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Project Summary

Re-Examination of Interim Estimates of Annual Sulfur Dry Deposition Across the Eastern United States

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During the summer of 1987 annual amounts of sulfur dry deposition were first estimated for more than 7,000 lakes in the eastern United States. These estimates, heretofore termed *interim estimates* since they were expected to be superceded in the near future, were derived from predictions of the Regional Acid Deposition Model (RADM) adjusted using the empirical data from two monitoring networks. Since that time, additional years of empirical data have become available and a portion of the previously available empirical data has been superseded. Consequently, the process of estimating annual amounts of sulfur dry deposition was repeated to determine whether these interim estimates should be revised, and if so, by how much. This study concludes that the interim estimates appeared to be too low by 13% and recommends that the interim estimates be systematically increased by the same amount.

A comparison of the revised estimates to empirically-derived sulfur dry deposition amounts suggests that there is some systematic error in the revised estimates. Adjusted RADM predictions of dry deposition tend to be biased low in the most significant source regions (where at least 200 ktonnes SO_2/yr are emitted within 80 km of the site). Conversely, in locations farther removed from significant sources (81-160 km) there is evidence that the estimates are biased high. However, in general, sulfur dry deposition estimates from adjusted model predictions are within ±60% of the empirical data.

This Project Summary was developed by EPA's Atmospheric Research and Exposure Assessment 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

Dry deposition rates are a function of the air concentration near the surface and the dry deposition velocity. Since dry deposition velocities cannot be measured directly, they are inferred from vertical mass flux measurements. In addition, since they are a function of atmospheric stability and surface attributes (e.g., vegetative type, roughness length, physical conditions, spatial fluctuations of terrain and surface roughness), dry deposition velocities can vary significantly across small areas (e.g., less than 1 km^2). Because of the potential for small spatial-scale variations in dry deposition velocity, there is considerable uncertainty in using a dry deposition estimate from an individual site to represent an average regional value.

In response to the needs of the Aquatic Effects Program of the National Acid Precipitation Assessment Program, *interim estimates* of annual sulfur dry deposition were derived in August 1987 for 7,000 lakes in the eastern United

States. Since that time, several developments have occurred that necessitated a re-examination of these interim estimates of sulfur dry deposition. First, the empirical data at the U.S. sites have been revised as a consequence of improvements to the dry deposition algorithm of the inferential model used to derive the empirical estimates. Secondly, in both the United States and Ontario, data are now available for additional years. The expanded data base not only has provided us with a more statistically representative sample, but also has enabled network staff to identify outliers and discard or correct erroneous values. Thirdly, improvements to the dry deposition module of RELMAP have greatly reduced its oversmoothing problem, thereby increasing this model's potential as an appropriate estimator of spatial patterns. Finally, the calculations from a third model, the Advanced Statistical Trajectory Regional Air Pollution (ASTRAP) model, became available.

Procedure

Estimation of a spatial pattern of sulfur dry deposition across the eastern United States is strongly hampered by the fact that there are only four U.S. sites at which empirical estimates are available. To circumvent this paucity of data, the predictions of regional-scale deposition models are used in conjunction with the available empirical data. The following procedure was developed for estimating annual amounts of sulfur dry deposition across the eastern United States:

1. Construct spatial patterns of annual amounts of SO_2 and sulfate dry deposition from available regional deposition models that relate emissions, transport, dispersion and transformation to dry deposition using dry deposition velocities assumed to represent the area of each model grid cell,
2. Adjust model predictions by a constant factor (based on the comparison of model predictions to site-specific empirical estimates) to correct for model bias,
3. Select the spatial pattern produced by the regional models with the smallest mean-square error,
4. Estimate dry deposition amounts at specific locations of interest by interpolating adjusted model predictions, and
5. Assess the uncertainty of these estimates by examining the correspondence between the model predictions and empirical estimates and characterizing the spatial and interannual variability of the empirical estimates.

