ANL/CP--73968

DE91 018545

REGIONAL ANALYSIS OF S EMISSION-DEPOSITION TRENDS IN NORTH
AMERICA FROM 1979 THROUGH 1988

Jack D. Shannon

Environmental Research Division
Argonne National Laboratory
Argonne, IL 60439 USA

INTRODUCTION

The United States recently enacted legislation that mandates reductions in SO_x emissions of about 50% by the end of the century and somewhat smaller reductions in NO_x emissions. Similar broad emission reductions are underway in Canada. Such large reductions are generally expected to lead to significant reductions in acid deposition loading; however, the expense of the required actions suggests that mid-course evaluations of the effectiveness of the emission control programs will be desirable.

Smaller but still significant changes in annual S emissions in the United States and Canada in recent years present an opportunity to test and evaluate methods to identify trends in source-receptor relationships. From 1979 through 1988, aggregated annual emissions of SO₂ decreased about 15% in both the United States (Kohout *et al.*, 1990) and Canada (Vena, 1989). One might anticipate that a similar decrease in sulfate wet deposition would be found, if integrated over a sufficiently large region (National Research Council, 1983). However, data for evaluating the deposition trend over the entire decade are limited. Although monitoring of precipitation chemistry was widespread by the end of the period, in the initial years many networks had much sparser station density and lower data capture. The number of wet deposition monitoring sites with suitable sulfate results for the entire period ranges from 26 to 35, depending upon the criteria for acceptance (Sisterson *et al.*, 1990). Although Sisterson *et al.* found that only a handful of sites exhibited statistically significant trends in deposition (some increasing, some decreasing), both annual wet deposition and precipitation-weighted concentration of sulfate decreased over the period at most sites, with a decrease of about 14% in the median PWC from 1979 through 1987.

The preponderance of decreases in deposition suggests that trends in ensemble statistics might be statistically significant. However, the appropriate approach for testing ensemble trends is not immediately obvious. First, ensembles must be defined. Techniques such as cluster analysis can group monitoring sites with similar deposition characteristics, but the sites do not necessarily have similar patterns of antecedent emissions. Second, data on emission trends are usually aggregated, either nationally, regionally, or across states and provinces, but emission rates of individual sources within a state or province do not change uniformly. Third, any underlying deposition trend is confounded with the effects of meteorological variability, primarily associated with changes in wind patterns and precipitation frequency and intensity. Increased rainfall tends to increase deposition and decrease concentration by dilution, but in neither case in a simple manner. One of the perceived advantages of examining the trends for ensembles of

The submitted manuscript has been authored by a contractor of the U. S. Government under contract No. W-31-109-ENG-38. Accordingly, the U. S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U. S. Government purposes.

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

monitoring sites rather than for individual sites is that the confounding effects of emission and meteorological variations over smaller scales should be reduced.

APPROACH

Defining the source region affecting a deposition region is not a simple matter. Unlike watersheds, airsheds do not have precise, easily determined boundaries. The air quality and deposition patterns produced by a particular emission source have no fixed outer limits but instead gradually decrease with distance. The deposition pattern tends to be elongated in one or more directions because of prevailing meteorological conditions. An elevated plume requires time and associated distance to diffuse to the surface and begin dry depositing or to be taken up in cloud or rain droplets, and oxidants may be limited locally; thus, peak deposition rates for a major source are probably located somewhat downstream. However, such perturbations tend to be on scales smaller than regional. Because deposition patterns are heterogeneous and because deposition is a function of source-receptor separation, each source has a unique potential per unit emission to affect deposition at any particular receptor. The approach followed here to define source and receptor regions is described below.

