5.3 RESPONSE OF SIMULATED SULFUR IN MODELS-3/CMAQ TO ALTERNATE CLOUD PARAMETERIZATIONS

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1. INTRODUCTION

Models-3/CMAQ—the air quality modeling system developed by the United States Environmental Protection Agency—simulates atmospheric sulfur chemistry using both homogeneous and heterogeneous reactions. Homogeneous gas-phase oxidation of SO_2 proceeds in response to available sunlight. Heterogeneous reactions in clouds oxidize SO_2 only when clouds and certain oxidants and/or catalysts are present. The atmospheric balance between SO_2 and sulfate affects the lifetimes of both species and influences source-receptor relationships. This paper describes a series of tests that examined the relationship between sulfur balances and cloud cover as simulated in CMAQ using both default and alternate cloud-related parameterizations.

2. OBSERVED CLOUD COVER

Archived National Weather Service (NWS) surface observations contain hourly data on fractional cloud cover for clouds below 3.7 km. This includes all the boundary layer and corresponds well with that portion of the atmosphere of greatest impact on the sulfur balance. All anthropogenic sources of SO_2 are located within a few hundred meters of the ground. Plume rise for large SO_2 sources rarely elevates plumes above 3 km.

Cloud cover fraction, f_c, is not reported when the sky is obscured by ground fog. No attempt was made to include fog in the comparison with model results. This is consistent with CMAQ which does not allow for cloud in the surface layer. For layers aloft, CMAQ cloud cover was compared with observations only when CMAQ layers below 3.7 km contained cloud. Data from 37 NWS stations scattered throughout the eastern U. S. and covered by a 36-km (grid cell size) model grid were compared with simulated f_c (Figure 1).

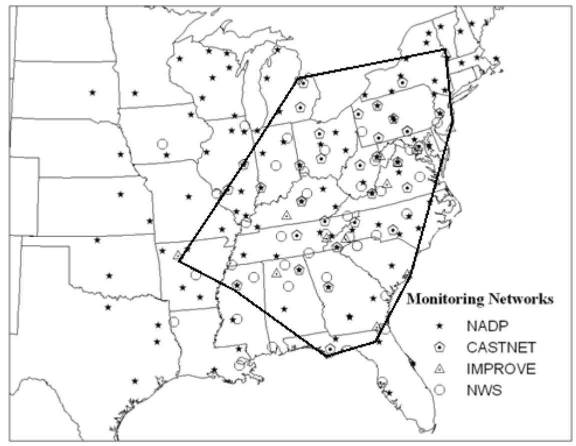

Figure 1. Locations of wet deposition (NADP), CASTNet, IMPROVE and cloud (NWS) measurement sites used in evaluating model performance for the 36-km grid. The large polygon represents the core study region.

Observed f_c for 32 simulation days (Section 4) revealed that clouds below 3.7 km were primarily an afternoon phenomenon (Figure 2a) although substantial nighttime cloud cover also occurred. In addition, f_c values—which ranged from 0 (no clouds) to 1 (complete overcast) were distributed

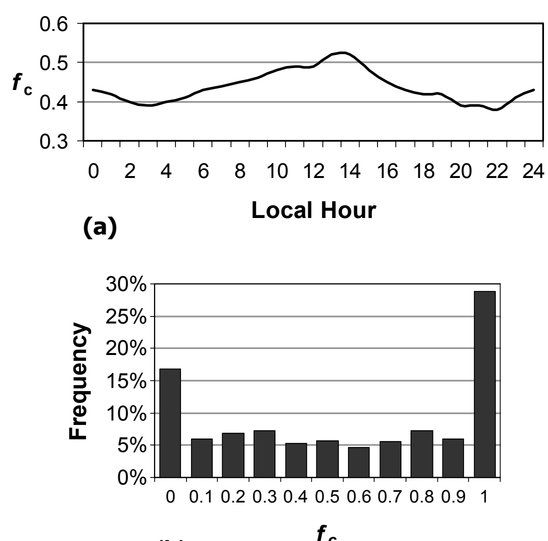

Figure 2. (a) Mean observed diurnal variation in f_c and (b) frequency distribution of f_c.

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such that 46 percent of observations indicated either clear or overcast skies. "Partly cloudy" conditions of varying degrees characterized the other hours (Figure 2b).