The first step of this approach was executed by constructing grids of annual sulfur dry deposition from each of three operational deposition models used by EPA — RADM, RELMAP and ASTRAP. As was the case described in the 1987 report, six three-day episodes of RADM output were averaged and normalized to construct one annual grid (Cases I, II and IV of the April 1981 Oxidizing and Scavenging Characteristics of April Rains (OSCAR) Experiment, the four-dimensional data assimilation run of OSCAR IV, the August 1979 Northeast Regional Oxidant Study (NEROS) case, and an October 1984 case). Unlike the RADM grid, those of RELMAP and ASTRAP were constructed from simulations of the entire year of 1980. The RELMAP results presented here were derived from the improved model version and differed from those presented in the 1987 report.

The second step was accomplished by first comparing interpolated model predictions with the annual means of the empirical estimates at each of four U.S. sites of the COre Research Establishment (CORE) Network and 18 sites of the Acidic Precipitation in Ontario Study (APIOS) Network. Empirical estimates at the CORE sites are expressed as the product of the weekly-mean, inferred dry deposition velocities and measured air concentrations of SO_2 and sulfate for the years 1985 to 1987, inclusive. In contrast, Ontario empirical data are based on a cruder method — the product of estimated annual mean dry deposition velocities and annual means of measured air concentrations of SO_2 and sulfate for the years 1982 to 1986, inclusive.

From the comparisons, mean-square errors were calculated and used as a measure of concurrence of interpolated predictions and empirical estimates. To minimize the bias of the annually-normalized RADM and the ASTRAP predictions, the ensembles of predictions were adjusted systematically by factors of 0.43 and 0.57, respectively. These factors are the regression coefficients of a linear regression of empirical estimates on model predictions forced through the origin. The RELMAP mean-square error was already small and could not be substantially reduced by systematic adjustments to the model predictions.

Based on the comparisons of adjusted/unadjusted predictions with the empirical estimates, the adjusted RAI predictions appeared to replicate best t characteristics exhibited by the empiri data. That is, the bias, root-mean-squ error, and average error for adjust RADM predictions are lower than those the other two models and the ov smoothing of the gradient acro southern Ontario is less severe than t other two models. Each model tends overpredict the sulfur dry deposition the empirical data range of 3 to 6 S/ha/yr. This is indicative of the mode slower rate of decreasing dry depositi away from the high emissions regions, in other terms, their smoothing tenden near steep gradients.

Uncertainty Assessment

The uncertainty in sulfur dry depositi amounts obtained from the estimati procedure described above is primal related to three main factors: (1) t accuracy with which the RADM captu the underlying spatial pattern of c deposition, (2) the accuracy of t empirical dry deposition estimates tl are used to adjust the predictions, and the potential systematic differences t tween the empirical dry deposition e: mates from recent years and the peri of interest.

Although RADM appears to repres an improvement over Lagrangian mod relative to oversmoothing in areas imn diately downwind of emission sour regions, there is still some estimati bias in these areas. It appears that t pattern of over/underestimation is link to the proximity of major source region:

The model predictions are greater th the empirical estimates at sites locat within 81 to 160 km of major sour regions. This suggests that the mo tends to oversmooth dry deposition g dients near source regions. The patt of overprediction does not, howev emerge at sites within 81 to 160 km the major sources in Sudbury, Ontario. fact, in this region the model performan appears to parallel that in regions witl 80 km of major source regions, w underpredictions and a single mod overprediction of 6%. One cause of tl difference in model behavior might related to the Sudbury stacks, which much taller than those elsewhere in No America. These tall stacks might depc sulfur compounds farther downwind th typical sources, with much of the depc tion occurring in the 81-160-km ran rather than within 80 km. Consequently would not be unreasonable to expect tl the model's behavior within 81-160 km the Sudbury source region might be similar to that observed in regions within 80 km of typical large source regions.

Oversmoothing of the spatial gradients is also evident in the groups of sites near less significant source regions. Within 160 km of source regions emitting between 20 and 125 ktonnes of SO_2, RADM estimates are considerably greater than the empirical estimates. With one exception, the relative error at these sites is of the same order as that at sites within 81 to 160 km of much larger sources. In contrast, the model predictions at the two sites removed from major source regions (i.e., <3 ktonnes/ yr within 160 km of the site) are within 10% of the empirical estimates.

