4.5 MODELING THE ROLE OF CLOUDS IN AEROSOL FORMATION

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1. INTRODUCTION

Sulfate aerosol is formed in the atmosphere from SO_2 by gas-phase oxidation involving the hydroxyl radical and from the reaction of SO_2 in aqueous phase with ozone or peroxides. Models that simulate aerosols must consider both formation mechanisms. Realistic modeling of heterogeneous (aqueous) chemistry requires that cloud information be passed from a meteorological model to the chemistry model. This describes the effectiveness and impact of one modeling approach.

2. MODELS AND ANALYSIS

The RAMS model, version 3a (Pielke *et. al*., 1992), simulated meteorological conditions during multi-day episodes. RAMS was run with a modified Kuo convective parameterization scheme. The following meteorological variables affected the treatment of clouds and precipitation scavenging in the chemical model: relative spatial coverage by convective clouds, fraction of precipitation from convection, convective cloud top height, stratiform cloud top height, total precipitation, temperature and water vapor mixing ratio. RAMS simulations were made using a series of nested grids. The finest grid, 12 km, was centered over the southern Appalachians.

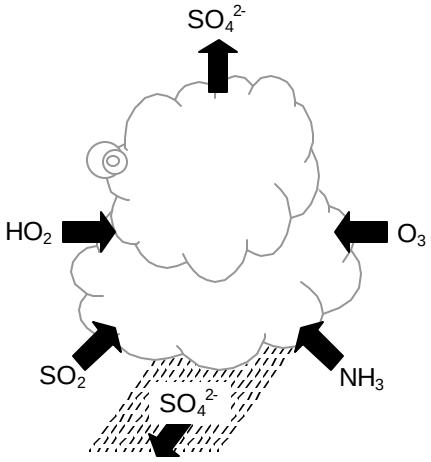

Figure 1. Clouds are chemical reactors that ingest SO_2(g) and other gases and particles. Sulfate forms within cloud and rain drops at a rate that depends on pH and the amount of peroxides and ozone present. Precipitation removes some sulfate while evaporation leaves behind additional aerosol.

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The URM-1ATM air quality model (Boylan *et. al*., 2001) simulated atmospheric chemistry and pollutant removal processes. This multi-scale, multi-pollutant model treats photochemistry, heterogeneous chemistry, pollutant scavenging by precipitation, and aerosol formation and growth. Heterogeneous chemistry and scavenging are treated by the Reactive Scavenging Module, RSM (Berkowitz *et. al*., 1989).

The role of clouds in aerosol chemistry is illustrated in Figure 1. The RSM computes the equilibria between gas- and aqueous-phase species, computes the chemical kinetic reactions that produce sulfates, vertically redistributes pollutant mass due to convective transport, and calculates the wet deposition of soluble species. In addition, cloud evaporation leaves behind suspended aerosols. The same set of chemical kinetic equations is used elsewhere in URM-1ATM to simulate heterogeneous reactions in non-precipitating clouds and hygroscopic aerosols. These conditions are diagnosed by the model based on local relative humidity. Thus, the model treats precipitating and non-precipitating clouds separately though similarly.

A series of URM-1ATM runs was made for each of 4 multi-day episodes. Of these, 3 involved significant periods of cloud cover and rainfall over the southern Appalachian region. Cloud observations made at 14

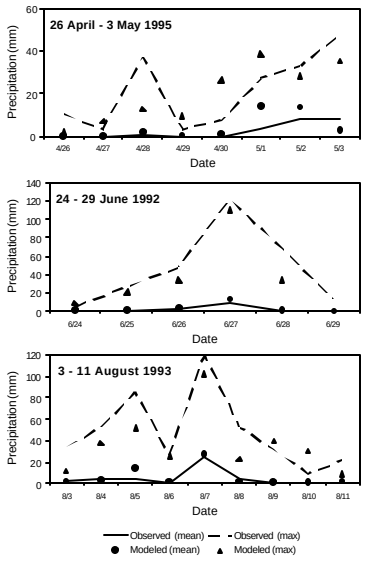

Figure 2. Time series of 24-h mean and maximum precipitation (observed and modeled) on the 12-km grid cell region for three episodes.

