# P1.14 AEROSOL-CLOUD-CHEMISTRY INTERACTIONS IN A LARGE EDDY SIMULATION

Graham Feingold[1] and Sonia M. Kreidenweis[2]

[1] NOAA Environmental Technology Laboratory, 325 Broadway, Boulder CO 80305

[2] Department of Atmospheric Science, Colorado State University, Fort Collins, Colorado 80523

## 1. INTRODUCTION

When assessing the aerosol indirect effect it is important to bear in mind that aerosol particles not only modify cloud drop size distributions but that clouds also modify aerosol size distributions. This occurs through numerous mechanisms including the addition of nonvolatile sulfate mass to aerosol particles via aqueous chemistry (e.g., Hegg and Hobbs, 1982; Hoppel et al., 1990; Bott, 1999; Feingold et al., 1998; Wurzler et al., 2000), drop collection (Hudson, 1993; Flossmann, 1994; Feingold et al., 1996), precipitation scavenging, and homogeneous nucleation of new particles in the vicinity of clouds. The two-way feedback between aerosol and cloud has implications for associated climate feedbacks. For example, aqueous addition of sulfate to aerosol particles means that aerosol size spectra can change the drop size distribution in subsequent cloud cycles, and either increase or decrease drop number concentrations (e.g., Bower et al., 1997; Feingold and Kreidenweis, 2000).

Here we explore cloud processing of aerosol through aqueous chemistry, collection and sedimentation in a large eddy simulation (LES). In particular we show how aqueous chemistry can either increase or decrease drizzle in a stratocumulus cloud and explore the conditions under which each response occurs. Details of this work are in Feingold and Kreidenweis (2002).

## 2. THE LES MODEL

The model is described in Feingold and Kreidenweis (2002). It uses the Regional Atmospheric Modeling system (RAMS) in its LES configuration as a dynamical framework. It incorporates coupled components of aerosol and drop size distributions (Feingold et al. 1996), a radiative transfer model (Harrington et al. 1999), as well as aqueous sulfate chemistry. The model carries prognostic variables for $SO_2$, $O_3$, $H_2O_2$, $NH_3$, and $HNO_3$, as well as their soluble counterparts. Diffusion-limited dissolution of the gases into drops is calculated using an ordinary differential equation solver. Oxidation of dissolved S(IV) occurs via $O_3$ and $H_2O_2$. Gas-phase chemistry is not simulated. The input aerosol is assumed to be a completely soluble ammonium sulfate aerosol, defined by a lognormal function resolved into 14 size bins. Aqueous chemistry is simulated in a cloud drop size class, and a drizzle size class. Off line simulations indicate that for the conditions simulated, these two size classes are adequate. In each of the size classes, we track the soluble gases and the ammonium and sulfate ions, their transfer to drizzle drops due to drop growth, and sedimentation to the surface. On complete evaporation, dissolved sulfate is returned to the aerosol population as described in Feingold et al. (1996). The initial sounding is based on one from the ASTEX first Lagrangian experiment. Further details can be found in Feingold and Kreidenweis (2002).

## 3. RESULTS

### 3.1 Sample of LES output

Two simulations are performed for an input CCN concentration of 100 $\mathrm{cm}^{-3}$, $r_g = 0.1$ $\mu$m, and $\sigma_g = 1.5$; one simulation does not include chemistry whereas the other does. Figures 1, 2, and 3 illustrate some of the model output with snapshots of various fields 1 h after initialization for the case that includes chemistry. The maximum cloud water mixing ratio $r_l$ is about 0.7 g $\mathrm{kg}^{-1}$. Soluble gases are depleted from their initial conditions to a degree consistent with individual parcel model calculations (Feingold et al., 1998) for clouds of similar average liquid-water content. Significant structure exists in these fields at scales of a few hundred meters; these structures are associated with similar structures in microphysical and dynamical fields. Droplet pH ranges from 2.5 near the cloud base where cloud water mixing ratio $r_l$ is minimal to 4.7 near the cloud top where most of the cloud water resides. In general, pH contours parallel $r_l$ contours with some exceptions (Feingold and Kreidenweis, 2002)

From the perspective of aerosol-cloud interactions, the effect of aqueous chemistry on the CCN size distribution is of interest. Figure 4 illustrates the total CCN mass for the time period corresponding to Figure 1. The increase in CCN mass in the sub-cloud air is significant ($\simeq$ 70%) so that this case can be viewed as one with strong aqueous sulfate production. Examination of the drizzle produced by this cloud shows (Figure 5) that for $N_{\mathrm{ccn}}$ =100 $\mathrm{cm}^{-3}$, there is virtually no effect of aqueous chemistry on drizzle. This is because there is already an active drizzle process for these conditions and aqueous production of sulfate has little effect.

