2.4 Interaction of aerosol particles with low-level, warm, precipitating stratiform clouds

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1. Introduction

Low-level boundary-layer clouds occur frequently and play a vital role in climate through radiative forcing and in atmospheric chemistry through wet deposition and aqueous-phase chemistry processes.

It has been known for many years that clouds developing over the continents differ substantially in their precipitation efficiency from clouds developing under maritime conditions. The radically different cloud condensation nuclei (CCN) size distributions and composition play a clear role in precipitation development, particularly in warm clouds. The effect of aerosols on cloud microphysics is significant and occurs on a global scale (Breon et al., 2002). Enhanced CCN concentrations that could be produced both through natural or anthropogenic processes would be expected to inhibit rain production (Radke et al., 1989; Durkee et al., 2000). However, modelling studies (Johnson 1982; Lahave and Rosenfeld, 2003) and recent observations (Rosenfeld et al. 2002) show that when giant CCN are ingested into non-precipitating clouds the precipitation rates are restored. Feingold et al. (1999) investigated the impact of giant and ultragiant CCN on drizzle formation in stratocumulus using a number of different models. They found that at low CCN concentrations, drizzle is often active and the addition of giant CCN has little impact. At higher CCN concentrations, drizzle development is slow and giant CCN has great potential for enhancing precipitation.

Aerosol scavenging by precipitation is one of several important removal mechanisms that need to be addressed carefully in air-quality models (Seinfeld and Pandis, 1997). Most, if not all, size-resolved aerosol models being developed use bulk representation of cloud microphysics, with little consideration of the dependence on the size distribution of the cloud microphysics. In order to link the bulk cloud properties produced by a meteorological model with a size-resolved aerosol scavenging process, both the relation between the bulk cloud properties and size-resolved ones, and the dependence of size-resolved aerosol scavenging on the size-resolved cloud properties have to be understood quantitatively, through experiments (Glantz and Noone, 2001; Sellegri et al., 2003) and theoretical studies with explicit cloud microphysics (Flossman et al., 1985; Pinsky et al., 2001).

The purpose of the present study is to investigate the effects of CCN and giant CCN on precipitation formation, and to investigate in- and below-cloud scavenging of aerosol particles by low-level, warm, stratiform clouds. A cloud microphysics model originally developed by Toon et al. (1988) and Ackerman et al. (1995) is modified to produce warm stratiform clouds and precipitation in a 1-dimentional (1-D) model framework.

2. Brief description of the model

Particle concentrations at time t and height z are described by *C (r, z, t)* (m^{-3} µm^{-1}), where *C(r)dr* (m^{-3}) represents the number of particles having radii between *r* and *r+dr*. Within each size bin, the model solves continuity equations for both aerosol particles (for simplicity, CCN is used to represent all aerosol particles through the text) and water drops. The continuity equation can be expressed as (Ackerman et al. 1995):

$$\frac{\partial C}{\partial t} = \frac{C}{\rho}\frac{\partial \rho w}{\partial z} - \frac{\partial}{\partial z}[(w - V_f)C] + \frac{\partial}{\partial z}[K_z \rho \frac{\partial (C/\rho)}{\partial z}] + S - R$$

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where ρ is the air density (kg m^{-3}), w is the vertical air velocity (m s^{-1}), $V_f(r)$ is the terminal velocity (m s^{-1}) of the particles and K_z is the vertical diffusion coefficient (m^2 s^{-1}). The first term on the right side represents the horizontal divergence that compensates for any changes in air density due to the vertical convergence if we make the anelastic approximation and assume ρ and C are independent of x and y. The second term represents the vertical flux due to advection and sedimentation, the third term represents turbulent diffusion, and the fourth and fifth terms represent sources (S) and sinks (R), respectively.

