particulate control devices are possible options for sulfate control under ESMS. EPA is currently encouraging the development of alternative HC and NO<sub>x</sub> controls. Implementation would be uniform nationally except for California, which generally has stricter controls mandated by the state.

#### 2.2.5 Other Clean Air Act Options

Although application of Abatement Conferences and to a lesser extent emergency powers is possible for sulfates, such actions are anticipated to be of minor significance in a national sulfate control

strategy. However, Abatement Conferences could be of use in dealing with the international long range transport of sulfates and acid rain between Canada and the United States.

The Clean Air Act was amended by the Energy Supply and Environmental Coordination Act of 1974 (ESECA). Under Section 119(d), a federally ordered prohibition resulting in the conversion of a fuel burning stationary source from gas or oil to coal may be suspended if resultant emissions of a non-regulated pollutant "will result in an increase which causes (or materially contributes to) a significant risk to health. . ." This provision is clearly applicable to sulfates. Implementation of this section of the Clean Air Act is currently being reviewed and will involve limiting increases in SO<sub>2</sub> emissions from converting plants in areas where elevated sulfate levels pose a public health concern.

#### 2.2.6 Alternate Regulatory Options

Because implementation problems in applying existing Clean Air Act options to sulfate regulation are anticipated in the next few years considerations should be given to modifications to the current approaches. The regulatory alternatives should attempt to allow efficient implementation of the most effective control strategies available without unnecessary adverse social and economic consequences. Also, some means for recognizing the impact of long range transport of sulfate is required. Consideration of two potential alternative regulatory mechanisms is presented in this section.

##### 2.2.6.1 Regional Emission Control

The implementation problems outlined above might be resolved by dividing the nation into sulfate control regions based on the severity of the sulfate problem and emissions transport patterns and by proposing regionally specific SO<sub>2</sub> emission standards for new and existing sources. Limited control resources could be used first in the regions with the most serious acid-sulfate problems to ensure greatest possible reduction of sulfur oxide emissions where it is most needed.

A regional sulfate control program incorporating the features outlined above could be implemented by a statutory mandate of  $\text{SO}_2$  emission reductions similar to the approach taken for hydrocarbon automotive reductions. For example, the 24-state region east of the Mississippi with the highest nonurban sulfate levels (Figure 6) could be required to install stringent continuous  $\text{SO}_2$  emissions control by 1982. In regions with less serious sulfate problems, control might be deferred 3 to 6 years later. The degree of control could reflect regional variations in transport, formation, and emissions density considerations. Mandated reductions might take the form of best available control technology (BACT) considering costs for both new and existing sources. Ultimately, as the effectiveness of control options (such as gasification and solar energy) increased,  $\text{SO}_x$  emissions in all regions would trend downwards as older sources with poorer control were phased out. Assuming requirements for health criteria are less stringent than for a NAAQS, a regional emissions reduction program might begin in 3 to 4 years.

In the short term, regional control differences might encourage expansion of sulfur oxide producing sources in areas where requirements are less strict. Even though revised NSPS would restrict this growth, regulations to prevent significant deterioration from sulfates might be necessary.

#### 2.2.6.2 Emissions Tax as Component of Sulfates Control Strategy

The concept of an emission tax has often been discussed as an efficient means of controlling sulfur oxides, although it has usually been dismissed as impractical. The unique characteristics of the sulfate problem previously discussed make such a tax disincentive worthy of serious consideration.

Generally speaking, an  $\text{SO}_x$  emissions tax might be useful in implementing an overall sulfate control strategy such as regional emission control that allows for a somewhat lengthy and flexible time for reduction of ambient concentrations. A sulfur oxides emissions tax would be desirable in this context because it would promote an orderly industry response to the availability of  $\text{SO}_x$  control technology. The tax would provide the



economic incentive necessary for voluntary implementation of available control technologies. In a practical vein, the tax would eliminate some of the problems EPA has faced in convincing utilities to undertake research on and installation of flue gas desulfurization equipment. With the tax provision, tactics to delay compliance would no longer be cost free.

