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RE-EXAMINATION OF INTERIM ESTIMATES OF ANNUAL SULFUR DRY DEPOSITION ACROSS THE EASTERN UNITED STATES

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ABSTRACT

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 superceeded 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years of empirical data have become available and a portion of the previously available empirical data has been superseded.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.

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ACKNOWLEDGEMENTS

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SECTION 1

INTRODUCTION

The Aquatic Effects Task Group of the National Acid Precipitation Assessment Program (NAPAP) required estimates of annual sulfur dry deposition across more than 7,000 lakes in the eastern United States. In August of 1987, in response to this need, the Atmospheric Sciences Research Laboratory (recently renamed the Atmospheric Research and Exposure Assessment Laboratory) provided estimates of sulfur dry deposition for these locations in an EPA Internal Report entitled "Interim Estimates of Annual Dry Sulfur Deposition for the Eastern United States for the Aquatics Research Program" (Dennis and Seilkop, 1987). *Interim* appeared in the title to reflect the impending improvements in our ability to estimate dry deposition amounts.

These interim estimates were derived by interpolating the spatial pattern of annual sulfur dry deposition amounts generated from annually-normalized predictions of Version I of the Regional Acid Deposition Model (RADM) (Chang *et al.*, 1987) that were adjusted using empirical data at 4 U.S. sites in the COre Research Establishment (CORE) Network (Hales *et al.*, 1987) and 18 Canadian sites in the Acidic Precipitation in Ontario Study (APIOS) Network (Ro *et al.*, 1988). The RADM results were preferred over those of Version 4D of the Regional Lagrangian Model of Air Pollution (RELMAP) (Eder *et al.*, 1986), since at that time the RADM spatial pattern exhibited less of an oversmoothing problem.

Since August 1987 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, as they appeared in the Interim

Report, 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 predictions from a third model, the Advanced Statistical Trajectory Regional Air Pollution (ASTRAP) model (Shannon, 1985), became available.

In this report we compare the dry deposition predictions from the three models (RADM, RELMAP and ASTRAP) to the available empirically-derived estimates to ascertain which of these models most accurately represents the spatial pattern of sulfur dry deposition. Using one of these models in conjunction with the available data, we then recommend systematic revisions to the interim estimates. The uncertainty and bias in these revised estimates are then characterized relative to interannual variability in the empirically-estimated amounts of sulfur dry deposition, and distance of sites from the emission source regions.

SECTION 2

REASONS FOR RE-EXAMINING THE INTERIM ESTIMATES

2.1 REVISION OF EMPIRICAL DATA AT U.S. SITES

Subsequent to the Interim Report, the inferential model developed by the National Oceanic and Atmospheric Administration's Atmospheric Turbulence and Diffusion Division (NOAA/ATDD) to estimate dry deposition rates was upgraded (Hicks *et al.*, 1987 and Hicks and Matt, 1988). Two significant changes resulted. First, more information about the nature of the surface has been incorporated into the calculation of the surface uptake resistance. In the previous version, only the one dominant plant species at each site was considered in the calculation of the deposition velocity. Currently, the model considers the two dominant plant species at each site. This was considered to be a necessary change since many sites are located within forests that are composed of both deciduous and coniferous species (e.g., Whiteface Mountain).

The second significant change to the inferential model was in its treatment of deposition to wet surfaces. The influence of surface wetness on deposition rates is an important consideration since, for example, vegetation in the southeastern United States is wet 15% to 20% of the time as a result of either dew or frost. In preceeding versions of the model, wet surfaces caused by rain or dew were considered to be stronger sinks for sulfur dioxide (SO_2) than were dry surfaces. However, recent empirical evidence from research projects conducted in the United States by NOAA/ATDD and in England (Mike Unsworth, University of Nottingham, personal communications) suggests that the enhancement of the deposition velocity occurs only for surfaces that are wet by dew. It has been suggested that for many locations, rainwater films enveloping leaves and stems are in equilibrium with the sulfur in the atmosphere. Thus, surfaces wet by rainwater constitute a relatively smaller sink compared to the same surfaces enveloped by dew. Other factors such as rainfall rates and the accumulation of particles on the leaves may be significant; however, their influence on uptake rates is not likely to be understood in the near future.

Some of the recent estimates of seasonal and annual dry deposition rates may have changed slightly as a result of minor corrections made to the concentrations analyzed from filterpack sampler data. These corrections compensate for irregularities in sampler flow rates, which since 1986 are periodically checked using mass flow controllers. In some cases, flow rates either have increased or decreased with time. In such instances, changes in the flow rates with time are assumed to be linear; adjustments are performed on a weekly basis. The most significant adjustments, a 50% reduction, were applied to pre-February 1985 concentrations at the Penn State site. Here, filter pack SO_2 concentrations were found to be twice those simultaneously measured by a colocated monitor. Adjustments to concentrations measured at other locations were much less significant. Similar discrepancies are also noted when comparing seasonally averaged SO_2 concentrations from 1985 to 1987.

2.2 AVAILABILITY OF ADDITIONAL DATA TO CHARACTERIZE INTERANNUAL VARIABILITY

Interannual variability of sulfur dry deposition must be considered in any estimation procedure that is based on data from a specific year or other time period. If there is significant interannual variability, estimates based on a particular year or other time period may not be applicable to another period of interest. This is precisely the situation that occurs in our estimation of sulfur dry deposition for the 12-month period chosen for studying aquatic effects (1 October 1983 through 30 September 1984). Since dry deposition data were not collected until after this time period, our annual estimates are therefore specific to this later time period, and could potentially misrepresent the earlier (aquatic effects) time period. To characterize the uncertainty in applying estimates based on data obtained after 1984 to the aquatic effects time period, it is therefore important to attempt to determine the magnitude of the interannual variability in sulfur dry deposition.

In early 1987, empirical data were available for at most two years. Consequently, at that time the annual variability of sulfur dry deposition could not be established with confidence. Now, however, for some sites in Ontario and in the eastern United States, data are available for as many as five years. Although more data are needed to better estimate the actual interannual variability of sulfur dry deposition, we are now able to derive a preliminary estimate of the variability with the understanding that additional data could expand the range of annual amounts. Therefore, these uncertainty estimates could be slightly underestimated.

The expanded empirical data base offers a second benefit in that we now have for the first time U.S. and Ontario empirical data for the same years (1985 and 1986). If interannual variability is indeed significant, then this is a substantial improvement, since empirical data at the sites are used to adjust the annual model predictions for bias. With this approach, it is therefore desirable to use empirical estimates for the same period so that differences between sites will not reflect the temporal variability of sulfur dry deposition.

Finally, the expanded data base, in addition to providing a more representative sample, enables us to identify outliers and discard or, if possible, correct erroneous values. The adjustment of the SO_2 concentrations at the Penn State site, previously discussed, is an example of the benefits derived from an expanded data base.

2.3 POSSIBLE IMPROVEMENT OF ESTIMATES USING ADDITIONAL MODELING RESULTS

In the August 1987 Interim Report, the annual sulfur dry deposition predictions derived from RADM's annually normalized results were deemed to be more representative of the empirical data than the pattern derived from RELMAP annual results. At that time, a comparison of empirical data and RELMAP results indicated that RELMAP underpredicted near the regions of high emissions and overpredicted across regions farther downwind. The recent improvements to RELMAP and the availability of annual predictions from a third model, the ASTRAP model (Shannon, 1985), warranted a second comparison of model predictions to the empirical data.