---

Corresponding author address: Graham Feingold, NOAA ETL, 325 Broadway, Boulder, CO 80305; e-mail: graham.feingold@noaa.gov

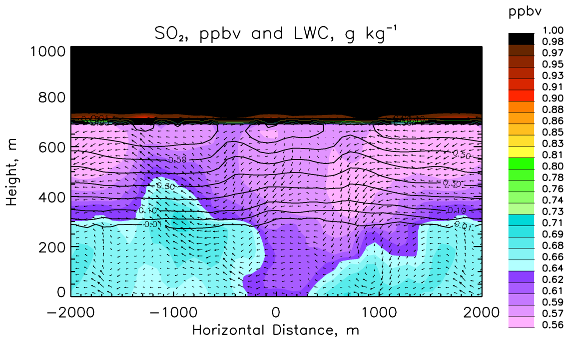

Figure 1: *Gas phase $SO_2$ (color flooded contours) superimposed on cloud water (contour lines) and vectors showing air motions.*

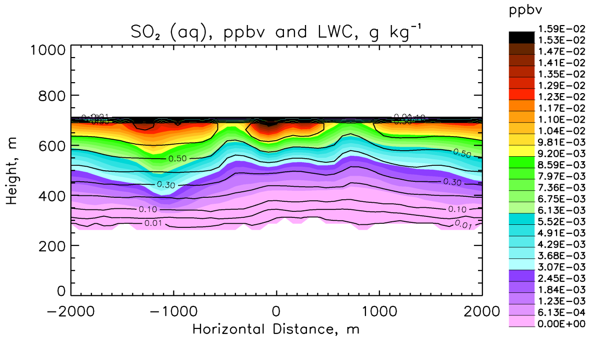

Figure 2: *As in Figure 1 but for dissolved $SO_2$ species.*

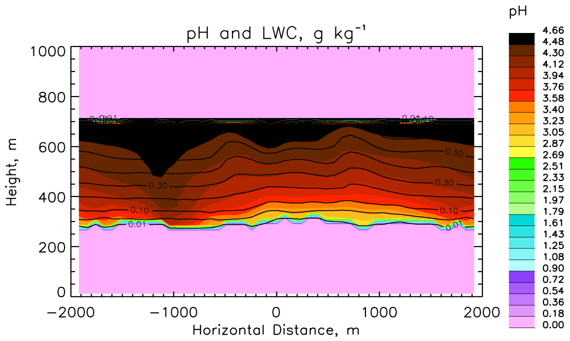

Figure 3: *As in Figure 1 but for droplet pH.*

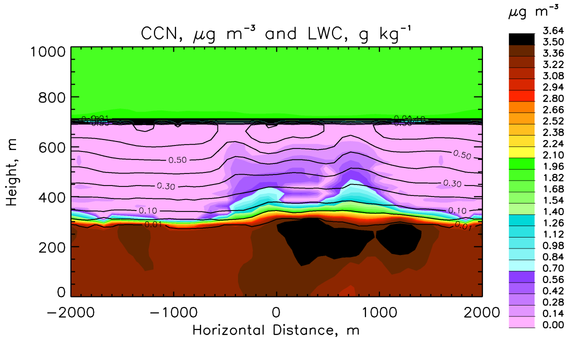

Figure 4: *As in Figure 1 but for CCN mass. Note the significant increase in sub-cloud CCN mass due to aqueous conversion.*

### 3.2 Two-dimensional eddy resolving model (ERM) simulations

3.2.1 Large median radius: $r_g$=0.1 $\mu$m, $\sigma_g$=1.5

Given the large computational burden of LES, we turn to 2-d ERM simulations to enable exploration of a broader parameter space. Simulations shown in section 3.1 are repeated for the range of CCN concentrations $N_{\rm ccn}$ from 50 cm$^{-3}$ to 200 cm$^{-3}$ with $r_g$ =0.1 $\mu$m and $\sigma_g$ =1.5, although only select results are shown. Simulations are terminated when the LWP is depleted by about 50 % of the maximum value; therefore cases with higher $N_{\rm ccn}$ are run for significantly longer than those with lower $N_{\rm ccn}$. Time series similar to Figure 5 confirm that, as in the case of the 3-d simulations, aqueous chemistry does not result in a significant change in these fields when $N_{\rm ccn}$ = 100 cm$^{-3}$. When $N_{\rm ccn}$ =150 cm$^{-3}$, a number of trends are noteworthy (Figure 6). Measurable surface precipitation takes much longer to develop and occurs only when the cloud LWP has reached about 300 g m$^{-2}$. This occurs when cloud top $r_l \simeq 1$ g kg$^{-1}$and the cloud top effective radius $r_e$ =14 $\mu$m. There is a clear trend for aqueous chemistry to enhance LWP and suppress precipitation: after 8 h $I$ is 28 % lower in the case with aqueous chemistry.

