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Project Summary

Sulfur Deposition Modeling in Support of the U.S./Canadian Memorandum of Intent on Acid Rain

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At the request of the U.S./Canadian Work Group 2 of the Acid Rain Memorandum of Intent, the Eastern North American Model of Air Pollution (ENAMAP-1) was applied to simulate the monthly wet and dry depositions and monthly averaged ambient concentrations of SO_2 and $SO_4^=$ for January and July 1978 across eastern North America. Using these model results, unit emissions (1.0 Tg S yr^{-1}) transfer matrices, which describe source/receptor relationships, were generated and a model performance study was undertaken. In addition, a model sensitivity study was conducted to examine the consequence of model input parameter uncertainties.

This Project Summary was developed by EPA's Environmental Sciences Research Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

In the mid 1970's, SRI International developed a Lagrangian-puff air pollution model, European Regional Model of Air Pollution (EURMAP) for the Federal Environment Office of the Federal Republic of Germany (Johnson, et al., 1978). This regional model was capable of calculating monthly SO_2 and $SO_4^=$ concentrations and dry and wet deposition patterns and international exchanges of sulfur across 13 countries of western and central Europe.

In the late 1970's, SRI International, sponsored by the U.S. Environmental Protection Agency (EPA), adapted and applied EURMAP to eastern North America. The adapted version of this model, Eastern North American Model of Air Pollution (ENAMAP), was capable of calculating monthly SO_2 and $SO_4^=$ concentrations and dry and wet deposition patterns and interregional exchanges of sulfur across a user-defined number of regions (Bhumralkar et al., 1980). Thus, it was possible to assess the impact of sulfur emissions from individual sites and provinces on the sulfur concentrations and depositions across the same regions.

In 1981, the Atmospheric Sciences and Analysis Work Group (Work Group 2) of the U.S./Canadian Memorandum of Intent on Transboundary Air Pollution included the ENAMAP-1 model as one of eight Lagrangian long-range sulfur pollution models to be applied. Work Group 2 requested ESRL to apply the ENAMAP-1 model using January and July 1978 input data to generate transfer matrices, assess model performance, and analyze model sensitivity in input parameters. This report summarizes the ESRL work.

Model Description

Parameterizations

In the design and development of any air quality simulation model, there are usually two conflicting goals: maximum realism and accuracy on one hand, and minimum computational requirements on the other. Greater realism and accuracy

usually require more detailed information and sophisticated formulations of physical processes, which in turn require more computer time and memory capacity. It was clear from the outset that the computer requirements could be severe for two reasons:

1. The model must treat a very large geographical area (2870 km north to south and 3220 km east to west) and yet preserve acceptable spatial resolution (70 by 70 km).
2. The model must compute monthly and annual mean concentration and deposition fields while preserving the original temporal resolution (12 h) of standard meteorological data; thus, the model must make repetitive calculations for long sequences of input data.

Accordingly, as a first step, it was desirable to design a very simple model having minimum computer requirements (i.e., a practical and economical model that would offer acceptable realism in simulating the most important processes involved in the transboundary sulfur pollution problem). More sophisticated parameterizations can replace the simplistic approaches described here as knowledge of the appropriate physical processes accumulates. Later, the simplistic parameterizations of vertical mixing, dry and wet depositions, and transformations of SO_2 will be replaced by more sophisticated parameterizations by the end of 1982. Also by the end of 1982, NO_x chemistry will be added to the model.

Results

Model Applications

To accomplish its goals, Work Group 2 requested that each of the eight long-range transport models generate the following model output:

1. annual 1978 unit transfer matrices of wet sulfur deposition, dry sulfur deposition, ambient SO_2 concentrations, and ambient $SO_4^=$ concentrations normalized by a 1.0 Tg S yr^{-1} emission rate from each of the 40 source/receptor regions and 9 sensitive receptors defined by Work Group 2, and
2. January, July, and annual 1978 wet sulfur depositions and average ambient $SO_4^=$ concentrations at monitoring sites selected by Work Group 2.