The RELMAP module for dry deposition was modified to better reflect the physical processes occurring across the spatial scales of the model grid cells (i.e., one degree latitude by one degree longitude). First and foremost, rather than injecting all of the SO_2 and sulfate emissions into the layers above the surface layer (i.e., 50 m at night and 200 m otherwise), the model now injects all area source emissions into the surface layer. This is more realistic since much of the area source emissions in the United States are emitted at or near the surface. Secondly, the order of the calculations of transport and dry deposition was reversed. The model now calculates the amount of dry deposition at the source before transporting pollutant mass away from it. The net effect of these changes has increased the amounts at the source cells and decreased them hundreds of kilometers downwind.

Finally the last significant modification enabled RELMAP to apply dry deposition velocities appropriate for each of 11 land-use categories. These deposition velocities were also updated by those recommended by Sheih *et al.* (1986). The model predictions are now based on the dry deposition across each land-use category, whereas before, the cell deposition was based on only the predominant land-use category. Thus, in theory, the new approach yields more representative predictions for the cell as a whole.

SECTION 3

SPATIAL DISTRIBUTION OF ANNUAL SULFUR DRY DEPOSITION

3.1 COMPARISON OF MODEL PREDICTIONS AND EMPIRICAL DATA

Dry deposition rates are a function of the air concentration near the surface and the dry deposition velocity. The latter cannot be measured directly but is inferred from vertical flux measurements. In addition, since dry deposition velocities are a function of atmospheric stability and surface attributes (e.g., vegetative type, roughness length, physical conditions, spatial fluctuations of terrain and surface roughness), they 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.

Estimation of a spatial pattern of sulfur dry deposition across the eastern United States is even more 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 (Table 1). As was the case described in the Interim 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

TABLE 1. MODELS USED TO CONSTRUCT GRIDS OF ANNUAL AMOUNTS OF SULFUR DRY DEPOSITION ACROSS EASTERN NORTH AMERICA

Model name	Model genre and approximate spatial resolution, km
Regional Acid Deposition Model (RADM)	Eulerian 80
Regional Lagrangian Model of Air Pollution (RELMAP)	Lagrangian 100
Advanced Statistical Trajectory Regional Air Pollution (ASTRAP) Model	Statistical-Lagrangian 130

year of 1980. The RELMAP results presented here were derived from the improved model version and differed from those presented in the Interim Report.

The second step was accomplished by first comparing model predictions with the annual means of the empirical data at each of 22 sites of the CORE and APIOS networks (Figures 1 and 2). Empirical data at the CORE sites were based on weekly mean dry deposition velocities and air concentrations for the years 1985 to 1987, inclusive. Dry deposition data from two CORE sites were not used here. A significant bias, as high as +50%, was suspected at West Point, New York due to "edge" effects (i.e., significant spatial gradients in terrain or surface roughness). In addition, the revised data from Bondville, Illinois were not available. In contrast, Ontario empirical data were based on a cruder, less accurate method -- since only annual mean dry deposition velocities were available, sulfur dry deposition amounts at the APIOS sites were determined from these deposition velocities and annual mean concentrations for the years 1982 to 1986, inclusive.

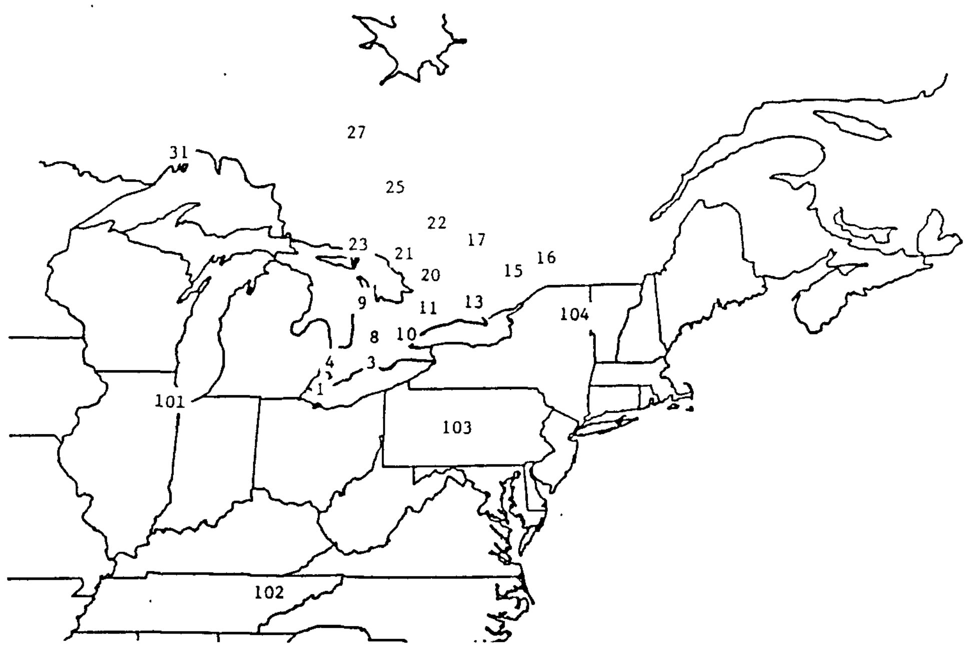

Figure 1. The location of 22 sites of the CORE and APIOS networks from which empirically-derived amounts of sulfur dry deposition were available for at least one year.

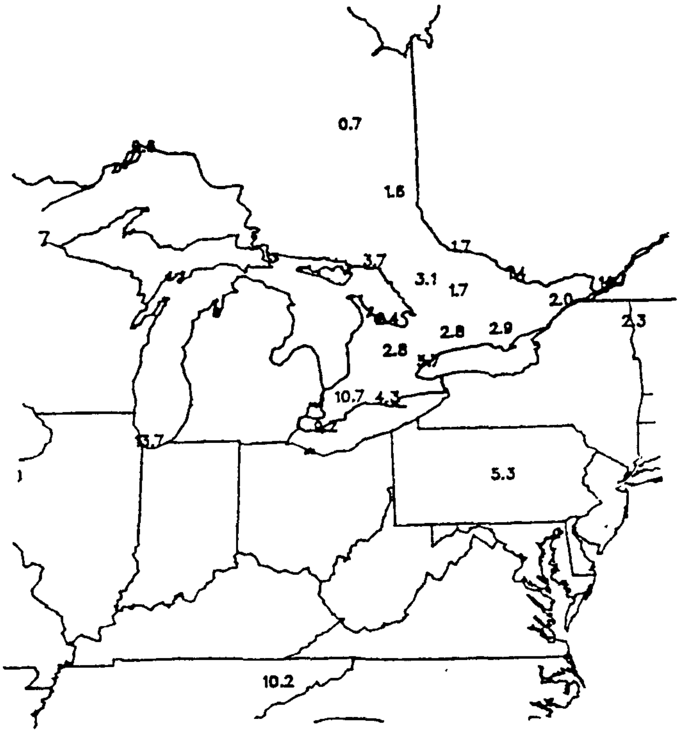

Figure 2. Empirically-derived annual amounts of sulfur dry deposition (kg S/ha/yr).

A comparison of the annual mean amounts of sulfur dry deposition at the CORE and APIOS sites (Figure 2) and the spatial distribution of 1980 anthropogenic SO_2 emissions (Figure 3) gives evidence of a strong correlation between the two. Specifically, sulfur dry deposition tends to be greatest across regions of high SO_2 emissions. Consequently, model predictions of spatial gradients of sulfur dry deposition are expected to generally reflect those of emissions.