The afternoon peak in f_c is associated with convection that occurs many days within and above the boundary layer, especially during the warmer months. These clouds, mostly of the cumulus type, interact with pollutants to drive various aqueous reactions involving SO_2 oxidation. Heterogeneous oxidation of SO_2 can enhance total sulfate formation rates. Several aqueous reactions are known to oxidize SO_2, with the most important being the reaction with O_3 at pH>5 and the reaction with H_2O_2 across a wide range of pH (Seinfeld and Pandis, 1998). The effective SO_2 oxidation rate in a cloud depends primarily on the liquid water content of the cloud, the presence of species affecting pH (such as HNO_3 and NH_3), aqueous concentrations of O_3 and H_2O_2, and cloud lifetime. Under optimal conditions clouds can effectively convert all ambient SO_2 into sulfate.

3. CLOUD PARAMETERIZATIONS

CMAQ version 4.2 was used in this study (U. S. EPA, 1999). More recent versions exist but the treatment of clouds appears to be fundamentally unchanged through version 4.4. CMAQ simulates the presence of both resolved and subgrid-scale clouds and their effects on atmospheric chemistry. Using module RESCLD, CMAQ first determines the presence of "resolved" clouds based on the total liquid water mixing ratio, Q_L, in each layer. Resolved clouds are assumed to be present whenever Q_L>0.05 g kg^{-1}. Heterogeneous chemistry is simulated within each layer if the cloud liquid water content, C_L exceeds 0.01 g m^{-3}. At sea level and 273 K, a value of Q_L=0.05 g kg^{-1} is equivalent to C_L=0.065 g m^{-3}. Hence, in the boundary layer (pressure ≥80 kPa over the eastern United States) the model is unlikely to activate the heterogeneous chemistry module from RESCLD unless C_L ≥0.05 g m^{-3}. Note that in the model Q_L and C_L represent grid layer average values. As the depth of a layer increases there is a greater risk that input values of Q_L will decrease as cloud layers are diluted with drier layers from above and below. Thus, the probability of identifying resolved cloud layers decreases inversely with vertical layer depth.

Next CMAQ calls module RADMCLD to diagnose the presence of subgrid-scale clouds. This module was originally designed for use in the Regional Acid Deposition Model, or RADM (National Acid Precipitation Assessment Program, 1990a; Dennis *et al*., 1993). Dennis *et al*. modified the subgrid-scale module so that it would diagnose the presence of non-precipitating clouds. Their work was motivated by an underestimation of cloud cover and a significant sulfate underestimation bias in RADM. These 1993 modifications improved model performance, including a reduction in sulfate bias.

RADMCLD does not appear to have changed much since its description by Dennis *et al.* (1993). The module first determines the presence of subgrid-scale precipitating convective clouds (RW). Their fractional coverage is based on a parameterization by Kuo (1974). The fractional coverage of RW clouds, denoted f_c(RW), is determined as that value required to balance the relation

$$f_c(RW)M_{cloud} = F_{mass}M_{subcloud}\ , \qquad (1)$$

where M_{cloud} is the total mass per unit area of air below cloud base, $M_{subcloud}$ is the total mass per unit area of air within the cloud, and F_{mass} (set to 0.5 in the model) is the amount of air below cloud base allowed to be convected upward into the precipitating cloud. Cloud base height is computed as the bottom of the model layer containing the lifted condensation level (LCL). The LCL determines which layers are used to compute $M_{subcloud}$. M_{cloud} is determined from cloud base up to the lower of (a) the layer in which the buoyant cloud parcel would lose buoyancy, (b) the layer containing the 60 kPa (600 mb) pressure level or (c) the first layer encountered with relative humidity below 65%. The value of f_c(RW) varies inversely with cloud depth.

An upper limit to non-precipitating (NP) cloud cover, f_c(NP), is based on (1) with F_{mass} set to 0.5 for NP clouds in cells without precipitation. In cells with precipitation, F_{mass} for NP clouds is limited so that they contain no more than 90% of the below-cloud air that is not convected into precipitating clouds. In practice, a large grid covering the eastern U. S. will have most cells free of precipitation for any given hour. Tests of CMAQ with a variety of summer meteorological conditions revealed that the upper limit to cloud fraction represented by f_c(NP) as defined above was rarely reached. In these cases, when the ambient saturation ratio, S, is at least 0.7 ($S=e/e_s$, e and e_s being the vapor pressure and saturation vapor pressure for water) then f_c(NP) is determined from

$$f_c(NP) = 0.9\left(\frac{S - 0.7}{0.9 - 0.7}\right),\ 0.7 < S < 0.9$$
$$= 0.9,\ S \geq 0.9\,. \qquad (2)$$

Otherwise, f_c=0 for S<0.7. In CMAQ, S=S_o, the latter being the saturation ratio of the "source" layer (see Section 4).

