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ANALYSIS OF SOURCE-RECEPTOR RELATIONSHIPS FOR SULFUR COMPOUNDS USING SPATIAL AND TREND TECHNIQUES

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1. INTRODUCTION

The National Acid Precipitation Assessment Program (NAPAP) recently published its Integrated Assessment, which describes the causes and effects of acidic deposition and presents a comparative evaluation of the effects of future emissions control scenarios (NAPAP 1990). One component of this Integrated Assessment is an empirical investigation of the relationship between emissions and deposition of sulfur compounds, using available data on sulfur dioxide (SO_2) emissions and wet sulfate ($SO_4^=$) deposition. The rationale for such an investigation is twofold: 1) the analyses provide observational evidence that links emissions and deposition, with cause and effect relationships inferred when atmospheric processes are considered, and 2) the analyses apply these relationships to policy questions on source attribution at specific receptors and the linearity between emissions changes and deposition changes.

This paper summarizes the analysis of the empirical relationship between sources and wet deposition of sulfur compounds presented in Question 2 of the Integrated Assessent. A spatial analysis of the relationship between emissions and wet deposition is presented for 1985. The relationship between trends in emissions and wet deposition is presented for the period from 1979 to 1987. The results of these analyses are discussed in the context of the role such information plays in assessing emissions control options.

2. SPATIAL ANALYSES

Comparison of spatial patterns of SO_2 emissions and wet $SO_4^=$ deposition over the eastern United States and southeastern Canada is possible for the year 1985, for which an extensive emissions inventory is available.

2.1 Data Sources

The 1985 NAPAP Emissions Inventory, described in detail by Placet et al. (1990), includes data on the emissions rates, physical characteristics (e.g., stack heights, exit velocities, and temperatures of the gases exiting the stacks), and locations of SO_2, NO_x, and VOC point sources emitting more than 100 short tons/yr. Area sources of these compounds are estimated collectively at the county level. Estimates of natural emissions of these species are included.

Wet deposition data from six monitoring networks in the United States and Canada were assembled to provide spatial patterns of analyte concentrations and deposition for a variety of compounds over a period of several years. The patterns are developed from the combined data sources by use of kriging as described by Sisterson et al. (1990).

2.2 North American Sulfur Budget

Regionally representative state and provincial wet sulfur (S) deposition was estimated from spatially interpolated wet S deposition fields (Sisterson et al. 1990). State and provincial dry S deposition was estimated by scaling the wet S deposition by the ratio of wet and dry S deposition obtained at 6 sites for which wet and dry S deposition estimates were available. State and provincial wet and dry S deposition were adjusted for urban areas. The urban contribution was assumed to be 1.75 times the regional wet S deposition and 5 times the dry S deposition. Droplet S deposition to the Appalachians above 610 m was assumed to be 3 times the regional wet plus dry S deposition. Uncertainties in the deposition fields for the eastern United States were estimated to be 10%, 40%, and 30% for wet, dry, and total S deposition, respectively, and are higher elsewhere.

Those estimates of total S deposition to the United States and Canada and emissions of S from this region (Placet et al. 1990) were combined with model-derived regional S transport to obtain the mass budget for S (Venkatram et al. 1990) shown in Fig. 1.

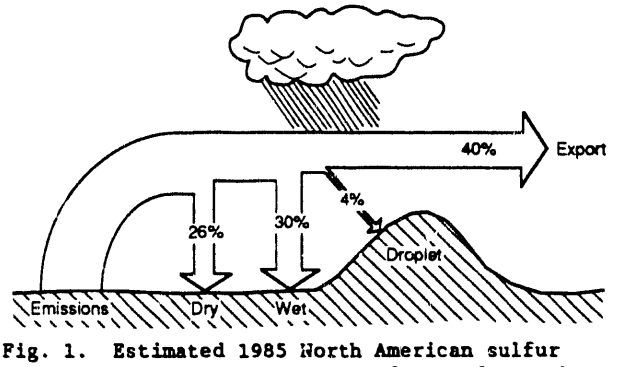

Fig. 1. Estimated 1985 North American sulfur budget showing the percentage of mass for each pathway (annual emissions are 13.3 million tons).

