Design of the Sulfate Regional Experiment (SURE)

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Dr. Morton Corn, University of Pittsburgh (Resigned October 1, 1975)
Mr. T. T. Frankenberg, American Electric Power Service Corporation
Mr. Charles E. Hickman, Southern Services, Inc. (Appointed June 20, 1975)
Mr. Marcus R. McCraven, United Illuminating Company
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EPRI FOREWORD

A planning study for EPRI's research program on the regional transport, conversion, and fate of air pollutants - Design of the Sulfate Regional Experiment (SURE) - has been conducted by Environmental Research & Technology, Inc. under EPRI contract number RP485. The results of this effort are presented in four reports:

- an executive summary
- general text describing the technical approach and interpretations of supporting analyses ("Volume 1")
- the proposed experimental design ("Volume 2")
- a compilation of technical appendices, including the emissions inventory, archives of collected air quality and meteorological data, and computer program listings.

In addition, two IBM-compatible computer tapes containing the emissions inventory, air quality, and meteorological data are available. Further information on the tapes may be obtained from the EPRI project manager. Requests for copies of the reports should be directed to EPRI, Records and Reports Center, P.O. Box 10412, Palo Alto, CA 94303.

It should be noted that while the proposed design and supporting documents have been reviewed by EPRI and approved for publication, such approval does not signify that the contents necessarily reflect the views and policies of EPRI, nor is it intended to describe the Institute's program.

As a final note, I wish to express my sincere appreciation to the members of the SURE Technical Advisory Committee and EPRI staff colleagues who have conscientiously reviewed the work plan, and drafts of the experimental design, and have provided comments to ERT and to me. Special thanks are extended to Gene Tong of ERT for handling the committee meeting arrangements.

Charles Hakkarinen
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EXECUTIVE SUMMARY

The purpose of the Sulfate Regional Experiment (SURE) is to develop an understanding of the relationships between sulfur oxide emissions and ambient air quality. In this context, air quality is measured in terms of the concentrations of sulfur dioxide and the water soluble sulfates suspended in the ambient air.

This report presents the data and reviews the analytical procedures which support design of the SURE. The design itself is Volume 2 of this report.

OBJECTIVES

The principal objectives of the study were to:

- Develop a special emissions inventory based on existing data, to indicate the relationship between the public utilities and other sources as contributors to the total sulfur oxide (SO_x) emissions originating from selected major urban centers and regional complexes.

- Review and evaluate the quality of, and possible interrelationships among existing observations of sulfur dioxide (SO_2), the sulfates (SO_4^2-), and selected aerometric variables.

- Evaluate and adapt present air quality models for use in predicting ambient concentrations of sulfur dioxide and the particulate sulfates, downstream of major sources.

- Use the results of the first three objectives, to create an experimental design for a major regional, air chemistry study of how particulate sulfate concentrations may relate to sulfur oxide emissions.

METHODS OF DATA ASSIMILATION AND ANALYSIS

The main components of the investigation included:
A review and analysis of daily sets of sulfur dioxide (SO₂) and filter collected airborne particles for a period of a year between 1974 and 1975 at rural sites. These data were taken for twelve stations in the northeastern quadrant of the United States. These stations were located in Indiana and Illinois through the Ohio Valley to Pennsylvania and New York. Three thousand, six hundred fifty filters were analyzed chemically for water soluble sulfates.

A review and analysis of the available observations of sulfur dioxide (SO₂), sulfate (SO₄²⁻), nitrogen oxides (NOₓ), nitrates (NO₃), and total suspended particulates (TSP) from the National Air Surveillance Network (NASN) and the Community Health Air Monitoring Program (CHAMP) and from some states. Available data for 1974 were selected.

A selection and analysis of National Weather Service data that characterized meteorological conditions in the United States near air quality stations of interest.

Construction of a specialized emissions inventory of SO₂, NOₓ, and TSP for stationary sources in the northeast quadrant of the United States, with spatial resolution of 80 kilometers (km) square and vertical resolution for atmospheric injection at 0-300, 300-700 and 700-1500 meters (m), effective stack height. The geographical extent of the emissions inventory is shown in Figure 1-1.

