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IN EASTERN NORTH AMERICA

by

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Abstract – An analytical model of long distance transport
of air pollutants (Fay and Rosenzweig, 1980) has been
adapted for the estimation of long term (e.g. annual) wet
sulfate deposition in eastern N. America. The model para-
meters have been optimized for best agreement with 1980-
1982 measurements at 109 monitoring sites in this region.
The mean residual of the model and measurement comparison
is 4 kg ha^{-1}y^{-1} (17% of the mean measured value). Trans-
fer coefficients were found to decrease exponentially with
source-receptor distance, having length scales between 1100
and 400 km depending upon whether the source is upwind or down-
wind of the receptor. Source apportionment calculated for
four sites from this model shows that about half of the
deposition is due to 7-8 of the largest source contributors
to each site (aggregated to the state and sub-province level).
A 17-year record of precipitation sulfate measured at Hubbard
Brook, New Hampshire, compares favorably with the model
calculation. Calculated U.S.-Canada transboundary fluxes
agree with previous estimates. Isopleths of 1980-82 yearly
depositions were determined, and the deposition effects of a
typical U.S. emissions reduction proposal were evaluated.

INTRODUCTION

While estimates of the transboundary transport of acid sulfate across the U.S.-Canada border (Galloway and Whelpdale, 1980; Olson, Voldner and Oikawa, 1982) are in reasonable agreement, the more detailed estimates of source contributions (by state and province) to wet deposition at sensitive receptors near the U.S.-Canada border are quite disparate among eight models examined by the U.S.-Canada Working Group (Schiermeier and Misra, 1983; Fay, Golomb and Kumar, 1984). This latter disagreement has engendered an opinion that none of the current models can be relied upon to provide accurate source-receptor relations (National Research Council, 1983). Because measurement of airborne or deposited acid material cannot distinguish among the large number of precursor sources, there can be no direct observational test of the validity of model predictions for source apportionment.

In contrast, the European studies of long range transport and deposition of acid material have converged on generally accepted values for the transnational transport and aggregate deposition within national boundaries (Eliassen and Saltbones, 1983). The national emissions, distances between sources and receptors, and number of emitters and receptors are all comparable to those of eastern North America when the latter are disaggregated at the state and province level. Indeed, some models have been found to perform equally well in both Europe and North America (Johnson, 1983; Fisher and Clark, 1983).

The apportionment of source contributions to deposition at a receptor is a key element for the analysis of acid deposition control strategies which seek to minimize the cost of attaining desired reductions in deposition (Fay, Golomb and Gruhl, 1983). Such receptor-oriented strategies distinguish among sources by their distance and direction from a target receptor. More importantly, the quantitative relation between source emissions and receptor deposition is essential to the minimization of costs and the optimum allocation of emission reductions. While no such strategies have been considered yet in Europe, they have been proposed for eastern North America (Fay et al., 1983; Lugar, 1983; Husar, 1983; Trisko, 1983).

A related problem of source-apportionment is the estimation of the change in deposition to be expected from the various proposed reductions in emissions (Streets, Knudson and Shannon, 1983; Fay et al., 1983). Quantitative estimates of expected benefits from emission control will depend upon the ability to forecast these changes.

Current long range transport and transformation models are linear; that is, the rates of chemical transformation and deposition are first order in the conserved pollutant species. As a consequence, the contribution of any source to a receptor deposition may be separately determined and subsequently aggregated to whatever level is desired. Although there is some dispute as to the validity of the linear assumption (National Research Council, 1983; Hidy et al., 1984), it is not easily resolvable in the absence of suitable nonlinear models.

A necessary requirement for a valid determination of source apportionment is that the method account for the measured spatial and

time dependence of acid deposition. It is known that such comparisons improve when longer time averages of measured and calculated quantities are used (Stewart, Morris and Liu, 1983). In this paper, we consider annual averages only. Such averages are pertinent for assessing the effects of various emission reduction proposals on the annual amount of deposition. We do recognize, however, that seasonal averaging may be required for evaluating intermittent emission control strategies.

There now exists an extensive record of precipitation chemistry in eastern North America (Watson and Olsen, 1984) covering several years and more than two hundred monitoring stations. In addition, there is a seventeen year record at Hubbard Brook, New Hampshire (National Research Council, 1983). Comparison of these observations with various long range transport models, such as those reviewed by the U.S.-Canada Working Group (U.S.-Canada, 1983; Schiermeier and Misra, 1983), would be a vast task. In lieu of such a comparison, we have adapted a model described by Fay and Rosenzweig (1980) to the calculation of long-term (annual or seasonal) averages of wet sulfate deposition. A major advantage of this model is its analytic form which permits very rapid calculation of deposition at the large number of stations for which data is now available. Furthermore, unlike other Eulerian models, it is not necessary to specify boundary conditions. Even more important, it is quite simple to optimize the few parameters of the model so as to best match the measured depositions. In this sense, the resulting source apportionment is empirically determined, although the basis of the model is, like most others, the physical and chemical processes believed to be important to the long range transport and transformation of pollutants.

· In the following sections, we discuss the features of the modified source apportionment model, the comparison of the model calculations with measurements of wet sulfate deposition, the use of this model to determine source apportionment at several sensitive receptor locations in N. America, and the applicability of the model to control strategies.

MODEL STRUCTURE

The source apportionment model is an adaptation of the time-averaged Eulerian model of Fay and Rosenzweig (1980). Like that model, it assumes first order chemical and deposition processes, a uniform height h of the mixed layer, two dependent variables X_p and X_s (primary and secondary sulfur concentrations) which vary with horizontal position only, and flow variables and rate constants which are constant throughout the flow field: mean wind speed w and resultant wind direction δ, horizontal diffusivity D_h, primary time constants for conversion, wet and dry deposition (τ_c, τ_{wp}, and τ_{dp}), and secondary time constants for wet and dry deposition (τ_{ws} and τ_{ds}). For a point source of sulfur of strength Q, the resulting primary and secondary sulfur concentrations at a receptor are:

$$X_p = \frac{Q}{2\pi h D_h} \left\{ \exp(wr\cos\theta/D_h) \right\} K_o(\gamma r) \tag{1}$$

$$X_s = \frac{Q}{2\pi h D_h^2 \tau_c} \left\{ \exp(wr\cos\theta/D_h) \right\} \frac{K_o(\alpha r) - K_o(\gamma r)}{\gamma^2 - \alpha^2} \tag{2}$$

where

$$\alpha^2 \equiv \left(\frac{1}{\tau_{ws}} + \frac{1}{\tau_{ds}} \right) \frac{1}{D_h} + \frac{w^2}{4D_h^2} \tag{3}$$

$$\gamma^2 \equiv \left(\frac{1}{\tau_{wp}} + \frac{1}{\tau_{dp}} + \frac{1}{\tau_c} \right) \frac{1}{D_h} + \frac{w^2}{4D_h^2} \tag{4}$$

r is the source-receptor distance and θ is the azimuthal angle of the receptor as viewed from the source and measured from the resultant wind direction. K_o is the modified Bessel function of zeroth order.

