A Variable Trajectory Model for Regional Assessments of Air Pollution From Sulfur Compounds

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February 1979

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SUMMARY

This report describes a sulfur oxides atmospheric pollution model that calculates trajectories using single-layer historical wind data as well as chemical transformation and deposition following discrete contaminant air masses. Vertical diffusion under constraints is calculated, but all horizontal dispersion is a function of trajectory variation. The ground-level air concentrations and deposition are calculated in a rectangular area comprising the northeastern United States and southeastern Canada.

Calculations for a 29-day assessment period in April 1974 are presented along with a limited verification. Results for the studies were calculated using a source inventory comprising 61% of the anthropogenic SO_2 emissions. Using current model parameterization levels, predicted concentration values are most sensitive to variations in dry deposition of SO_2 , wet deposition of sulfate, and transformation of SO_2 to sulfate. Replacing the variable mixed-layer depth and variable stability features of the model with constant definitions of each results in increased ground-level concentration predictions for SO_2 and particularly for sulfate.

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INTRODUCTION

Because of potential health and ecological effects of air contaminants,⁽¹⁾ the U.S. Department of Energy (DOE) and the Electric Power Research Institute (EPRI) are currently sponsoring programs to assess, on a multistate scale, the impact of sulfur oxide emissions from fossil-fuel-fired generating plants and other industrial emission sources.^(2,3)

This document presents a regional-scale, assessment model designed for economical computation of long-term averages of ground-level air concentration and deposition rate of sulfur dioxide (SO_2) and of sulfate ($\text{SO}_4^{=}$). The term "regional" embraces horizontal scales of 25 to 2500 km;⁽⁴⁾ the final recommended averaging time is 1 mo or more. The model also uses hourly real-time precipitation and estimates rainfall pH, averaged over the total assessment period.

This discussion provides a complete model description, including model formulation and test case and sensitivity studies. Section 2 discusses model description. Test-case input data, test-case results and sensitivity analysis results follow in Section 3.

The test case includes 29-day averages computed over a 1360 by 2040 km grid over the northeastern United States, using an inventory of 60 sources and stepping in 1-hr increments. Thus used, the model achieves an 11,000 to 1 ratio of real time to running time on the CDC 7600 computer.

CONCLUSIONS

A model test case was evaluated and model results were compared to observed ground-level air concentration data for sulfate at 107 stations throughout the northeastern United States. Using these data for regression of predicted results against observed results, a correlation of 0.43 was obtained. Furthermore, using an emissions inventory believed to account for approximately 61% of the SO_2 emissions in the predicted sulfate concentration accounts for an average of 53% of the corresponding observed results. Nonrepresentative observational data used to describe regional pollutant behavior are believed to contribute to this relatively low correlation value. However, substantial improvements can be made in the model by more sophisticated parameterization of removal and transformation, including the appropriate variation of these parameters with physical conditions.

Sensitivity estimates made directly from the numerically specified equations for removal and transformation of mass indicate: if parameter levels for each of these processes are varied within the same ratio limits of presently assumed values, the ground-level concentrations are more sensitive to dry deposition of SO_2 , wet deposition of sulfate, and transformation than to wet deposition of SO_2 and dry deposition of sulfate. Sensitivity tests also indicate that with this model, constant stability increased both predicted levels of SO_2 ground-level air concentrations and deposition by 22%, while the predicted sulfate ground-level air concentration increased 73%.

The ratio of real time to execution time for the 60-source inventory, 29-day run described is greater than 11,000 to 1 on the CDC 7600 computer. The required execution time was 219 seconds.

MODEL DESCRIPTION

CALCULATION GRIDS

Model calculations are performed on a rectangular grid, which is a subset of the National Meteorological Center (NMC) Northern Hemisphere grid. The area for calculation in the test case, presented in the following section (see Figure 1), covers the northeastern United States and a portion of southern Canada. It extends 2040 km on the abscissa and 1360 km on the ordinate.

Two grids are defined on this area. The advection grid (see Figure 1) is formed by dividing the area into a 12 by 8 array of squares so that each square is 1/2 NMC grid unit long, with an average linear dimension of 170 km. Because of the polar stereographic projection of the NMC grid, the actual grid-square dimensions are a function of latitude; the dimensions of the advection grid unit (ΔG_a) are:

$$\Delta G_a = 190.5 \frac{1 + \sin \phi}{1 + \sin(\pi/3)} \text{ km}, \quad (1)$$

where ϕ is the latitude in radians.

The second or sampling grid has elements with linear dimensions 1/5 the advection grid spacing. Therefore, the sampling grid divides the predictive area into an array of 60 by 40 squares.

TRANSPORT CALCULATIONS

Trajectories are simulated for plume elements released hourly from several emission source locations. Emission rate at the source position determines the initial sulfur dioxide mass of each plume element. The initial sulfate mass is zero. Subsequent horizontal transport of these plume elements is in hourly steps using the layer-averaged winds at each advection-grid, intersection point. An objective analysis scheme developed for mesoscale purposes⁽⁵⁾ interpolates wind data in space and time to provide the hourly winds at each particle position. The current transport scheme has no vertical component. It is assumed that the concentrations of sulfur oxides are averaged over a time period in which the effects of vertical atmospheric

FIGURE 1. Wind Station Distribution with Reference to the Advection Grid (Inner Rectangle) and a Portion of NMC Octagonal Northern Hemisphere Grid

processes can be adequately expressed by the parameterized vertical component of diffusion and by the mixing depth constraints. It is also assumed that the effects of vertical transport on horizontal transport can be neglected over the long assessment time period.