Probably the most cogent argument for use of an  $\text{SO}_x$  emissions tax for sulfates control lies in its cost efficiency. An emissions tax, if given sufficient time to work, should result in an aggregate level of emissions reduction at uniform marginal control cost and, consequently, minimum total cost. The cost involved in achieving levels of sulfate control approaching a low "threshold" health effects standard makes the attribute of economic efficiency very important. A further argument for the efficiency of the tax derives from the regionality of the sulfate problem. In the case of  $\text{SO}_2$  abatement, the objective of specific control strategies is to control localized ground level concentrations. This localized feature of the  $\text{SO}_2$  problem would tend to reduce the value of a uniform regional  $\text{SO}_x$  emissions tax because the "regions" of control should be quite small, with source parameters playing a major role in tax determination. An optimum  $\text{SO}_2$  control tax would be determined on a source specific basis. Assuming long-range transport, however, the wide area nature of the sulfates problem should reduce the "error" on a uniform tax structure.

Problems with this approach include the possibility that utilities would pass the tax on the costs to consumers without installing controls, the increased costs of electrical production above that required for control, the political palatability of an added tax, and the difficulty in altering tax rates to reflect changing cost-benefit information.

To summarize, an emissions tax sulfate control strategy could provide a relatively cost-efficient means to gradually reduce sulfate concentrations through orderly development and introduction of  $\text{SO}_x$  controls and should be investigated as a possible strategy. The National Academy of Sciences<sup>2</sup> also recommends that this approach be the subject of further study.

### 3. POLICY IMPLICATIONS

In view of the available data, it is the judgment of EPA that an air quality standard or other major regulatory program for sulfates is not supportable at this time. In order to fill the information gaps described earlier, EPA is expanding its sulfate research effort. The research will focus on improving monitoring capability to permit identification of particle size and chemical form of toxic sulfates, developing more comprehensive health effects data, and characterizing the long-range transport and transformation mechanisms. The research program will require several years to complete; consequently, it is doubtful that a comprehensive regulatory program specifically for sulfates could be initiated before the end of the decade. The recent reports by the National Academy of Sciences<sup>2</sup> and the Science Advisory Board<sup>1</sup> support EPA's position that considerable research and data development must precede such a regulatory program for sulfates.

Nevertheless, until further research makes a comprehensive regulatory program possible, EPA must respond to the potential sulfate problem suggested by the preliminary sulfate/health effects information cited earlier. Although considerable uncertainty exists concerning the relationship between ambient sulfate concentrations and adverse health effects, the preliminary health effects information can be useful in identifying areas of potential health concern.

As described previously, a large portion of the northeastern United States is experiencing annual sulfate concentrations that are relatively higher than concentrations observed throughout the remainder of the country. Figure 17 illustrates a 24-state region in which nonurban concentrations have averaged in excess of  $10 \mu\text{g}/\text{m}^3$ . The average of urban concentrations has been about  $14 \mu\text{g}/\text{m}^3$  (annual average). The boundary of this 24-state region has been drawn on the basis of observed sulfate patterns,  $\text{SO}_2$  emission density, and likely prevailing transport patterns. This area of high sulfate concentrations also correlates spatially with high rainfall acidity patterns and a high density of power plant locations. Furthermore, the region exhibits widespread violation of the national primary ambient air quality standard for suspended particulate matter, a potential precursor agent in the