At large distances, the primary and secondary airborne sulfur concentrations given in (1) and (2) decay exponentially with distance from the source. At the source, the primary concentration becomes logarithmically infinite, a physically unrealistic result which can be avoided by adding a small nominal distance of about 10 km to the source-receptor distance r. This represents a typical distance for the emissions from an elevated point source to mix vertically throughout the mixed layer.

The local wet deposition rate of sulfur W is set proportional to the local precipitation rate R:

$$W = \left(\frac{\chi_p}{\tau_{wp}} + \frac{\chi_s}{\tau_{ws}} \right) \frac{hR}{R_o} \tag{5}$$

where R_o is the average precipitation rate throughout the flow field. Note that upon substituting (1) and (2) in (5), the wet deposition rate

is independent of the mixed layer height h. In addition, the sulfur concentration in precipitation, W/R, is independent of both h and R. In using (5) to determine wet sulfate deposition, we assume that all wet sulfur appears in the form of sulfate.

The assumption that the local wet deposition of sulfur is proportional to the local precipitation rate is not exactly consistent with the solution (1) and (2), which requires that the wet deposition depends only upon the sulfur concentration and not the local precipitation rate R. But because the precipitation varies about the area-wide mean within a length scale which is smaller than the predominant length scales of (1) and (2) (which we found to be about 1000 km), the correction to the local concentration and also the deposition W due to this discrepancy would be small and would balance out over the domain of the solution.

The averaging time for which the solution (1) and (2) is valid must be sufficiently long that the time dependent terms in the mass conservation equation will average nearly to zero. In order for the parameters to be uniform, the averaging time must include several significant deposition episodes throughout the flow field. Therefore, the minimum averaging time ought to be several months. In this paper, we consider only one to three year averages. We note, however, that there is a significant difference between summer and winter wet sulfur deposition rates (MAP3S/RAINE, 1982), and therefore expect that seasonal averaging would be useful.

As is the case for all linear models, there is some uncertainty about the approximation of linear representation of what may be essentially nonlinear processes. While it is possible to include suitable

time-averaged nonlinear effects in the Eulerian model of Fay and
Rosenzweig (1980), an analytic solution cannot be found, and the ability
to easily optimize the model parameters is lost. Instead, we first seek
to determine the degree by which the linear model succeeds in describing
the spatial and temporal variation of wet deposition measurements before
elaborating the model.

The mass flux F across a boundary line in the horizontal plane can be
found from:

$$F = h \int \vec{n} \cdot (X\vec{w} - D_h \; grad \; X) ds \qquad (6)$$

in which $\vec{w}$ is the average wind vector, $\vec{n}$ is the unit vector normal to the
boundary line in the positive direction of the flux, grad is the gradient
operator in the horizontal plane, and s is the distance along the
boundary line. We have found it easier to calculate the gradient
numerically than to evaluate it in closed form using the solution (1) and
(2).

The simplicity of this model greatly enhances its usefulness for
source apportionment. Once the optimum values of the eight parameters
have been determined, it is quite easy to superpose the contributions of
a large number of sources to the deposition at a particular receptor. In
addition, source-receptor transfer coefficients are readily determined.

MODEL OPTIMIZATION

The long-range transport model was optimized to achieve the closest fit with measurements of wet sulfate deposition by varying the values of the parameters of the model. The degree of fit with the measurements of wet sulfate deposition is defined quantitatively according to a least squares error criterion. A root mean square error E is defined by

$$E^2 = \Sigma(\text{observation} - \text{prediction})^2 / \Sigma(\text{observation})^2 \qquad (7)$$

where the summation is performed over all observations. Thus, during the optimization, eight parameters of the model (excluding h) are determined so as to minimize E.

Three years (1980-82) of observational data of wet sulfate deposition (Watson and Olsen, 1984) were used in this optimization. The 252 precipitation chemistry monitoring stations contributing to this data set include the APIOS, APN and CANSAP networks in Canada and the MAP3S, NADP and UAPSP networks in the U.S. Data used in this comparison consisted of the cumulative wet sulfate deposition at each monitoring site for each calendar year. The sampling period at some APIOS and all CANSAP sites was one month, at the remaining APIOS and all APN sites one day, at the NADP sites one week, at the UAPSP sites one day, and at the MAP3S sites by event. Although the land area covered by these networks includes all of the North American continent, this modeling exercise was confined to an eastern region within roughly 30-50° N. Lat. and 70-90° W. Long. The

locations of the monitoring sites within these boundaries are shown in Figure 1.

In addition to wet sulfate deposition, the data included total annual precipitation. However, the measurements were not always complete. The data collected during a sampling period was counted when the amount of precipitation was correctly measured, or when it was definitely known that no precipitation occurred during that period. If neither of these conditions was satisfied, then the data was not counted. The percentage of days of the year during which data was counted was defined as the "precipitation coverage length". In this data analysis only data from sites with precipitation coverage length exceeding 75% were used.

An annual emission inventory for the years 1976-1982 aggregated at the state level was obtained from the National Emissions Data System (Colon-Velez, 1984). A Canadian emission inventory for 1980 aggregated by 15 sub-province regions was taken from U.S.-Canada (1983). Each state in the U.S. and sub-province in Canada was assigned an emission centroid, taken from U.S.-Canada (1983). Thus, with 49 sources in the contiguous U.S. and 15 sources in Canada, a 64 point source emission inventory was created. (In a trial optimization, results were barely affected when the number of emission centroids was increased by subdividing each state or province into 5 sectors.)

A quasi-Newton optimization method was used. All parameters were allowed to float during the optimization procedure except the dry deposition time constant of the primary species, SO_2, which was fixed at $\tau_{dp} = 2 \times 10^5$ s, corresponding to a deposition velocity of about 0.5 cm s^{-1}. When this parameter was allowed to vary in the optimization procedure, physically unrealistic values were obtained.

. As indicated in (5), the model wet deposition values are proportional to the ratio of local precipitation R to the domain-wide average R_o. The domain-wide average was computed by averaging the observed annual precipitation over all the sites in the modeling domain. If this proportionality is not used, the error E increases by a factor of 1.4.