The unit transfer matrices, although strongly influenced by the meteorological scenarios used in the simulation period, were to be used by another work group to assess the merits of several emission scenarios. From these transfer matrices, the effects of emissions from individual regions on the sulfur depositions and ambient concentrations across sensitive receptor areas (Boundary Waters, Algoma, Muskoka, Quebec, southern Nova Scotia in Canada and northern New Hampshire, the Adirondacks, central Pennsylvania, and Great Smoky Mountains in the United States) and 40 source/receptor regions of Canada and the United States could be determined.

The January and July 1978 meteorological data used to generate the transfer matrices were analyzed and gridded by preprocessors as described in the previous section. The emissions from each grid cell within a given source/receptor region were multiplied by a constant factor so that the total annual sulfur emissions from that region equaled 1.0 Tg.

Since the meteorological data and analyses for the entire year of 1978 were unobtainable in the rather brief time frame imposed upon the work group, it was agreed that ENAMAP-1 would be applied using only January and July 1978 input data. Annual estimates of the transfer matrices would be based only on matrices for those two months. In addition, since the ENAMAP-1 domain did not include western North America, only 35 instead of 40 source/receptor regions were considered.

The ENAMAP-1 January and July 1978 unit transfer matrices are presented in two appendices in the final report. The transfer matrices indicated that the sources within any 1 of the 35 regions contributed significantly to the sulfur depositions and concentrations within that region. In January, an average of 68% of SO_2 wet deposition in a particular region resulted from SO_2 emissions from that region. Similarly, in July, an average of 64% of the SO_2 wet deposition in a particular region resulted from SO_2 emissions from that region. The ENAMAP-1 results indicated that much of the sulfur wet deposition consisted of SO_2 wet deposition (in some cases, wet deposition of SO_2 was an order of magnitude greater than that of $SO_4^=$) and that "local" sources are significant in sulfur wet deposition.

Model Evaluation

An ideal assessment of the performance of any regional sulfur model would require an extensive data base of dry sulfur depositions, wet sulfur depositions, and ambient SO_2 and $SO_4^=$ concentrations measured during all seasons of the year across all portions of the modeling domain that are removed from the effects of local sources. Unfortunately, such a data base does not exist. However, daily average concentrations of SO_2 and $SO_4^=$ from 1 Canadian and 53 U.S. sites were available from the Electric Power Research Institute's Sulfur Regional Experiment (EPRI-SURE). Monthly sulfur wet deposition data for these periods were available from several Multistate Atmosphere Power Production Pollution Study (MAP3S) sites in the northeastern U.S. and from about a dozen Canadian Network for Sampling Precipitation (CANSAP) sites. Together, these data formed the best available regional sulfur modeling evaluation data base for North America during this period.

Work Group 2 screened the data on ambient $SO_4^=$ and sulfur wet deposition for January and July 1978. For the MAP3S network, precipitation samples with a catch of less than 50% of a nearby rain gauge measurement were ignored. Otherwise, the precipitation sample amount was adjusted to the rain gauge measurement. Valid monthly samples required a minimum 90% capture. For CANSAP, 7 of the 16 operational sites were ignored because of operational or siting problems. Valid monthly samples required a minimum operational time of 20 and a minimum collection efficiency of 25%. Collection efficiency was defined as the ratio (%) of the precipitation recorded by the sampler to that measured by a colocated rain gauge. Since even rain gauges do not collect all of the precipitation (precipitation collection efficiency is a function of gauge type, site exposure, and wind velocity), the rain gauge precipitation data from both networks were adjusted.

The SO_2 emission data used in the evaluation study were obtained from several sources, since a complete 1978 emission inventory was not available. Because much of the SO_2 was emitted from electric power plants, it was imperative to accurately define the 1978 power plant emissions. With this in mind, Environment Canada prepared a 1978 SO_2 emission inventory for large point sources. Emissions from all other sources were assumed to be the same as in 1976. The 1978 SO_2 emissions from the U.S. power plants were estimated from fuel-use records, while the 1978 emissions from all other sources were assumed to

be the same as in 1980, the year for which emissions data were available.

The emission data set was more precise than the precipitation data set. Due to the lack of extensive precipitation measurements in Canada in the winter, the 3-h precipitation amounts used in the ENAMAP-1 applications were extrapolated for much of the Canadian portion of the grid domain. The data from the Canadian sites showed less agreement with the monthly ENAMAP-1 precipitation amounts across the grid cells encompassing each site.

