

Cloud Processing of Aerosols and Their Effects on Aerosol Radiative Properties

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Introduction

The scavenging of aerosols by clouds and their removal from the atmosphere by precipitation are important sinks for atmospheric aerosols. It is estimated that, on the global scale, precipitation removes about 80% of the mass of aerosols from the atmosphere, and gravitational settling (dry fallout) accounts for the rest (Wallace and Hobbs 1977). Before aerosols are removed by precipitation, their parameters are substantially modified by cloud processing. According to Pruppacher and Klett (1978), the cloud materials, on a global scale, go through about ten condensation-evaporation cycles before the cloud eventually precipitates. The effect of the repeated cycling of cloudy air is the decrease of small aerosol particles and the increase of aerosol mass at sizes larger than 0.1 micron. The cloud recycling also produces a minimum at a size determined by maximum supersaturation in cloud. The increased mass of larger particles is the result not only of coalescence of cloud drops, but also Brownian scavenging of interstitial aerosol, as well as gas-to-particle conversion inside cloud drops.

Cloud processing of aerosols has been studied by Hudson and Frisbie (1991) and Hudson (1993) using extensive airborne measurements of cloud condensation nucleus (CCN) spectra and concentrations of total particles during the First ISCCP^(a) Regional Experiment (FIRE) and Hawaiian Rainband Project (HaRP) experiments. The FIRE observations carried out over the 4-day period showed the decrease in the boundary-layer aerosol concentration consistent with the in-cloud scavenging process. Hudson and Frisbie (1991) emphasize the importance of “coalescence scavenging.” It reduces the CCN concentration according to the number of cloud droplet captures, as well as transforms the CCN size distribution. The more active nuclei (corresponding to lower critical supersaturations) are not only preferentially used to form cloud droplets, but also increase in size due to the recycling that follows coalescence and evaporation of cloud droplets.

The efficiency of the in-cloud scavenging processes can be estimated from the measured reduction of CCN concentration in the boundary layer below the inversion and by comparing the CCN activation spectra in a cloud-free air with those in the air within and around the clouds. Observations by Hudson (1993) confirm that concentrations of CCN are often lower within the boundary layer, and they are also composed of larger particles with lower median critical supersaturation. Under conditions with few or no clouds, the spectra below and above the temperature inversion are similar.

In non-precipitating clouds, the decrease of aerosol particle concentration due to drop coalescence will lead to reduction of the total aerosol surface area, thus influencing the aerosol extinction coefficient which is defined as

$$\beta_e = \int_0^\infty Q_e(r) \pi r^2 n(r) dr$$

where Q_e is the extinction efficiency factor, r is the aerosol radius, $n(r)$ is the aerosol number distribution.

In the present paper, we study the change of the aerosol radiative properties due to cloud processing of aerosols (coalescence scavenging only) using a new version of the Cooperative Institute for Mesoscale Meteorological Studies' (CIMMS) large-eddy simulation (LES) model that allows us to track CCN particles inside the cloud.

CIMMS LES Cloud Model

The physical formulation of the CIMMS LES cloud model is described in Kogan et al. (1995). The current version of the model employs a new dynamical framework based on the finite difference discretization. The advective transport of momentum is computed using a flux-conserving advection scheme analogous to Tremback's (Tremback et al. 1987) with an option to select the order of spatial accuracy from the second to the fifth. The time integration is performed using

(a) International Satellite Cloud Climatology Project

the third-order Adams-Bashforth (AB) scheme. The numerical scheme is described in detail in Khairoutdinov and Kogan (1997).

The numerical and physical formulation of microphysics is described in Kogan et al. (1995). The recent improvement in numerical formulation is the implementation of the new variational minimization method for remapping drop spectra during condensation/evaporation calculations. The method (Liu et al. 1995; Liu et al. 1996) conserves four moments of the drop size distribution function and significantly minimizes the numerical diffusion of the drop spectra without sacrificing the computational efficiency of the code.