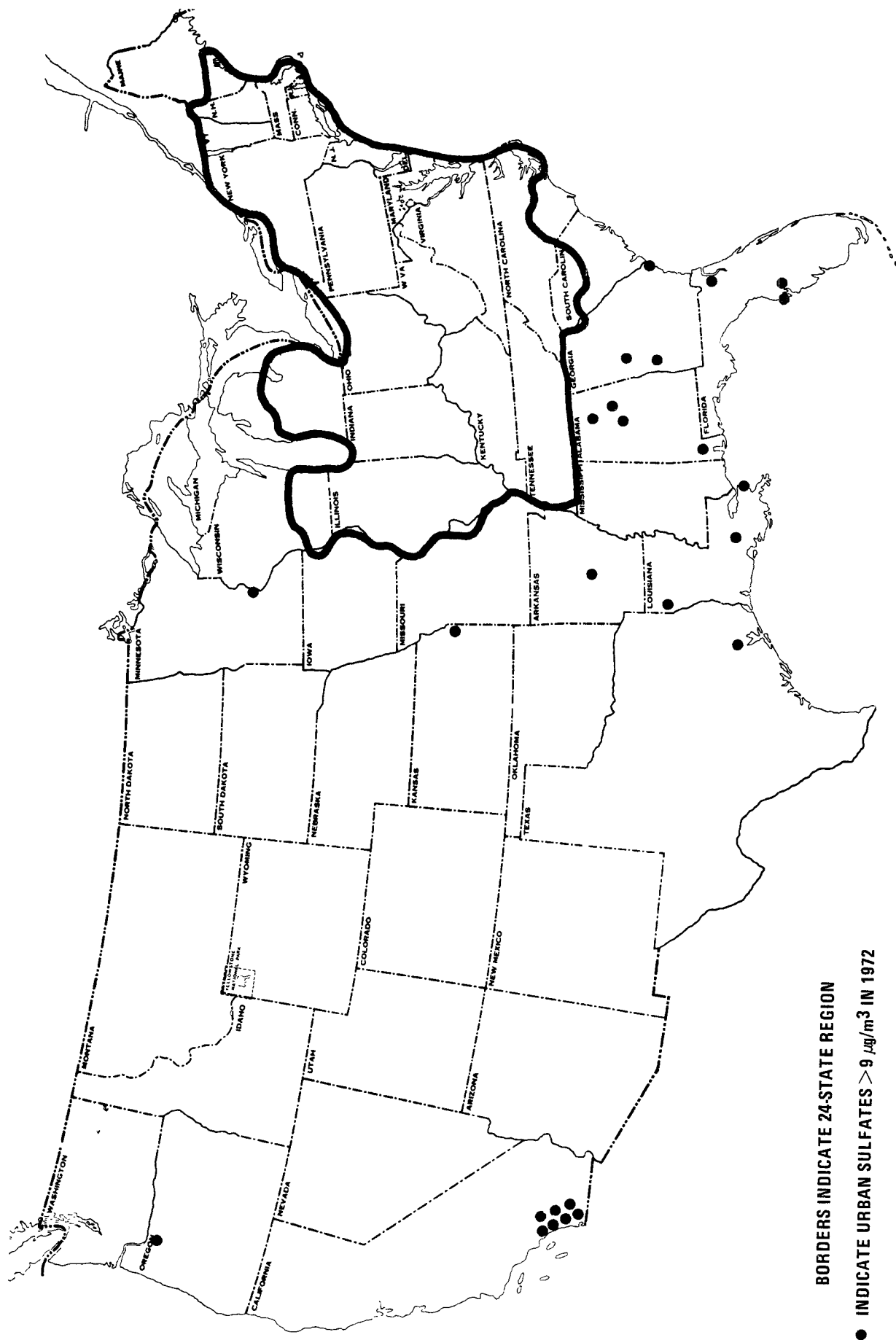


Figure 17. 24-state region with high sulfate levels.

formation of sulfates. The relatively high levels of nonurban sulfate and rainfall acidity in this region suggest the existence of a general sulfates problem. In addition, these high nonurban concentrations are potentially indicative of the background sulfate levels entering urban areas. Recognizing that these more populous urban areas add locally formed sulfate to this general background gives further support to the concern for the health impact of sulfates in this 24 state region. High sulfate concentrations have also been observed in several isolated urban areas in the remainder of the country.