Because the measured data set was inadequate for concrete conclusions, Work Group 2 computed the values of selected statistical parameters to assess the performance of the models for January, July, and annual 1978. Models applied to only the two months were not evaluated for the annual period. This evaluation exercise was expected to identify models that consistently performed unacceptably.

Table 1 presents the values of the statistical parameters calculated for the ENAMAP-1 results. The residuals were determined by subtracting the calculated value from the observed value. The mean residual is a good measure of a model's ability to correctly calculate the higher and lower observed values. In general, a model is said to overpredict the observations when the mean residual is less than zero. For a "good" model, the correlation between the residual and the calculation is low.

Based on the correlation of the residual and calculation, the model performed best for July sulfur wet deposition and January ambient $SO_4^=$ concentrations. Furthermore, the values of the mean residuals were comparable.

Table 1. Values of Statistical Parameters Calculated for the ENAMAP-1 Results

Parameter	Total Sulfur Wet Deposition (kg ha^{-1}) January 1978	July 1978
Sample size	5	11
Mean of observation	0.84	1.20
Mean of calculation	1.00	0.87
Mean residual	-0.16	0.33
Standard deviation of residual	0.31	0.46
Correlation of residual and calculation	-0.62	0.18
	Average Ambient $SO_4^=$ Concentrations ($\mu g\ m^{-3}$) January 1978	July 1978
Sample size	29	47
Mean of observation	6.8	11.6
Mean of calculation	6.3	11.8
Mean residual	0.5	-0.2
Standard deviation of residual	1.8	4.9
Correlation of residual and calculation	0.14	-0.36

Model Sensitivity Study

Every numerical simulation model produces results that do not concur exactly with observations. These discrepancies are due partially to uncertainties in the values of model input parameters (e.g., dry deposition rates, scavenging coefficients, mixing heights, transformation rates, etc.). Atmospheric scientists cannot reach a consensus on a single appropriate value for each of these model input parameters, but a consensus can be reached on a general range of "acceptable" values.

Since there are many "acceptable" values, the modeler is faced with the problem of selecting one value to use in model applications. The magnitude of this problem is proportional to the model sensitivity, or the degree of observed change in the model calculation from a unit change in the model input value. If the model proves sensitive to a certain model input parameter, the modeler must carefully select the value of that parameter. A model sensitivity study identifies those model input parameters whose values must be chosen carefully.

In a sensitivity study, the model is applied many times; the value of one of the model input parameters is changed each time, and the resultant changes in model output are examined. Typically, this involves the consumption of considerable computer time. To minimize this expense, this sensitivity study used an abbreviated version of ENAMAP-1 and only 3 values of each of the 15 model input parameters (Table 2). Two of the three values represented the low and high values of the "acceptable" range of each parameter, while the third value (base case) was the value used in past ENAMAP-1 applications.

The abbreviated version of ENAMAP-1 considered (1) only one emission source, which emitted puffs of SO_2 and $SO_4^=$ at 12-h increments, (2) a continuous 1.0 mm h^{-1} precipitation rate, and (3) a uniform transport wind from the southwest. All other parameterizations were preserved, with the exception of the SO_2 and $SO_4^=$ dry depositions, which reflected the parameterization used in a version of ENAMAP-1 currently under development. This parameterization is based on atmospheric stability and land-use characteristics. The region between southern Ohio and eastern Quebec was selected. The Adirondacks sensitive area is located in the middle of this region, 700 km from the source.

Conclusions

Any conclusions drawn from only two 1-month periods would not likely apply to much longer periods. However, for this evaluation data set, the model performed rather well. The ENAMAP-1 January mean sulfur wet deposition was slightly greater than the monthly mean of the measurements at the five sites. The ENAMAP-1 July mean was slightly less than the mean measurement. The mean ambient $SO_4^=$ concentrations calculated by the model (6.3 and 11.8 $\mu g\ m^{-3}$ for January and July, respectively) compared very favorably with the mean measurements (6.8 and 11.6 $\mu g\ m^{-1}$ for January and July, respectively). The mean residuals for both January and July were less than 1.0 $\mu g\ m^{-3}$. Except for the case of sulfur wet deposition in January, the absolute values of the correlation between the residuals and the model calculations were less than 0.40, which indicated that the model performed well for the two 1-month periods.