For the aerosol processing study, the microphysical formulation has been significantly modified by introducing a two-dimensional Drop and Salt Size Distribution (DSSD) function, $N(m,n)$, which allows us to follow the salt (CCN) particles transformations as they are processed by cloud drops. The function $N(m,n)$ represents drop mass distribution such that $N(m,n)dmdn$ is the number of particles per unit volume of air in the drop mass range from m to $m+dm$ and the salt mass range from n to $n+dn$. Let N_{ij} denote the DSSD function in the discrete form with the indices i and j referring to the drop and the CCN mass categories, respectively. The governing equation for the 2D drop spectrum is Houze (1993):

$$\frac{\partial N_{ij}}{\partial t} + \frac{\partial}{\partial x_k} (u_k N_{ij}) = R_{ij} + D_{ij} + C_{ij} + S_{ij} + \frac{\partial}{\partial x_k} (K_m \frac{\partial N_{ij}}{\partial x_k})$$

The changes in the concentration N_{ij} are the result of nucleation (R_{ij}), vapor diffusion (D_{ij}), collection (C_{ij}), sedimentation (S_{ij}), and turbulent diffusion.

For the collection term (C_{ij}), we assume that the collection kernel depends only on the cloud drop mass, and the nuclei are coalesced whenever two parent drops coalesce. The cloud droplet mass m and the nuclear mass n of the newly formed drop are determined by adding the m 's and n 's of the coalescing drops, respectively. Therefore, the stochastic collection equation can be written as

$$C_{ij} = \frac{1}{2} \int_0^{m_i} \int_0^{n_j} W(m_i - m', m') N(m_i - m', n_j - n') N(m', n') dn' dm' - \int_0^\infty \int_0^\infty W(m_i, m') N(m_i, n_j) N(m', n') dn' dm'$$

where $W(m, m')$ is the collection kernel defined in Kogan (1991).

In addition to drop particles, we also account for CCN particles which do not form cloud drops. We specify the mass distribution function $f(n)$ such that $f(n)dn$ is the number of CCN particles per unit volume of air in the mass range between n and $n+dn$. The governing equation for $f(n)$ (f_j in the discrete form) is

$$\frac{\partial f_j}{\partial t} + \frac{\partial}{\partial x_k} (u_k f_j) = - \sum_i R_{ij} + \sum_i E_{ij} + \frac{\partial}{\partial x_k} (K_m \frac{\partial f_j}{\partial x_k})$$

The right hand side of the equation represents changes in the CCN concentration f_j as the result of nucleation (R_{ij}), evaporation of cloud drops (E_{ij}) and turbulent diffusion.

Simulation Results

We show the simulation results based on the sounding taken during flight A334 in the Monterey Area Ship Track (MAST) field experiment. We use 25 bins for cloud drops in the size range from 1 to 256 micron and 14 bins for CCN in the size range from 0.01 to 10 micron. The total number of prognostic equations in the model is 370, which includes 5 equations for the thermodynamical variables, one for the turbulent kinetic energy (TKE), 14 for CCN, $350=25 \times 14$ for cloud drops. The simulation was made in a domain using 40×40 grid points with grid sizes $\Delta x = 100m$ and $\Delta z = 20m$. The model was first run in the bulk mode for 90 minutes until the turbulence was fully developed. Then the model was run for 30 minutes in the detailed microphysics mode without coagulation; afterwards the coagulation was started and the model was run for another 60 minutes.

Figure 1 shows the 2D bar-diagram of the DSSD averaged over the horizontal plane at $z=0.5$ km level. A quantitative idea about the DSSD at various cloud levels is given by Figures 2a, b, and c, which show the cross-sections of the 2-D DSSD along the CCN size and drop size axis, respectively. More precisely, Figure 2a shows the liquid water distribution integrated over all drop bins as a function of CCN size, while Figure 2b shows the same distribution (horizontally averaged and integrated over all CCN bins) as a function of drop size. Near cloud base ($z=0.20$ km), the liquid water is more or less evenly distributed over all activated CCN categories. However, at higher levels, the maximum of the liquid water

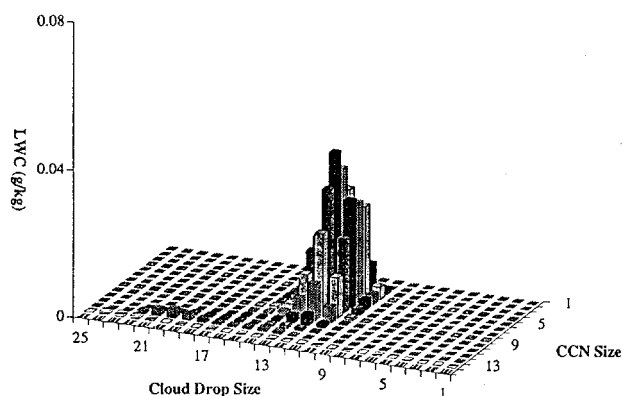


Figure 1. The horizontally averaged DSSD spectrum at $z=0.50$ km.