Given the significant potential for sulfate related health risk due to the multiple influence of high sulfate concentrations, high precursor concentrations, and high SO<sub>2</sub> emission density, prudence dictates that EPA adopt a policy of avoiding aggravation of existing conditions by minimizing increases in the relatively high sulfate levels in the northeastern United States and other more localized problem areas. In addition, close attention must be paid to sulfate trends in areas of lower sulfate concentration. Although the goal of avoiding sulfate increases will primarily be achieved by minimizing SO<sub>2</sub> emission increases, existing programs for control of pollutants such as oxidants and particulates may provide some measure of sulfate control by limiting formation processes.

Opinions differ over where, and to what extent, SO<sub>2</sub> emissions should be limited to adequately address the potential sulfate problem. There is general agreement that SO<sub>2</sub> emissions increases should be avoided in or near urban areas where ambient concentrations of sulfates, SO<sub>2</sub>, or total suspended particulates are high. The NAS<sup>2</sup> places high priority on abating SO<sub>2</sub> emissions from sources located in or near urban areas with high concentrations of sulfur dioxide and sulfates. In addition, the NAS<sup>2</sup> is concerned about the effects of area-wide SO<sub>2</sub> emissions increases on regional sulfate levels and in a separate report<sup>60</sup> cautions "further increases in the ambient concentration of these pollutants through the relaxation of standards of emissions and air quality would pose an unacceptable risk for the population." The SAB, however, does not share this same concern for the impact of area-wide SO<sub>2</sub> emissions. Rather, the SAB suggests that increased SO<sub>2</sub> and other sulfate precursor emissions may have primarily a local impact on sulfate formation, and "that increases in exposure to sulfur oxides or particulates in localities

where sulfur dioxide and/or total suspended particulates exceed primary standards would be viewed with grave concern."

EPA considers the points addressed in both reports as important and essentially compatible with the Agency's assessments. Current efforts to attain the primary standard for the criteria pollutants are directly responsive to the SAB's primary concerns. In addition, EPA believes that the area-wide concern must be addressed by minimizing increases in SO<sub>2</sub> and other sulfate precursor emissions in the areas of maximum sulfate impact. A strategy of minimizing regional and local increases in SO<sub>2</sub> emissions can be implemented through existing regulatory options such as State Implementation Plans (SIPs) and New Source Performance Standards (NSPS).

A policy of minimizing SO<sub>2</sub> emission increases is generally consistent with other Agency policies previously announced. These policies include the Clean Fuels Policy, the application of intermittent control systems (ICS), the significant risk aspect of oil-to-coal conversions, and the recent deferral of more restrictive hydrocarbon/carbon monoxide vehicle emission standards.

The EPA Clean Fuels Policy is intended to make it unnecessary for plants to switch to lower sulfur fuels to comply with state regulations where such compliance is not needed for attainment and maintenance of the national health-related standards for SO<sub>2</sub>. By revising SIPs appropriately, plants currently in areas meeting primary air quality standards could continue to burn currently available fuels; no switch to higher sulfur fuel is intended. Therefore, sulfur emissions from these sources should not increase.

With respect to EPA policy on intermittent controls systems (ICS), a limited number of isolated power plants may be permitted to use intermittent emission control to meet air quality standards, temporarily deferring installation of costly continuous emission controls. Eligible plants are already burning coal and, under ICS, will continue to burn existing fuel except during adverse meteorological conditions when they will reduce emissions by switching to a lower sulfur fuel or shifting generation load. Again, total sulfur emissions from these sources should not increase and may actually be slightly reduced.