Model optimization was performed for each of the three years as well as for a three-year average. For the multiyear analysis, only data from 109 sites that had a three year continuous record with average coverage greater than 75% was used. For this analysis a scatterplot comparing the predicted with measured annual deposition is shown in Figure 2. Note that the values of wet sulfate deposition presented are those corresponding to the actual coverage at each site, which may be smaller than the annual depositions. For the data of Figure 2, the error E as defined above is 17% and the correlation coefficient is 0.87, indicating that the model explains more than 75% of the variance. This error corresponds to a root mean square value of the wet deposition residual of 4 kg $ha^{-1}y^{-1}$. Similar scatterplots for the individual years 1980-82 gave errors in the range 18-22%, and correlation coefficients 0.81-0.87.

The model parameters that gave the least error E for the multiyear analysis are listed in Table 1. They all lie within "conventional" value ranges. For example, the optimized horizontal diffusivity D_h = 4.3 x 10^6 m^2s^{-1} is in the range 10^6 - 10^7 estimated by Durst et al. (1959); the average wind speed w = 7.1 ms^{-1} and the mean direction δ = $214°$ (southwesterly) is typical for the 850 mb level over much of eastern North America, especially in summer (Whelpdale, Low and Kolomeychuk, 1984). The time constants (inverse rate constants) are also in the range used in other acid deposition models (U.S.-Canada, 1983).

Table 1 lists also the sensitivity of the error E to the individual parameters. Because E has been minimized, it will vary about its minimum only as the square of the variation of a parameter p. We therefore define a sensitivity S as:

$$S = \frac{1}{2} \left(\frac{p^2}{E} \right) \frac{\partial^2 E}{\partial p^2} \qquad (8)$$

where p is any parameter. The sensitivities are listed in Table 1. It can be seen that the error is more sensitive to D_h and δ and less sensitive to the remaining parameters. The sensitivity of δ is large because the range 2π is much less than that of the other parameters (zero to infinity). Since sensitivity and uncertainty bear an inverse relationship, one may consider the former parameters to be more accurately determined than the latter.

SOURCE APPORTIONMENT

Transfer coefficients

The transfer coefficient T_{ij} is defined as the ratio of the annual wet sulfur deposition at the receptor j to the annual sulfur emission from the source i. It is obtained from (1)-(5) as a function of the optimized model parameter set p, the distance r_{ij} and the angle θ of the receptor as seen from the source, measured with respect to the resultant wind direction.

To illustrate the dependence of T_{ij} on the source-receptor distance r_{ij} and direction θ , we show in Figure 3 curves of T_{ij} vs. r_{ij} for

three source orientations with respect to the resultant wind direction:
(a) upwind, (b) crosswind, and (c) downwind of the receptor. In the
linear sections of the curves, the 1/e-th length scale for upwind sources
is about 1100 km, crosswind 600 km, and downwind 400 km. As one would
expect, sources lying upwind have a longer action radius than downwind
sources.

Deposition contours

The amount of wet deposition at a receptor j from all sources i is
obtained from

$$D_j = \Sigma T_{ij} Q_i \qquad (9)$$

where Q_i is the strength of source i, i.e., the annual emissions of
sulfur. In Figure 4 we show the modeled annual wet sulfate deposition
isopleths over eastern N. America for 1980-82. The higher deposition
contours (30 kg $ha^{-1} y^{-1}$ or larger) occur over parts of Ohio, W.
Virginia, Pennsylvania, New York, Quebec, Ontario and Michigan, extending
northeast from the largest emission sources. Based on the error
analysis, the isopleth values are uncertain by about 4 kg $ha^{-1} y^{-1}$.
Figure 4 also shows the measured depositions. The values are annual
averages extrapolated to 100% coverage from the 3-year record 1980-82.
When two or more stations are clustered, the average deposition at those
stations is given. The residuals between predictions and observations
appear to be fairly randomly distributed.

Trend analysis

In N. America, one monitoring station located at Hubbard Brook, New
Hampshire, operated for about 20 years with consistent data quality. The
annual average concentration of sulfate in precipitation at Hubbard Brook

from 1964 through 1981 was reported by the National Research Council
(1983). In Figure 5, a comparison is made between the reported and model
results. In constructing this comparison, we used the multiyear transfer
coefficients and the 5-yearly emission inventory of Hidy et al. (1983)
for the period 1960-80. It is seen that the model results capture quite
well the time trend, although individual years show some fluctuations
about the mean trend. This is probably due to a greater variability of
yearly meteorological factors than is accounted for by the model. The
rise in the calculated annual average sulfate concentration in the 1960's
and the decline in the 1970's reflects the overall emission trend of
SO_2 in the northeastern U.S.

Source apportionment

The fractional contribution of source i to the wet sulfate deposition
at receptor j can be determined from:

$$S_{ij} = \frac{Q_i T_{ij}}{\sum_i Q_i T_{ij}} \tag{10}$$

The source contributions of states and provinces to wet sulfate
deposition at four receptors are presented in Figure 6. The receptors
are Whiteface Mountain, New York; Mt. Mitchell, North Carolina; Quebec
City, Quebec; and Boundary Waters-Canoe Area on the border of Minnesota
and Ontario. These receptors were designated sensitive to acid
deposition by the U.S.-Canada Working Group on Transboundary Air
Pollution (U.S.-Canada, 1983). The percent contributions of Figure 6 for
the states and sub-provinces are based upon the 1980-82 source inventory
and the parameters of Table 1. For the cases studied, the 7-8 largest
contributors account for about 50% of the annual wet sulfate deposition.

Assessment of a sulfur emission reduction proposal

We have used the model to assess the deposition reduction that would ensue from limiting sulfur emissions according to the requirements of the bill introduced by Senator Mitchell (S.1706) in the 1982 U.S. Congress. The bill would require a 9.1 million ton (metric) per year SO_2 emission reduction out of a total of 20.4 Mty^{-1} emitted in 1980 in 31 states east of or bordering on the Mississippi River. The emission reduction would be allocated to the states so as to achieve approximately an equal average state-wide sulfur emission per fuel heat value. Using the estimated state reductions of Friedman (1981) and the multiyear transfer coefficients, we calculated the expected percent reduction of wet sulfate deposition over eastern N. America, as shown in Figure 7. The largest deposition reductions of 45% would be obtained over the Ohio River Valley, and an average of 35% in the sensitive areas of the northeastern U.S. and southeastern Canada.

The pattern of deposition reductions illustrated in Figure 7 depends both upon the transfer coefficients and the allocation of emission reductions. Other allocations of emission reductions would give different patterns. Fay, Golomb and Gruhl (1983) estimated that a deposition reduction in the Adirondacks area equal to that of Figure 7 could be obtained with only a 6.4 Mty^{-1} reduction in emissions if the latter were concentrated more at those sources closer to the Adirondacks. Determination of deposition reduction patterns similar to Figure 7 are easily accomplished using the source apportionment model described above.