content (LWC) falls into the CCN category, which has the maximum number of activated CCN particles (dashed line with solid diamonds in Figure 3). The mean drop radius at the same cloud level (Figure 2c) does not significantly depend on the CCN category, which shows that cloud drops become rather quickly diluted, and the effect of the salt on the drop growth is insignificant, except for drops formed by very small or very large aerosols. Very small aerosols can only be activated in a very high supersaturation environment and therefore tend to grow faster, while very large aerosols have large initial wet radius.

The averaged cloud drop spectra shown in Figure 2b for different levels are still unimodal. We note that coagulation was run in the experiment only for 60 minutes. At longer simulation times, the second (drizzle) mode will eventually develop and result in a more significant transformation of the CCN spectra.

The effects of coagulation on the CCN spectra after 60 minutes are summarized in Figure 3. The CCN spectrum at 60 minutes (averaged over the entire domain) that would result if all cloud drops evaporate is shown by the solid line. Part of CCN particles are activated by nucleation (dashed line with solid diamonds) and part of CCNs remain as interstitial aerosol (dash-dotted line with circles). We can see that the percentage of activated CCN particles varies from 0 to 100% depending on the CCN category. For example, at bin 4 (CCN size equal to $0.02 \mu\text{m}$ corresponding to critical supersaturation 0.3%) only about 50% of CCNs are activated. The partial activation is due to the large scatter in vertical velocities and, hence, the large scatter in supersaturations at the cloud base. As a result, some number of large CCN particles will remain unactivated at certain locations and exist as interstitial

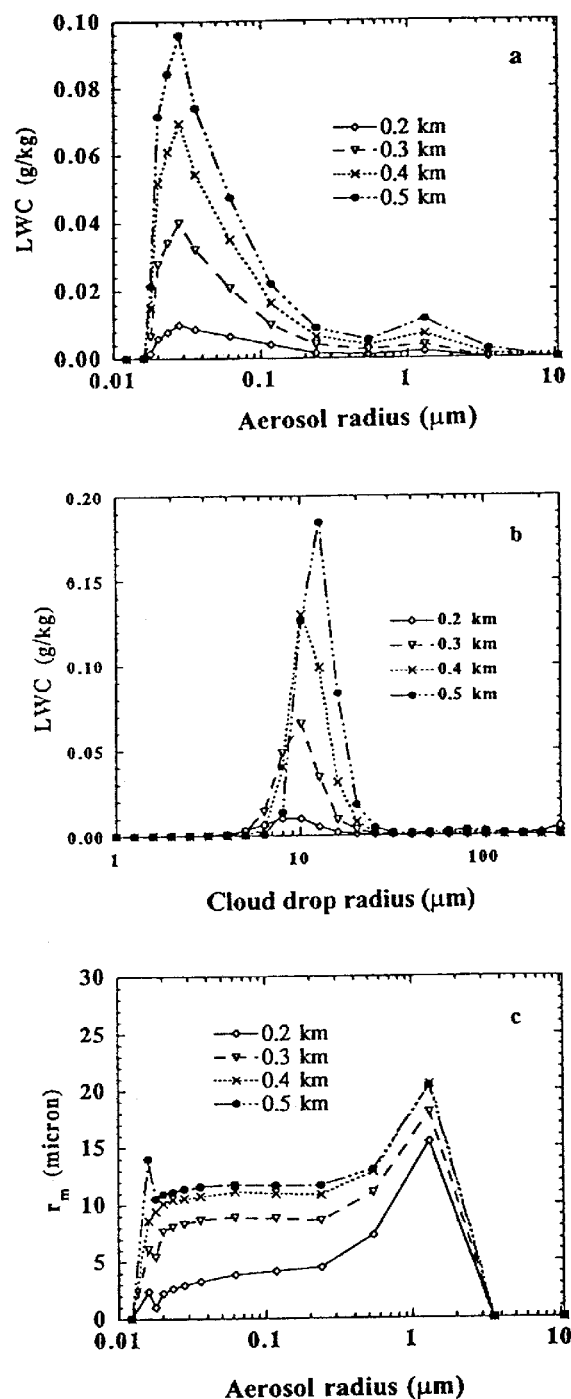


Figure 2. Figures (a) and (b) show the horizontally averaged LWC distribution as functions of the CCN size (integrated over all cloud drop bins) and cloud drop size (integrated over all CCN bins), respectively, (c) shows the horizontally averaged mean drop radius as a function of the CCN size.

