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# Project Summary

# ENAMAP-1A Long-Term $SO_2$ and Sulfate Air Pollution Model: Refinements of Transformation and Deposition Mechanisms

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The ENAMAP-1 model for long-range air pollution transport has been modified in several ways to produce the newer version, ENAMAP-1A. The modeled domain has been increased to include more of Southeastern Canada; the meteorological and emissions data for this area have been added to the United States data base; the transformation rate for $SO_2$ to $SO_4^=$ and the deposition rates for $SO_2$ and $SO_4^=$ have been changed to reflect variations in space and time; and the transformation rate has been parameterized to be a function of latitude and season. The new transformation rate is, on the average, several times larger than the former 1 percent per hr rate. In ENAMAP-1A, the dry deposition rate has been parameterized to be a function of the underlying terrain and vegetation, the thermal stability in the boundary layer, and the time of day. The wet deposition rate has been changed to be a function of rainfall rate and cloud process type (convective, warm process, or Bergeron process).

For this project, the ENAMAP-1A model was run for each day of January and August 1977 to produce monthly averaged values of airborne concentrations, dry deposition, and wet deposition of $SO_2$ and $SO_4^=$. These values have been compared to values generated by the previous version of the model. The boundary exchanges of $SO_2$ and $SO_4^=$ have been computed for each of 41 states or provinces and also for 12 smaller areas of special interest. The course of pollution from emission to deposition is documented in the form of maps and tables. For brevity, only the comparisons are presented and discussed in this summary. The remaining results are discussed in the final report, EPA-600/3-82-063. In contrast to the previous computations using ENAMAP-1, the new computations showed much larger concentration and deposition amounts of airborne $SO_4^=$, while the amount of $SO_2$ deposition was decreased. The total sulfur deposition ($SO_2$ and $SO_4^=$ combined) was approximately 40 percent of the previously computed value in winter and 70 percent of the previous value in summer. Scatter diagrams of calculated and observed concentrations showed reasonably good agreement for $SO_2$; however, computed $SO_4^=$ concentrations were significantly greater. This may be interpreted as evidence that the new model transformation rate was on the average too large and/or that $SO_4^=$ deposition was too low.

***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

Under contract to the U.S. Environmental Protection Agency (EPA), SRI International developed and evaluated an Eastern North American Model of Air Pollution (ENAMAP-1). The ENAMAP-1 model was adapted from the SRI-developed European Model of Air Pollution (EURMAP). The ENAMAP-1 model was designed to study the long-term transport and deposition of airborne sulfur pollutants and to calculate ambient sulfur concentrations for monthly, seasonal, and annual periods over the eastern United States and Canada. The model has been used to calculate exchanges of airborne sulfur among various United States and Canadian regions. ENAMAP-1 was further tested to study the variability of the model's calculations of seasonal sulfur concentrations and depositions due to year-to-year changes in the wind and precipitation patterns.

Under another contract with EPA, work was continued to develop ENAMAP-1 further. A new version of the model, ENAMAP-1A, has been developed by expanding the domain of ENAMAP-1 to include the area bounded by 29°N and 55°N latitude and 60°W and 104°W longitude. Other modifications include

1) treating most states and Canadian provinces as separate receptor and emitter areas and adding 12 smaller receptor areas; and
2) incorporating deposition and transformation parameterizations expressed as functions of variables theorized to be factors governing the physical processes.

ENAMAP-1A has been applied to emissions and meterological data for January and August, 1977 and the results have been compared with the measured concentrations as well as with the results of ENAMAP-1. Comparisons of ENAMAP-1 and ENAMAP-1A are discussed in this summary. These comparisons show that the new transformation rate is too high and/or the $SO_4^=$ deposition rate is too low.

As the research effort continues, the model input values will be reassessed. In addition, the boundary layer is to be divided into the distinct sublayers with vertical mixing parameterized as a function of stability (the previous model version assumed instantaneous, complete vertical mixing at the source). Also, the transport wind speed near mountainous regions will be adjusted to account for terrain effects and concentrations and depositions of nitrogen compounds will be calculated by the model.