The Energy Supply and Environmental Coordination Act of 1974 (ESECA) provides the Federal Energy Administration with the authority to prohibit

a power plant from burning oil or natural gas subject to certification by EPA of the power plant's ability to burn coal in compliance with certain environmental requirements. Two requirements relate to a plant's ability to burn coal without contributing to a violation of primary standards for total suspended particulate or sulfur dioxide. In this regard, if certain legal criteria are satisfied (regional limitation), EPA may specify alternate emission requirements (primary standard conditions) to be met temporarily by converting plants. An additional requirement of ESECA states that conversions cannot result in an increase in the emission of unregulated pollutants or pollutant precursors at levels that may result in a significant risk to public health. Based on currently available health effects information, EPA has decided to apply this "significant risk" provision only with respect to sulfates. Under the significant risk provision, EPA plans to restrict emissions of sulfate precursor pollutants--sulfur dioxide or particulate matter--if a converting plant is located in an area with high sulfate concentrations and with concentrations of sulfur dioxide or particulate in excess of primary standards. This policy recognizes the concerns of both the NAS and the SAB and is believed to provide adequate protection against increased health risk due to sulfates by significantly limiting potential SO<sub>2</sub> emission increases.

EPA has analyzed the effect of existing regulations, such as SIPs and NSPS, and the policies described above on sulfur dioxide emissions in the 24-state region. Table 12 summarizes the assumptions made in the analysis. Emission levels have been evaluated for 1972, the most recent year for which area sulfate information is available, and 1980, the time at which a sulfate regulatory control program might begin. Table 13 lists the regional emissions for 1972, broken down into utility and non-utility sources, and illustrates the effect of each regulatory option on emissions in 1980. Each subsequent option listed assumes that each of the previous options was implemented for the relevant sources.

During the period 1975 to 1980, it is assumed that both utility and non-utility sources will move to compliance with SIP regulations. Because most non-utility point and area sources are located in urban areas, these sources are assumed to comply with SIPs by continuous emission

Table 12. SUMMARY OF ASSUMPTIONS USED FOR SO<sub>x</sub> EMISSIONS  
PROJECTIONS OF TABLE 13

- 1972 Emissions from NEDS<sup>25</sup> and SASD files
- Growth:
  - Utilities<sup>67</sup> - Federal Power Commission Data
  - Non-utilities - Based on state-specific OBERS<sup>68</sup> population and economic activity projections
- Effect of SIPs on 1975 non-utility emissions determined using state-specific fuel use and SIP requirements
- Effect of SIPs on utility emissions determined using 1980 plant-specific SIPs. Future revisions in SIP limit were assumed for some plants in Illinois, Indiana, Kentucky, and Ohio.<sup>67</sup>
- NSPS in effect on utility growth after 1976
- ICS Case:
  - Certain isolated plants continue to burn existing coal meeting ~~SO<sub>2</sub>~~ standards by use of ICS if considered enforceable according to a range of criteria [29 plants (25,500 MW) to 63 plants (54,500 MW) qualify under the assumed criteria]
  - All other sources meet SIPs
- Oil to coal conversions: 48 plants convert and meet primary standard conditions

Table 13. 24-STATE REGION SO<sub>x</sub> EMISSIONS  
(10<sup>6</sup> tons/yr)

	Utilities	Non-utilities	Total
1972 Emissions	14.3	8.1	22.4
1980 Emissions			
Regulatory options:			
Enforcement of SIPs, NSPS	10.9	7.7	18.6
ICS (29 to 63 plants)	12.5 - 14.9	7.7	20.2 - 22.6
Conversions <sup>a</sup> (48 plants)	12.5 - 14.9	7.7	20.2 - 22.6

<sup>a</sup>The conversion program requires all candidates to meet SIP before 1980. Thus the maximum impact of the program (0.7 million tons SO<sub>2</sub>/yr) is assumed to last from 1976 to 1979 and is not apparent in 1980.

control methods and thus are not affected by the other policies. Even when accounting for source growth,  $\text{SO}_2$  emissions from these sources are reduced by 0.4 million tons/yr in 1980 through compliance with SIPs. Similarly, if all utilities were to meet applicable SIPs and NSPS, substantial emissions reductions (about 5.9 million tons/yr) would result. However, because control options in the near term are limited and costly, the Clean Fuels Policy has been developed to allow existing power plants to continue the use of currently available coal supplies while still meeting primary standards. If affected states revise their SIP regulations to conform with this policy, emissions would still be reduced by 3.4 million tons/yr in 1980. If additional SIP revisions were to occur in this region, part or all of the scheduled reductions in power plant emissions could be eliminated. SIP revisions do not affect the requirement that new power plants meet NSPS, which will significantly limit emissions growth past 1975-1976.