3.2.2 Small median radius: $r_g$=0.05 $\mu$m, $\sigma_g$=1.8

Because previous simulations were for a relatively large median radius of 0.1 $\mu$m, the second aerosol mode created by the addition of sulfate mass, which usually resides at a similar radius (Hoppel et al., 1990) will not be separated substantially from the original aerosol mode. A subset of simulations is now repeated for $r_g$ = 0.05 $\mu$m and $\sigma_g$ = 1.8. Results for $N_{\rm ccn}$ = 100 cm$^{-3}$ (not shown) again indicate only slight differences in precipitation between the two simulations. Figure 7

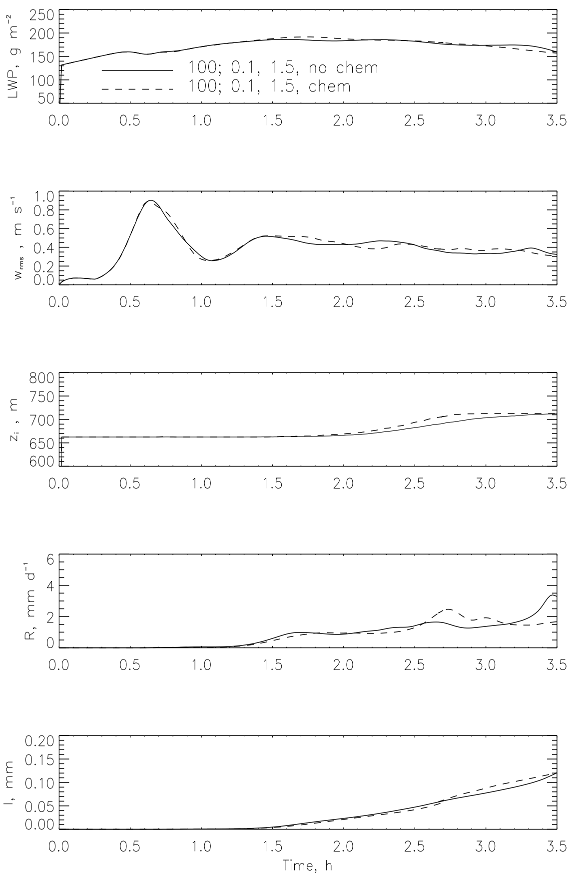

Figure 5: *Time series of various fields for LES runs with and without aqueous chemistry (a) Liquid water path LWP, (b) maximum root-mean-square vertical velocity $w_{\rm rms}$, (c) boundary layer top $z_i$, (d) surface rain rate R, and (e) accumulated surface precipitation I.*

shows time series of key fields for $N_{\rm ccn}$ = 150 cm$^{-3}$. In contrast to Figure 6, where precipitation was suppressed by aqueous chemistry, here precipitation is increased by as much as 37 % after 8 h.

## 4. DISCUSSION

The results suggest that even for a case with rather strong aqueous production of sulfate, the effects on boundary layer dynamics, cloud microphysics, and precipitation are likely to be felt over a limited range of conditions. For the simulation presented here, in which $N_{\rm ccn}$ is relatively small ($<$ 100 cm$^{-3}$), the collection process is quite efficient, and aqueous production of sulfate does not appear to be able to modify this process.

The simulations indicate that when $N_{\rm ccn} >$ 150 cm$^{-3}$ and the CCN size distribution has a relatively large median size, the addition of sulfate suppresses precipitation. Under these conditions, the contrast between the mode of the original CCN size distribution and the mode associated with the mass addition is small, and aqueous chemistry does not change the size of particles sufficiently to generate a significant number of larger drops. In this case, the net effect of aqueous chemistry is to increase the number of cloud drops. When the CCN size distribution has a relatively small median size, the contrast is much larger, and the particles that are produced are able to produce higher concentrations of larger drops (relative to the initial size distribution) that help initiate collection.