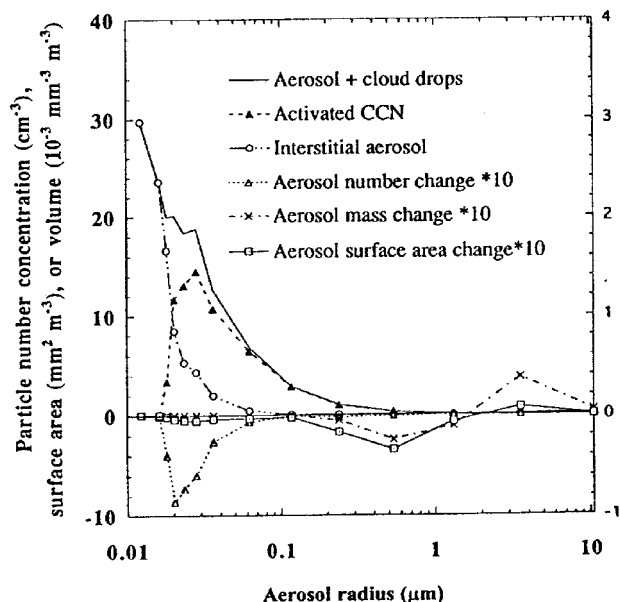


Figure 3. Summary of the effect of the coagulation process on the aerosols and cloud drop spectra: solid line shows the aerosol spectrum if the cloud would completely evaporate at 60 minutes; dashed line with solid diamonds shows the activated CCN distribution; dashed-dotted line with circles shows the interstitial aerosol distribution; dashed line with open diamonds shows particle number loss due to coalescence process (multiplied by 10). Dashed line with crosses shows the aerosol mass change (multiplied by 10). Solid line with boxes show the total aerosol surface area change (multiplied by 10). All spectra are averaged over the whole cloud domain where LWC is greater than 0.1 g/kg.

aerosols. For example, particles in bin 7 (CCN size equal to 0.34 μm , which corresponds to critical supersaturation 0.1%) have not been activated and most likely have been brought into the cloud layer by relatively weak updrafts or by entrainment from the cloud top.

The dashed line with open diamonds shows the particle loss through the coalescence process. The loss in terms of particle number is most significant for the smallest activated CCNs.

For the largest CCN categories, there is a very small gain in concentration, as formation of one large CCN particle may require hundreds or even thousands of small CCN particles. In terms of CCN mass, the loss of the small CCNs equals to the gain of the large CCNs (dashed line with crosses). One important change is the net loss in the total aerosol surface area (solid line with boxes). Because of the total surface area loss, aerosol extinction coefficients are reduced by $\sim 1.2\%$.

Since this result is only for 60-minute simulations, (aerosol number concentration is 157.4 cm^{-3} , maximum LWC in the cloud layer $\sim 0.3 \text{ g/kg}$), we can expect that aerosol extinction coefficients can be reduced by more than 50% with two days in such clouds because the coalescence rate increases exponentially as drop concentration decreases.

Conclusions

We presented preliminary results from a new version of the CIMMS LES model that is able to track the CCN particle transformations as a result of drop coagulation. The cloud microphysical processes are formulated based on a two-dimensional distribution function that depends on the cloud drop size and the aerosol soluble mass inside each drop. This new feature significantly enhances our previous approach based on two separate one-dimensional functions for CCN and cloud drop spectra and allows us to study the aerosol history and transformation of the aerosol spectra as a result of cloud drop collisions and scavenging effects.

The simulation of the coagulation effects in a stratocumulus cloud-topped boundary layer has demonstrated the modification of the salt distribution inside cloud drops and the transformation of the interstitial aerosol. The quantitative results, so far, are limited only to 60 minutes of coagulation time. Longer time 3-D simulations have been completed and their analysis is now under way.

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