DIFFUSION AND DIFFUSION CONSTRAINTS

The following assumptions are made on atmospheric diffusion:

1. Horizontal diffusion is a function of synoptic scale wind variation only. Tennekes,⁽⁶⁾ Bolin and Persson,⁽⁷⁾ and Prahm and Christensen⁽⁸⁾ support this assumption. All state that either horizontal turbulent diffusion about the plume center line is relatively unimportant or that it is less important than plume meandering.
2. The vertical-turbulent diffusion parameterization is a function of stability in a specified diurnal cycle for all plume elements within the daytime mixed layer or the nocturnal stable layer.
3. The depth of the daytime mixed layer is represented by a diurnal cycle in which a daytime layer increases from a minimum depth at sunrise to a maximum depth in the afternoon. A nocturnal layer builds to a lower depth.
4. Depth of the boundary layer (mixed layer in daytime or nocturnal layer) for each hour determines which of two vertical dispersion regimes - mixed layer or above mixed layer - will be applied to a given plume element. Those elements released within the depth of the layer expand according to the stability of the hour. Those released above the current depth of the layer expand as if the atmosphere were extremely stable. An exception to this method occurs when releases have expanded to the maximum depth of the mixed layer during the previous day; in this case, their expansion continues according to a fixed function of travel time independent of stability or layer depth.
5. Depth of the mixed layer provides a vertical constraint for releases at heights within this layer. The depth of the nocturnal boundary does not serve as an analogous constraint. Carson⁽⁹⁾ supports this assumption

by depicting the mixed-layer depth as a function that increases during the daytime, then loses its definition. This picture is in relative agreement with modeling results by Venkatram and Viskanta⁽¹⁰⁾ and with experiment results by Kaimal et al.,⁽¹¹⁾ who find that about 1 hr before sunset the convective layer disintegrates rather abruptly, although remnants of the capping inversion persist through the development of the nocturnal boundary layer.

6. At the present stage of model development, terrain effects are neglected.

Diffusion equations are best explained by relating them to final concentration and deposition calculations where the average ground-level air concentrations of pollutant (n) over grid square (i,j) are given by summing time-integrated concentrations [integrated over time interval (Δt)] and dividing by total assessment time (T). Thus,

$$\bar{X}_n^{ij} = T^{-1} \sum_{m(t)} \Delta X_{mn}^{ij}(\Delta t; t) . \quad (2)$$

where ΔX_{mn}^{ij} is the time-integrated ground-level air concentration over (i,j) for pollutant (n) carried by each plume element (m) that is located over (i,j) at time (t).

ΔX_{mn}^{ij} is computed at the end of each sampling interval (Δt) for each pollutant constituent of each plume element that is on the grid by

$$\Delta X_{mn}^{ij}(\Delta t; t) = \frac{Q_{mn}(t)\Delta t}{A^{ij} Z_m} , \quad (3)$$

where

- $Q_{mn}(t)$ is the mass of n carried by m at t
- A^{ij} is the area of (i,j)
- Z_m is the vertical distribution factor for m calculated by assuming Gaussian vertical distribution modified by reflection from the ground and from the top of the mixed layer.

If the vertical constraint height is L and the effective stack height is H , Z_m^{-1} is:

$$Z_m^{-1} = \frac{\sqrt{2/\pi}}{(\sigma_z)_m} \left\{ e^{-\frac{1}{2} \left(\frac{H}{\sigma_z} \right)^2} + \sum_{k=1}^{\infty} \left[e^{-\frac{1}{2} \left(\frac{2kL - H}{\sigma_z} \right)^2} + e^{-\frac{1}{2} \left(\frac{2kL + H}{\sigma_z} \right)^2} \right] \right\}, \quad (4)$$

where $(\sigma_z)_m$ is the vertical standard deviation for plume element m .

If σ_z is small compared to L , the term represented by the entire summation over k may be neglected. If σ_z is approximately equal to or larger than L , the vertical distribution is approaching uniformity. Therefore, instead of calculating Z^{-1} by a wasteful summation, Z is set equal to L :

$$Z = L \quad (5)$$

L is the height of a reflection effective for transport at height H .

In the model, the depth in meters of the daytime mixed layer (L_d) is given by:

$$L_d(t_h) = 200 + 1300 \sin \left[0.05\pi(t_h - 6) \right] \quad (6)$$

when

$$6 \leq t_h \leq 18;$$

and the depth of the nocturnal boundary layer (B_n) is given by:

$$B_n(t_h) = 100 + 300 \sin \left[0.05\pi((t_h - 18)) \right] \quad (7)$$

when

$$18 < t_h < 30$$

where t_h is the hour of the day unless t_h is greater than 24, in which case t_h is specified as the hour of the day plus 24 to provide a continuous function.

These formulae were drawn up to approximate graphic results given by Carson.⁽⁹⁾ They are advanced only for purposes of the calculations of this model, because the typical daytime mixed layer increases from 200 m at sunrise to 1500 m by late afternoon, and a nocturnal boundary layer develops to about 400 m.

All plume elements not spread beyond the mixed-layer depth (L) expand vertically as a function of travel distance. The increase of σ_z is calculated using

$$\sigma_z(x + \Delta x) = \sigma_z(x) + \Delta x \left. \frac{\partial \sigma_z}{\partial x} \right|_{x + 1/2 \Delta x, S} \quad (8)$$

where S indicates the derivative differs according to the stability that is hourly specified in the model.

The formulae thus differentiated are those of Eimutis and Konicek⁽¹²⁾ If H is greater than the mixed-layer depth (L_d or B_n), whichever applies at the time, the plume elements expand at the rate of 60% of that for Pasquill Stability 6.^(a) This expansion rate results because mixing by mechanical turbulence is not effective above the mixed layer; therefore, the rate of vertical expansion is minimal.