With regard to the analysis of the intermittent control system option, the majority of existing fossil-fuel-fired power plants in the 24-state region (about 360 plants representing 160,000 MW in 1975) are located in urban or other areas in which continuous emission control is required to meet  $\text{SO}_2$  standards and SIPs. However, a small number of coal-fired power plants are in isolated locations and could defer the use of limited and costly continuous emission controls by using intermittent emission control systems (ICS) to meet  $\text{SO}_2$  ambient standards. Assuming that such plants would continue to burn current fuels except during periods of poor atmospheric dispersion,  $\text{SO}_x$  emissions should not increase from these plants. Using a preliminary screening procedure, 29 plants (25,500 MW) to 63 (54,500 MW) in the 24-state region have been identified as possible candidates for the use of ICS. The range of emissions resulting if all of the possible candidates used ICS is presented in Table 13. Under these assumptions, total emissions in 1980 would still remain at or below 1972 levels.

Concerning the analysis of the potential  $\text{SO}_2$  emission increase due to the ESECA program for conversion of power plants from oil or gas to coal operation, 49 plants, representing 18,300 MW of generating capacity, are assumed to convert by 1980 in the 24-state region. Because technical and economic considerations may limit the number of conversions



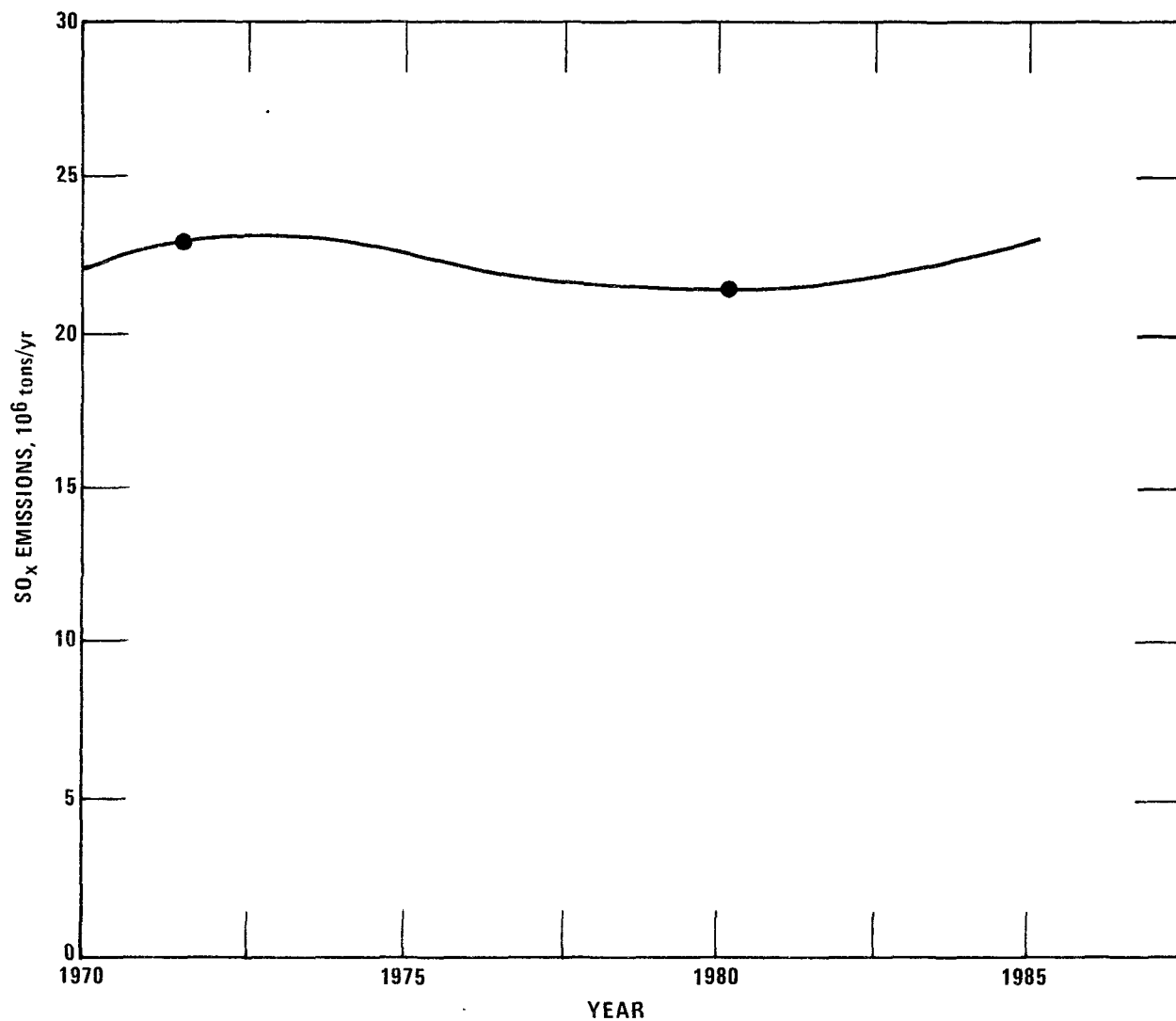


Figure 18. Projected SO<sub>x</sub> emissions from 24-state region, 1970-1985.

increases in atmospheric sulfate levels. Present EPA policies that affect sulfur dioxide emissions are consistent with a sulfate-related policy of minimizing sulfur dioxide emissions increases in high sulfate regions. The "significant risk" policy for converting power plants, the vigorous enforcement of state implementation plans for the control of sulfur dioxide and particulate, and the increasing application of new source performance standards to power generating facilities are the vital components of a broad strategy that should