## Description of the ENAMAP-1A Model

ENAMAP-1 has been updated in various ways in the course of developing ENAMAP-1A. Algorithms for wet and dry deposition rates and transformation rates have been developed to account for temporal and spatial variability in the parameters and have been incorporated in the latest version of the model. These algorithms have been formulated on the basis of an extensive literature search; they represent the state-of-the-art of the treatment of dry and wet deposition in long-range transport models. A description of the model's basic structure, including grid cell sizes and the puff advection and diffusion scheme, can be found in the final report, EPA-600/4-80-039.

Figure 1 shows the boundaries of ENAMAP-1A model domain. This region covers the area between 29°N and 55°N latitude and 60°W and 104°W longitude. It represents the addition of 560 km (eight 70-by 70-km grid cells) to the northern side and 210 km (three 70-by-70-km grid cells) to the eastern side of the previous grid. The model has been modified to calculate interregional exchanges of sulfur pollution between 41 states and provinces (as opposed to 13 EPA and Canadian regions in ENAMAP-1).

The transformation rate for $SO_2$ to $SO_4^=$ is expressed as the sum of two components: a homogeneous transformation rate and a heterogeneous transformation rate. For the homogeneous rate, the rate constant is calculated theoretically as a function of solar insolation (i.e., latitude and season). The theoretical rates were based on tests made in a relatively clean atmosphere; therefore, the rates were doubled in ENAMAP-1A because of the greater number of pollutants and reactions occurring in the actual atmosphere. The transformation coefficients used in the homogeneous conversion are presented in Table 1. An additional term for the heterogeneous conversion of $SO_2$ to

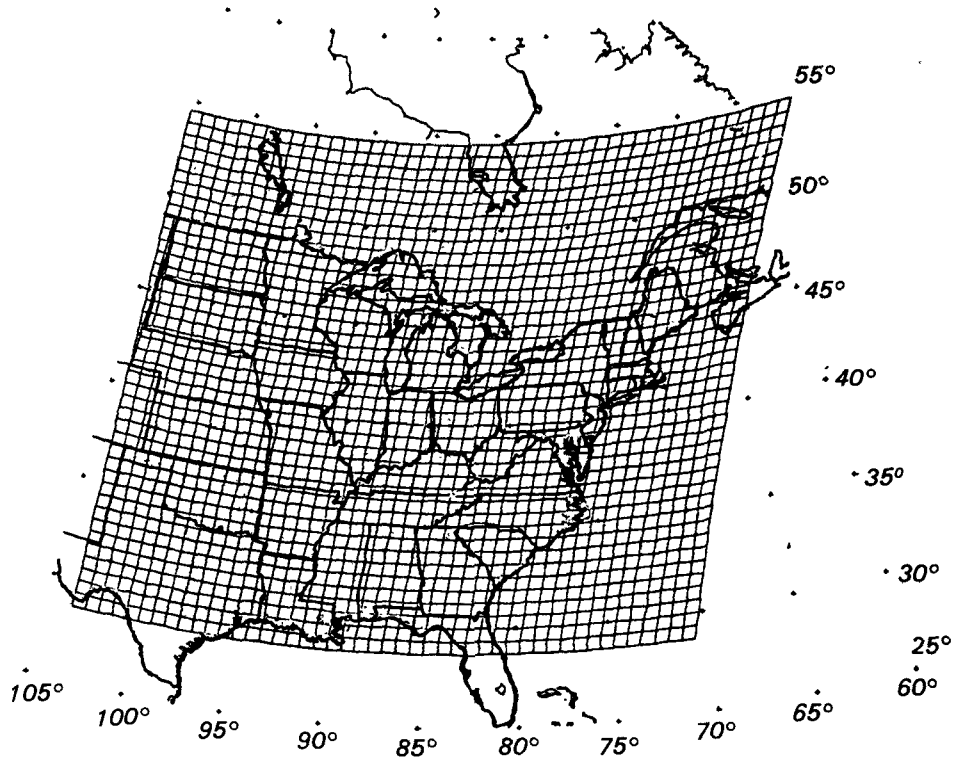


*Figure 1. The 70 km grid used in the ENAMAP-1A model.*

$SO_4^=$ is included. There seems to be some question in the literature as to the relative importance of the various heterogeneous conversion mechanisms, particularly the differentiation between the strong oxidizing agents (such as $H_2O_2$ and $O_3$) and the effect of metallic catalysts on conversion. Because of the difficulty in determining the relative importance of the various heterogeneous conversion mechanisms, a constant conversion rate of 0.005 (0.5 percent $h^{-1}$) for the heterogeneous conversion is used in the model. The total transformation rate varies from approximately 0.01 in winter to 0.04 in summer.