From the comparisons of empirical estimates and model predictions, mean-square errors were calculated and used as a measure of concurrence. To reduce 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. Similar adjustments to RELMAP predictions were unnecessary, as the estimated regression coefficient was almost identical to one. Thus,

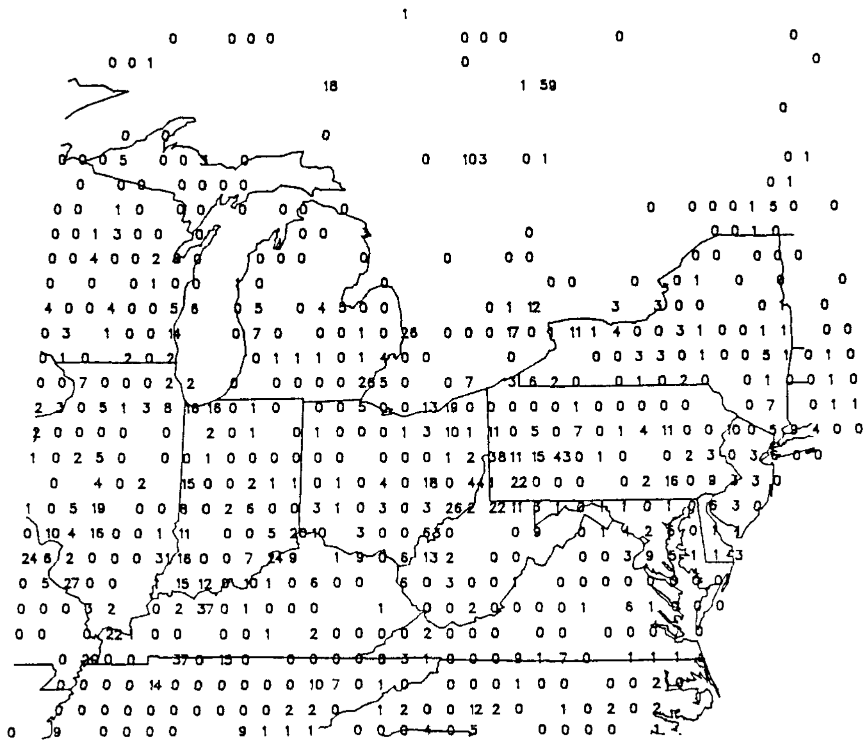

Figure 3. Annual 1980 SO_2 emissions (10 ktonnes/yr) from anthropogenic sources (NAPAP, 1987).

unlike the other models, RELMAP gives no evidence of bias. Compared to the results of the earlier version of RELMAP (presented in the Interim Report) these results represent a significant improvement.

For each of the 22 sites, Table 2 compares the empirically-derived amounts of annual sulfur dry deposition with the adjusted RADM predictions, unadjusted RELMAP predictions and adjusted ASTRAP predictions. As the descriptive statistics in Table 3 indicate, the adjusted RADM predictions best replicate the empirical data; its bias, root-mean-square error, and average error are lower than those of the other two models. This is also supported by the scatter plots of Figure 4. The scatter plots also illustrate the tendency of the models to overpredict the sulfur dry deposition in the empirical data range of 3 to 6 kg S/ha/yr. This is indicative of the models' slower rate of decreasing dry deposition away from the high emissions regions, or in other terms, their smoothing tendency near steep

TABLE 2. COMPARISON OF MODEL PREDICTIONS AND EMPIRICALLY-DERIVED ESTIMATES OF ANNUAL SULFUR DRY DEPOSITION AT THE 22 SITES

Site ID	Site Name	Mean Empirical Estimate, kg S/ha/yr (No. of years)	Predictions of annual sulfur dry deposition, kg S/ha/yr (Relative deviation#, %)		
			Adjusted RADM	Unadjusted RELMAP	Adjusted ASTRAP
CORE Sites (U.S.)					
101*	Argonne, IL	13.74 (3)	15.72 (+14.5)	10.48 (-23.7)	9.32 (-32.2)
102*	Oak Ridge, TN	10.20 (3)	7.98 (-21.8)	7.73 (-24.2)	8.07 (-20.8)
103*	Penn State, PA	5.34 (2)	7.99 (+49.6)	9.52 (+78.3)	11.37 (+113.0)
104*	Whiteface, NY	2.31 (2)	2.40 (+4.0)	5.78 (+150.3)	4.23 (+83.2)
APIOS Sites (Ontario)					
1*	Colchester	9.24 (3)	7.18 (-22.3)	6.96 (-24.7)	9.94 (+7.6)
3*	Port Stanley	4.32 (5)	6.90 (+59.8)	5.80 (+34.3)	8.03 (+85.9)
4*	Wilkesport	10.66 (2)	9.39 (-11.9)	8.12 (-23.8)	8.64 (-18.9)
8*	Palmerston	2.82 (3)	4.28 (+51.8)	5.71 (+102.9)	5.59 (+98.5)
9	Shallow Lake	2.45 (4)	3.84 (+57.0)	2.26 (-7.7)	3.67 (+50.1)
10*	Milton	5.69 (1)	5.13 (-9.8)	11.16 (+96.2)	10.85 (+90.6)
11	Uxbridge	2.82 (3)	5.46 (+93.6)	3.70 (+31.0)	5.01 (+77.8)
13	Campbellford	2.90 (5)	2.59 (-10.6)	3.81 (+31.3)	4.79 (+65.3)
15*	Smith's Falls	2.01 (3)	2.64 (+31.7)	4.24 (+111.2)	3.65 (+82.2)
16	Dalhousie Mills	1.71 (5)	2.41 (+40.4)	5.65 (+229.8)	3.53 (+106.2)
17	Golden Lake	1.43 (5)	1.57 (+9.6)	2.61 (+82.7)	3.52 (+146.3)
20	Dorset	1.69 (5)	1.82 (+7.7)	2.75 (+62.1)	3.83 (+125.9)
21	McKellar	3.06 (4)	2.06 (-32.7)	2.95 (-3.5)	3.83 (+25.3)
22*	Matawa	1.69 (5)	1.79 (+6.2)	2.71 (+60.4)	4.12 (+143.9)
23	Killarney	3.68 (5)	2.16 (-41.5)	1.81 (-50.9)	11.13 (+202.3)
25	Gowganda	1.65 (5)	1.50 (-7.3)	1.63 (+0.5)	2.24 (+37.9)
27	Moonbeam	0.72 (4)	1.42 (+96.8)	0.67 (-7.6)	0.78 (+8.7)
31	Dorion	0.59 (2)	0.92 (+57.7)	0.30 (-49.2)	0.49 (-16.2)

[(Prediction - Empirical Estimate) / (Empirical Estimate)] x 100

* Sites used in the 11-site subset (Table 3).