limit growth of ambient sulfate levels. As the previous EPA analysis indicates, these regulations and policies should prevent major SO<sub>2</sub> emission increases through 1980 in regions of maximum sulfate impact. Until information is available to support the enforcement of a more rigorous sulfate regulatory program, the use of these currently applicable regulatory measures should provide reasonable protection against increased health risk from sulfates.

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15 SUPPLEMENTARY NOTES		
16. ABSTRACT <p>Atmospheric sulfates as measured include a variety of chemical entities. Toxicological evidence indicates that certain sulfates, particularly fine particulate acid sulfates, are more potent respiratory irritants than sulfur dioxide alone. Preliminary epidemiological studies suggest that measured sulfates are associated with a variety of health indicators. Sulfates may also be related to damage to the environment by direct deposition or by formation of acid rain and can cause visibility deterioration. Although natural sulfur emissions are important on a global scale, sulfates in industrialized regions are largely produced by atmospheric reactions of manmade sulfur oxides emissions. Sulfates may be transported long distances from source areas and result in high ambient levels over broad regions. This is apparently the case in a 24 state region in the eastern U. S. Considerations of chemistry and transport suggest that reductions in regional SO<sub>2</sub> emissions would produce reduction in sulfates, although the reductions would be less than one to one.</p> <p>Considering the uncertainties in scientific information, it will take 3 to 5 years of additional research before a major sulfate regulatory program is possible. Until then, present concerns dictate that EPA follow a policy of avoiding increases in SO<sub>2</sub> emissions and consequent sulfate increases in the 24 state region and other areas subject to high sulfate levels. This policy can be implemented through the existing authority of the Clean Air Act.</p>		
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