CMAQ puts various limits on subgrid-scale cloud formation:

- Clouds are not allowed in the first layer above the surface (i.e., ground fog is ignored).
- RW clouds are allowed only when convective precipitation is ≥0.1 mm h^{-1}.
- NP clouds in cells without precipitation may not exist if cloud base exceeds 1500 m.
- NP clouds in cells with precipitation may not exist if cloud base exceeds 3000 m.
- Cloud top never extends above 50 kPa or the height of any layer with S<0.65.
- Tops of NP clouds in cells without precipitation are not allowed to exceed the top of the layer containing the 1500 m height level.

4. MODEL SIMULATIONS

Meteorological conditions were simulated by the meteorological model MM5 (Grell *et al.*, 1995). MM5 performance was evaluated against observed surface wind speed, temperature (T), and water vapor mixing ratio (Q). Some biases were identified and adjustments were made to minimize biases in all three variables and lessen the impact of MM5 performance on CMAQ clouds.

MM5 fields were input to CMAQ using standard Models-3 protocols and software. All CMAQ simulations were made with an added capability for writing cloud fields [resolved cloud presence, subgrid-scale f_c(RW) and f_c(NP), and cloud base and top heights] to a special file. Cloud cover at selected NWS stations was compared directly with CMAQ f_c for grid cells containing the station.

Although all MM5 simulations were made with a 31-layer vertical grid structure, CMAQ simulations examined grid structures having 7- and 29-layer configurations. The 7-layer configuration mirrors grid layering typical of 1990s applications whereas the 29-layer grid is more typical of current advanced modeling practice (15-20 layers are now typical for most regulatory applications).

In the CMAQ RADMCLD module, the source layer is the layer nearest the ground in which originates a perturbed air parcel that is convectively buoyant when compared to higher layers. The source layer is usually the first or second layer above the surface. An air parcel lifted from the source layer rises until it reaches the LCL which determines cloud base. The saturation ratio of the source layer, S_o, is used in CMAQ to determine f_c for subgrid-scale clouds as determined in (2). Newer models (for example, Collins *et al.*, 2004) use S from different layers—such as the layers in which a cloud forms—to estimate f_c.

Diagnosing the presence of subgrid-scale convection requires RADMCLD to "initiate" convection by computing the potential buoyancy of locally perturbed air parcels. This is done by introducing perturbation temperature (T') and mixing ratio (Q') values that are added to the state variables, T and Q, in each layer. Air parcels with conditions of T+T' and Q+Q' are evaluated for convective instability. The magnitude of the perturbed variables determines the presence of conditional instability, the LCL and the source layer. These conditions all influence diagnosed f_c.

Various CMAQ simulations were made to test the influence of different parameterizations. Tests were done to examine model sensitivity to

- layer structure,
- limits on cloud base and top heights,
- S_o,
- T' and Q',
- Q_L/C_L thresholds for resolved clouds.

Other cloud related parameters were also tested but model results were generally not sufficiently sensitive to warrant additional investigation.

Simulations were made for four evaluation periods characterized by a variety of meteorological conditions. The periods were 26 April-3 May 1995 (Apr-95), 24-29 June 1992 (Jun-92), 11-19 July 1995 (Jul-95) and 3-11 August 1993 (Aug-93). These dates do not include model start-up days that preceded the evaluation periods.

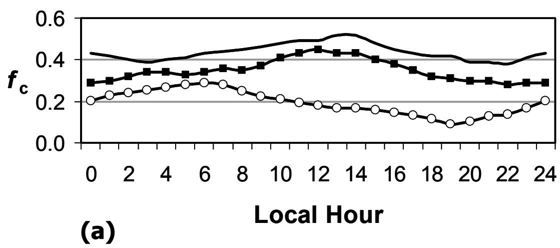
5. RESULTS

CMAQ performance was evaluated using observed f_c along with data on ambient SO_2 and sulfate. Ambient ground-level sulfur data were obtained from the CASTNet (U. S. EPA, 1998; Baumgardner *et al*., 1999) and IMPROVE (Malm *et al*., 1994) archives. CASTNet data include weekly average SO_2 and sulfate concentrations. IMPROVE data include 24-h sulfate concentrations measured twice each week.