Adaption of two models for regional air quality analysis that takes into account semi-empirically atmospheric oxidation of sulfur oxides and surface removal processes. The two approaches used were:

-- An air trajectory box model simulation
-- A three dimensional, fixed grid numerical model

Data available on the chemistry of precipitation collected in the United States have been surveyed, but have not been examined in detail as part of the project.

To establish empirical relationships between sulfate concentrations and certain variables such as humidity, temperature
Figure 1-1 Map of Northeastern Sector of the United States Which was Considered for Air Quality Modeling.
and SO₂ or TSP concentrations, statistical analyses and other investigations have been conducted on the data collected. These analyses examine annual behavior as well as seasonal and shorter term influences. The airborne sulfate data was grouped according to classes of traditional air mass identifiers. Furthermore, cases of extreme sulfate concentration were examined for high sulfate, low sulfate, precipitation, fair weather, frontal disturbances and air mass stagnation over two to five-day sequences.

The aerometric observations were examined for evaluating the significance of large scale transport over the northeast quadrant. Air mass trajectory analysis was undertaken for several cases, assuming the wind at six hundred meter height was representative of motion over hundreds of kilometers. Using the trajectory model developed as part of this study, the resulting concentrations arriving at Albany, New York, and Wheeling, West Virginia, were calculated. Comparisons were made between observed and calculated sulfur oxide levels at these locations for two different episodes, October 2-5, 1974 and May 18-20, 1975, using the emission inventory developed for the study.

The alternate approach of estimating the ground level sulfur oxide concentrations, taking into account winds at three heights and vertical dispersion, was undertaken using the three dimensional air quality model. The test case was the episode on October 2-5, 1974. Estimates were generated for the northeast quadrant (shown in Figure 1-1) for emissions calculated from the inventory, and compared with observations at twelve stations located in this region.

In both regional models the oxidation of sulfur dioxide to sulfate was simulated by a first order relation in ambient SO₂ concentrations. The assumed rate constant was one percent per hour. Removal processes at the ground were also
simulated by assuming them to be first order in ambient SO$_2$ and SO$_4^{2-}$ concentrations. The deposition coefficients were equivalent to about 1 cm per sec for SO$_2$ and about 0.1 cm per sec for SO$_4^{2-}$. A sensitivity analysis for the significance of variations in these parameters was conducted for the trajectory model. The results are sensitive to the SO$_2$ chemical transformation rate, but relatively insensitive to the removal rates.

The governmental data bank that was examined included selected measurements in several geographic areas shown in Figure 1-2. The air quality data were evaluated for potentially significant geographical differences by qualitatively comparing them with meteorological data and with 1972 data compiled for each Air Quality Control Region (AQCR) in the National Emissions Data System (NEDS).

PRINCIPAL FINDINGS

Analysis and evaluation of available aerometric and emissions data combined with air quality modeling, yielded the following findings:

- Of the five aerometric variables measured (dew point, relative humidity, temperature, SO$_2$ concentration, and particulate concentration), temperature and dew point, are the most strongly correlated with concentrations of the sulfates
- Sulfate concentration at ground level appears to be maximum in maritime tropical (mT) air masses in the northeast quadrant of the United States. These air masses are generally defined by high temperature and high absolute humidity (high dew point)
- Sulfate concentrations appear to be minimum in continental polar cold air (cPk) masses in the northeastern United States
Figure 1-2. Areas of the United States Considered in the Study
• Patterns of sulfate formation differ in specific geographical regions. For example:

-- A high incidence of ambient sulfate concentrations appears to occur together with photochemical smog in California

-- By contrast, the southwest has low ground level sulfate concentration despite very high SO₂ emission densities from isolated sources

-- The southeast appears to have seasonally uniform moderately high sulfate levels. Although some strong SO₂ emission sources exist, they are more isolated than those in the northeast