Numerical schemes for vertical advection and turbulent diffusion were described in detail by Toon et al. (1988). For boundary conditions for particles, the approach used in Ackerman et al. (1995) is also used here with no-fluxes at upper boundary and outflow fluxes decided by gravitation setting velocity at the lower boundary. Vertical advection is specified by a prescribed vertical velocity including an oscillating component and the vertical diffusion coefficient for momentum, K_z, is modified from an empirical formula developed by Brost and Wyngaard (1978). Details of w and K_z can be found in Zhang et al. (2004). The microphysical processes considered for source and sink terms include activation/deactivation of CCN to cloud drops, condensation/evaporation of cloud drops, and collision-coalescence among CCN and water drops. In addition there are evolution equations for potential temperature and water vapour (Ackerman et al., 1995). The radiative transfer processes, which are used to calculate radiative heating rates in the thermodynamic equation, are also included based on a sub-model described by Toon et al. (1989).

Two types of particles, CCN and water drops, are considered in the cloud microphysics model. 37 size bins are used for both CCN and water drops ranging from 0.0025 to 10 µm (in radius) for CCN and from 1.0 to 500 µm for hydrometers. A constant volume ratio between successive bins is used (2.0 for CCN and 1.7 for hydrometers). The CCN size distribution is taken as a sum of three lognormal distributions (Zhang et al. 2004). CCN size distributions from different origin (e.g., marine, rural, remote continental, urban) are used in the present study. For some marine clouds, the CCN distributions are partially affected by CCN originating from land. A combination of marine and continental CCN is used to account for this situation. This is done by summing the marine CCN and a percentage (10%) of rural CCN. For marine CCN, the vertical distribution is assumed to be constant since both increases and decreases of CCN number density with height have been observed (Jaenicke 1993). For continental CCN, most observations show an exponential decrease of CCN with height. Here CCN number density is also assumed to decrease exponentially with a scale height of 2 km. The chemical composition of CCN is chosen as $(NH_4)_2SO_4$ for soluble fraction and SiO_2 as an insoluble substance. The soluble fraction is taken as 100% for marine CCN for all size bins. For other cases, the soluble fraction is taken as 100% for the smallest size bin and then decreases linearly to 45% for the largest size bin.

A 1-D model is used with 60 uniformly spaced vertical levels at 30 m resolution between the surface and 1,800 m for all tests. Initial cloud base is chosen at around 700 m and cloud top at 1,300 m, though the cloud base gradually decreases with the development of rain. These values are chosen based on a review of observations of low level marine and continental stratiform clouds by Miles et al. (2000). The initial temperature is chosen to be higher than 0^{o}C everywhere in the cloud. The initial field decreases with height at the dry adiabatic rate from the surface up to the pre-chosen cloud top height. An inversion layer is assumed to lie above the cloud layer. Initial relative humidity (RH) is given as 98.5% within the pre-chosen cloud layer and is assumed to be lower above and below the cloud layer. RH is taken as 95% for most part of below cloud except at the ocean surface where RH is not allowed to follow below 98%. The model is integrated forward in time for 8 hours at a time step of 0.5s starting from midnight (t=0) for all tests from prescribed initial conditions. Radiation is calculated at a 60s time step.

3. Results

3.1 A precipitating pristine marine cloud moves over polluted regions

CCN distribution representing pristine marine situation is chosen as initial condition to produce a precipitating marine cloud (here after called marine base case). The same marine cloud is chosen to move over polluted regions to study the effect of below-cloud high CCN concentration on precipitation properties. This is done by replacing the below-cloud marine CCN by a rural and an urban CCN distribution at a time when precipitation has already fully developed.