The model sensitivity study assessed the changes in the model output at 100-km increments downwind of a single source due to changes in the values of one of the model input parameters. Some significant conclusions of this study were:

1. The 3-h time step used in previous ENAMAP-1 applications led to a saw-toothed distribution in the model output for moderate and high wind speeds (> 20 km h^{-1}).
2. A 2-h time step led to a saw-toothed distribution in the model output for

Table 2. *Model Input Parameter Values Considered in the Sensitivity Study*

Parameter	Case 1	Base Case	Case 2
Computational time step (h)	1.0	3.0	2.0
Wind speed (km h^{-1})	8.1	24.2	40.3
Precipitation rate (mm h^{-1})	0.05	0.10	0.15
Puff expansion rate (km^2 h^{-1})	29.0	36.0	42.0
Transformation rate (% h^{-1})	0.50	1.00	1.50
Mixing height (km)	1.00	1.45	1.90
Initial puff size (km)*	60.0	70.0	80.0
SO_2 wet deposition rate (% mm^{-1})	10.0	28.0	46.0
$SO_4^=$ wet deposition rate (% mm^{-1})	1.0	7.0	13.0
SO_2 dry deposition factor #	0.67	1.00	2.22
$SO_4^=$ deposition factor #	0.167	1.000	1.830
SO_2 emission rate (kton h^{-1})	0.29	0.36	0.43
$SO_4^=$ emission rate (kton h^{-1})	0.018	0.000	0.036
SO_2 loss rate from mixed layer top (kton h^{-1})	0.005	0.000	0.010
$SO_4^=$ loss rate from mixed layer top (kton h^{-1})	0.005	0.000	0.010

**The initial area of each puff is defined as the area of a square of sides EMISCELL.*
#The land-use, stability-dependent dry deposition rates are multiplied by these factors.

only the high wind speed (> 35 km h^{-1}).

3. A 1-h time step did not remove the saw-toothed distribution in the model output for the high wind speed, but it did reduce the amplitude of the fluctuations.
4. Within 700 km of the source, SO_2 wet deposition was sensitive to the SO_2 wet deposition rate; at a distance of 100 km from the source, SO_2 wet deposition increased about 200 to about 800 mg m^{-2} resulting from an increase in the SO_2 wet deposition rate of 0.10 to 0.46% mm^{-1}
5. Beyond 200 km from the source, $SO_4^=$ wet deposition was sensitive to the SO_2 wet deposition rate; at a distance of 1500 km from the source, $SO_4^=$ wet deposition decreased about 25 to about 10 mg m^{-2} for the same increase in the SO_2 wet deposition rate.
6. Beyond 100 and 300 km, the ambient concentrations of SO_2 and $SO_4^=$, respectively, were sensitive to the SO_2 deposition rate.
7. The SO_2 and $SO_4^=$ wet deposition rates affected wet deposition and concentrations more than the changes in the SO_2 dry deposition rate (from 67 to 220% of the base case value).
8. Changing the $SO_4^=$ dry deposition rate (from 16.7 to 183.0% of the base case value) affected the $SO_4^=$ concentrations and wet depositions in the same way as changing the SO_2 wet deposition rate.
9. The consideration of a small $SO_4^=$ emission rate, 18 t h^{-1} or 5% of the base case SO_2 emission rate, resulted in significant increases in $SO_4^=$ concentrations and wet deposition; at 200 km from the source, deposition and concentration increased 35 and 48%, respectively.

The model sensitivity study also assessed changes in model output at 100-km increments downwind of a single source due to changes in all the model input parameters except those relating to meteorological and emission scenarios.

This assessment showed that the $SO_4^=$ wet depositions and SO_2 concentrations calculated using the base case values were very similar to those calculated using the high case values. Furthermore, the $SO_4^=$ concentrations beyond 500 km of the source calculated for the base case were greater than those calculated for the other two cases.

References

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The complete report, entitled "Sulfur Deposition Modeling in Support of the U.S./Canadian Memorandum of Intent on Acid Rain," (Order No. PB 84-122 837; Cost: $14.50, subject to change) will be available only from:
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