2.3 Relationship Between Emissions and Deposition Patterns

The 1985 SO_2 emissions and wet $SO_4^=$ deposition data were transformed into a common visual format so that the deposition and emission fields over the eastern United States could be compared. Emissions and wet deposition data were incorporated into a grid of hexagonal cells, with each cell encompassing an area of about 2700 km^2. Each data set was smoothed twice, the smoothed value for a cell being the average of the values for that cell and its six neighbors. This approach eliminated sharp discontinuities in the visual presentation of the emissions data, particularly near coastal regions.

The spatial distribution of the smoothed, area-averaged SO_2 emissions and wet $SO_4^=$ deposition data is shown in Fig. 2. The shading scheme was chosen to highlight regions of high and low emissions and high and low deposition. Shading changes from white to light gray when deposition exceeds 20 kg ha^{-1} (the median value of the distribution of deposition levels for all cells). Shading changes from white to a moderate gray when emissions exceed 28 kg ha^{-1} (the median value of the distribution of emission strength for all cells). Cells for which emissions exceed 28 kg ha^{-1} and wet deposition exceeds 20 kg ha^{-1} are shaded a dark gray.

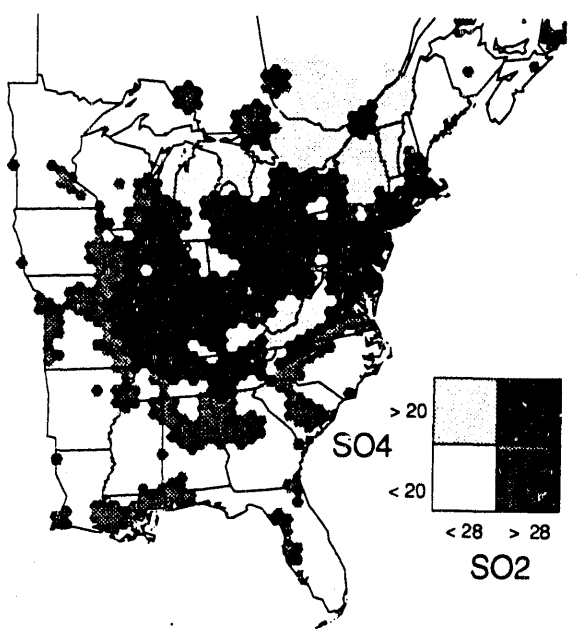

Fig. 2. Relationship between 1985 SO_2 emissions and wet $SO_4^=$ deposition.

Fig. 2 clearly shows the spatial relationship between the region of high SO_2 emission strength in the Upper Midwest and Ohio River valley, and the region of high wet $SO_4^=$ deposition extending to the northeast from the Ohio River valley. The highest wet deposition occurs to the east and north of the major emissions sources in the eastern United States; this shift has an approximate length scale of 200 to 500 km.

The displacement of the areas of highest deposition and emissions is partially explained by the eastward and northward movement of storm systems in the region and the fact that several hours residence time is required for SO_2 to be converted to $SO_4^=$ before it is readily removed by precipitation systems. Thus, a qualitative relationship is established between the major SO_2 emissions regions and the areas of high wet $SO_4^=$ deposition in the eastern United States. However, attribution of specific sources to specific receptors cannot be established from this analysis alone.

3. TRENDS ANALYSIS

Comparison of trends in the emissions of sulfur compounds and their wet deposition was possible for the years 1979 to 1987, a period for which emissions and wet deposition data are concurrently available for the United States.

3.1 Trend Data Bases

The temporal and spatial resolution of SO_2 emissions needed for comparison to wet $SO_4^=$ deposition is provided by the Month and State Current Emissions Trends (MSCET) data base (Kohout et al. 1990). Time series of state-level and national annual SO_2 emissions over the 1979 to 1987 period are shown in Fig. 3 for the upper and lower quartiles, the median, and the average of the 48 contiguous states of the United States and for the combined (USA) 48 contiguous states (divided by 100 for plotting purposes).