5.1 *Clouds*

A large difference was found in f_c for 7- and 29-layer simulations (Figure 3). Results produced with only 7 layers (dubbed CMAQ-7 runs) fell well short of reproducing the observed amount of cloud cover. The modeled short fall in f_c was found across all hours and episodes. In addition, the peak in the f_c diurnal pattern occurred at 0600 local time instead of in the afternoon as was observed. CMAQ-7 runs produced only about a third of the cloud cover observed for hours between noon and 1800.

CMAQ-29 (29-layer) results, in contrast, produced a more realistic f_c diurnal pattern and overall more cloud cover than did CMAQ-7 runs, although average cloud cover was still less than observed. Despite the improvement, CMAQ-29 continued to seriously overestimate the occurrence of clear hours (compare Figures 2b and 3b) while somewhat underestimating the frequency of overcast hours. CMAQ primarily failed to diagnose "partly cloudy" conditions in place of clear conditions. The difference between observed and CMAQ-29 frequencies for f_c=0 was equivalent to about 3 percent for each frequency bin for $0.1 \le f_c \le 0.9$.



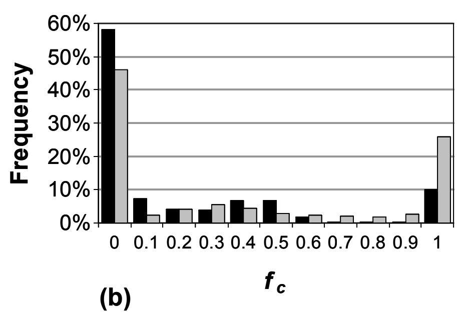


Figure 3. Summary of modeled clouds for all evaluation days: (a) mean observed (solid line, no symbols), CMAQ-7 (open circles), and CMAQ-29 (solid squares) diurnal variation in f_c; (b) f_c frequency distribution for CMAQ-7 (black) and CMAQ-29 (gray) simulations.

Improvement in f_c resulting from an increase in the number of model vertical layers is directly related to the ability of the model to resolve shallow cloud layers. Deep layers dilute simulated (by MM5) cloud decks by averaging Q_L in cloudy and adjacent dry layers. An artifact of modeling with a small number of layers is reflected in the limits in RADMCLD placed on cloud base and top heights. These limits were designed to prevent older models from diagnosing extremely deep cloud layers in place of shallow cumulus layers. The limits also prevented formation of convective clouds with extremely high bases. As the use of more layers becomes commonplace the somewhat arbitrary cloud height limits become an unnecessary and often restrictive artifact.

Test simulations were made that loosened height restrictions for subgrid-scale clouds diagnosed in RADMCLD. This was done by increasing the base height restriction from 1500 m to 2500 m for unstable NP clouds within grid cells having no precipitation. The base height limit on NP clouds in precipitating cells was raised to 3500 m. Significant improvements resulted in CMAQ-7 runs with more afternoon clouds. CMAQ-29 results actually produced too many afternoon clouds. Morning and nighttime cloud cover was still too low in both sets of runs. From this it is seen that the adverse influence of the cloud height restrictions on model performance can be reduced by increasing the number of layers though results can still be unsatisfactory.

Despite changes in layer structure CMAQ still underestimated total cloud cover, overestimated SO_2 and underestimated the fraction of total sulfur in the form of sulfate (R_S) for every period modeled. This was interpreted, in large part, as a failure to adequately simulate the heterogeneous oxidation of SO_2 in clouds. Source layer saturation ratio, S_o, was identified as another source of problems. S_o varies with Q and inversely with T in the source layer. Temperature is usually at a maximum and S_o at a minimum during the afternoon. Tests revealed that this was a primary source of the low bias in afternoon cloud cover. It also contributed to an unrealistically large amount of cloud cover over water surfaces.

In estimating f_c, S_o is meant, in part, to represent the level of moisture in the environment in which clouds form and provide a limit on cloud cover due to lateral dry air entrainment. However, as used in RADMCLD, S_o only represents moisture near the surface and does not represent moisture aloft. A more realistic approach—and one adopted in other models—is to use a value for S_o that represents the layers in which clouds are computed to exist. This finding was the motivation for replacing the diagnostic cloud fraction methodology in RADMCLD with one that uses a different method for estimating S_o and distinguishes between low clouds formed above land and marine surfaces. The alternate approach, described in Collins *et al.* (2004), produced fewer low marine clouds and a slight increase in cloud fraction over land.