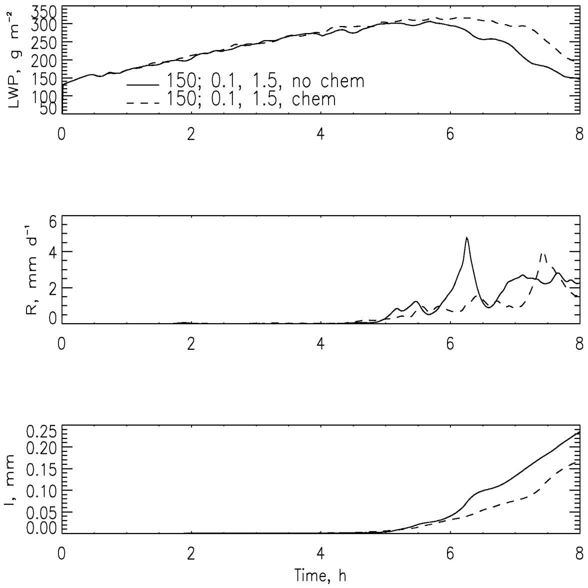

Figure 6: *As in Figure 5 but for ERM runs and $N_{\rm ccn}$ =150 cm$^{-3}$.*

Illustrations of the effect on aerosol size spectra $n(r)$ are shown in Figs. 8 and 9; processed CCN size distributions as well as the background (initial) distributions are displayed for the $N_{\rm ccn}$ = 150 cm$^{-3}$ cases and ($r_g$ ; $\sigma_g$ ) = either (0.1 $\mu$m; 1.5) or (0.05 $\mu$m; 1.8). The processed size spectra are horizontally averaged at a height of 300 m and at a simulation time of 2 h. The larger % increase in mean radius for the (0.05 $\mu$m; 1.8) case is again apparent. Although the size spectra lack detail, they do show the creation of a mode at $\simeq$ 0.1 $\mu$m to 0.2 $\mu$m, and do represent the feedback of aqueous chemistry processing to the microphysical/dynamical model.

## 4. SUMMARY

A large eddy simulation of the marine boundary layer that includes size-resolved treatment of aerosol and cloud drop size distributions, and a coupled aqueous chemistry component has been presented. The model has been used to simulate a stratocumulus cloud deck from a case study based on the ASTEX first Lagrangian experiment. Twin LESs are performed for a relatively low CCN concentration of 100 cm$^{-3}$. In one case, aqueous

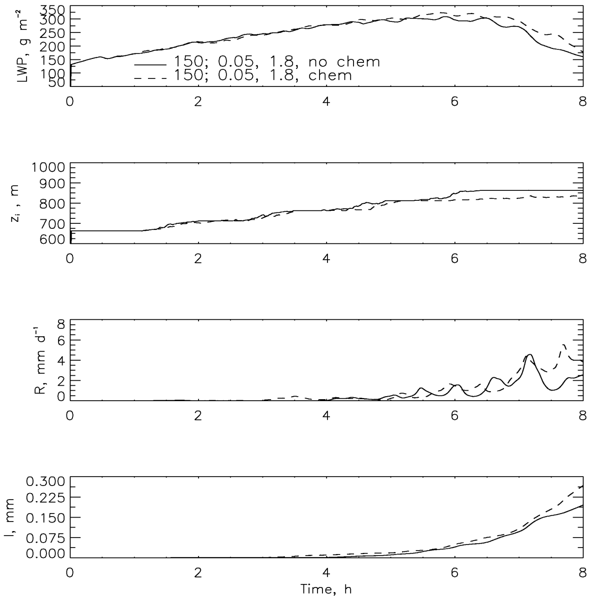

Figure 7: *As in Figure 5 but for ERM runs and* $N_{\rm ccn}$ *=150 cm*$^{-3}$, $r_g$ *=0.05 μm, and* $\sigma_g$ *=1.8.*

chemistry is included, whereas in the second it is not. For the conditions studied here, although aqueous chemistry produces a higher sulfate mass, it is not sufficient to modify the amount of precipitation produced by the cloud, apparently because at low CCN concentrations there is a fairly active collection process. However, a series of 2-d simulations for a wider range of CCN concentrations suggests that aqueous chemistry may either enhance or suppress drizzle. Precipitation may be enhanced when the initial CCN size distribution has a relatively small mode. In this case, the larger particles produced by aqueous chemistry (about 0.1 μm radius) are more easily activated, tend to generate larger drops, and therefore enhance drizzle production. Suppression of drizzle occurs when the input CCN size distribution has a larger mode that is similar to that at which sulfate is produced by clouds. In this case, the added sulfate mass translates to higher drop concentrations, smaller drops, and reduced collection.