Because of the natural variability of dry deposition, ENAMAP-1A treats it as a function of land-use type, stability, and time of day. Land-use type is defined by the surface characteristics (land type or water) and the type of vegetation. Land-use type was gridded to each 70-by-70-km receptor cell to incorporate dry deposition variability at this resolution. Deposition velocities for each land-use type for $SO_2$ and $SO_4^=$ for January (winter) and August (summer) for stability classes 1 through 6 (very unstable through very stable) are applied in ENAMAP-1A. The values vary from 0.05 cm $s^{-1}$ for cities to approximately 1.0 cm $s^{-1}$ for swamps. To account for the low absorption by plant surfaces at night, $SO_2$ and $SO_4^=$ deposition velocities have been reduced to 0.07 cm $s^{-1}$ during nighttime hours. The length of night is adjusted for each season.

Wet deposition is treated as a function of season and rainfall rate (mm $h^{-1}$). The removal rates are based primarily on considering the washout ratio as a function of precipitation rate and three cloud types: cold clouds in which nucleation of rain is essentially caused by the Bergeron or ice growth process; warm or maritime clouds; and convective clouds. For the model, the following was assumed: winter precipitation follows the Bergeron process, fall and spring precipitation are warm cloud phenomena, and summer precipitation is confined exclusively to the convective type of precipitation. The semi-empirical representation of these removal rates for use in the model is presented in Table 1 for both $SO_2$ and $SO_4^=$. The seasonal variation in the parameters a and b for $SO_2$ were adopted to reflect the variation obtained by Scott for sulfate. They may be revised at a later date when data can be obtained to make a more appropriate distinction between summer and winter gaseous $SO_2$.

The choice of these parameters has resulted in a significant difference in the treatment of wet deposition between ENAMAP-1 and ENAMAP-1A. In ENAMAP-1, the washout ratio is four times greater for $SO_2$ than for $SO_4^=$ for both January and August. The washout ratio for $SO_2$ in ENAMAP-1 is approximately 40 times lower for January and 5 times lower for August than in ENAMAP-1. The washout ratio for $SO_4^=$ in ENAMAP-1A is approximately five times lower for January and two times higher for August than in ENAMAP-1. Naturally, these changed rates produce large differences in the wet deposition patterns and statistics, as will be shown below.

## Results of Model Application for January and August 1977

To determine the effects of using the new sulfur deposition and transformation algorithms, the more complete Canadian emissions data, and the increased model domain, the model was run for January and August 1977. Separate runs were made for the states and provinces for which emissions data were available. For each month, fields of $SO_2$ and $SO_4^=$ concentrations, dry deposition, and wet deposition resulting from sulfur emissions in each of the individual areas were then combined into maps showing the monthly area totals. Interregional exchange tables were also generated, but are not presented in this summary. For comparison purposes, plots of $SO_2$ and $SO_4^=$ deposition and concentration patterns generated from applications of the previous version of the model (ENAMAP-1) are included in this section.