First, 33 wet deposition monitoring sites with essentially complete records of precipitation chemistry during 1979-1988 are grouped by region (Fig. 1). The site groups, determined subjectively, are termed South (9 sites), Northeast (9 sites), Midwest (9 sites), North Central (4 sites), Colorado (2 sites), and All Sites (33 sites). Single Canadian sites are included in the Northeast and North Central regions, while a site in northern California with otherwise suitable data is omitted from this regional-scale analysis because it cannot be grouped conveniently with other sites. Although one Midwest site is quite close to some of the South sites, they are separated by major Appalachian ridges. The group defined by the Colorado site pair is much smaller than is desirable, but geographical isolation makes grouping with other sites potentially misleading. In addition, the Colorado sites are among those examined in empirical source-receptor studies related to smelter emission trends in the West (e.g., Oppenheimer *et al.*, 1986; Shannon and Lesht, 1988).

Next, a weighted emission region is determined for each deposition region from modeled source-receptor matrices for annual wet S deposition at the sites in the region. Sources are gridded in multiple levels for cells of approximately 120 km x 120 km horizontal dimensions for source-receptor calculations, but they are subsequently aggregated in the matrices to state or provincial totals. The matrices have been produced with the ASTRAP model (Shannon, 1985) for 1980-1988 climatology and 1985 emissions. The climatological data set excludes 1979 because the available wind and precipitation analyses have somewhat different resolution than the analyses for the other years, and that resolution difference can change model output somewhat (Shannon, 1991). The emission field for 1985, widely used in modeling studies for the Final Assessment of the National Acid Precipitation Assessment Program (e.g., Venkatram *et al.*, 1990), was selected because it was known in the greatest spatial and seasonal detail. The deposition contributions from the emissions of each state or province are totaled for all of the sites within a particular region; the total wet deposition is then scaled by the inverse of the state or provincial emission total. To extend the matrices to other years, a (10 x 59) matrix of emissions by state and province for each year of 1979-1988, produced from emission inventory trends from Kohout *et al.* (1990) and Vena (1989), is multiplied by the (59 x 1) normalized source-contribution vector for each region to produce a weighted-emission trend that can be nondimensionalized through scaling by the inverse of its mean value over the period.

The primary purpose of using a model to develop the emission weighting functions is to define emission source regions objectively. If another regional acid deposition model, even a sophisticated Eulerian model such as the Regional Acid Deposition Model (RADM), were used as outlined here, results might differ in detail but should be broadly similar. Unless otherwise noted, the terms "regional emissions" or "weighted emissions" as used below imply all sources weighted by their contribution to a particular deposition region and not just the emission sources within that geographical region.

Dimensionless trends of regional annual sulfur wet deposition (WD) are produced by averaging arithmetically the annual deposition at all sites within a region, then scaling by the inverse of the ten-year regional average. Similar calculations are made for annual PWCs. The focus is on means rather than on medians because some regional groups are small. Geometric averaging of observations was also examined but little difference was noted in most instances. Before averaging, PWC and WD are adjusted by subtracting an assumed SO_4^{2-} contribution from natural sources, 2 mmol/L (4 mmol/L for Colorado) or its deposition equivalent, consistent with the estimates of Galloway *et al.* (1984).

RESULTS

In Fig. 2a-f the trends of WD, PWC, and weighted emissions are compared by deposition region. All emission trends show a general decrease over the period, although the decrease for the emissions affecting the South is quite small. Overall weighted emissions peak in 1979, the first year of the trend, decrease noticeably through 1982, and have a minor secondary peak in 1984 and a minimum in 1987, much like the trend in aggregated U.S. and Canadian emissions. Regional trends are generally similar to the overall trend, but some noticeable differences exist, particularly for the peripheral regions. Weighted emissions peak in 1980 for the South and in 1981 for Colorado, and the secondary maximum in 1984 is absent for those regions. Minimum emissions occur in 1982 in the South.

All regions experienced decreases in WD and PWC as well, although some trends are insignificant. Overall WD peaks in 1979, while overall PWC peaks in 1980. Both show general decreases with time but have secondary maxima in both 1984 and 1986. Weighted emissions explain more than 50% of the variance only for Midwest, Colorado, and All Sites WD and for Northeast, Midwest, North Central, Colorado, and All Sites PWC.