To identify the cloud base from the marine base case so that rural and urban CCN can be introduced into the model domain, vertical profiles of maximum supersaturation within every half-hour are extracted and some of them are presented in Figure 1a (numbers in the figure representing the ending time of that half-hour). Maximum supersaturation exceeds 0.2% during the first half-hour and then decreases to below 0.1% for the next three hours and then recovers a little bit. With the cloud development, below-cloud supersaturation increases due to diffusion of water vapor and evaporation of falling droplets. Cloud-base height gradually decreases to as low as 400 m (based on positive supersaturation values). Noticeable raindrops reach surface after t=3h (Figure 1b). Precipitation intensity then rapidly increases during the period of t=4-6h and then gradually decreases. The rural and urban CCN are introduced at t=4h (when precipitation already reaches surface) to any height below 400 m to study the below-cloud CCN effect on precipitation properties.

Figure 2 shows that below-cloud rural or urban CCN are gradually scavenged by falling droplets and are also transported upwards by vertical diffusion and advection. In-cloud interstitial CCN number concentration increases gradually while below cloud CCN decreases. In-cloud interstitial CCN mass (Figure not presented) increases little because major CCN mass are associated with large CCN and are nucleated into cloud droplets. As a result, cloud droplet number increases substantially and the effective droplet radius decreases (Figure 3a, b). This results in a decrease of precipitation intensity at all heights as can be seen from a comparison with the marine base case (only surface-level PR is shown in Figure 3c). The more cloud droplets there are, the smaller are the effective radii and the weaker are the precipitation rates below cloud. Model results presented here explains recent observations of Rosenfeld (1999), suggesting that large concentrations of small aerosols to suppress coalescence and precipitation.

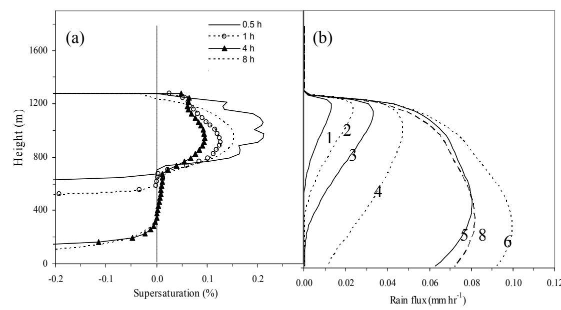

Figure 1. (a) Vertical profiles of maximum supersaturation during half-hour periods ending at t= 0.5, 1, 4 and 8h; (b) Vertical profiles of half-hourly averaged rain flux ending at t=1, 2, 3, 4, 5, 6 and 8h.

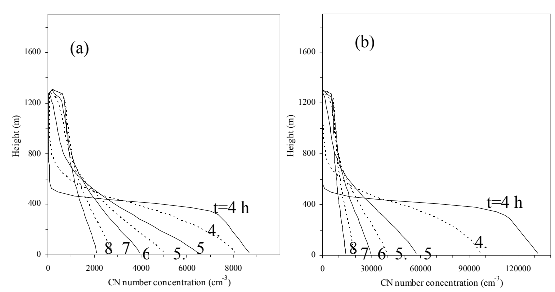

Figure 2. Vertical profiles of half-hourly averaged (ending at t=4.5, 5, 5.5, 6, 7 and 8h) interstitial CCN number concentration when a marine cloud moving over a rural (a) and an urban (b) region at t=4h.

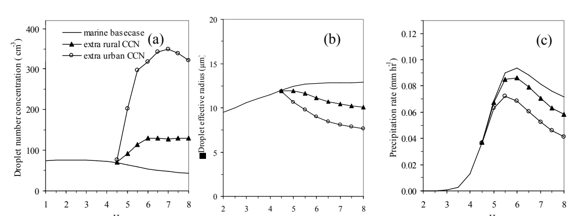

Figure 3. Time series of half-hourly averaged droplet number concentration (a) and effective radii (b) at height of 1005 m, and surface precipitation rate (c).