National Weather Service (NWS) stations within the 12-km grid cell region were used to evaluate simulated cloud cover and cloud ceiling height. Precipitation data provided a benchmark for how well modeling reproduced rainfall.

Time series of 24-h precipitation on the 12-km grid cells (Figure 2) show large differences in rainfall characteristics across the 3 rainy episodes. The April-May 1995 episode had the least precipitation and the poorest model performance. Figure 3 plots time series of fractional cloud cover (FCC) on the 12-km grid cells for the same episodes. Again, model performance was weakest for the April-May episode. Frequencies of observed and modeled daytime (0900-1800 LST) cloud ceilings (FCC>0.5) within each model layer are shown in Figure 4. Frequencies of periods without ceilings are well represented by the modeling. The model does indicate higher frequencies of ceilings in the lowest three layers that fall below 500 m above the surface. This is caused by the cloud diagnostic criterion in the model which selects 90% relative humidity as the lower threshold for activating heterogeneous chemistry in hygroscopic aerosols. Hence, the criterion used for cloud presence as it relates to atmospheric chemistry does not conform to meteorological convention.

3. CONCLUSIONS

Cloud treatment in the URM-1ATM model appears reasonable. Accuracy problems were most notable for one episode. The need to simulate heterogeneous chemistry requires a criterion for determining cloud presence that does not conform to meteorological convention. Despite this the model representation of cloud presence and cloud amount matched observations well. The role of heterogeneous chemistry varied greatly according to the meteorological circumstances. Heterogeneous oxidation produced less than a quarter of the sulfate during a rainy summer period, but two-thirds of the sulfate during an extremely dry summer period. Regions modeled to have low sulfate levels were less likely to have active heterogeneous chemistry than areas computed to have high sulfate. Overall, heterogeneous sulfate formation accounted for about half of the total sulfate produced.

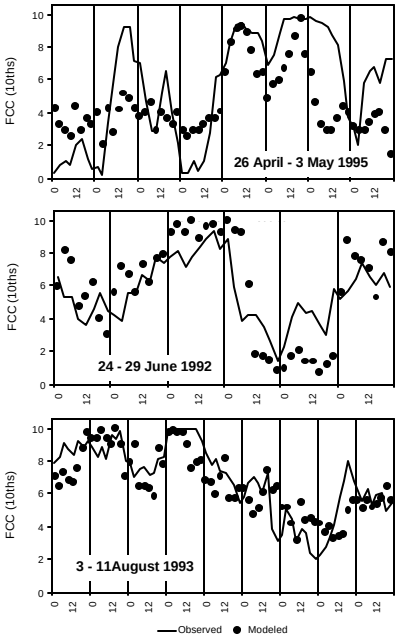

Figure 3. Time series of modeled and observed fractional cloud cover averaged for 12-km grid cells.

4. ACKNOWLEDGEMENTS

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5. REFERENCES

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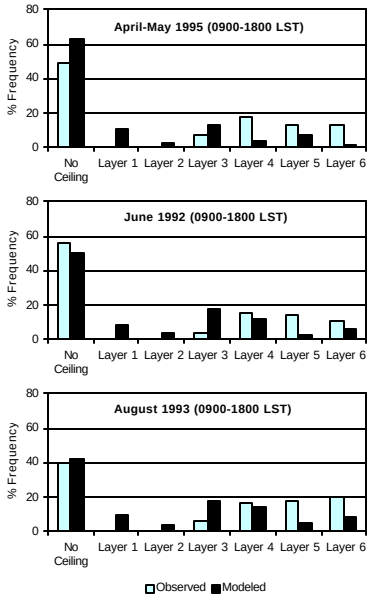

Figure 4. Frequencies of observed and modeled daytime cloud ceilings for 12-km grid cells.