Transboundary flux

The model can be used to estimate the transboundary sulfur flux between the U.S. and Canada east of the Mississippi River. From Eq. (6) and based on the multiyear data set, we estimate the transboundary fluxes given in Table 2. It is seen that our estimates are in good agreement with the figures of Olson et al. (1982) and Galloway and Whelpdale (1980).

CONCLUSIONS

The analytic model developed in this paper is easily applied to the calculation of annual (or multiyear) wet sulfate deposition isopleths for any spatial distribution of sulfur sources. Parameters of the model chosen to minimize the difference between calculation and measurements for 109 monitoring sites in eastern North America over the period 1980-1982 have values commensurate with those used in other models. The mean difference between the model calculation and measured deposition is 4 kg ha^{-1}y^{-1} of SO_4, or 17% of the mean measured deposition rate.

When used in source apportionment analyses, the model supports many useful assessments: the trend in deposition at a single location over many years; the transboundary fluxes due to sources on either side of a boundary; the fractional contribution of each source to any receptor deposition; and the deposition decrements resulting from various national emission reduction strategies.

The model transfer coefficients, assumed to be independent of source or receptor location within eastern North America, decay exponentially

with source-receptor distance. The decay length varies between 400 and 1100 km, depending upon the orientation of the source-receptor direction with respect to the resultant wind direction (SW). Upwind sources contribute much more to receptor deposition than downwind ones of equal strength.

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Table 1. Optimized model parameters

Parameter	Optimized value	Sensitivity (S)
D_h	$4.3 \times 10^6 \ m^2 s^{-1}$	3.6
w	$7.1 \ ms^{-1}$	0.4
δ	214 degrees	35.3
τ_c	$1.9 \times 10^5 \ s$	1.0
τ_{wp}	$11.3 \times 10^5 \ s$	0.3
τ_{ws}	$0.6 \times 10^5 \ s$	0.8
τ_{dp}	$2.0 \times 10^5 \ s$	(fixed)
τ_{ds}	$12.5 \times 10^5 \ s$	1.1

Table 2. 1980 Eastern North American
transboundary fluxes

	This paper	Galloway and Whelpdale (1980)	Olson et al. (1982)
Flux to Canada from U.S. sources* $(Tg \ S \ y^{-1})$	1.7	2.0	1.2
Flux to U.S. from Canadian sources* $(Tg \ S \ y^{-1})$	0.4	0.7	0.35

*Eastern U.S. sources = $10.2 \ Tg \ S \ y^{-1}$; Eastern Canadian sources = $2.15 \ Tg \ S \ y^{-1}$