3.2 Seeding giant CCN into a non-precipitating cloud

Recent observations show that injecting giant CCN into existing clouds containing high concentration of small droplets can restore precipitation (e.g., Rosenfeld et al. 2002). Also, cloud seeding practices around the world have produced artificial rain to solve the extreme drought problems. Though most of these phenomena happen in convective clouds, investigation of similar processes in stratiform clouds will help us to better understand them theoretically. Here giant CCN (radius >1.0 µm) are artificially seeded into an existing non-precipitating cloud. The urban CCN is chosen here as an example since its concentration is very high and thus can produce a cloud with high concentration of small cloud droplets. The cloud forms in less than 5 minutes with the current model configuration and giant CCN are seeded into the cloud starting at t=4h and lasting for 10 min. Giant CCN are seeded only into the lower part (375-975m) of the cloud at a rate of 0.0013 cm^{-3} s^{-1} distributed into bins 28-37 (decreasing from bin 28 to bin 37). This approach is intended to be close to real world situations (e.g., sea spray particles, Lahav and Rosenfeld 2003).

For the seeding cloud, extra giant CCN are nucleated into droplets and decrease the supersaturation, this in turn causes evaporation of small droplets and reduces the total droplet number density (note that during the second-half oscillation period, supersaturation is below zero and small droplets evaporate). For example, droplet number density reduces from 400 cm^{-3} to 250 cm^{-3} in less than an hour after giant CCN seeding (Figure 4a). The effective radius of cloud droplets increases rapidly due to the decrease of total droplet number (Figure 4b). Droplet spectra at three levels (two inside the cloud and one at the cloud base) are wider for the seeding case that the original cloud (Figure 5). As a result, a non-precipitating cloud starts to produce drizzle precipitation shortly after giant CCN being seeded in (Figure 6). The giant CCN override the precipitation suppression effect of the large number of small droplets. This explains the restored precipitation by sea spray when clouds, which formed from polluted air and have high droplet concentration, moved from land to sea areas (Rosenfeld 2002). These results also agree qualitatively with modelling studies presented in Lahav and Rosenfeld (2003), though their studies are on convective clouds with a parcel model while the present study is on stratiform cloud with a 1-D model.

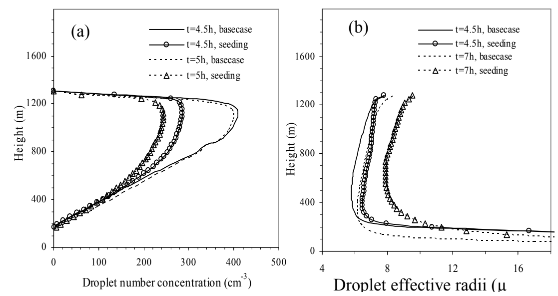

Figure 4. Vertical profiles of droplet number concentration (a) and effective radii (b) for urban case with and without giant CCN seeding.

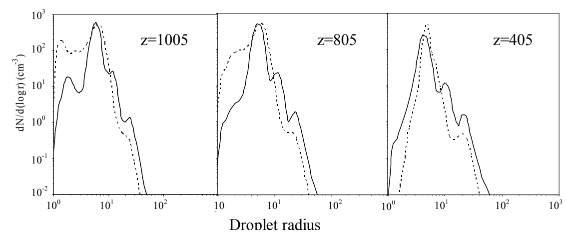

Figure 5. Droplet spectra at three heights at t=5 h for urban base case (dashed lines) and urban case with giant CCN seeding (solid lines).

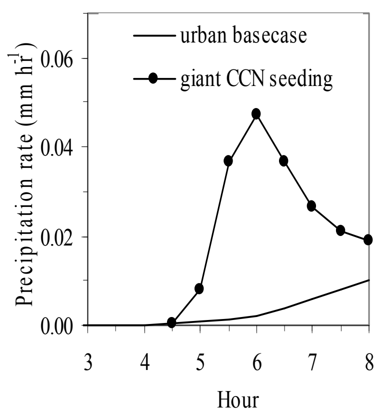

Figure 6. Time series of surface precipitation rate from urban base case and urban cloud with giant CCN seeding.

3.3 The effects of background giant CCN

To study the effects of background giant CCN on precipitation formation, two sets of tests are done with and without giant CCN using three CCN distributions (marine, rural, marine+10%rural).