All plume elements, which have expanded vertically to fill the mixed layer at its maximum depth, are released from the mixed-layer constraint at

(a) D. C. Powell, H. L. Wegley and T. D. Fox MESODIF-II: A variable trajectory plume segment model to assess ground-level air concentrations and deposition of routine effluent releases from nuclear power facilities. To be published by the U.S. Nuclear Regulatory Commission.

sunset when the boundary-layer structure collapses. These puffs expand as a function of time according to

$$\sigma_z(t+\Delta t; L_{dm}) = \sigma_z(t; L_{dm}) + \Delta t \sqrt{K_z/2t} , \quad (9)$$

where K_z is the vertical diffusivity.

Heffter and Ferber⁽¹³⁾ recommend a value of $5 \text{ m}^2 \text{ s}^{-1}$ for K_z . The plume elements not subject to the mixed-layer constraint after sunset include almost all those emitted during the previous 24 hr. Since these plumes all have the same vertical distribution at sunset, taken to be 1800, they are all assigned a common value of t , 12 hr. If L_{dm} is the maximum depth of the mixed layer at $t_h = 18$ for each of these plume elements, then

$$\sigma_z(12 \text{ hr}; L_{dm}) = \sigma_z \bigg|_{t_h=18} = \frac{L_{dm}}{\sqrt{\pi/2}} . \quad (10)$$

A Gaussian plume calculation with $\sigma_z = L_{dm} \sqrt{2/\pi}$ yields the same ground concentration as calculations for evenly distributed plumes over a layer of depth L_{dm} . Further plume expansion in time interval (Δt) is given by

$$\sigma_z(12 + \Delta t) = \frac{L_{dm}}{\sqrt{\pi/2}} + \Delta t \sqrt{\frac{K_z}{2(12 + \Delta t)}} . \quad (11)$$

The daytime plume expansion is illustrated in Figure 2. The plume at the bottom left is a low-level release, which expands at a rate corresponding to the stability of the hour to a height not greater than that of the mixed-layer depth. Farther to the right, an elevated plume is released above the mixed layer. It expands very slowly so that it does not contribute to surface concentrations. Above both plumes, the depth of the boundary-layer is shown by L_d . At a later hour when L_d rises above the stack, the second plume will also expand according to the stability of the hour. Above L_{dm} , σ_z of a

FIGURE 2. Daytime Illustration of Stability Definition and Vertical Constraints.

plume released on a previous day is shown. It expands at the rate given by Equation (9). This expansion rate reflects the effects of convection averaged over a 24-hr period at the higher levels.

The hourly stability array used for the diurnal cycle generally allows for stable conditions at night, unstable conditions in the afternoon and neutral transition hours. The portion of time allotted to each classification corresponds to the climatology of stability for the spring season over the northeastern United States discussed by Doty, Wallace and Holzworth.⁽¹⁴⁾ During unstable hours in the afternoon, all plume elements quickly become evenly distributed throughout the mixed layer.

TRANSFORMATION AND REMOVAL MECHANISMS

The transformation of SO_2 to sulfate and the removal of SO_2 and sulfate from the atmosphere by dry and wet deposition are calculated by a set of ordinary linear differential equations that are solved for each plume element. These equations assume that:

- transformation is a linear, first-order process.
- Dry-removal rate is obtained by multiplying the deposition velocity by the vertical distribution factor Z^{-1} .
- Wet-removal rate is directly proportional to the local precipitation rate.

The following definitions are used in the equations:

$Q_1(t), Q_2(t)$ = masses of SO_2 and sulfate in a plume element at time, t , μg

$D_1(t), D_2(t)$ = masses of SO_2 and sulfate deposited from a plume element up to time, t , μg

d_1, d_2 = coefficients of dry deposition of SO_2 and sulfate, hr^{-1}

w_1, w_2 = coefficients of wet deposition of SO_2 and sulfate, hr^{-1}

r_{12} = transformation rate of SO_2 to sulfate, hr^{-1}

In this notation $Q_1(t)$ is equivalent to $Q_{m1}(t)$ in Equation (3). For the sake of simplicity, the m notation is suppressed.

Given the above definitions,

$$\frac{dQ_1}{dt} = - (d_1 + w_1 + r_{12})Q_1, \quad (12)$$

$$\frac{dQ_2}{dt} = -(d_2 + w_2)Q_2 + 1.5 r_{12}Q_1, \quad (13)$$

$$\frac{dD_1}{dt} = (d_1 + w_1)Q_1, \quad (14)$$

$$\frac{dD_2}{dt} = (d_2 + w_2)Q_2. \quad (15)$$

The 1.5 factor in Equation (12) accounts for different masses associated with equimolar quantities of sulfate and SO_2 . Solutions of these equations are expressed by the change of mass for each plume element (m) over the advection time (Δt) and by the mass to be deposited over grid unit (i,j) from m over the same time interval, usually 1 hr.

The calculated deposition is expressed in terms of mass $\text{m}^{-2} \text{yr}^{-1}$. Thus, the annual deposition (F) of pollutant (n) applying to i,j may be written as:

$$F_n^{ij} = Y^{-1} T^{-1} \sum_{m(t)} \Delta F_{mn}^{ij}(\Delta t; t) = Y^{-1} T^{-1} \sum_{m(t)} \frac{D_{mn}^{ij}(\Delta t; t)}{A^{ij}} \quad (16)$$

where Y is 1 yr in units of T and ΔF_{mn}^{ij} is the deposition of pollutant n, mass per unit area, over time interval (Δt) over grid square i,j because of plume element m at time (t).