TABLE 3. DESCRIPTIVE STATISTICS FOR ADJUSTED RADM, UNADJUSTED RELMAP, ADJUSTED ASTRAP PREDICTIONS, AND EMPIRICAL ESTIMATES OF ANNUAL SULFUR DRY DEPOSITION

	Empirical Estimates	RADM	RELMAP	ASTRAP
AT ALL 22 U.S. AND ONTARIO SITES:				
Mean	4.12	4.42	4.83	5.76
Bias		7.3%	17.2%	39.8%
RMSE*		1.42 (34%)	2.41 (58%)	3.04 (74%)
Average Error		1.11 (25%)	1.90 (46%)	2.42 (59%)
AT 11 SITES (4 UNITED STATES AND 7 ONTARIO SITES):				
Mean	6.18	6.49	7.11	7.62
Bias		5.0%	15.0%	23.3%
RMSE*		1.69 (27%)	3.08 (50%)	3.38 (55%)
Average Error		1.42 (23%)	1.90 (31%)	2.99 (48%)

* Root-mean-square error

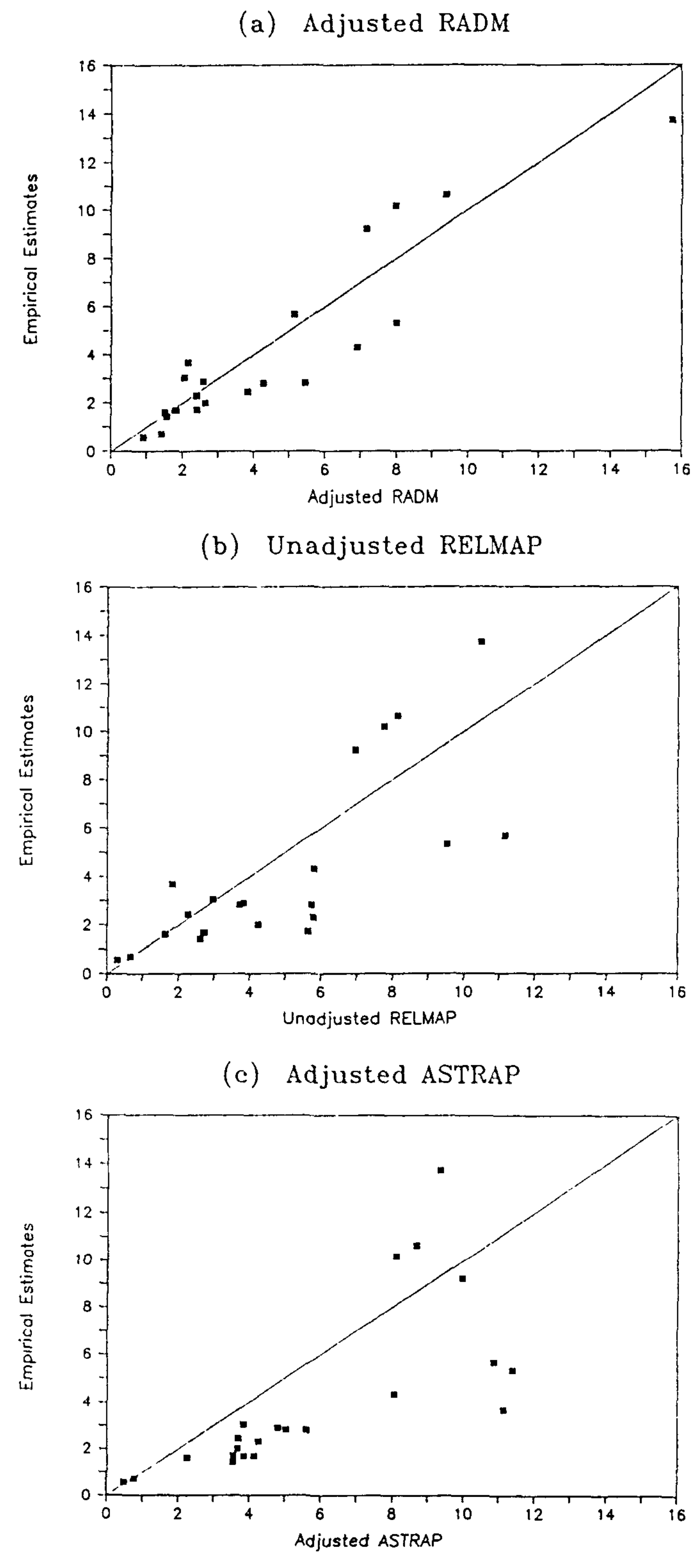

Figure 4. Scatter plots of empirically-estimated annual amounts of sulfur dry deposition (kg S/ha/yr) versus: (a) adjusted RADM, (b) unadjusted RELMAP, and (c) adjusted ASTRAP predictions at the 22 sites.

gradients (Figures 2 and 5). Moreover, the degree of smoothing appears to be a function of the spatial resolution of the models (Table 1). That is, the coarser the spatial resolution, the more smoothing is evident.

Descriptive statistics were also computed using only the empirical estimates at four of the U.S. sites and seven Ontario sites (Table 3) to provide a more equal balance between the available U.S. and Ontario data, thereby reducing the possibility of overweighting discrepancies in Ontario relative to those found at United States sites. The Ontario sites, identified in Table 2, were selected on the basis of their position relative to the steep gradient both in terms of sulfur emissions and sulfur dry deposition from Lake Huron to eastern Ontario. By computing descriptive statistics from this subset, one can assess: (1) the performance of the models in replicating steep gradients, and (2) the degree of smoothing intrinsic to each model.

As was the case for the entire data set, adjusted RADM results best replicate the empirical data of the 11-site subset. The scatter plots of Figure 6 also support this conclusion. Therefore, as was the case in the Interim Report, the adjusted RADM predictions appear to provide better estimates of the annual sulfur dry deposition amounts than those obtained from the simpler linear models. Figure 7 illustrates the spatial pattern of annual sulfur dry deposition derived from the adjusted RADM predictions.