### *$SO_2$ and $SO_4^=$ Concentrations*

Calculated and observed $SO_2$ January 1977 concentrations were in good agreement, except that calculated concentrations were low (<10 $\mu g/m^3$ versus >32 $\mu g/m^3$) in Minnesota and

***Table 1.*** *Comparison of ENAMAP-1 and ENAMAP-1A Transformation and Deposition Parameters*

| | *ENAMAP-1* | *ENAMAP-1A* |
|---|---|---|
| *Transformation* | *1%/h* | *(2[a+b ln (latitude)]+0.5)%/h, where in winter, a=2.5 and b=-0.61; in spring/fall, a=4.4 and b=-1.0; and in summer, a=6.3 and b=-1.4.* |
| *Dry Deposition* | | |
| *$SO_2$* | *1.18 cm/s (winter)<br>1.34 cm/s (spring/fall)<br>1.49 cm/s (summer)* | *0.05 - 0.95 cm/s, depending on stability and land use.* |
| *$SO_4^=$* | *0.22 cm/s (winter)<br>0.25 cm/s (spring/fall)<br>0.28 cm/s (summer)* | *0.15 - 0.95 cm/s, depending on stability and land use.* |
| *Wet Deposition* | | |
| *$SO_2$* | *28.0 R %/h, where R is the precipitation rate (mm/h).* | *100 ($aR^b$) %/h, where in winter, a=-0.009 and b=0.70; in spring/fall, a=-0.036 and b=0.53; and in summer, a=-0.140 and b=0.12.* |
| *$SO_4^=$* | *7.0 R %/h where R is the precipitation rate (mm/h)* | *100 ($aR^b$) %/h, where in winter, a=-0.021 and b=0.70; in spring/fall, a=-0.091 and b=0.27; and in summer, a=-0.390 and b=0.06.* |

Wisconsin. Concentrations calculated by ENAMAP-1A were higher at the northern and southern boundaries of the model domain than those calculated by ENAMAP-1. The concentrations from the new model run were closer to the measured values in this respect; previously the calculated concentrations were too low in Alabama and Georgia. The slightly higher $SO_2$ concentrations calculated by ENAMAP-1A were due to its much lower wet and dry removal rates, which, to some degree, counteract the higher transformation rate.

The calculated $SO_2$ concentrations for August 1977 were similar to the measured concentrations in pattern and in magnitude. The new model results from the northern and southern states were higher than the previous model results, which made them closer to the measured values in these areas. The higher $SO_2$ concentrations calculated by ENAMAP-1A were due to the lower wet and dry removal rates, even though the transformation rate was higher.

The ENAMAP-1A January 1977 $SO_4^=$ concentrations were approximately twice as large as both the previously calculated values and the measured values, but the pattern of the isopleths was very similar to the earlier run. The reason for the higher $SO_4^=$ concentrations calculated by ENAMAP-1A was the higher transformation rate and lower wet removal rate, which overshadow the higher dry removal rate.

As in the January 1977 $SO_4^=$ model results, the August $SO_4^=$ concentrations from ENAMAP-1A increased by a factor of two over the previously calculated concentrations. This made them much higher than the measured values. The new calculated concentrations were higher than the previous values because of the higher transformation rate, although the wet and dry removal rates were also several times higher than before.

Table 2 compares the modeling results using the former and revised wet and dry removal and transformation rates and the $SO_2$ and $SO_4^=$ emissions from Illinois, Indiana, and Ohio. For January 1977, the revision of wet removal rates led to a significant reduction, nearly 20-fold for $SO_2$ and 4-fold for $SO_4^=$. Dry deposition of $SO_2$ decreased by a factor of nearly three, while dry deposition of $SO_4^=$ increased by a factor of three. Transformation of $SO_2$ to $SO_4^=$ increased by a factor of 2.6.

As was the case for January 1977, the wet $SO_2$ deposition calculated by ENAMAP-1A for August 1977 decreased, but not nearly as much. However, unlike the January results, the wet $SO_4^=$ deposition for August increased (by a factor of nearly five). Dry $SO_2$ deposition decreased as they did in the January case, but this time by a factor of nearly four. Dry $SO_4^=$ depositions increased by a factor of nearly five. Transformation increased by a factor of nearly 4.5.