Over a time span sufficiently long for annual variations to be small relative to the long-term trend, WD and PWC should be equally likely to reflect a long-term emission trend. Over a shorter time, however, both WD and PWC are subject to considerable variation. Year-to-year differences in the annual precipitation amount at a site can result from variations of either precipitation frequency or precipitation intensity or from variations in both. If precipitation frequency changes, WD should be more affected; if precipitation intensity changes, PWC should be more affected because of the dilutive effect of heavy precipitation. The annual changes in WD and PWC are of opposite sign about one third of the time.

Both WD and PWC exhibit more annual variability than do weighted emissions. Variability in annual deposition produced by meteorological variability over scales smaller than are resolved in regional deposition model input would not be expected to be explainable by the weighted-emission trend as determined here, nor would the effects on deposition of emission variations over unresolved spatial and temporal scales.

A preliminary version of this approach appeared in Venkatram *et al.* (1990). Data from the same deposition monitoring sites were used in the earlier study, but the analysis extended only through 1987, and the deposition regions other than Colorado and All Sites were defined differently. Another difference was that the modeling procedure to develop weighted emissions used meteorology only from 1983.

Certain aspects of model performance can be examined by comparing the trend statistics for modeled deposition and concentration, both normalized, to the observed regional trends. While inclusion of annual meteorological variability improves the explanation of WD variance relative to climatologically weighted emissions in the South, Northeast, and North Central regions and PWC variance in the South, variance explanation decreases elsewhere, particularly in the Midwest, as shown in Table I. This implies that including annual meteorological variability in the modeling merely increases the noise in the approach, perhaps because (1) the deposition is being totaled January through December while the meteorological input to the model is organized December through November, (2)

some meteorological data are missing in the years for which calculations were made, (3) resolution in the emission trends is limited, or (4) subgrid-scale variations occur in wet deposition. The decrease in variance explanation may also indicate that highly parameterized models such as ASTRAP are most suitable for use with long-term climatology. ASTRAP explains more of the PWC variability than the WD variability because the monitored annual precipitation at the site is used to scale the modeled WD in order to calculate modeled PWC.

TABLE I: Explanation of variance (R-Square) of regional WD and PWC with each approach.

DEPOSITION REGION	CLIMATOLOGICALLY WEIGHTED EMISSIONS		ANNUAL ASTRAP SIMULATIONS		SAME STATE/PROV EMISSIONS	
	WD	PWC	WD	PWC	WD	PWC
SOUTH	0.15	0.32	0.42	0.68	0.00	0.62
NORTHEAST	0.22	0.51	0.27	0.44	0.19	0.61
MIDWEST	0.69	0.66	0.35	0.42	0.57	0.40
NORTH	0.30	0.72	0.38	0.67	0.00	0.17
CENTRAL						
COLORADO	0.83	0.59	0.42	0.60	0.01	0.00
ALL SITES	0.66	0.72	0.56	0.67	0.73	0.73

Another issue that can be investigated here is suitability of the definition of the emission regions. If the emission regions are defined as the states or provinces within which the monitoring sites of a particular region are located, such as Florida, Georgia, North Carolina and Virginia for the South region, then explanation of variance decreases, in some cases to essentially zero, relative to the climatologically weighted emission regions for all cases except PWC in the South and Northeast and both PWC and WD in All Sites. The poor performance in the peripheral regions is because most of the deposition results from emissions from outside the immediate region. The good performance of local emissions in the All Sites region may be a manifestation of "what goes up must come down," because almost one-half of the total U.S. and Canadian S emissions are produced by sources in the 16 states and provinces in which the monitoring sites are located. Because all 16 aggregated sources are used to explain the variance at each of the 33 receptors, the emissions should no longer be thought of as local.