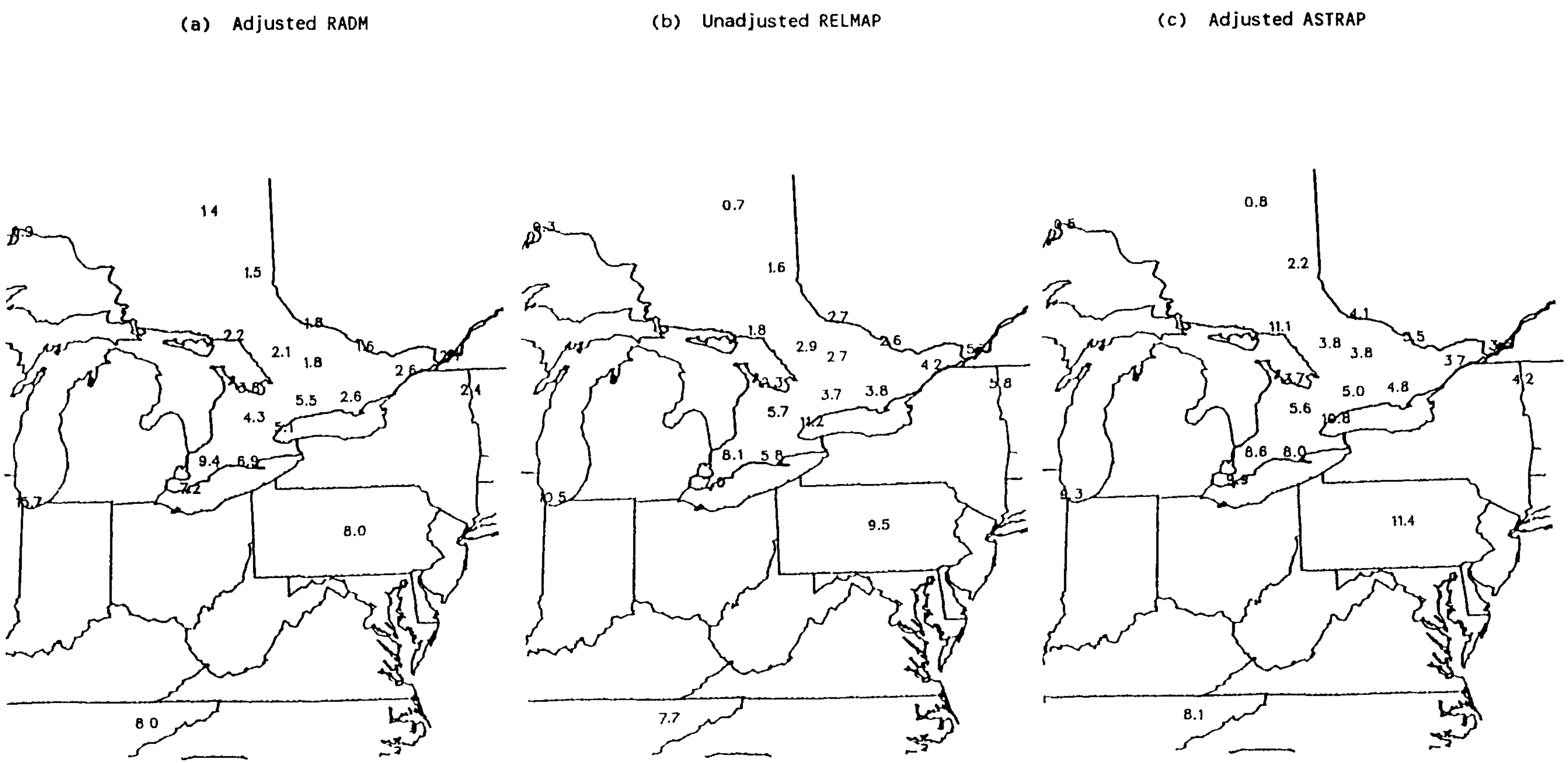

Figure 5. Annually-normalized RADM predictions adjusted for bias (a), annual RELMAP predictions (b), and annual ASTRAP predictions adjusted for bias (c) of sulfur dry deposition (kg S/ha/yr) at the 22 sites.

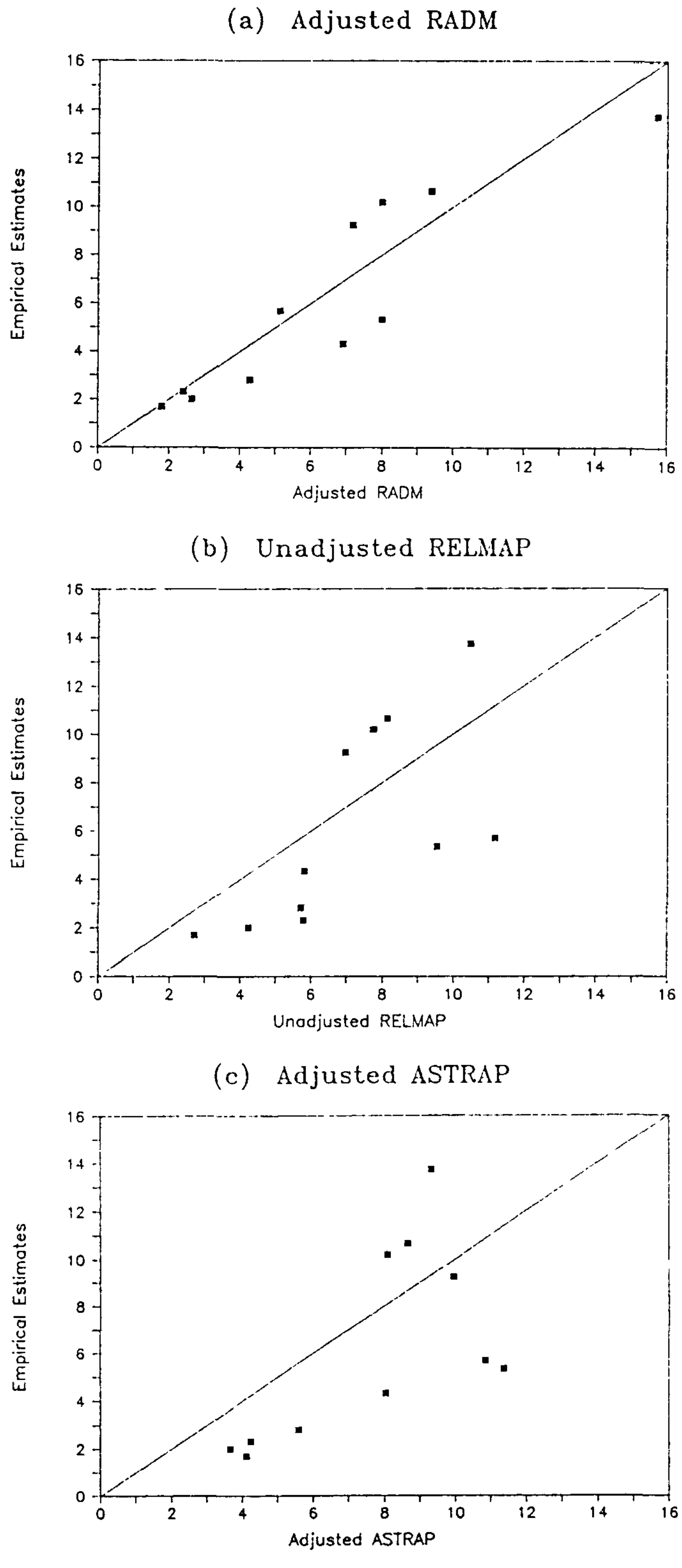

Figure 6. Scatter plots of empirically-estimated annual amounts of sulfur dry deposition (kg S/ha/yr) versus: (a) adjusted RADM, (b) unadjusted RELMAP, and (c) adjusted ASTRAP predictions at the four U.S. sites and seven Ontario sites.