***Table 2.*** *Comparison of ENAMAP-1A and ENAMAP-1 Results (KTON) for Illinois, Indiana, and Ohio Emissions for January and August 1977.*

| | *January 1977* | | *August 1977* | |
|---|---|---|---|---|
| *Emission* | *ENAMAP-1* | *ENAMAP-1A* | *ENAMAP-1* | *ENAMAP-1A* |
| $SO_2$ | | | | |
| *Total $SO_2$ emitted* | *638.7* | *645.1* | *576.8* | *882.9* |
| *Wet deposition* | *145.6* | *72* | *210.6* | *164.7* |
| *Dry deposition* | *372.7* | *213.5* | *286.4* | *68.1* |
| *Flux** | *19.7* | *165.4* | *2.4* | *7.5* |
| *Transformation ($SO_2$ -> $SO_4^=$)* | 100.7 | 259.0 | 77.4 | 342.3 |
| $SO_4^=$ | | | | |
| *Total $SO_4^=$ emitted and transformed* | *165.9* | *403.8* | *127.6* | *525.9* |
| *Wet deposition* | *44.6* | *117* | *65.5* | *305.7* |
| *Dry deposition* | *44.3* | *131.7* | *36.6* | *170.7* |
| *Flux** | *77.0* | *260.4* | *25.5* | *49.5* |

**Flux is the amount of $SO_2$ or $SO_4^=$ that was transported out of the model domain by the wind.*

## *Dry and Wet Depositions*

Dry depositions calculated by ENAMAP-1A were much lower than the ENAMAP-1 results, which displayed a closed isopleth of 1024 mg/m$^2$ of $SO_2$ deposition over eastern Pennsylvania (absent in ENAMAP-1A results). The January $SO_2$ wet deposition results indicate that the wet deposition was drastically reduced by using the new coefficients in ENAMAP-1A. This was the largest change of any of the $SO_2$ or $SO_4^=$ wet or dry deposition results.

The new dry deposition of $SO_4^=$ increased by a factor of approximately two, similar to the change in $SO_4^=$ concentration. The wet $SO_4^=$ deposition results were reduced in ENAMAP-1A because of the lower wet removal rate. The increases or decreases in deposition from ENAMAP-1A compared to ENAMAP-1 are shown in Table 2.

## Summary and Concluding Remarks

This project summary describes a new version of the ENAMAP-1 model, ENAMAP-1A, of long-range airborne pollution transport and removal. The new version covers a larger geographical area and includes emission data and weather observations from southeastern Canada as well as from the eastern United States.

The transformation and deposition parameterizations in ENAMAP-1A have been modified to be functions of those variables theorized to be factors in governing the relevant physical processes. The new transformation rate for $SO_2$ to $SO_4^=$ in ENAMAP-1A varies with solar insolation (i.e., it is dependent on latitude and season). It is several times larger than the rate used previously. This factor, combined with greater $SO_4^=$ deposition rates, significantly increased the $SO_4^=$ deposition amounts, while the deposition amounts for $SO_2$ correspondingly decreased. In ENAMAP-1A the total monthly sulfur depositions for August were much less than those for ENAMAP-1. Unfortunately, measured deposition data to compare with the simulations were not available. However, the computed $SO_2$ and $SO_4^=$ concentrations can be compared to air quality data. The $SO_2$ concentrations yielded by ENAMAP-1A for January were closer to measured values than previous computations, particularly in the northern and southern parts of the domain. The pattern of August $SO_2$ concentrations were very similar to previous computations. The ENAMAP-1A $SO_4^=$ concentrations were too large for both January and August, however. Because of the lack of $SO_2$ and $SO_4^=$ deposition measurements, it was not possible to assess the overall accuracy of the ENAMAP-1A results. As such data become available, total model evaluation will become feasible. The present overestimation of $SO_4^=$ concentrations can be taken as evidence suggesting that the new transformation rate was too high and/or the new $SO_4^=$ deposition rates were too low.

Research under the existing contract is continuing. ENAMAP-1A is undergo-

ing further refinement to include the effects of terrain on the wind flow and to divide the boundary layer into three sublayers with vertical mixing among them. This modification will enable emissions from near-ground sources to be injected into sublayer one, the lowest sublayer, and tall stack emissions to be injected into sublayer two. The effects of these changes on the computations can then be determined.

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*The complete report, entitled "ENAMAP-1A Long-Term $SO_2$ and Sulfate Air Pollution Model: Refinements of Transformation and Deposition Mechanisms," (Order No. PB 82-237 017; Cost: $10.50, subject to change) will be available only from:*
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