NETWORK
+ = APIOS
O = APN
▽ = CANSAP
△ = MAP3S
□ = NADP
◇ = UAPSP
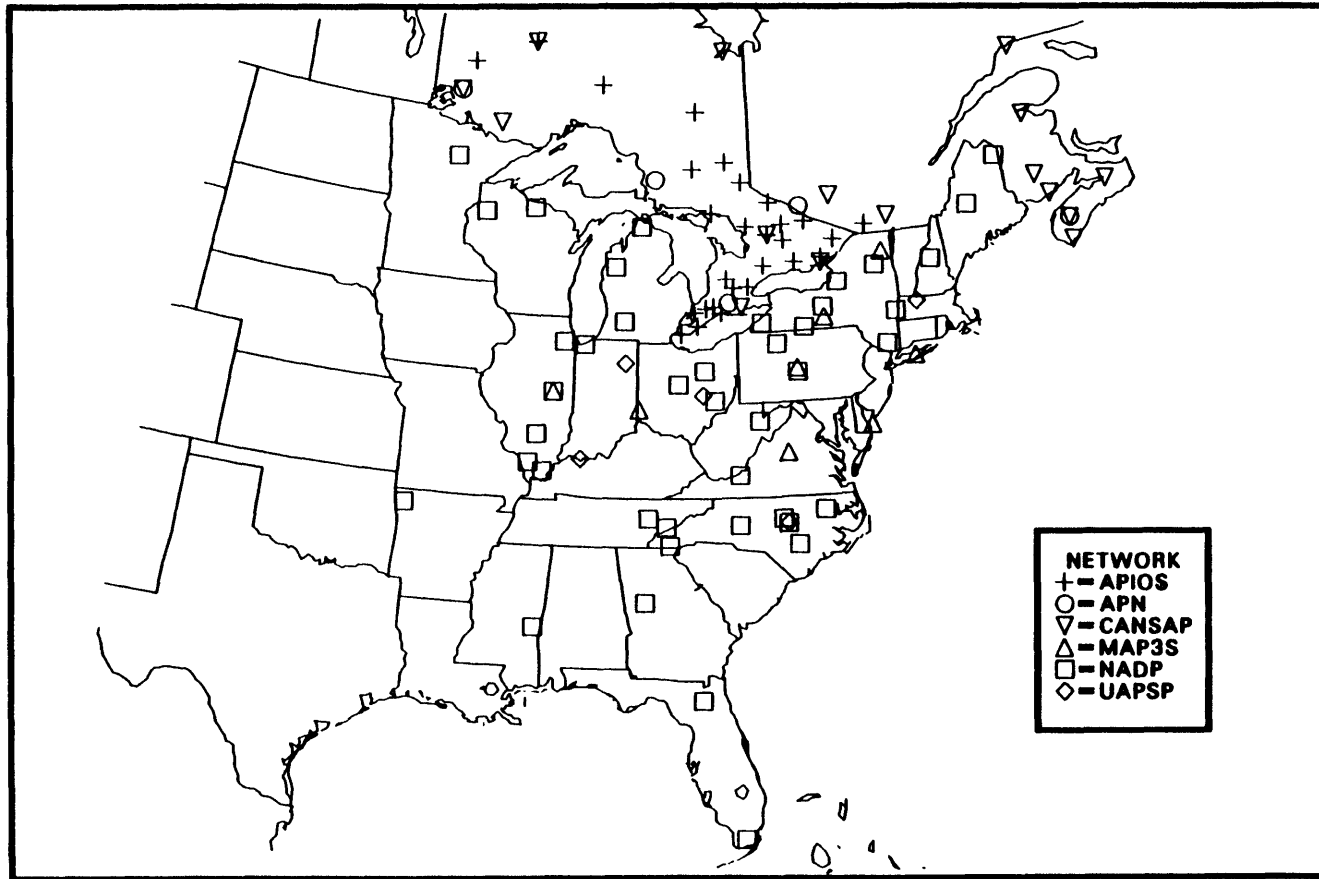
Fig. 1   Precipitation chemistry monitoring stations used in this
               study.
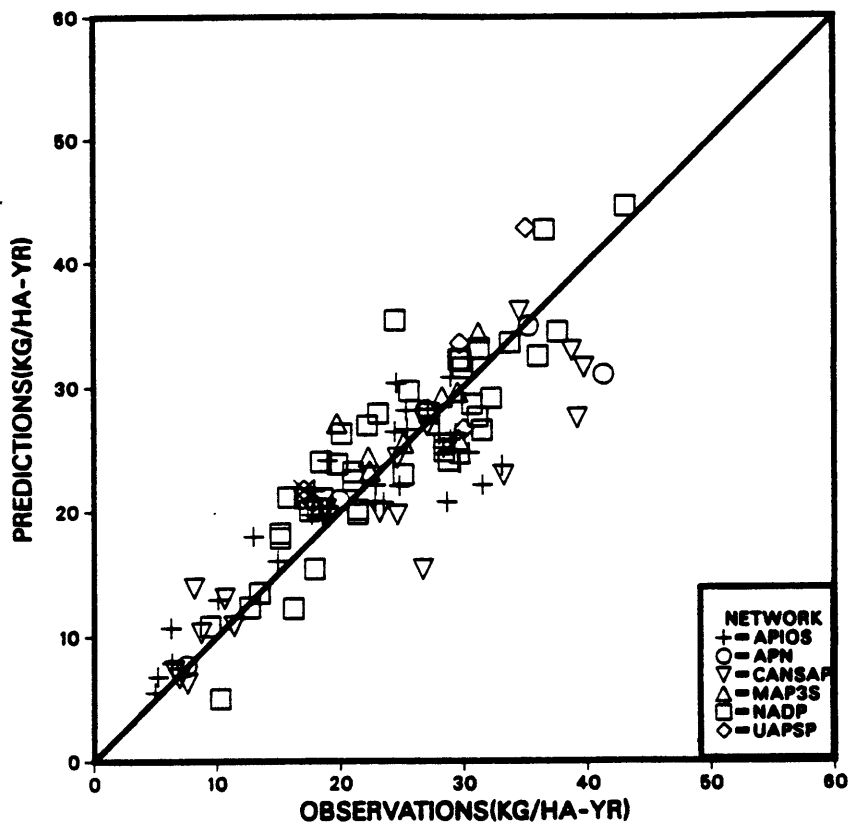
Fig. 2 Predicted vs. observed wet sulfate deposition at 109 sites for 1980-82.
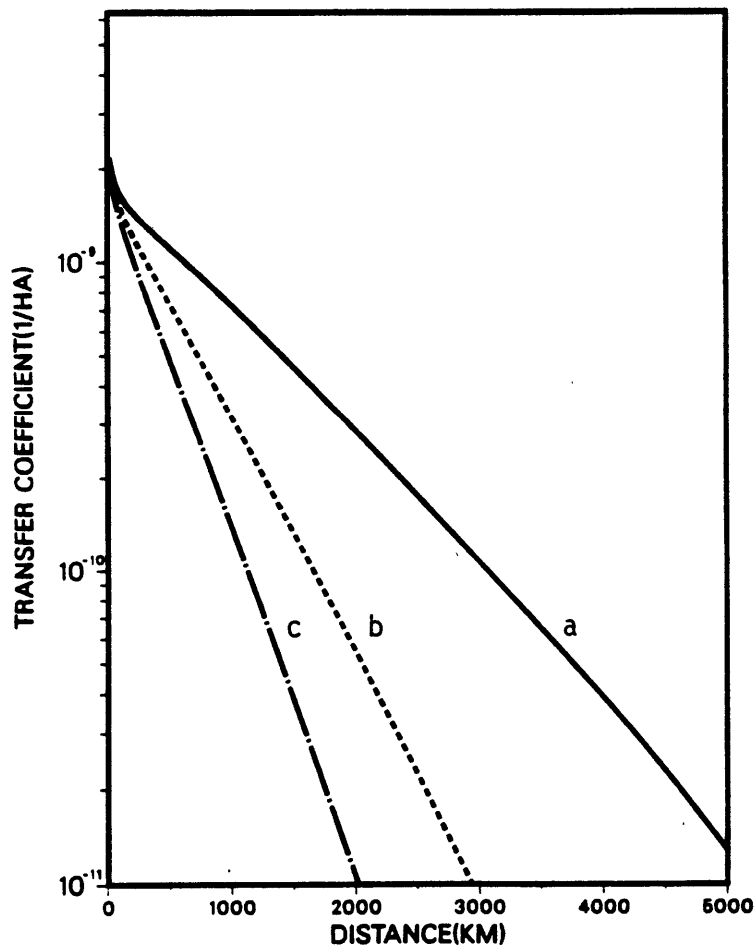
Fig. 3 Transfer coefficient as a function of source-receptor distance