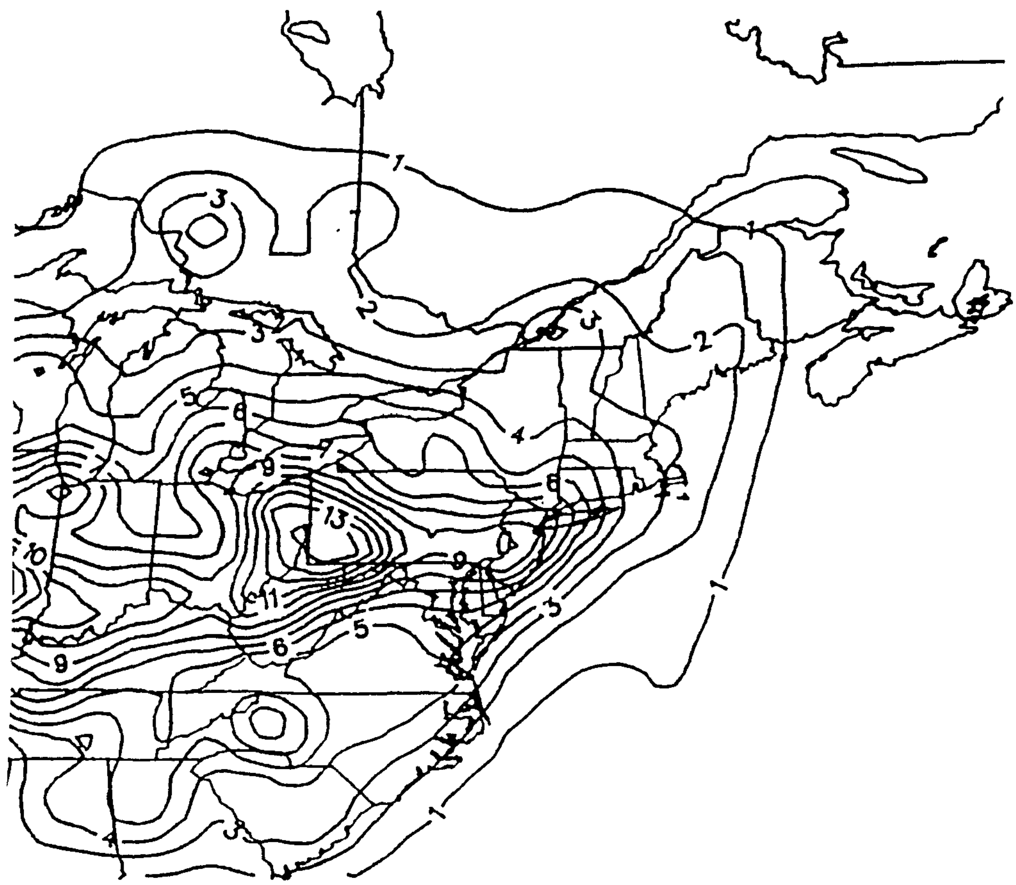

Figure 7. The spatial pattern of annual sulfur dry deposition (kg S/ha/yr) derived from adjusted RADM predictions.

3.2 UNCERTAINTY ASSESSMENT OF THE REVISED ESTIMATES

The uncertainty in sulfur dry deposition amounts obtained from the estimation procedure described above is primarily related to three main factors: (1) the accuracy with which the RADM captures the underlying spatial pattern of dry deposition, (2) the accuracy of the empirical dry deposition estimates that are used to adjust the predictions, and (3) the potential systematic differences between the empirical dry deposition estimates from recent years and the period of interest. This section discusses these factors and their effects on the uncertainty in estimated regional patterns of sulfur dry deposition.

3.2.1. Uncertainty of Adjusted RADM Predictions

To some extent, we have already investigated the uncertainty in the adjusted RADM predictions by comparing them to the empirical estimates (Tables 2 and 3 and Figures 4 and 5). Some random differences between the adjusted RADM and the empirical estimates are expected because of differences in spatial scales. For example, the RADM is a regional model that is not expected to simulate spatial gradients on scales below its grid cell resolution of approximately 80 km. Similarly, empirical estimates from an individual site imperfectly represent a regional average. Because we have so little data from the United States against which to compare the adjusted RADM predictions, it is important to attempt to determine whether any of the observed difference is related to model error. In particular, examination of model performance relative to emission patterns can provide insight into the model's ability to accurately represent physical processes, and give a sense of the degree of confidence that can be placed in its predictions.

There is some evidence to suggest that the RADM exhibits biases relative to the emissions pattern. Although it appears to represent an improvement over Lagrangian models relative to oversmoothing in areas within approximately 200 km downwind of significant emission source regions, there is still some estimation bias in these areas. As illustrated by Figure 8, the largest positive errors occur in southwestern Ontario and to the northeast of Toronto; sites in this region are 100 to 200 km northeast of major sources. In addition, the adjusted model estimate at the Penn State site, downwind of a major source region (Figures 3 and 4), is 50% greater than the empirical estimate.

It appears that the pattern of over/underestimation is linked to the proximity of major source regions, as indicated in Table 4 and Figure 9. In regions within 80 km of major sources (i.e., those emitting more than 200 ktonnes of SO_2 per year), RADM tends to underestimate dry deposition.

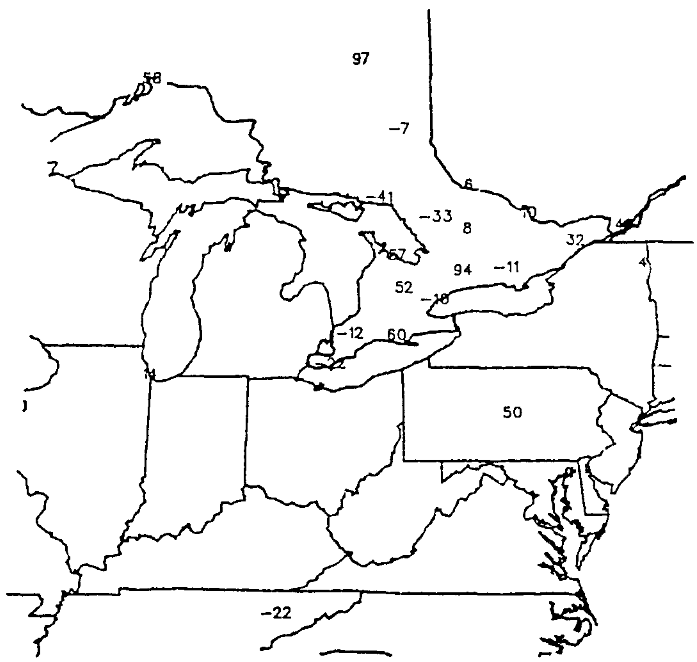

Figure 8. Percent errors in the adjusted RADM predictions of annual sulfur dry deposition.

TABLE 4. DEVIATIONS OF ADJUSTED RADM PREDICTIONS FROM EMPIRICALLY-DERIVED DRY DEPOSITION ESTIMATES RELATIVE TO ANNUAL SO_2 POINT SOURCE EMISSIONS

	SO_2 emissions, ktonnes/yr		Sulfur dry deposition, kg S/ha/yr		Relative deviation*, %
Site ID	Within 80 km	Within 81-160 km	RADM	Empirical estimates	
GROUP A SITES (>200 KTONNES WITHIN 80 KM)					
1	421	746	7.2	9.2	-22
101	409	345	15.7	13.7	14
10	304	205	5.1	5.7	-10
4	297	803	9.4	10.7	-12
102	209	169	8.0	10.2	-22
GROUP B SITES (<125 KTONNES WITHIN 80 KM BUT >125 KTONNES WITHIN 81-160 KM)					
103	95	904	8.0	5.3	50
3	<1	957	6.9	4.3	60
8	3	620	4.3	2.8	51
11	102	382	5.5	2.8	94
13	<1	146	2.6	2.9	-11
9	2	137	3.8	2.4	57
GROUP C SITES (<125 KTONNES WITHIN 160 KM)					
16	27	69	2.4	1.7	40
104	64	60	2.4	2.3	4
15	4	33	2.6	2.0	32
31	14	16	0.9	0.6	57
27	8	13	1.4	0.7	97
17	<1	2	1.6	1.4	10
20	0	<1	1.8	1.7	8
GROUP D SITES (WITHIN 160 KM OF SUDBURY, ONTARIO)					
23	896	135	2.2	3.7	-41
25	0	1,633	1.5	1.7	-7
21	0	1,040	2.1	3.1	-33
22	9	1,031	1.8	1.7	6