Although the simulations presented here cover only a handful of scenarios and only one set of aqueous chemistry conditions, the study has demonstrated that aqueous chemistry may, under some conditions, modify drop concentrations, cloud optical properties, and drizzle formation in stratocumulus. It suggests that the myriad feedbacks that exist in the aerosol-cloud-chemistry system present a formidable challenge to our attempts to quantify the aerosol indirect effect. Future work will explore these feedbacks in greater detail.

## ACKNOWLEDGMENTS

Sonia Kreidenweis acknowledges support from NOAA/OGP under grant # NA17RJ1228.

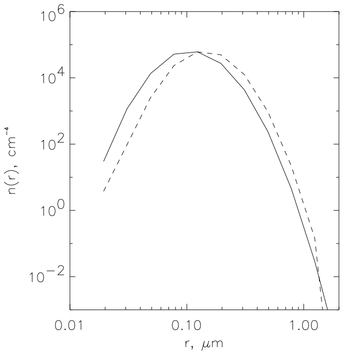

Figure 8: Processed (solid) and unprocessed (dashed) CCN size distributions for $N_{\rm ccn}$ =150 cm$^{-3}$, $r_g$ =0.1 μm, and $\sigma_g$ =1.5, i.e., the case when drizzle is reduced by aqueous chemsitry.

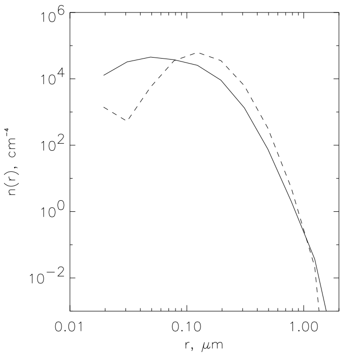

Figure 9: *As in Figure 8 but for* $N_{\rm ccn}$ *=150 cm*$^{-3}$, $r_g$ *=0.05 μm, and* $\sigma_g$ *=1.8, i.e., the case when drizzle is increased by aqueous chemistry.*

## REFERENCES

Bott, A., 1999: A numerical model of the cloud-topped planetary boundary-layer: chemistry in marine stratus and the effects on aerosol particles. *Atmos. Environ.,* **33,** 1921–1936.

Bower, K. N., and T. W. Choularton, 1993: Cloud processing of the cloud condensation nucleus spectrum and its climatological consequences. *Quart. J. R. Meteor. Soc.*, **119,** 655–679.

Bower, K. N., and 26 coauthors, 1997: Observations and modelling of the processing of aerosol by a hill cap cloud. *Atmos. Environ.,* **31,** 2527–2543.

Feingold, G., S. M. Kreidenweis, B. Stevens, and W. R. Cotton, 1996: Numerical simulation of stratocumulus processing of cloud condensation nuclei through collision-coalescence. *J. Geophys. Res,* **101,** 21,391–21,402.

Feingold, G., S. M. Kreidenweis, and Y. Zhang, 1998: Stratocumulus processing of gases and cloud condensation nuclei: Part I: trajectory ensemble model. *J. Geophys. Res.,* **103,** 19,527–19,542.

Feingold, G., and S. M. Kreidenweis, 2000: Does heterogeneous processing of aerosol increase the number of cloud droplets? *J. Geophys. Res.,* **105,** 24,351–24,361.

Feingold, G., and S. M. Kreidenweis, 2002: Cloud processing of aerosol as modeled by a large eddy simulation with coupled microphysics and aqueous chemistry. In Press, *J. Geophys. Res.*

Flossmann, A. I., 1994: A 2-D spectral model simulation of the scavenging of gaseous and particulate sulfate by a warm marine cloud. *Atmos. Res.,* **32,** 233–248.

Harrington, J. Y., T. Reisin, W. R. Cotton, and S. M. Kreidenweis, 1999: Cloud Resolving Simulations of Arctic Stratus. Part II: Transition-Season Clouds.*Atmos. Res.,* **51,** 45–75.

Hegg, D. A., and P. V. Hobbs, 1982: Measurements of sulfate production in natural clouds. *Atmos. Environ.*, **16** , 2663–2668.

Hoppel, W. A., J. W. Fitzgerald, G. M. Frick, and R. E. Larson, 1990: Aerosol size distributions and optical properties found in the marine boundary layer over the Atlantic ocean. *J. Geophys. Res.,* **95,** 3659–3686.

Hudson, J. G., 1993: Cloud condensation nuclei near marine cumulus. *J. Geophys. Res.,* **98,**2693–2702.

Wurzler, S., T. G. Reisin, and Z. Levin, 2000: Modification of mineral dust particles by cloud processing and subsequent effects on drop size distributions, *J. Geophys. Res.,* **105,** 4501–4512.