In $D_{mn}^{ij}(\Delta t; t)$, n = 1,2 is a more complete notation for the deposited mass. The D_{mn}^{ij} are divided into dry and wet deposition:

$$D_{mn}^{ij} = Q_{mn} D_{mn}^{ij}(\Delta t; t), \quad (17)$$

where $D_{mn}^{ij}(\Delta t; t)$ is given by:

$$D_{mn}^{ij}(\Delta t; t) = 1 - \text{EXP} \left\{ \left[- \frac{\overline{Vd}_n}{Z_m(t)} - \lambda_n p^{ij}(t) \right] \Delta t \right\}. \quad (18)$$

where

- $\overline{Vd}_n$ is the deposition velocity for n.
- $Z_m(t)$ is the vertical distribution factor for m.
- $p^{ij}(t)$ is the precipitation rate at t over i,j.
- λ_n is an inverse length scavenging parameter for n.

The model currently assumes that Vd_n and λ_n are constants.

Relating Equations (14) and (15) to Equation (18) yields

$$d_n = \overline{Vd}_n / Z_m(t) , \quad (19)$$

$$w_n = \lambda_n p^{ij}(t) . \quad (20)$$

A constant value for each of these parameters was selected even though they are all known to vary in space and time with changing physical conditions. The object is to establish a performance level for the model by comparing predicted results with observed results as a base case for sensitivity studies (see page 28 of this report).

The parameter values chosen for the model test case are:

$$\overline{Vd}_1 = 1.0 \text{ cm s}^{-1} , \quad (21)$$

$$\overline{Vd}_2 = 0.1 \text{ cm s}^{-1} , \quad (22)$$

$$\lambda_1 = 5 \text{ m}^{-1} , \quad (23)$$

$$\lambda_2 = 250 \text{ m}^{-1} , \quad (24)$$

$$r_{12} = 0.02 \text{ hr}^{-1} . \quad (25)$$

The dry-deposition velocities used are the same as those used by Sheih.⁽¹⁵⁾ Values were obtained on the basis of observation and experiment given by Owers and A. W. Powell,⁽¹⁶⁾ and Smith and Jeffries.⁽¹⁷⁾

The value of λ_1 , the scavenging parameter for SO_2 , was selected from calculations made with experimental data by Dana, Hales and Wolf.⁽¹⁸⁾

Values calculated from the data from independent experiments ranged from 2 m^{-1} to 10 m^{-1} .

The value of λ_2 , the scavenging parameter for sulfate, was derived by using simultaneously reported water concentration and average air concentration values of sulfate ($2.9 \text{ mg liter}^{-1}$ and $8.3 \text{ } \mu\text{g m}^{-3}$, respectively) in

the Air Quality and Stationary Source Emission Control Report⁽¹⁾ for a New Hampshire location. Using an average value of $0.7 \times 10^{-3} \text{ m}^{-1}$ for Z^{-1} (see Equation (5) and Table 2), a value of 250 m^{-1} is obtained for λ_2 . This value is consistent with the value obtained by Dana and Hales⁽¹⁹⁾ where frontal-type rainfall is assumed to have values of 0.1 and 2.5 mm for geometric mean particle radius and geometric standard deviation.

The transformation rate used is consistent with those used by other modelers. Egan, Rao and Bass⁽²⁰⁾ show their model predicts too much sulfate if a transformation rate of 0.02 hr^{-1} is used and not enough if 0.01 hr^{-1} is used. Sheih⁽¹⁵⁾ uses 0.023 hr^{-1} . Values inferred from analysis of experimental data (see Roberts and Friedlander⁽²¹⁾ or Alkezweeny and Powell⁽²²⁾) were found by Levy, Drewes and Hales⁽²³⁾ to range from 0 to 0.55 hr^{-1} . The higher values were generally deduced from data taken from urban areas. Obviously the mechanisms of transformation will have to be better understood, as well as their distribution in time and space, before such experimentally derived results can be directly used.

CONTRIBUTION OF H_2SO_4 DISSOCIATION TO RAINFALL ACIDITY

The Air Quality and Stationary Source Emission Control Report,⁽¹⁾ contains a map of annual averages of pH of rainfall from several measuring points in the eastern United States for a period within 1965-66. A similar map for 1973-74 is given by Likens.⁽²⁴⁾ Minimum values shown on these maps are 4.2 and 4.1, respectively.

The pH values appearing on these maps are long-term averages. Similarly the equation for pH to be derived below is intended for long-term averages over many individual showers. To derive the equation, the hydrogen ion mass from one plume element is calculated as shown, along with the diluting water volume from rain. Then, this calculation is summed over the many plume elements passing over a given sampling grid square during rains and is added to a base level to obtain a pH number for each sampling grid unit for the assessment period.

The hydrogen ion content is assumed to come from 100% dissociation of atmospheric H_2SO_4 into $2H^+$ and $(SO_4^{=})$ and from a specified level that is derived from the presence of other compounds.

The rate of wet removal of sulfate from one plume element is:

$$Q_2(t) \lambda_2 P^{ij}(t) \quad (\mu g \text{ hr}^{-1}) . \quad (26)$$

The mass of sulfate removed by rainfall in one sampling period (t) is:

$$Q_2(t) \lambda_2 P^{ij}(t) \Delta t \quad (\mu g) . \quad (27)$$

Since there are 96 grams of sulfate per mole, the number of moles removed is:

$$Q_2(t) \lambda_2 P^{ij}(t) (10^{-6}/96) = 1.04 \times 10^{-8} Q_2(t) \lambda_2 P^{ij}(t) \Delta t . \quad (28)$$

Since there are two hydrogen ions for each sulfate ion, number of moles of hydrogen ions in the same water volume is:

$$2.08 \times 10^{-8} Q_2(t) \lambda_2 P^{ij}(t) \Delta t \quad (\text{moles } H^+) . \quad (29)$$