In summary, a comparison of the revised estimates to empirically-derived amounts of sulfur dry deposition suggests that there is a systematic error in the revised estimates. Adjusted RADM predictions of dry deposition tend to be biased low in the most significant source regions (where at least 200 ktonnes SO_2/yr are emitted within 80 km of the site). Conversely, in locations farther removed from significant sources (81-160 km) there is evidence that the estimates are biased high.

It is noteworthy that the adjusted model predictions are generally within ±60% of the empirical estimates. Relative errors are generally less than 40% (in absolute value) at high deposition sites (where empirical estimates exceed 9 kg S/ha/yr). At sites with moderate deposition (where empirical estimates are between 2 and 6 kg S/ha/yr), which are fairly close to significant source regions (e.g., Penn State, south-central Ontario, and northeast of Toronto), estimation errors are generally between 45% and 60%, with one error approaching 100%. For other sites with moderate deposition, errors range ±40%. However, some of the greater deviations (in absolute value) in this group of sites might also be explained by the relationship of the sites and their distance from the sources.

With the very limited available data, it is impossible to separate and quantify the three sources of error of the empirical estimates (i.e., model bias, subgrid-scale variability, and empirical estimation errors). Therefore, the best that we can do in characterizing the uncertainty in our dry deposition estimates is to consider the aggregate of all these errors, as reflected in the distribution of RADM deviations from empirical estimates. These deviations suggest that the adjusted RADM predictions of dry deposition generally are expected to lie within ±60% of the actual values. Although there is some evidence that the magnitude and direction of the errors in model-predicted dry deposition might be related to distance from significant source regions, we do not feel that the available data allow us to further refine the ±60% estimate of uncertainty.

Conclusions and Recommendations

Three regional models, RADM, RELMAP and ASTRAP, were applied to construct grids of annual sulfur dry deposition. Comparison of RELMAP predictions, adjusted RADM and ASTRAP predictions with empirical data at 22 sites indicated that RADM best replicated the steep gradient downwind of a significant emissions source region. Although each model exhibited a tendency to smooth the gradient, the degree of smoothing appeared to be a function of the spatial resolution of the model. Based on the model comparisons with available empirical data, the adjusted RADM predictions appear to be the best estimates to date of the spatial distribution of annual sulfur dry deposition in the eastern United States.

Since the RADM adjustment factor here was 13% greater than that used in the Interim Report, an identical systematic increase in the interpolations appearing in the Interim Report is recommended. The difference in adjustment factors was a result of using as many as five years of Canadian data and two to three years of U.S. data, as opposed to only one to two years of data that were available at the time of the Interim Report.

The comparison of the revised estimates to empirically-derived amounts of sulfur dry deposition suggests that there is a systematic error in the revised estimates. Although adjusted RADM predictions of dry deposition are generally within ±60% of the empirical estimates, they tend to be biased low in the most significant source regions (where at least 200 ktonnes SO_2/yr are emitted within 80 km of the site). Conversely, in locations farther removed from significant sources (81-160 km) there is evidence that the estimates are biased high.

Because of the anticipation of data from additional sites and periods and impending improvements to the algorithms that calculate dry deposition velocities, it is recommended that this procedure be repeated at a later time. Therefore, these estimates of the annual sulfur dry deposition across the eastern United States could be revised in the future.

The EPA authors, **Terry L. Clark** *and* **Robin L. Dennis** *(also the EPA Project Officers, see below), are on assignment to the Atmospheric Research and Exposure Assessment Laboratory, Research Triangle Park, NC 27711 from the National Oceanic and Atmospheric Administration; Steven K. Seilkop is with Analytical Sciences, Incorporated, Research Triangle Park, NC 27713.*
The complete report, entitled "Re-Examination of Interim Estimates of Annual Sulfur Dry Deposition Across the Eastern United States," (Order No. PB 89-233 464/AS; Cost: $13.95, subject to change) will be available only from:
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