for source: (a) upwind (b) crosswind and (c) downwind from receptor.
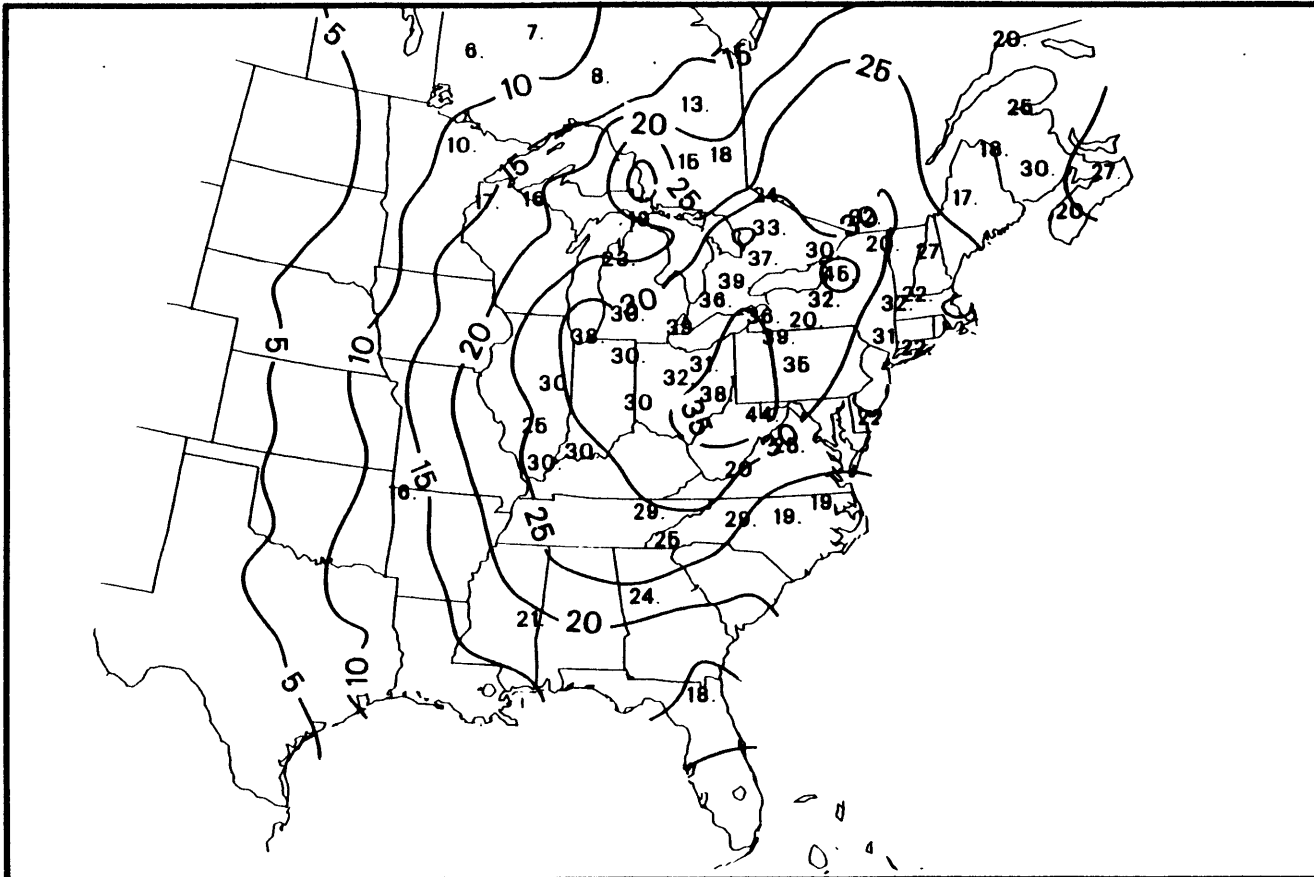
Fig. 4   Modeled annual wet sulfate deposition isopleths
for eastern North America and observed depositions
(small numbers) for 1980-82 (kg SO_4 $ha^{-1}y^{-1}$).
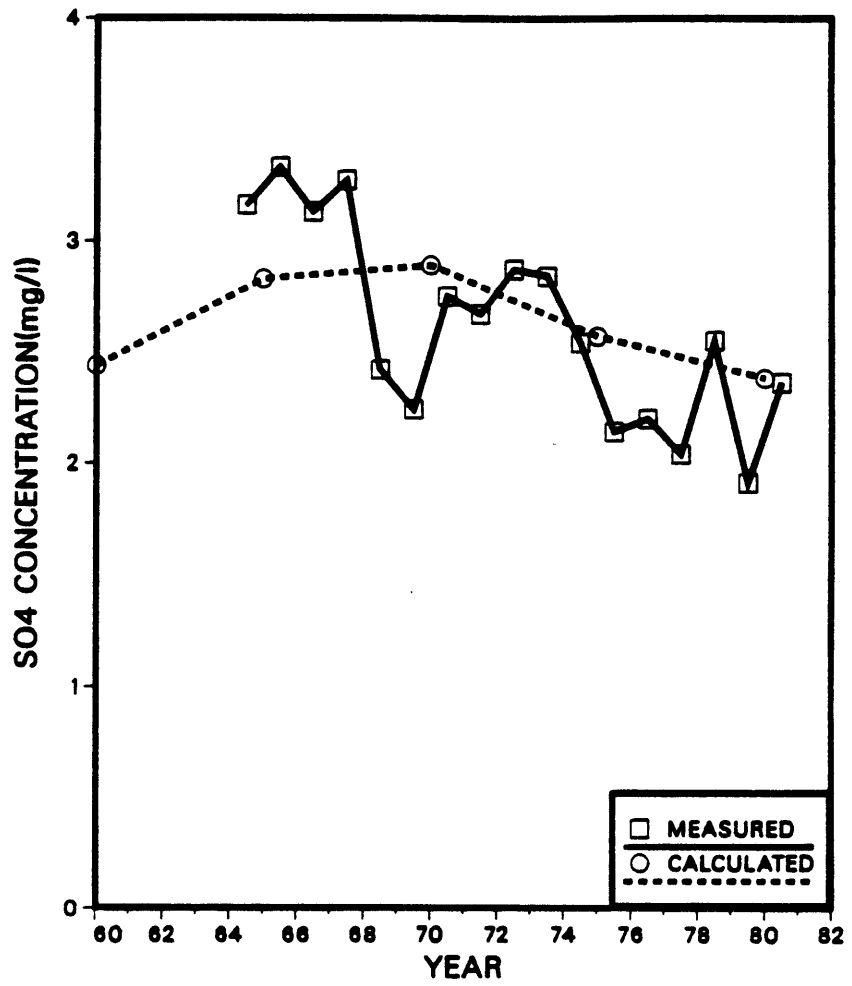
Fig. 5   Calculated and measured annual mean sulfate concentration
          (mg ℓ^{-1}) in precipitation at Hubbard Brook, New Hampshire.