To calculate the volume of water involved, the horizontal dimension of each plume element is assumed to be the area of the grid square it is located over. Using this assumption, the amount of water is $A^{ij} P^{ij}(t) \Delta t (m^3)$ or $10^3 A^{ij} P^{ij} \Delta t$ (liters). Thus, the hydrogen ion concentration is given by:

$$\begin{aligned} [H^+] &= \frac{2.08 \times 10^{-8} Q_2(t) \lambda_2 P^{ij}(t) \Delta t}{10^3 A^{ij} P^{ij}(t) \Delta t} \\ &= \frac{2.08 \times 10^{-11} Q_2(t) \lambda_2}{A^{ij}} \left(\frac{\text{mole}}{\text{liter}} \right) . \end{aligned} \quad (30)$$

For the entire simulation, the pH at grid square i,j is calculated by summing the contributions of many plume elements that happened to be over the square during the hours of rain. Each of these contributions must be weighted by the rainfall rate of the hour, $P^{ij}(t)$. Because a base level of hydrogen ion concentration from other constituents is involved, the final equation for pH can be written as follows:

$$pH^{ij} = -\log_{10} \left[\text{Base} + 2.08 \times 10^{-11} \lambda_2 \frac{\sum_t Q_2(t) P^{ij}(t)}{A^{ij} \sum_t P^{ij}(t)} \right] \quad (31)$$

Equation (31) states that under the employed assumptions, pH is a function of vertically integrated sulfate concentration and scavenging efficiency. Since the number of plume elements passing over the area A^{ij} depends on the magnitude of the area, the apparent dependence on A^{ij} should cancel out if the assessment period is divided into a sufficiently large number of individual sampling periods.

The base concentration selected for these runs was $10^{-5.6}$ moles of hydrogen ion per liter, which is given by Likens⁽²⁴⁾ as a representative value for the contribution of CO_2 alone to acidity. Another significant contribution, that from nitrates, is taken into account later.

MODEL RESULTS

TEST SIMULATION DATA

The model required three different types of data: wind, precipitation, and emission. In addition to discussing these data, this section discusses model testing observations.

Wind Data

Transport calculations were made with radiosonde data from selected groups of stations shown in Figure 1. The winds at each grid point were calculated by computing an average wind at each station through the layer 100 to 1000 m above the surface and then interpolating values to the grid intersections. The interpolation technique is based on the weighted average wind values of the three-to-five stations closest to each grid intersection. The weighting factor selected was the inverse square of the distance of the station from the grid intersection. Horizontal winds from any other interpolation scheme could be used.

Precipitation Data

All simulations presented made use of the sequential hourly precipitation data for April 1974. The data used were derived from records of 777 cooperative observing stations of the National Climatic Center (NCC) and a small number of Atmospheric Environment Service stations (AES) in Canada as shown in Figure 3. Data for each hour were processed, using an inverse-square distance-weighting interpolation program discussed above, to provide values for each square on the 60 by 40 grid within a resolution of approximately 34 km.

Emissions Data

A 1974 SO₂ emissions inventory was developed from 1973 National Emissions Data System data⁽²⁵⁾ and 1974 data from the Federal Power Commission.⁽²⁶⁾ The inventory listed 60 sources: 40 utility and 20 nonutility. This inventory contains the largest SO₂ emission sources in the U.S. portion of the grid, which accounts for approximately 65% of emissions over the grid.

FIGURE 3. U. S. Precipitation Station Distribution and Sampling Grid

Owing to lack of data and high source-to-source variability of sulfate emissions, all sulfate in this test was assumed to be generated by SO_2 transformation.

This study of SO_2 considered all sources to be point sources with a specified emission height. Nonutility sources were modeled as surface sources and utilities were modeled with an effective release height of 400 m. Each utility and nonutility source in the inventory represents one actual emission source or a combination of emission sources located close together. Inventory approximations reduced computational costs during model development. The model can use different emission heights for each source.

Figure 4 gives the locations and relative strength of the SO_2 emission sources. Emission strengths are depicted in the figure by circle diameters. The largest sources are on the order of 650×10^3 tons/yr.

Observations for Model Testing

Average sulfate concentrations for April 1974, published as part of EPRI Design of the Sulfate Regional Experiment (SURE) I study,⁽²⁷⁾ were selected for preliminary model comparisons. The sulfate data were obtained from the National Aerometric Sampling Network (NASN) and the EPRI SURE I monitors. The EPRI SURE I monitors were selected from an existing monitoring network to represent regional sulfate patterns. For initial analysis SO_2 data were not considered since observations were more likely to be dominated by local sources unresolved by the regional model.

Using monthly sulfate concentrations limits application of statistical comparisons with model results to qualitative guidelines. This is because of monitoring site location, data quality, small sample sizes, and model predictions close to the noise level of ambient measurements. In this study almost 85% of the monitoring sites were located in urban areas where higher concentrations of sulfate are caused by effects not explained by the model. For example, higher SO_2 -to-sulfate conversion rates occur in urban areas⁽²²⁾ than in rural.⁽²⁸⁾ Measurement uncertainty caused by SO_2 interference on the hi-vol filters used in sulfate monitoring is another problem with the sulfate data.⁽²¹⁾ Monthly average concentrations taken from 24 observations

FIGURE 4. Source Distribution: Utility; Nonutility

2-to-5 days/mo contributed to NASN data uncertainty. This uncertainty must be considered when interpreting verification statistics for comparison with other models. Results presented in this study are based on monthly averages of hourly values rather than predictions during short episodes when pronounced variations in spatial concentration fields would be expected. Finally the interpretation and effectiveness of verification statistics are limited by using predicted results from periods when observations are close to the noise level.