T' and Q' are applied to ambient conditions to produce local instability and initiate convection. In RADMCLD, T'=1.5 K and Q'=0.0015. The same values of T' and Q' are used to test for conditional instability in all layers below 65 kPa. CMAQ is fairly insensitive to larger values of T' and Q' up to a point. For T' =4.5 K the model diagnoses more cloud fraction but the increase in f_c occurs during late afternoon and early evening and results in an overestimate of cloud cover during those times. Two changes were tested in the way RADMCLD uses T' and Q'. First, Q' was tied to T' so that the saturation ratio in the perturbed parcel was nearly the same as that in the environment. This avoided simulating a parcel that was dramatically drier or more moist than ambient air. A higher default maximum value (4.5 K) for T' was introduced but values of T' and Q' were required to decrease exponentially with height in the atmosphere to zero above 300 m. In addition, the maximum value of T' was tied to the solar cycle, reaching its greatest value at local noon and decreasing to 0.5 K at night. These changes enabled a stronger coupling in the model between convective cloud formation and daytime heating.

Despite the previously described modifications CMAQ-29 results continued to show a bias in the sulfur balance for the cloudiest evaluation period (Aug-93). Two additional modifications were tested. First, the threshold Q_L used to establish the presence of resolved clouds was lowered from 5×10^{-5} (0.05 g kg^{-1}) to 2.5×10^{-5}. The justification for this was that the MM5 cloud cover, when compared to observations, was actually greater than that estimated in CMAQ and the revised version of CMAQ continued to underestimate cloud cover at night and with large-scale weather systems present during Aug-93. The lower Q_L threshold improved f_c under these conditions.

A second new feature was added to CMAQ to improve performance for daytime, subgrid-scale convective cloud cover. This feature was designed to increase f_c in grid cells where clouds were already present but the coverage was too low. This was a persistent problem, even after all the other changes were implemented, resulting in a significant underestimate in afternoon f_c for some evaluation periods. The new feature is referred to here as “convective enhancement.” It is based on the assumption that some conditionally unstable grid cells fail to experience the expected increase in afternoon convective clouds. This is because the modeled moisture fields do not adequately represent local (subgrid-scale) increases in moisture within active cloud layers due to the cyclic formation and evaporation of convective clouds. Small convective clouds have a limited lifetime of 30 minutes to an hour or so (one hour is the assumed lifetime in CMAQ). After entraining ambient air the clouds evaporate and leave behind their water content, leading to local increases in S_o. This local moistening of the cloud layer is not represented in CMAQ/RADMCLD. An option was added for a local convection enhancement parameter (C_E) that increases the value of S_o used to diagnose f_c, but only in persistent convective cloud layers within a given grid cell. The maximum effective increase in S_o allowed is 0.5. C_E is reset to zero whenever convection ceases to support cloud formation. This additional feature, when tested on the problematic Aug-93 period, increased f_c in a small subset of grid cells. It produced more afternoon cloud cover for Aug-93 and was found to have little effect on the other evaluation periods.

At this point the layer designation is dropped from the model version designation because all further comparisons are done only with the 29-layer configuration. The modified model versions are referred to here as $CMAQ^{+}$ (for the version that included all modifications except C_E) and $CMAQ^{++}$ (with C_E activated). Figure 4 compares the mean observed and modeled diurnal f_c pattern across all simulated days for each model version. The best overall agreement with observations was achieved with the $CMAQ^{++}$ version. Distributions of f_c are compared in Figure 5 for observations and all model runs. Compared to observations, all model runs produced too many grid cells with no clouds, but $CMAQ^{++}$ had the smallest bias.

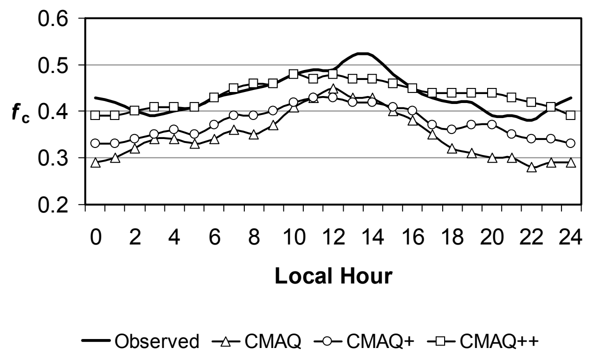


Figure 4. Mean observed and simulated diurnal f_c patterns averaged over all hours and all evaluation days.