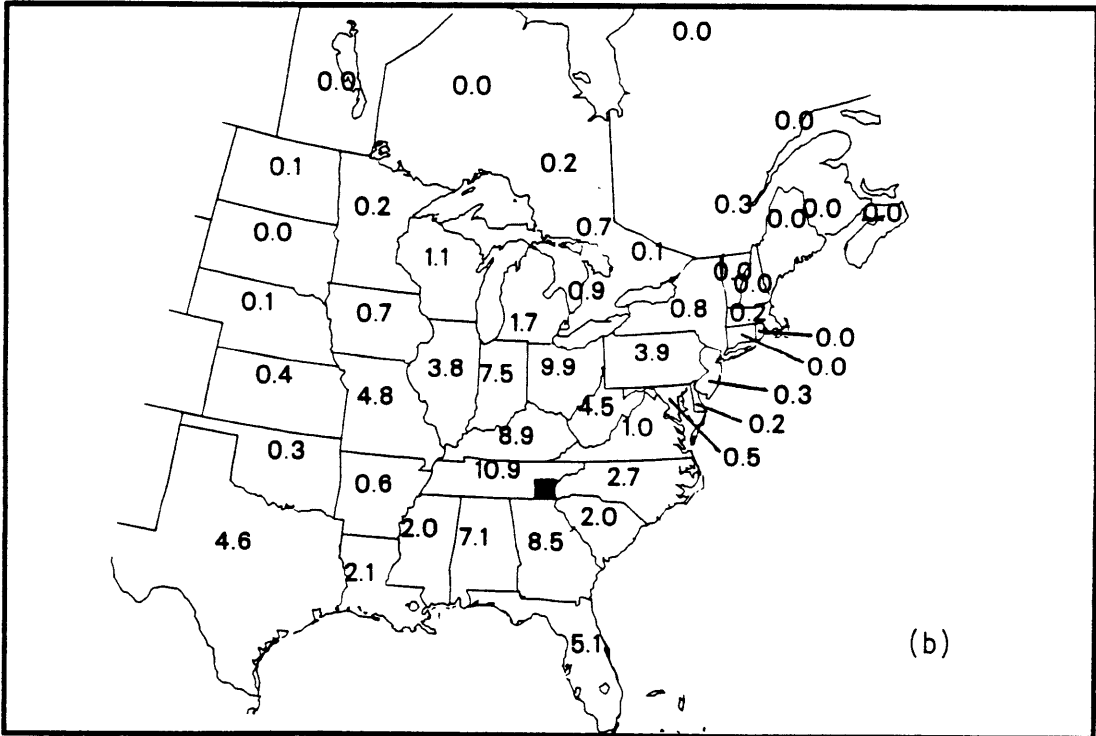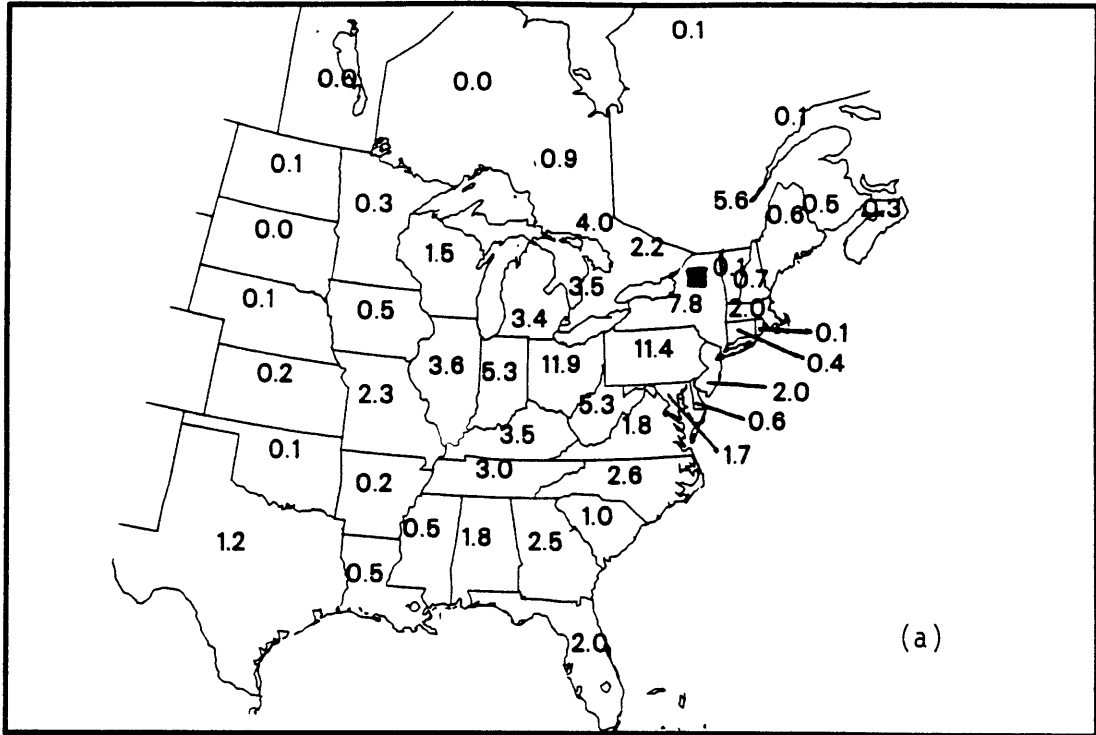
Fig. 6   Source apportionment of wet sulfate deposition (percent per state or sub-province) at (a) Whiteface Mountain, New York, (b) Mt. Mitchell, North Carolina, (c) Quebec City, Quebec, and (d) Boundary Waters, Ontario.
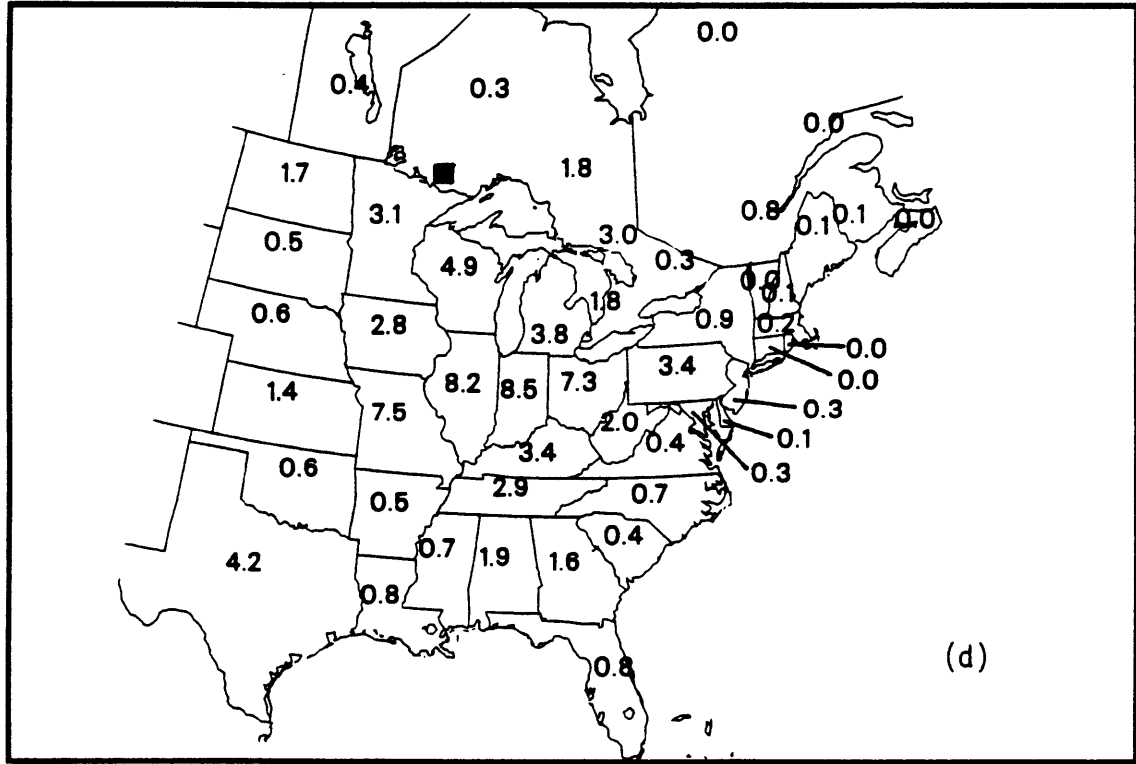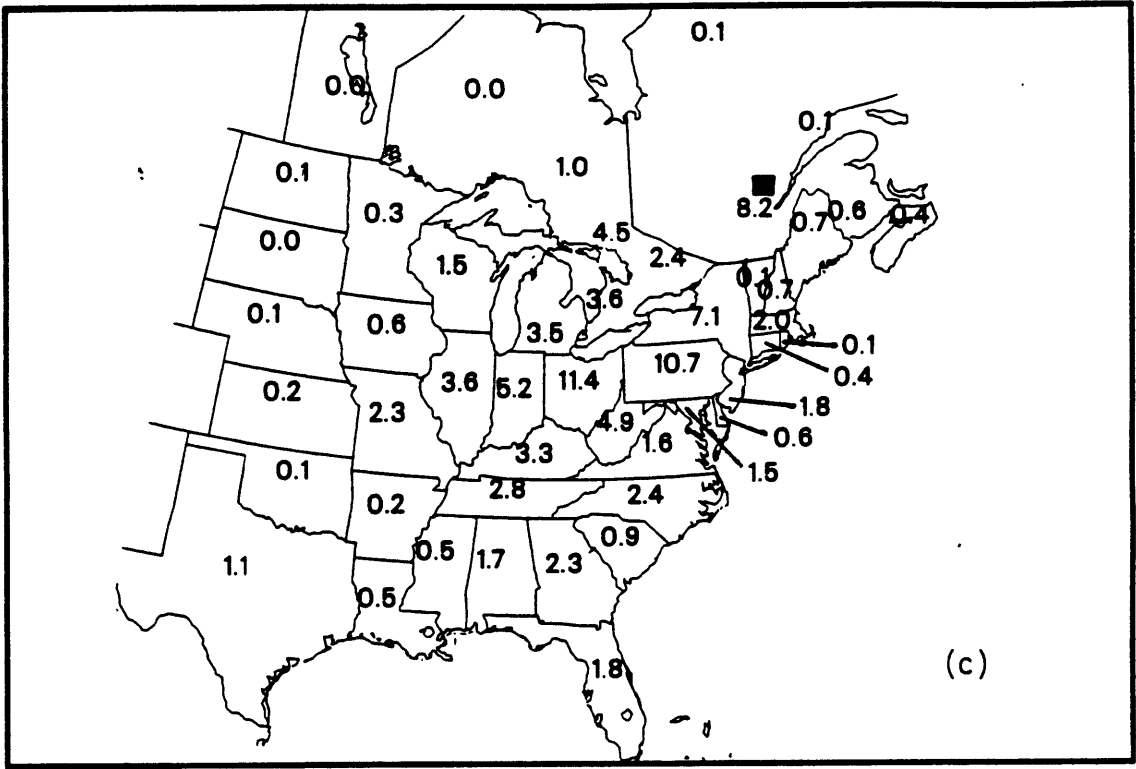
(c)