* [(Prediction - Empirical Estimate) / (Empirical Estimate)] x 100

DEVIATIONS OF ADJUSTED RADM PREDICTIONS

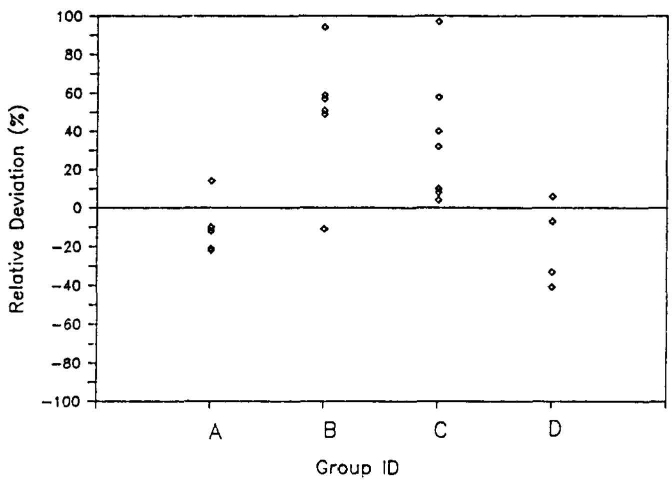

Figure 9. The deviations of adjusted RADM predictions from empirically-derived annual estimates of sulfur dry deposition relative to the four groups of sites in Table 4.

The one exception is the Argonne site (101), which, unlike the other Group A sites, is normally situated on the upwind side of the source region.

The model predictions are greater than the empirical estimates at sites located within 81 to 160 km of major source regions. This suggests that the model tends to oversmooths dry deposition gradients near source regions. The pattern of overprediction does not, however, emerge at sites within 81 to 160 km of the major sources in Sudbury, Ontario. In fact, in this region the model performance appears to parallel that in regions within 80 km of major source regions, with underpredictions and a single modest overprediction of 6%. One cause of this difference in model behavior might be related to the Sudbury stacks, which are much taller than those elsewhere in North America. These tall stacks might deposit sulfur compounds farther downwind than typical sources, with much of the deposition occuring in the 81-160-km range rather than within 80 km. Consequently, it would not be unreasonable to expect that the model's behavior within 81-160 km of 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 (Group C). 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 (Figure 10). 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, Figure 8 illustrates the consistency of model behavior with regard to over and underestimation, with errors ranging ±40%. However, as noted previously, 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.

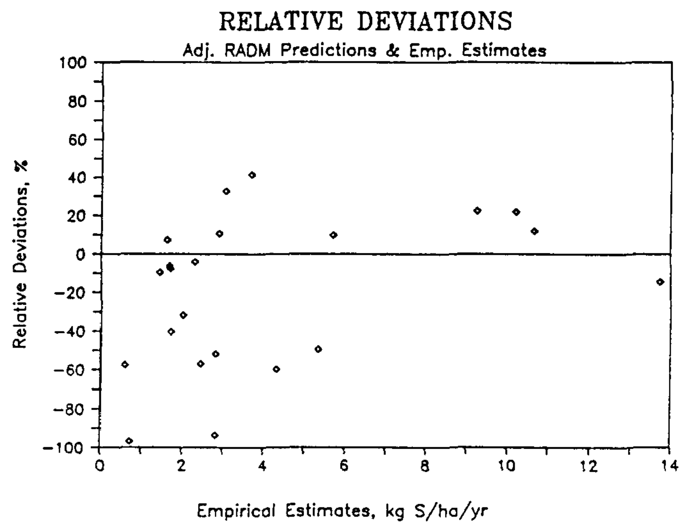

Figure 10. Comparison of the relative error in the adjusted RADM predictions and the empirically-derived annual estimates of sulfur dry deposition (kg S/ha/yr).

3.2.2 Uncertainty of the Empirical Estimates

Since appropriate techniques are not yet developed to directly measure dry deposition, we must rely on either inferential techniques that estimate dry deposition from measurable parameters [such as vertical fluxes of pollutants near the surface -- the method used at the U.S. sites (Hicks and Matt, 1988)], or the less desirable method, used at the Ontario sites, based on long-term mean concentration measurements and estimates of dry deposition velocities. Specifically, the Ontario estimates of annual dry deposition were computed from annual mean measurements of SO_2 and sulfate concentrations and annual mean dry deposition velocities. The dry deposition velocities were estimated via the method first reported by Masse and Voldner (1983) and updated by Voldner *et al.* (1986). In contrast, the U.S. amounts were computed from weekly mean measurements of SO_2 and sulfate concentrations and mean dry deposition velocities inferred from flux measurements for the same week. Regardless of which of these two approaches are used, errors arise and contribute to the deviations illustrated in Figure 8.

Since dry deposition inferential methods rely directly on vertical flux measurements of both SO_2 and sulfate, the accuracy of these measurements directly determines the accuracy of the U.S. empirical estimates. For individual measurements this could translate into significant deviations between model predictions and empirical estimates. Fortunately, however, the empirical estimates at the U.S. sites are actually annual averages based on enough weekly measurements to virtually eliminate the effects of random measurement errors. Therefore, only a bias in the measurement technique would affect the accuracy of the U.S. estimates. If there are biases in the empirical deposition estimates, it is unlikely that they would be related to the emissions pattern, as the deviations of adjusted model predictions from empirical estimates appear to be.

The key sources of uncertainty of the Ontario estimates are related to the uncertainties arising from the estimation of the climatological dry deposition velocities and the assumption that the product of the long-term mean concentrations and deposition velocities is representative of the sum of the products of the short-term concentrations and deposition velocities. These uncertainties have yet to be assessed.

With the very limited available data, it is impossible to separate and quantify the three sources of error (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.

3.2.3 Interannual Variability of the Empirically-Derived Estimates

Since the Aquatics Effects Program of NAPAP focuses on the period 1 October 1983 to 30 September 1984, it then follows that our estimates of annual sulfur dry deposition should be confined to that same period. However, since empirical data in the United States exist only after this period, we must base our estimates on the years subsequent to the period of interest. Therefore, one component of the uncertainty of our best estimates for this 12-month period relates to the interannual variability of sulfur dry deposition as determined from the empirical data.

To assess the magnitude of this uncertainty, the interannual variability was estimated at the Ontario APIOS sites using all available data passing the screening criteria. Figure 11 illustrates the annual variations in sulfur dry deposition for five years (1982-1986) along a cross section of the APIOS network from Site 1 near Detroit to Site 16 in eastern Ontario.

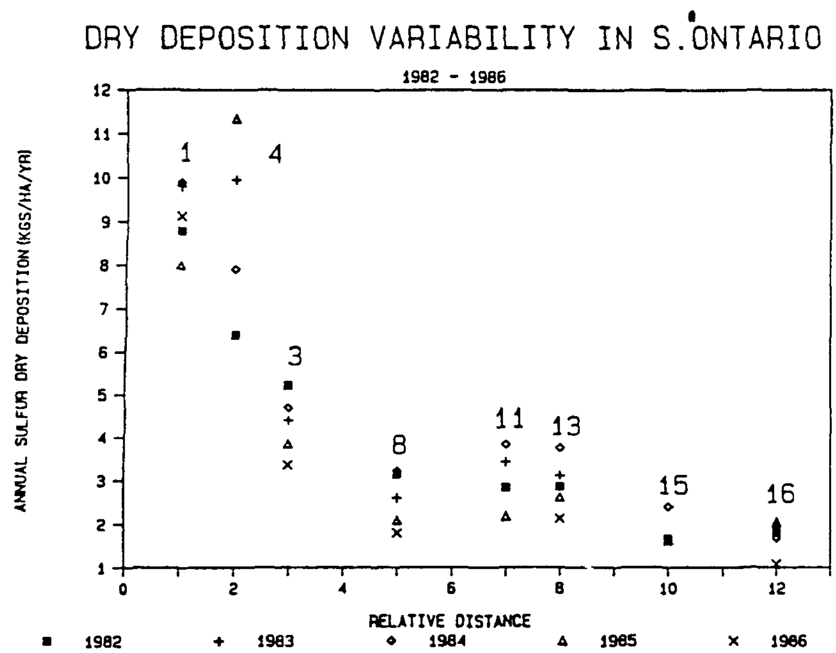

Figure 11. Variations of the annual means (1982-1986) of sulfur dry deposition along a cross section of southern Ontario sites.