CONCLUSIONS

Variations in SO_x emissions weighted by a regional deposition model are shown to account for most of the variance in regionally averaged annual wet deposition (WD) and precipitation-weighted concentrations (PWCs) of sulfate in the United States and Canada during the period 1979-1988. Total emissions decreased about 15% during that time. For all sites combined, weighted emissions accounted for about two-thirds of the variance, with slightly better performance for PWC than for WD. Restricting the emission regions to the states containing the monitoring sites in each region led to very mixed results, with good explanation of WD and PWC variance in the Midwest and in all sites combined but generally poor explanation elsewhere, particularly for WD. In some regions, local emission densities are quite low, and most deposition would be expected to result from outside sources, so no strong relationship between local emission rates and deposition would be expected. Including annual meteorological variability in the emission weighting improved variance explanation in several of the peripheral regions, but reduced variance explanation elsewhere, particularly in the Midwest, the region of highest emission density.

ACKNOWLEDGMENT: This work was supported by the Office of Health and Environmental Research of the U.S. Department of Energy's Office of Energy Research under contract W-31-109-Eng-38, as a part of the Atmospheric Chemistry Program.

REFERENCES

- Galloway, J. N., Likens, G. E., and Hawley, M. E., 1984, Acid precipitation: natural versus anthropogenic components, *Science* **226**, 829-831.
- Kohout, E. J., Miller, D. J., Nieves, L. A., Rothman, D. S., Saricks, C. L., Stodolsky, F., and Hanson, D. A., 1990, *Current Emission Trends for Nitrogen Oxides, Sulfur Dioxide, and Volatile Organic Compounds by Month and State: Methodology and Results*. Argonne National Laboratory Report ANL/EAIS/TM-25.
- National Research Council, 1983, *Acid Deposition: Atmospheric Processes in Eastern North America*, National Academy Press, Washington, DC.
- Oppenheimer, M., Epstein, C. B., and Yuhnke, R. E., 1986, Acid deposition in the Western United States, *Science* **233**, 12-14.
- Shannon, J. D., 1985, *User's Guide for the Advanced Statistical Trajectory Regional Air Pollution (ASTRAP) Model*. United States Environmental Protection Agency Report EPA/600/8-85/016 (NTIS PB85-236784/XAB).
- Shannon, J. D., 1991, Modeled sulfur deposition trends since 1900 in North America, in *Air Pollution Modeling and Its Application VIII* (H. van Dop and D. G. Steyn, eds.) Plenum Press, New York, 61-68.
- Shannon, J. D. and Lesht, B. M., 1988, Modeling the relationship between sources and receptors of wet deposition of S and NO_x-N in the intermountain west of the United States, in *Air Pollution Modeling and Its Application VI* (H. van Dop, ed.) Plenum Press, New York, 333-343.
- Sisterson, D. L., Bowersox, V. C., Olsen, A. R., Meyers, T. P., Vong, R. J., Simpson, J. C., and Mohnen, V., 1990, *NAPAP SOS/T Report 6, Deposition Monitoring: Methods and Results*, National Acid Precipitation Assessment Program, Washington, DC.
- Vena, F., 1989: personal communication.
- Venkatram, A., McNaughton, D., Karamchandani, P. K., Shannon, J., Sisterson, D. L., and Fernau, M., 1990, *NAPAP SOS/T Report 8, Relationships between Atmospheric Emissions and Deposition/Air Quality*, National Acid Precipitation Assessment Program, Washington, DC.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Figure 1: Regional ensembles of monitoring sites used in trend analyses of emissions and deposition. Regions are designated South (open circles), Northeast (solid circles), Midwest (diamonds), North Central (solid squares), Colorado (open squares), and All Sites.

Figure 2: Trends in annual wet deposition (dashed) and precipitation-weighted concentrations (dotted) of sulfate and weighted emissions of SO_x (solid) for the South, Northeast, Midwest, North Central, Colorado, and All Sites regions (a-f, respectively). Note that the vertical scale is different for Colorado.

Figure 2: Contd.