TEST-CASE RESULTS

A base case simulation was performed to provide both a demonstration of model capabilities and also a reference point for sensitivity studies. Input parameters, including emissions and meteorological data, for the simulation and data used for comparison are discussed in the previous sections. Two important features of the analysis are the use of a limited emissions inventory, which accounts for approximately 61% of U.S. SO_2 emissions on the grid and also the use of a constant SO_2 -to-sulfate transformation rate of 2%/hr.

Model simulated spatial patterns coming from the data fields are presented in Figures 5 through 8. These figures depict contoured results on a calculation-grid region map. The contours were smoothed by using a weighted average concentration of the grid square in question and the eight surrounding grid squares.

Sulfur dioxide concentration patterns shown in Figure 5 have areas of high concentrations that correspond to the area of highest emissions (see Figure 4). Low concentrations are near the grid boundaries. Peak concentrations of 30 to $60 \mu\text{g m}^{-3}$ occur in the vicinity of the largest ground-level or nonutility sources in the Chicago-Gary area, New York City, Philadelphia, the Cleveland-Akron area, and near elevated utility sources in southwestern Pennsylvania. Higher concentration levels are also predicted in the vicinity of other large utility sources. These results are consistent with expectations of SO_2 plume behavior, which produces maximum concentrations within about 10 km of the sources in most cases.

FIGURE 5. Predicted Average Ground-Level Air Concentrations for SO_2 , $\mu\text{g m}^{-3}$

FIGURE 6. Predicted Average Deposition Rate for SO_2 , $\text{g m}^{-2}\text{yr}^{-1}$

FIGURE 7. Predicted Average Ground-Level Air Concentrations of Sulfate, $\mu\text{g m}^{-3}$

FIGURE 8. Predicted Average Deposition Rate for Sulfate, g m⁻²yr⁻¹

Predicted monthly average SO_2 concentrations, which have a maximum monthly average of $61 \mu\text{g m}^{-3}$, fall within the same range as observed concentrations. (This can be compared to an annual National Ambient Air Quality Standard of $80 \mu\text{g m}^{-3}$.) Selected observed data from the EPRI SURE I report⁽²⁷⁾ and predicted SO_2 concentration decrease with regard to distance from individual sources. Therefore, most SO_2 maxima can be considered subgrid scale effects relative to the model.

SO_2 deposition patterns (see Figure 6) demonstrate the same source-related behavior as SO_2 concentrations. Maximum deposition values of 10 to $20 \text{ g m}^{-2}\text{yr}^{-1}$ are predicted near the largest ground-level emission sources. The close correspondence between predicted concentrations and deposition patterns indicates that in the current model configuration constant dry-deposition velocity effects dominate SO_2 removal rather than wet processes.

Average sulfate concentrations (see Figure 7) are distributed in a broad pattern that cannot be related to a single source complex. The dominant feature in the figure is a large area of concentrations in the range of 6 to $10 \mu\text{g m}^{-3}$, which covers most of the mid-Atlantic states and parts of New England and Ohio. The relative smoothness of the pattern, as compared to that of SO_2 , occurs because sulfate is formed as a secondary pollutant along the trajectories.

April 1974 test-case results were compared to observed concentrations. Data from a mix of 107 urban and nonurban sites provided a spatial average concentration of $9.8 \mu\text{g m}^{-3}$. Predicted concentrations at the same locations averaged $5.2 \mu\text{g m}^{-3}$. Thus, 53% of the observed sulfate is predicted from 61% of the SO_2 emissions inventory. The maximum observed and predicted monthly average concentrations differed by a similar factor with values of 18 and $10.5 \mu\text{g m}^{-3}$, respectively. A linear correlation-regression analysis indicated a positive correlation of 0.43 between observed and predicted concentrations and a regression equation with a slope of 0.26 and an intercept of $2.65 \mu\text{g m}^{-3}$. Fifty-seven percent of all predicted concentrations were within a factor of two of observed value, and 87% were within a factor of

three. The remaining 10% of the values were predictions at sites on the western boundary of the emission grid where no upwind sources were given for the prevailing wind direction.

Much of the variation between observed and predicted concentrations can be attributed to the basic model concept and the emissions used as model input. Because the model is Lagrangian, no allowance is made for pollutant mass entering the sampling grid from outside the grid boundaries. In addition, the model is not capable of predicting subgrid scale (~ 30 km) air quality effects as pointed out in the discussion of maximum SO_2 concentrations. Also, the Lagrangian model makes verification for the complete inventory prohibitive because of individual treatment of all sources. The 60-source base inventory represents approximately 60% of the U.S. SO_2 emissions on the grid. As the number of ranked sources increases, the rate of change for the fraction of total emission decreases. For example, increasing the number of sources from 60 to 70 increases the emission fraction only a few percentages.

By examining the rough analysis of observed sulfate data for April 1974 (see Figure 9) an additional, more qualitative form comparison can be made. As noted earlier, predicted concentrations are lower than observed concentrations, although the patterns are similar. The primary pattern difference is: higher observed concentrations on the Atlantic Coast between New York City and Baltimore, probably because of urban influences in the data unresolved by the model.

Maximum values for sulfate deposition patterns shown in Figure 8 predominate over the northeast quadrant of the grid rather than over the central portion as was the case for sulfate concentrations. This observation, as well as the irregular sulfate-deposition pattern, indicates the importance of wet-removal processes in the modeling of sulfate deposition. Modification of rainfall pH also shows the same pattern (see Figure 10) with high rainfall acidity in the northeast grid quadrant. The distribution correlates fairly well with the 1972-73 map given by Likens.⁽²⁴⁾ However, there is a considerable extension of low pH down into the Ohio River areas shown by Likens that does not appear on this prediction map.