Figure 7 shows that clouds with background giant CCN form precipitation later than those without giant CCN. The time of delay is around 0.3, 0.4 and 0.8h (if taken 0.02 mm hr^{-1} as the start of precipitation) for clouds formed from marine, marine+10%rural and rural CCN, respectively. It implies that the effects of giant CCN on precipitation formation are larger for clouds with more CCN (i.e., continental) than for clouds with less CCN (i.e., marine). On the other hand, differences in precipitation intensities between cases with and without giant CCN are quite small at later times, implying the very limited effect of giant CCN on precipitation formation.

By comparing the cloud droplet number concentration (Figure not presented), it is found that for marine and marine+10%rural situations, cases with background giant CCN have 2-5% more cloud droplets than cases without giant CCN during the first several hours, while for rural situation, the opposite has been found. However, the effective radii for all three clouds without giant CCN are larger than those with giant CCN (Figure 8). The larger effective radii from cases without giant CCN are caused by the higher supersaturation compared to cases with giant CCN. Though giant CCN only make up a very small percentage in numbers, they make up a larger fraction of total mass. A small number of giant CCN takes up a lot of the available water vapour and reduces the supersaturation, so tests without giant CCN have higher supersaturation than cases with giant CCN. Cases without giant CCN develop more large drops through the condensation process compared to cases with giant CCN due to the higher supersaturation and thus have broader droplet spectra, which lead to earlier precipitation formation. The differences in total droplet number are very small between cases with and without giant CCN and thus play a very minor role.

The differences in supersaturation seem to be important in broadening the droplet spectra, similar to results found in section 3.2. Cloud droplets nucleated from small CCN can quickly grow into the size of 10 µm by the condensation process (within minutes) and droplets activated from giant CCN (radius of 1-10 µm) also fall into this size range. Thus, the inclusion of the background giant CCN, which has a very low concentration of very large particles (e.g., >5 µm) does not necessarily broaden the droplet spectra, except during a short initial period (the first several minutes). Higher supersaturation is more effective for broadening the droplet spectra, which results in an earlier precipitation formation.

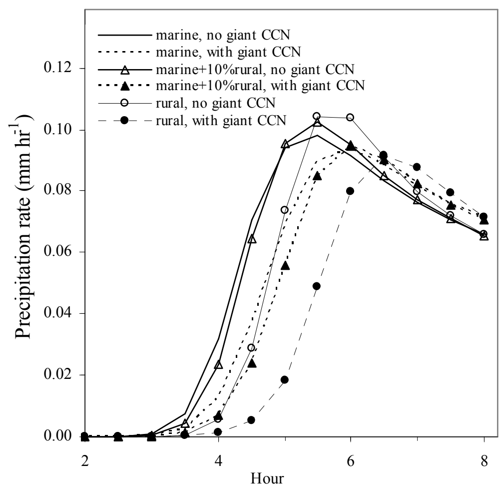

Figure 7. Time series of surface precipitation rate from clouds with and without background giant CCN.

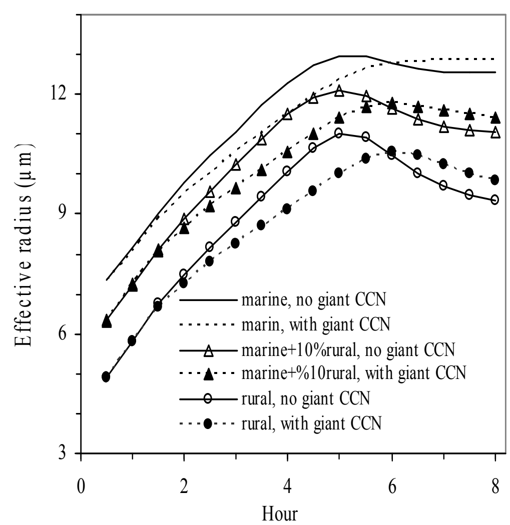

Figure 8. Time series of effective radii at the middle height of clouds with and without background giant CCN.