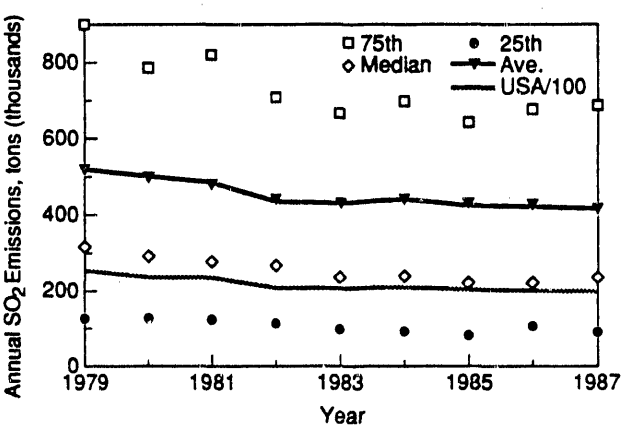

Fig. 3. Temporal pattern of SO_2 emissions for the 48 contiguous United States for 1979 to 1987.

Wet deposition data in the 1979 to 1987 period were obtained for 35 sites in the United States and Canada that met the data completeness criteria defined by Simpson and Olsen (1990) for trends analyses. The locations of these sites are shown in Fig. 4. Time series of wet $SO_4^=$ deposition are shown in Fig. 5 for the upper and lower quartiles, the median, and the average of the trend sites.

Figures 3 and 5 indicate that, nationally, both SO_2 emissions and wet $SO_4^=$ deposition decreased during the 1979 to 1987 period.

3.2 Analytical Approach

The trend sites were grouped into seven geographic regions, shown shown in Fig. 4, to reduce the spatial variability in the annual wet $SO_4^=$ deposition data. The yearly deposition for all sites in a region was averaged to obtain a yearly regional wet $SO_4^=$ deposition.

Emissions trends can be calculated for any

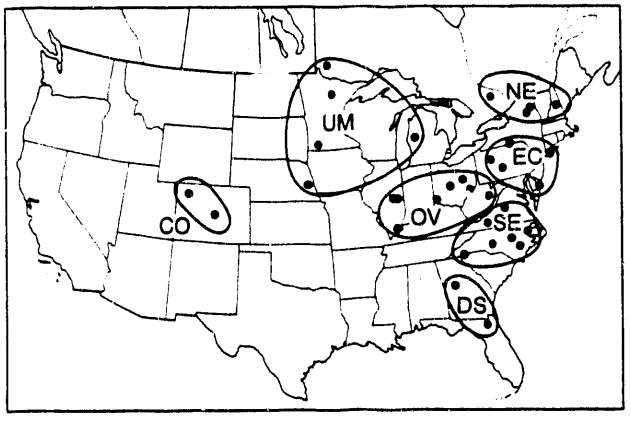

Fig. 4. Location of wet $SO_4^=$ deposition sites qualifying for trends analysis and groups of sites used to define regional deposition trends.

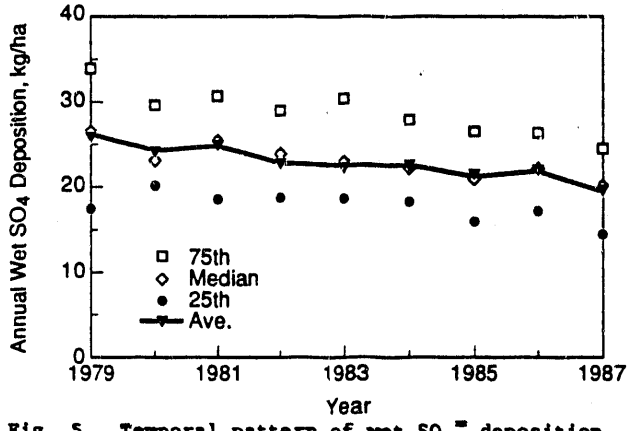

Fig. 5. Temporal pattern of wet $SO_4^=$ deposition at qualifying trend sites for 1979 to 1987.

geographically defined area. However, defining that area is difficult because it is not known, from measurements, which sources contribute to the deposition at a site. Therefore, the Advanced Statistical Trajectory Regional Air Pollution (ASTRAP) model (Shannon 1981) was used to determine the approximate attribution of wet deposition for each regional group of sites by SO_2 state-level emissions sources over the 1979-1987 period. The mean source attribution calculated by ASTRAP gave the weights that were used to form a weighted sum over all states of the yearly MSCET SO_2 emissions by state that affected each region.

The trend in regional wet deposition and emissions was estimated by applying least-squares linear regression to the time series of yearly regional wet $SO_4^=$ deposition and weighted-sum SO_2 emissions normalized by their respective long-term means over the 1979-1987 period.