FIGURE 9. Observed Average Ground-Level Air Concentrations of Sulfate, $\mu\text{g m}^{-3}$

FIGURE 10. Predicted Average pH of Rainfall

Chiefly because of differences in the Ohio Valley, the average pH on Figure 10 is approximately 0.3 higher than the average pH on the map by Likens. This difference is acceptable since the sulfur-compound-emissions inventory used represents only 61% of the anthropogenic sulfur-compound emissions, and since sulfur compound emissions according to Likens⁽²⁴⁾ constituted only 66% of the total contribution to ambient acidity, 30% being from nitrates. Therefore, the SO₂ emissions inventory used consists of not more than 40% of the total contribution to ambient acidity and average predicted pH value based on this inventory should be about 0.4 higher than the ambient pH. To aid in the interpretation of sensitivity test results, discussed in the following section, a mass-balance summary for the base test-case has been compiled in Table 1. The table shows SO₂ and sulfate mass left in the air over the grid at the completion of simulations, total deposition, and the pollutant mass that escapes the grid for elevated utility sources and for the total inventory.

Table 1, which involves utility as well as total inventory effects, shows the final fate of sulfate emissions with roughly 52% of emissions being

TABLE 1. Mass Balance Summary

	Fraction of SO ₂ Emissions, %	
	<u>All Sources</u>	<u>Elevated Utility Sources</u>
Mass remaining on computational grid		
SO ₂	2.4	2.8
Sulfate	1.0	1.1
Deposition		
SO ₂	43.3	37.9
Sulfate	9.1	10.1
Transport off grid		
SO ₂	21.9	23.6
Sulfate	22.4	24.5

deposited on the grid and 44% being transported beyond the grid boundaries. Figure 6 shows most of the SO₂ deposition is near the emission source while sulfate deposition is at a greater distance from the source. Deposition values in Table 1 indicate that SO₂ deposition fraction increases as ground-level sources are added, which probably indicates increased SO₂ removal. Emission transport off the grid demonstrates the same effects. With elevated sources, SO₂ deposition is less, allowing more SO₂ and sulfate mass to escape the grid.

SENSITIVITY ANALYSIS RESULTS

Sensitivity testing indicates which physical processes need to be specified with greater sophistication and which modeling approximations are more tolerable than others. Sensitivity testing also provides some insight into how predictive capabilities can be improved.

The first sensitivity testing approximation for ground-level concentrations is a simple study of the differential equations for the time rate-of-change of mass [Equations (12-15)] with values substituted from Equations (19-25), so that

$$\frac{dQ_1}{dt} = -\left(36Z^{-1} + 5P + 0.02\right)Q_1 \quad (32)$$

$$\frac{dQ_2}{dt} = -\left(3.6Z^{-1} + 250P\right)Q_2 + 0.03Q_1, \quad (33)$$

when time is in hours. The first terms on the right are the dry-deposition rate; the second terms are the wet-deposition rate; and the third terms are the chemical-transformation rate.

Both typical and extreme magnitudes of the separate terms need to be considered. Average Z^{-1} values differ according to whether the release is an elevated utility release or a ground-level nonutility release. Average Z^{-1}

values were calculated using the 10 utility and the 10 nonutility sources with the highest emission rate (see Table 2).

TABLE 2. Average Values of Vertical Distribution Factor (Z^{-1})

	H meters	$\frac{Z^{-1} 10^{-3} \text{ meter}^{-1}}{\text{for } \text{SO}_2 \quad \text{for } (\text{SO}_4^-)}$	
Utility	400	0.53	0.58
Urban	0	1.7	1.1

Average values can be substituted into Equations (32) and (33), along with a climatological precipitation rate of $10^{-4} \text{ m hr}^{-1}$, so that

$$\frac{dQ_1}{dt} = - \left(\begin{Bmatrix} 0.02 \\ 0.06 \end{Bmatrix} + 0.0005 + 0.02 \right) Q_1, \quad (34)$$

$$\frac{dQ_2}{dt} = - \left(\begin{Bmatrix} 0.002 \\ 0.004 \end{Bmatrix} + 0.025 \right) Q_2 + 0.03 Q_1, \quad (35)$$

This parameterization implies that for the climatological case wet deposition of SO_2 and dry deposition of sulfate can be neglected. However, for extreme episodes these two magnitudes could be important. For example, if the precipitation rate during a particular hour is 0.025 m/hr , the wet-removal rate for SO_2 is 0.0125 m hr^{-1} , which is comparable to the climatological dry-removal rate. Another extreme case for dry deposition occurs when the mean wind speed is small, about 2 m sec^{-1} , and very stable conditions prevail. In this case, the dry-removal rate for sulfate from ground-level source is calculated at 0.07 for the first hour (if an hourly time step is used). From these observations all five of the removal parameters are important for episode-type assessments. On the other hand, model sensitivity is greater for transformation rate, dry-removal rate of SO_2 , and wet-removal rate of sulfate than for wet deposition of SO_2 or dry deposition of sulfate in both long and short-term predictions.

Actual modeling sensitivity tests include studies of nine modeling changes as compared to the base-test case previously discussed. These changes include:

- using a fixed mixing depth rather than a variable one
- lowering the maximum mixing depth from 1500 m to 1000 m while using the variable mixing depth
- using neutral stability during all time periods
- studying the effects of changes in dry deposition-velocity for SO_2 over the range from 2.0 cm sec^{-1} to 0.5 cm sec^{-1}
- varying sulfate dry-deposition velocity from 0.1 cm sec^{-1} to 0.2 cm sec^{-1}
- varying the SO_2 wet-removal coefficient from 5 m^{-1} to 30 m^{-1}
- studying the effects of reducing the sulfate wet-removal coefficient from 250 m^{-1} to 150 m^{-1} and to 25 m^{-1}
- varying the SO_2 to sulfate transformation rate in the range 0.01 hr^{-1} to 0.04 hr^{-1}
- increasing the transformation rate during precipitation periods from 0.02 to 0.07 hr^{-1} , and decreasing it to 0.01 hr^{-1} .