3.4 Sensitivity tests on giant CCN concentration

Results in section 3.3 show that giant CCN at background concentration do not play a major role in enhancing precipitation formation. Here, sensitivity tests are done for different initial giant CCN concentrations to investigate their role on precipitation formation. The rural CCN distribution is chosen as the base case since it has high droplet number concentration so the effect of giant CCN is large. Many tests have been done by using different concentrations of giant CCN (size bins 28-37) from 0.5 to 20 times the background concentration. Here only several representative tests are presented. Test 1 uses only half the giant CCN of the background value (base case value), Test 2 uses 10 times the background value (this value is just slightly higher than urban background giant CCN shown in Figure 1), Test 3 uses 20 times the background value, and Test 4 uses a value equivalent to the total giant CCN seeded in Section 3.2. Note that Test 4 has far fewer total giant CCN, but more CCN of the largest three size-bins than Test 2 (See Figure 9 for giant CCN distributions for Tests 1-4).

Figure 10 shows that precipitation forms slightly earlier in clouds with only half number of giant CCN compared to the base case, which agrees with results presented in 3.3. Precipitation forms slightly later if giant CCN are doubled of the base case (Figure not presented), which also agrees with results presented in 3.3. With further increase of giant CCN concentration, the effect of giant CCN on precipitation formation gradually changes from slightly delay to slightly enhance. As can be seen from Test 2, precipitation forms earlier than the base case when the giant CCN concentration is increased to 10 times the base value, though the precipitation intensity is lower during the next several hours than the base case. When giant CCN concentration is increased to 20 times the base case value (total sea-sprayed giant CCN seems to be able to reach such high concentration, see Lahav and Rosenfeld 2003), precipitation forms much earlier and precipitation intensity is also much higher than Test 2. Though the total giant CCN in Test 4 are much less than in Test 2, there are slightly more particles of 5-10 µm in radius (the largest 3 size-bins), and precipitation is formed earlier in Test 4 than that in Test 2, implying that the largest CCN has dominating effects on precipitation formation.

Clouds having fewer giant CCN have more small droplets (due to the higher supersaturation) preventing them from forming large enough droplets that can fall through the clouds and reach the surface. Unlike cases presented in 3.3, for which the differences in droplet number concentration are small and supersaturation dominates the differences in precipitation formation time, cases presented in this section have large differences in droplet number concentration due to higher giant CCN concentration used. Thus both droplet number concentration and supersaturation have big impact on droplet spectra and thus precipitation formation. Sensitivity tests presented here suggest that giant CCN can have different effects on initiating precipitation depending on their concentration, and the largest CCN dominate the precipitation formation time.

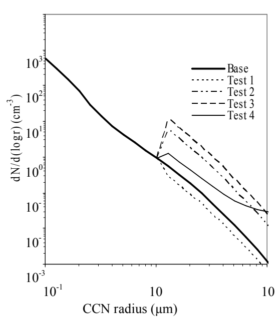

Figure 9. Initial giant CCN size distribution.

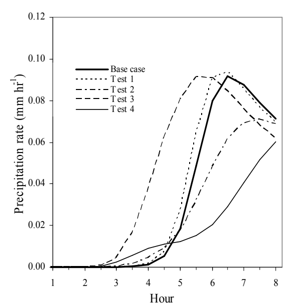

Figure 10. surface precipitation rate from rural cloud sensitivity tests.

3.5 Aerosol scavenging

A pristine marine cloud is integrated for 8 h. Vertically integrated CCN number and mass concentrations within and below the cloud layer (taking 400 m as cloud base according to Figure 1a), respectively, are shown in Figure 11. In-cloud CCN concentrations (in both number and mass) are rapidly reduced due to the activation process during the first hour. CCN are then continuously removed by impaction scavenging while the CCN mass changes little. On the other hand, below-cloud CCN number and mass concentrations are reduced gradually during the course of the cloud development. In-cloud processes scavenge 80% of the CCN by number and almost all the CN mass. Below the cloud rain scavenges around 50% CCN by number and 80% by CCN mass.