CMAQ^{++} had the most cells with overcast skies, but this was offset by its underestimate of cells with $0<f_c<1$. It is obviously difficult for the model to produce partly cloudy conditions, with clear and overcast being the preferred states. However, it is important for the model to produce a reasonable mix of conditions so that the sulfur chemistry is not overly dependent on one or the other SO_2 oxidation mechanisms.

5.2 *Sulfur*

This section summarizes the relationship between simulated cloud cover and sulfur from three versions of CMAQ using the 29-layer configuration. Model performance for atmospheric sulfur is summarized in Table 1. Metric R_S is defined as

$$R_S = \frac{\frac{1}{3} C_{SO4}}{\frac{1}{2} C_{SO2} + \frac{1}{3} C_{SO4}} , \qquad (3)$$

where C_{SO2} and C_{SO4} represent air concentrations of SO_2 and sulfate particles. Only the CASTNet data provide sufficient information to compute this ratio. Thus, R_S represents weekly average ratios for each CASTNet site.

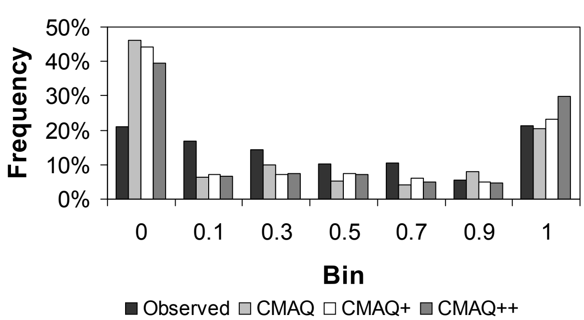


Figure 5. Frequency distributions of observed and simulated f_c. Labels for bins >0 and <1 represent bin midpoint values.

Table 1. Summary of model performance for simulated sulfur across all evaluation periods.

Metric Description[a]	Sulfur Performance Metrics by Model Version		
	CMAQ	CMAQ$^+$	CMAQ^{++}
Median Weekly C_{SO2} Bias	71%	56%	34%
Median Weekly C_{SO4} Bias	2%	-1%	5%
Median Weekly Bias in S_T[b]	40%	27%	23%
Median C_{SO4} Bias (all sites)[c]	-1%	0%	6%
Median Obs. R_S	0.49	0.49	0.49
Median Mdl. R_S / Median Obs. R_S[d]	0.80	0.88	0.98

[a]All biases (average errors) have been normalized by the observed values.
[b]S_T is sum of sulfur in both C_{SO2} and C_{SO4}.
[c]Combines CASTNet and IMPROVE C_{SO4} data, each measurement weighted by number of days in its averaging period.
[d]The ratio of model to observed median R_S, with perfect agreement equal to a value of 1.

Median normalized biases in C_{SO2} and S_T—the denominator in (3)—were consistently positive for all model versions. In contrast, median bias in C_{SO4} was always near or slightly greater than zero. This indicates that the model tended to overestimate SO_2 levels despite fairly good performance for sulfate. A large part of the C_{SO2} bias was apparently due to an underestimate of the oxidation of SO_2 to sulfate because the bias in C_{SO2} decreased as more clouds were diagnosed by the modified models, CMAQ$^+$ and CMAQ^{++}. With little bias in cloud cover, CMAQ^{++} reduced the C_{SO2} bias by half and produced a very good balance between SO_2 and sulfate as indicated by the ratio of median modeled to observed R_S being near unity.

Ordinarily it would be sensible to assume that the CMAQ high bias in S_T would increase the likelihood of the model overestimating sulfur wet deposition. However, comparing CMAQ wet deposition estimates with data from the deposition monitoring network (http://nadp.sws.uiuc.edu/) revealed that CMAQ underestimated sulfur wet deposition by an average 14 percent across the eastern U. S. These estimates were made by interpolating measured sulfur wet deposition data from the NADP (National Acid Deposition Program) monitoring sites to the CASTNet monitoring locations using kriging analysis so that deposition bias is directly comparable to biases in C_{SO2} and C_{SO4}. This finding suggests that the positive biases in C_{SO2} and S_T were caused in part by insufficient wet scavenging in the model. By comparison, wet sulfur deposition bias in CMAQ^{++} results was only 6 percent, indicating that the underestimate of wet scavenging was removed with a more accurate portrayal of cloud cover. In fact, wet scavenging of pollutants by subgrid-scale clouds is computed in RADMCLD. When RADMCLD underestimates the presence of subgrid-scale clouds it apparently can also underestimate wet sulfur deposition. The small CMAQ^{++} positive bias in wet sulfur deposition was likely associated with the remaining 34 percent overestimate in C_{SO2} once cloud cover bias was corrected. Remaining biases in C_{SO2} and S_T could be associated with an underestimate of dry SO_2 deposition, but data are not available to test that hypothesis.