This cross section was selected to relate annual variability to the pattern of sulfur emissions, which along this line exhibits an emissions decrease of two orders of magnitude from sites 1 and 4 to 3. Except for Site 4, where annual amounts varied as much as 5 kg S/ha/yr, a variation of 1 to 2 kg S/ha/yr is apparent. This translates to an annual variation of 10% to 30% of the average annual amount of sulfur dry deposition.

Since data for only two or three years are available at the U.S. sites, our ability to quantify interannual variability at the U.S. sites is limited. However, as Figure 12 illustrates, from the available data, only small interannual variations, on the order of 1 kg S/ha/yr, are observed during the period 1985 to 1987. At Argonne and Oak Ridge this amounts to only a 10% interannual variation, considerably less than that at the APIOS sites. Less than a 10% variation occurs in the two years of data at Penn State and Whiteface Mountain.

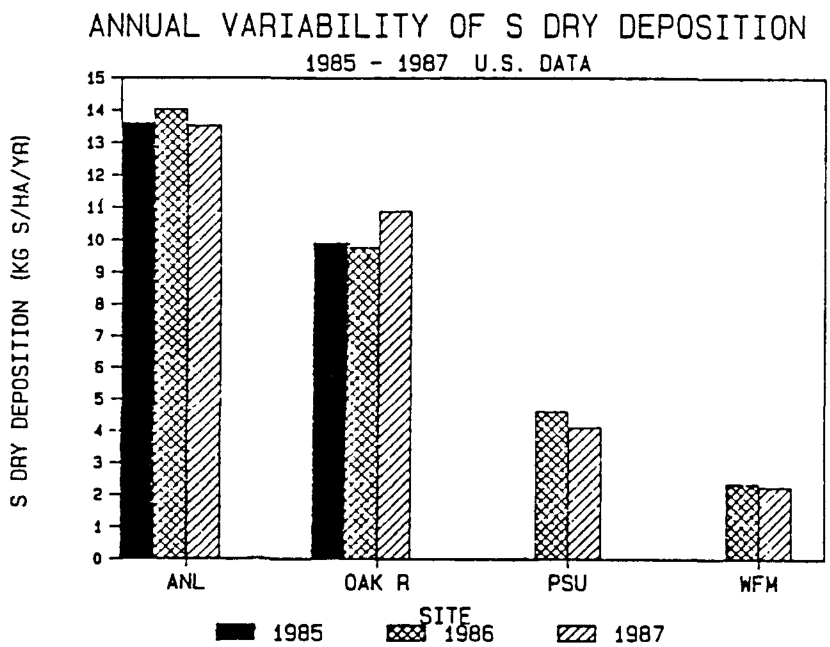

Figure 12. Variations of the annual means (1985-1987) of sulfur dry deposition at four U.S. sites.

The apparent differences in the interannual variabilities at the Ontario and U.S. sites can possibly be explained by the significant differences in the approaches of estimating sulfur dry deposition, as discussed in the preceeding section. Since the latter approach is theoretically more precise, the interannual variability apparent in the Ontario annual estimates might be related to the uncertainty imposed by the estimation method and not an indication of the true annual variability. Therefore, until additional U.S. data are available, a ±10% interannual variability is assumed. Since the magnitude of this variation is very small compared to the ±60% estimate of the uncertainty discussed previously, our uncertainty estimate remains at ±60%.

3.3 ESTIMATED SEASONAL DISTRIBUTION OF SULFUR DRY DEPOSITION

In addition to estimates of total annual sulfur dry deposition, some aquatic effects researchers might need to know the distribution of dry deposition amounts across seasons. This distribution was determined using 1985 to 1987 empirical estimates at the U.S. sites. As Figure 13 shows, approximately 30% of the annual sulfur dry deposition occurs in each the spring and summer and approximately 20% in each the winter and autumn at Argonne, Oak Ridge and Penn State. The seasonal variation of dry deposition was less at Whiteface Mountain; this site differed from the other three U.S. sites in that it was located in a mixed conifer/deciduous forest and, therefore, had a higher wintertime dry deposition velocity for SO_2.

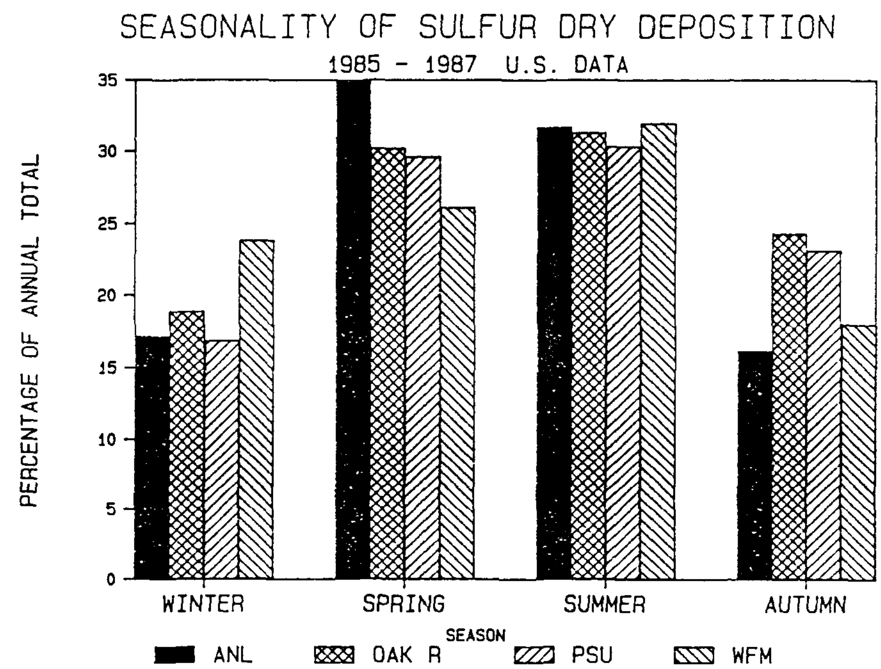

Figure 13. The seasonal distribution of annual sulfur dry deposition at four U.S. sites (Argonne -- ANL and Oak Ridge -- OAK R: 1985-1987; Penn State -- PSU and Whiteface Mountain -- WFM: 1986-1987).

SECTION 4

CONCLUSIONS AND RECOMMENDATIONS

In August 1987 annual sulfur dry deposition to more than 7000 lakes in the eastern United States were estimated from interpolated model calculations anchored to the empirical data. Since that time, several developments have occurred to warrant a second examination of these best estimates. The procedure described in the Interim Report was repeated; however this time, the predictions of an additional model and an expanded data base of empirical estimates were available.

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.

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