• In the northeast, a geographically widespread summer maximum of sulfate concentration has been noted, but this has not been observed in the southeast or in the southwest. The maximum in the northeast does not appear to be related directly to seasonal differences in SO₂ emissions. The data for central California show both late summer and winter maxima in sulfate concentrations. The data for the California South-coast Air Basin show both a summer and early fall maximum

• There is a widespread persistence in 24-hour average sulfate concentrations in rural areas of the northeast, showing simultaneous maxima and minima over distances of the order of several hundred kilometers. The episodes with maximum concentrations are of a few days duration, and occur several times each year, usually under conditions of stagnation

• High correlations of daily ground-level sulfate measurements occur among air monitoring stations spaced from 10 to 300 km apart. The annual average observations in the Ohio Valley and in the northeast show significant correlation as far as 400 km
The region of sulfate conversion and maximum ground-level exposure appears to extend over a distance confined to 300 km from major sulfur oxide (SOₓ) sources. The zone of influence of an individual source on the concentration values for the sulfur oxide particulate complex (SPC) appears to be 200 to 300 km weighted toward the downwind direction. The intensity of the sulfate impact in the northeastern United States can be construed to come from the result of a cumulative increase in SOₓ levels along the principal direction of air transport. Other areas have substantially lower source densities and consequently suffer less SOₓ exposure.

The highest nonurban sulfate concentrations have been found in West Virginia and in western Pennsylvania, which are in the zone of highest SOₓ emission density.

From the limited information available about airborne nitrates, it appears their behavior differs significantly from that of the airborne sulfates. Maximum concentrations of nitrates differ both on a geographical and a diurnal basis from the maximum concentrations of the sulfates. For example, nitrates in California are at a maximum in winter, but the maximum values for the sulfates occur in late summer.

The relationship between precipitation and sulfate levels measured at the ground is not well defined in the data set examined.

No useful relationship has been established between the existing sulfate data and local (up to 100 km) SO₂ emissions (elevated or surface). It is therefore not possible to define quantitatively zones of influence for major sources of SO₂ emissions. However, air quality model calculations show promise for developing such relationships. Two models have been tested:

-- A trajectory-based box model

-- A more sophisticated three-dimensional numerical scheme (EGAMA). This model has been particularly useful for testing the impact of elevated sources, and for determining the sensitivity of resulting concentrations to the geographical distributions of sources.
Twenty-four hour averaged samples provide inadequate resolution for evaluating the conversion of SO$_2$ emissions into the sulfates as a functions of meteorological events. Shorter sampling periods (perhaps as short as two hours) appear desirable.

Existing data are not adequate to describe fully the chemical conversion from SO$_2$ to the sulfates and the removal processes of sulfur oxides. There is a great need for time-dependent three-dimensional measurements of many aerometric quantities to characterize the evolution and fate of the atmospheric SO$_x$.

During the course of this study, the use of existing models, which account for the transport processes, oxidation chemistry and removal parameters in a relatively simple manner, suggests that modeling is a promising technique for developing an air quality control strategy.

RECOMMENDATIONS

On the basis of the above findings, we recommend:

- Plans for a major regional study of the relationship between air quality and sulfur oxide emissions be implemented immediately. The present information on sulfur oxide behavior in the atmosphere is inadequate for developing a rational approach for achieving air quality improvement.

- As a second-level priority, the regional experiment should be extended to include a study of the relationship between nitrogen oxides and the nitrates, which are reported to be possibly more hazardous to human health than the sulfates.

- The regional experiment should be conducted in the northeast quadrant of the United States, where the sulfate exposure problem is most widespread and intense.

- The experimental design should take into account the need for resolving particulate measurements over time periods shorter than 24 hours.
• The experimental plan should provide adequate three-dimensional characterization of aero-
metric behavior to be useful on a regional basis.

• The experimental design should provide ample resources for analysis and interpretation of
the measurements, including the development and testing of a useful, practical air qual-
ity model to relate sulfur oxide emissions to ambient sulfur oxide levels over dis-
tances extending as far as 1000 km.