The first three changes are modifications in the model that cannot be approximated by a simple parameter change. The last six changes represent effects caused by changes in pollutant removal parameters. Table 3 summarizes results of the sensitivity studies. The table lists the percent of change of average concentration and deposition values for the modified cases and corresponding values for the base test-case for all sources and for utility sources only.

Discussion of this table is limited to the most important features and focuses on utility emissions analysis. The most interesting comparison is between the variable stability base test-case and the constant (neutral) stability run. The SO_2 ground-level air concentration and deposition are each

TABLE 3. Sensitivity Test Results

Case	Fractional Parameter Change	Percent Change in Average Values From the Base Case							
		SO ₂ Concentration		SO ₂ Deposition		Sulfate Concentration		Sulfate Deposition	
		All		All		All		All	
		Sources	Utility	Sources	Utility	Sources	Utility	Sources	Utility
Fixed Mixing Depth		1.0	3.3	0.8	3.0	17.3	20.6	3.2	3.1
Maximum Mixing Depth	0.66	10.7	12.9	10.3	12.3	20.3	21.7	-0.2	-0.1
Constant Stability (Neutral)		22.8	28.2	21.8	27.7	73.5	68.4	4.8	2.8
Dry Deposition Veloc- ity for SO ₂	2.00	-30.5	-26.4	41.8	47.4	-21.7	-18.5	-21.1	-18.7
	0.50	25.3	19.8	-37.3	-39.3	17.5	13.7	16.7	13.4
Wet-Removal Coefficient for SO ₂	6.00	-4.7	-5.4	9.2	11.6	-2.9	-3.1	-6.9	-7.0
Dry Deposition Veloc- ity for Sulfate	2.00	0.0	0.0	0.0	0.0	-4.1	-3.7	14.3	13.6
Wet-Removal Coefficient for Sulfate	0.10	0.0	0.0	0.0	0.0	14.8	15.4	-59.4	-60.2
	0.60	0.0	0.0	0.0	0.0	3.8	4.0	-16.1	-16.3
SO ₂ -to-Sulfate Transformation	0.50	14.5	17.5	13.7	16.8	-43.6	-43.1	-43.4	-43.2
	2.00	-20.7	-24.5	-19.8	-23.7	63.0	60.5	62.1	61.1
SO ₂ -to-Sulfate Transformation Rate During Precipitation	3.50	-6.6	-7.8	-6.5	-7.9	11.1	10.7	43.6	43.0
	0.50	1.7	2.1	1.7	1.7	-3.3	-2.8	-11.4	-11.2

increased about 28% in the variable stability case, while the sulfate air concentration is increased 68%; however, the sulfate deposition is increased only 3%. Further runs were made, using the largest 10 utility sources, to determine what combination of parameterization caused the difference between the increase in SO_2 deposition and sulfate deposition.

The results of the additional runs indicated that constant stability increases the predicted ground-level SO_2 concentrations. This difference in predicted ground-level concentrations occurs because most of the sulfate concentrations are from plume elements that passed through the afternoon period of unstable conditions, thus becoming more vertically extended. At the onset of constant stability (C), the vertical distribution factor (Z^{-1}) drops to about 60% of its former value. Because most of the plume elements are already producing significant ground-level concentrations even though their release height is 400 m, the sudden vertical extension decreases the ground-level sulfate concentrations. On the other hand, much of the significant ground-level SO_2 concentration is produced by plume elements that have not undergone this expansion.

By leaving the stabilities as originally prescribed, comparisons of ground-level air concentration of constant stability modeling to varying stability modeling can be changed by manipulating the deposition velocities. For example, if the deposition velocity for sulfate is increased to 1.0 cm sec^{-1} (thus equaling that for SO_2 , constant stability modeling increases both the SO_2 and sulfate ground-level air concentrations by 29%. However, this comparison retains the inequality between wet-deposition rates. Since the inequality exists, the two increased ratios cannot be said to be equal if the dry-and wet-deposition rates for both compounds are the same. If the deposition rates are the same, the ratio of increase for sulfate would be greater.

From these investigations of model sensitivity, the differences between ratio increases, when constant stability modeling is compared with variable stability modeling, are not likely to vanish unless the parameterization is uncharacteristic of atmospheric behavior.

Table 3 also shows that the greatest sensitivity to parameter changes occurs with transformation and dry deposition of SO_2 . When the transformation rate in the ranges of experimental values is doubled from 2% to 4%, the increase of sulfate ground-level air concentration and sulfate deposition is 60%.

Another feature of the increases when constant stability modeling is compared to variable stability modeling is the difference between the increases of concentration and deposition when comparison is made between SO_2 and sulfate. Both total concentration and deposition for SO_2 increase by 28%, but for sulfate the concentration increases 68% and the deposition 3%. The reason for this difference is that dry deposition, which is proportional to ground-level concentration, dominates SO_2 deposition, while wet deposition, which is proportional to the vertically-integrated concentration, dominates sulfate deposition. Therefore, the increase of deposition of SO_2 matches the increase of ground-level air concentration. However, since there is more deposition of SO_2 , there is less of it available for transformation to sulfate. Therefore, wet deposition of sulfate is less while dry deposition of sulfate, being proportional to the ground-level air concentration, is more. Evidently the two effects largely cancel each other out.

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