Impaction scavenging is usually thought not important compared to the activation process within the cloud layer. A sensitivity test was conducted by turning off the coagulation process between CCN and droplets and comparing results with the case discussed above (Figure 12). It is found that the coagulation process (or impaction scavenging) within the cloud layer is important for reducing the CCN number but having little effect on CCN mass since large particles, which have most of the mass, are rapidly scavenged by the nucleation process. Activation alone reduces by 40% the CCN in number, while impaction scavenging reduces by another 40% or more.

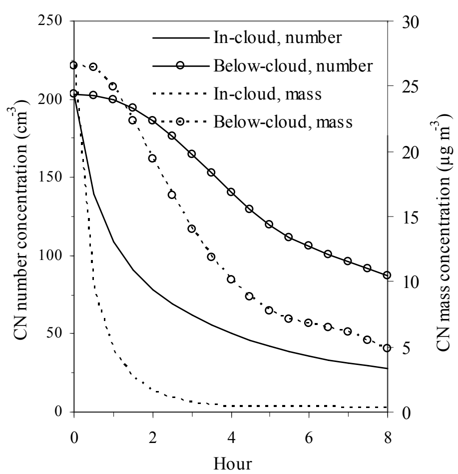

Figure 11. Time changes of half-hourly averaged in- and below cloud interstitial CN number and mass concentrations.

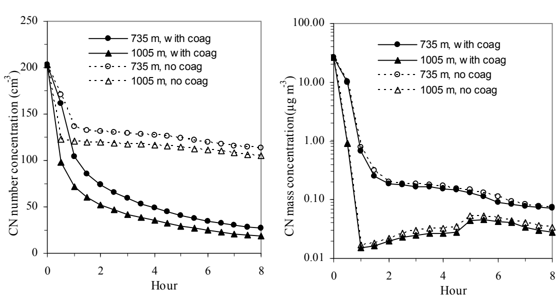

Figure 12. Time serious of half-hourly averaged interstitial CN number and mass concentration at two heights inside cloud layer with and without coagulation between CN and droplets.

Scavenging coefficients are found to depend on precipitation intensity, total droplet surface area, mean radii, initial CCN size distribution, etc. Details of results on scavenging coefficients can be found in Zhang et al. (2004).

4. Conclusions

The effects of CCN and giant CCN on drizzle properties in warm, stratiform clouds and precipitation have been numerically studied. When a precipitating cloud moves to a polluted region, precipitation is suppressed due to extra CCN activated into cloud droplets. When giant CCN are injected into non-precipitating clouds, precipitation can be initiated. Giant CCN are not essential in the development of precipitation in marine clouds, as was found from several earlier studies. Inclusion of background giant CCN delays precipitation formation compared to cases with no giant CCN present; however, opposite effects are found when giant CCN concentration are increased. Difference in precipitation formation time between clouds with and without background giant CCN are caused by differences in supersaturation rather than the small differences in droplet number concentration.

Numerical studies presented in this paper confirm that the activation processes dominate in-cloud aerosol mass scavenging; however, impaction scavenging is important for removing small CCN and tracking CCN numbers. Due to the long-lasting precipitation, though with a very weak rate, below-cloud scavenging is important for both CN number and mass distributions. Below-cloud scavenging can remove 50% of CCN in just a few hours. Bulk below-cloud scavenging coefficients depend on both CCN and droplet distributions. Size-resolved scavenging coefficients are needed in order to correctly track the CCN number and mass evolution. It seems necessary to include other precipitation properties (e.g., droplet surface area, mean radii and/or total droplet number) besides precipitation intensity when parameterizing bulk or size-resolved scavenging coefficients.

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