3.3 Relationships Between Trends

Figure 6 shows the relationship between the trends in normalized wet $SO_4^=$ deposition and SO_2 emissions by region. The horizontal and vertical bars give the estimated standard error of the trend obtained by the linear regression. The diagonal is the line of equal trends in wet $SO_4^=$ deposition and SO_2 emissions.

Several factors affect the relationship between trends in emissions and trends in wet deposition. First, the source attribution estimates used to weight the regional emissions do not

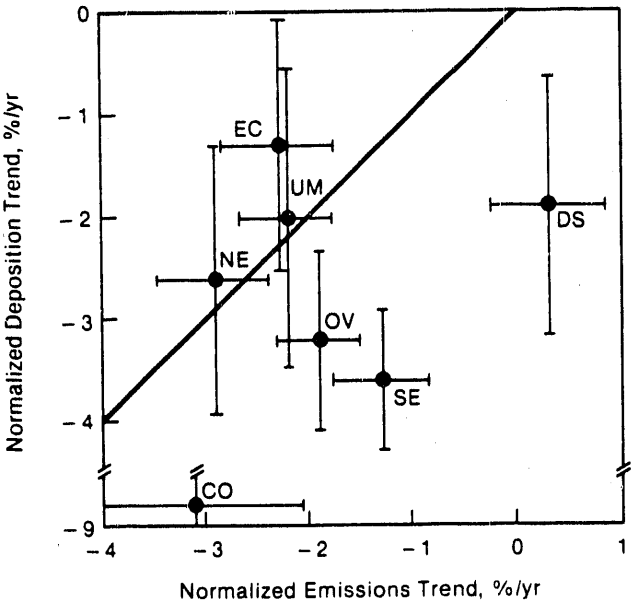

Fig. 6. Regional trends in normalized wet $SO_4^=$ deposition and normalized SO_2 emissions over the 1979 to 1987 period. The regions are identified in Fig. 4.

account for the yearly variability in source-receptor relationships due to year-to-year differences in wind fields and precipitation patterns. Second, source attribution is more complex than the simplified parameterizations in ASTRAP and other long-term regional deposition models. Third, state-level spatial resolution of yearly emissions may be inadequate to capture year-to-year changes in the spatial distribution of sources. Fourth, the response of wet deposition to emission changes may be a nonlinear function of the emissions of not only SO_2 but also of other pollutants that affect the production of oxidants.

The regional trends of wet $SO_4^=$ deposition and SO_2 emissions are consistent with the national pattern of decreasing wet $SO_4^=$ deposition and SO_2 emissions. For most of the northeastern United States, Fig. 6 indicates that, within the standard error of the trend estimate, the trends in wet $SO_4^=$ deposition and SO_2 emissions are similar. However, the magnitude of the standard errors and the scatter of the mean trend values around the line of equal trends in emissions and deposition indicates that extrapolating these empirical relationships to future emissions is unreliable for predicting future regional deposition.

4. SUMMARY AND CONCLUSIONS

A simple analysis of recent SO_2 emissions and wet $SO_4^=$ deposition data indicates that a causal link exists between major regions of SO_2 emissions and areas of high wet $SO_4^=$ deposition, with a source-receptor length scale of 200 to 500 km. The relationship between regional changes in deposition over a 9-year period and changes in emissions contributing to the deposition in those regions over the same period further establishes this link.

These empirical analyses are useful for estimating the predictability of future deposition changes to emissions changes and contribute to our understanding of the processes controlling source-receptor relationships. Nevertheless, these

analyses in and of themselves are inadequate for directly predicting future deposition. Spatial analyses do not identify the attribution of specific sources at a specific receptor, information necessary for evaluating the most effective emissions control options. Trends in factors such as storm paths, precipitation amount, concentrations of other pollutants, and source strengths and locations contribute to trends in regional wet deposition but have not been explicitly accounted for in this empirical trend analysis and, thus, the relationship between emissions and wet deposition trends cannot be confidently used to predict future wet deposition. These shortcomings of empirical analyses of the relationship between wet deposition and emissions support the decision by NAPAP to use numerical models that explicitly account for those factors for assessing future emissions control options.

5. ACKNOWLEDGMENTS

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