6. CONCLUSIONS

CMAQ treatment of subgrid-scale clouds was based on the methodology used in the Regional Acid Deposition Model (RADM) developed in the 1990s. This methodology has become outdated as computer technology has improved to allow higher vertical definition in the grid structure. Modeling the atmospheric sulfur cycle with too few layers can fail to properly simulate cloud cover and adversely impact estimated levels of SO_2 and sulfate. Merely increasing the number of layers does offer some improvement in cloud cover but does not guarantee better performance for ambient sulfur. Modifications are needed to the CMAQ module that simulates subgrid-scale clouds to ensure that cloud cover is diagnosed more accurately (with the proper diurnal pattern) and to minimize biases in the simulated balance between SO_2 and sulfate particles. The different lifetimes of SO_2 and sulfate influence the estimated impacts attributable to receptors downwind of large sources of SO_2. Thus, it is important to estimate these source impacts with a model that minimizes bias in its simulated balance of atmospheric sulfur.

7. ACKNOWLEDGEMENTS

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8. REFERENCES

Baumgardner, R. E., Jr., S. S. Isil, J. J. Bowser, and K. M. Fitzgerald, 1999: Measurements of rural sulfur dioxide and particle sulfate: analysis of CASTNet data, 1987 through 1996. *J. Air & Waste Manage. Assoc.* **49**, 1266-1279.

Collins, W. D., P. J. Rasch, B. A. Boville, J. J. Hack, J. R. McCaa, D. L. Williamson. J. T. Kiehl, B. Briegleb, C. Bitz, S.-J. Lin, M. Zhang, Y. Dai, 2004: Description of the NCAR Community Atmosphere Model (CAM 3.0). National Center for Atmospheric Research, Boulder, CO. (NCAR/TN-464+STR)

Dennis, R. L., J. N. McHenry, W. R. Barchet, F. S. Binkowski, and D. W. Byun, 1993: Correcting RADM's sulfate underprediction: discovery and correction of model errors and testing the corrections through comparisons against field data. *Atmos. Environ.* **27A**, 975-997.

Grell, G., J. Dudhia, and D. Stauffer, 1995: A Description of the Fifth-Generation Penn State/NCAR Mesoscale Model (MM5), National Center for Atmospheric Research, Boulder, CO. (NCAR/TN-398+STR)

Kuo, H. L., 1974: Further studies of the parameterization of the influence of cumulus convection on large-scale flow. *J. Atmos. Sci.* **31**, 1232-1240.

Malm, W.C., J. F. Sisler, D. Huffman, R. A. Eldred, and T. A. Cahill, 1994: Spatial and seasonal trends in particle concentration and optical extinction in the United States. *J. Geophys. Res.* **99**, 1347-1370.

National Acid Precipitation Assessment Program, 1990: Report 4 - The Regional Acid Deposition Model and Engineering Model. In *Acid Deposition: State of Science and Technology*; Irving, P. M., Ed.; Vol. 1: Emissions, Atmospheric Processes, and Deposition; Office of the Director: Washington, DC.

Seinfeld, J. H., and S. N. Pandis, 1998: *Atmospheric Chemistry and Physics.* John Wiley & Sons, Inc., New York, 363-380.

U. S. EPA, 1998: Clean Air Status and Trends Network (CASTNet) Deposition Summary Report (1987-1995). Washington, DC. (NTIS: 600/R-98/027)

U. S. EPA, 1999: Science Algorithms of the EPA Models-3 Community Multiscale Air Quality (CMAQ) Modeling System, D. W. Byung and J. K. S. Ching, Ed. U. S. Environmental Protection Agency, Office of Research and Development, Washington, DC, 22 pp. (NTIS: 600-R-99-030)