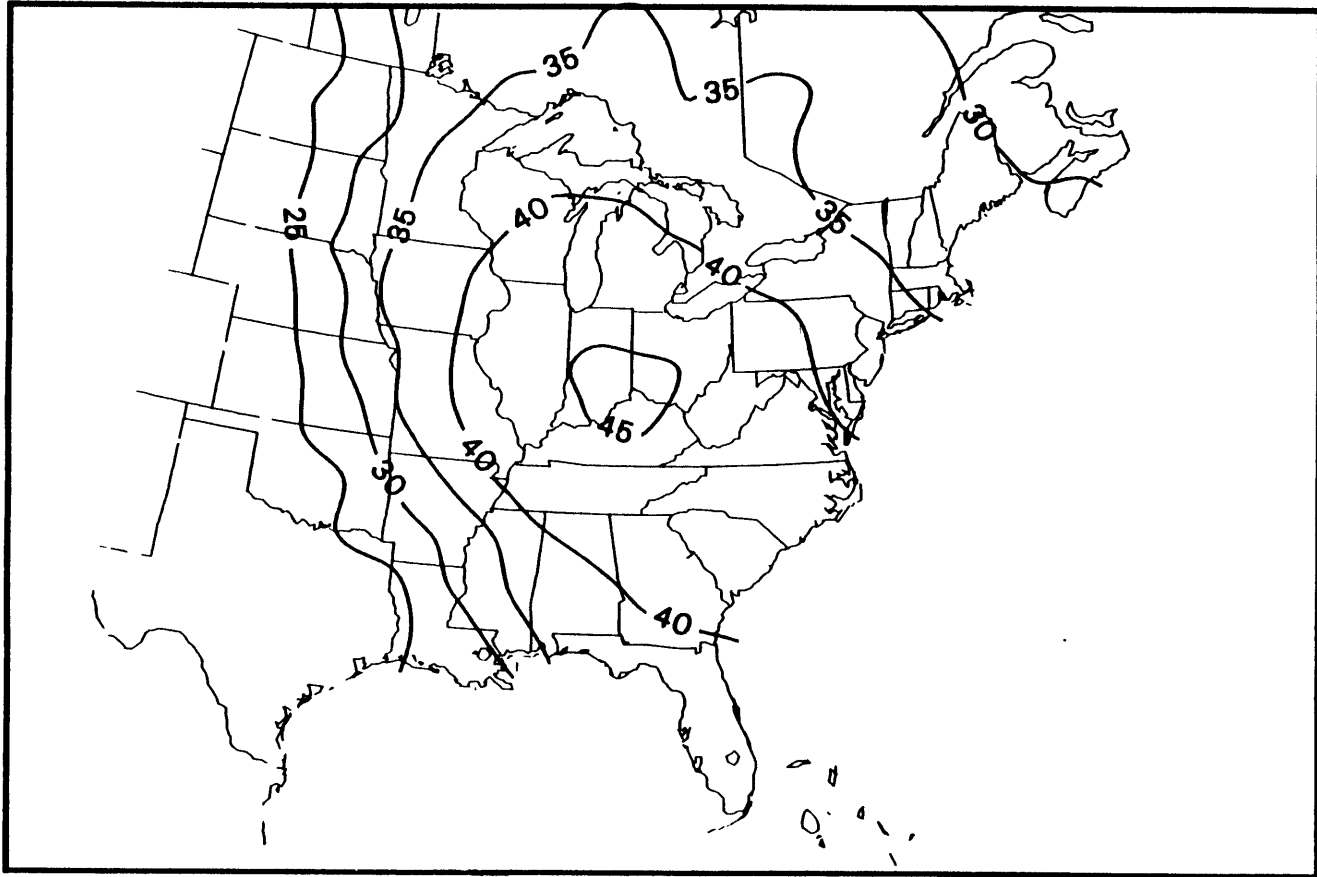(d)

Fig. 6   continued

Fig. 7   Calculated wet sulfate deposition reduction for Mitchell
          bill (percent below 1980-82 values).