Freezing Drizzle Formation in Stably Stratified Layer Clouds. Part II: The Role of Giant Nuclei and Aerosol Particle Size Distribution and Solubility

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(Manuscript received 3 May 2004, in final form 15 October 2004)

ABSTRACT

This paper investigates how the characteristics of aerosol particles (size distribution and solubility) as well as the presence of giant nuclei affect drizzle formation in stably stratified layer clouds. A new technique was developed to simulate the evolution of water drops from wet aerosol particles and implemented into a detailed microphysical model. The detailed microphysical model was incorporated into a one-dimensional parcel model and a two-dimensional version of the fifth-generation Pennsylvania State University–National Center for Atmospheric Research (PSU–NCAR) Mesoscale Model (MM5). Sensitivity experiments were performed with the parcel model using a constant updraft speed and with the two-dimensional model by simulating flow over a bell-shaped mountain. The results showed that 1) stably stratified clouds with weak updrafts (<10 cm s\(^{-1}\)) can form drizzle relatively rapidly for maritime size distributions with any aerosol particle solubility, and for continental size distributions with highly insoluble particles due to the low number of activated cloud condensation nuclei (CCN) (<100 cm\(^{-3}\)), 2) drizzle is suppressed in stably stratified clouds with weak updrafts (<10 cm s\(^{-1}\)) for highly soluble urban and extreme urban size distributions, and 3) the presence of giant nuclei only has an effect on drizzle formation for the highly soluble continental aerosol size distributions.

1. Introduction

It is becoming increasingly evident that aerosol particles play an important role in the processes leading to precipitation formation. A number of studies have shown that the chemical composition and size distribution of aerosol particles directly affect the number and size of cloud droplets activated from cloud condensation nuclei (CCN), which in turn impacts the formation of precipitation (e.g., Feingold et al. 1999; French et al. 2000; Eichel et al. 1996). Typically, bulk microphysical schemes do not explicitly simulate the activation of CCN or the time evolution of the drop spectra, whereas detailed bin microphysical schemes may be used to investigate these processes. In the 1980s, Flossmann et al. (1985) investigated wet deposition of atmospheric aerosol particles using a detailed bin model in a steady-state framework. Later studies allowed for droplet and aerosol mass to vary independently and, as a result, the change of the chemical composition and redistribution of the aerosol particles could be simulated (Chen and Lamb, 1994; Wurzler et al. 2000). However, this technique requires a very large computer capacity and calculations are mostly made with one-dimensional steady-state or air parcel models.

In the current study, we examine the role of aerosol particles on precipitation formation through the use of a one-dimensional drop size distribution and two-dimensional cloud model (Feingold et al. 1996). Previous research concentrated primarily on convective clouds or boundary layer clouds such as stratocumulus (e.g., Feingold et al. 1996; Bott 2000). In this paper we investigate how the size distribution and the water-soluble fraction of aerosol particles affects the formation of drizzle in stably stratified clouds. These clouds are distinct from the previously mentioned clouds by their formation mechanism (upglide over frontal surfaces or orographic barriers) and the weakness of their updrafts, typically <10 cm s\(^{-1}\). As mentioned in our previous study (Rasmussen et al. 2002, hereafter Part I), drizzle in stably stratified clouds is hazardous to aircraft in flight because of the icing hazard it poses. It can also impact the radiative properties of these widespread clouds.
Our previous research reported in Part I showed that the efficiency of drizzle formation strongly depends on the CCN concentration. In this study, we examine the effect of aerosol composition (soluble fraction of aerosol particles), shape of the size distribution, and the presence of giant nuclei (giant nuclei are defined in this study as dry aerosol particles larger than 1.5 \mu m radius) on drizzle formation using a detailed microphysical model. To perform this study, we extended the model presented in Part I to simulate the evaporation of aerosol particles into water drops during diffusional growth.

The techniques most widely used to simulate this process are based on the calculation of the critical drop size associated with activated CCN particles. These activated droplets are then put into the appropriate drop size category (e.g., Flossmann et al. 1985). This method may correctly simulate drop formation in convective clouds where the updraft is relatively strong and micron-sized drops can form in a relatively short time compared with the model time step. Feingold et al. (1999) assumed that 20-\mu m sized drops formed on giant nuclei during a single time step of the model. With the low supersaturations typical of stratified layer clouds (less than 0.1%), it takes on the order of 100 s to form micron-sized drops, requiring a different methodology. In this paper, we calculate the evolution of wet aerosol particles into water drops by the diffusional growth equation rather than mapping the activated aerosol particles into water drop bins.

A description of the model used in this study is given in section 2. One-dimensional parcel model calculations are given in section 3. Section 4 provides results from two-dimensional simulations over an idealized bell-shaped mountain and conclusions are summarized in section 5.

2. Model description

The detailed microphysics scheme used in the current research is the same scheme discussed in Part I with the addition of an improved treatment of water drop formation on CCN particles and a few other improvements mentioned below. The formation of water drops on CCN particles is achieved by adding 40 size bins to the 13 provided in Part I. The first bin for CCN particles starts at a mass of 10^{-6} kg (about 0.015 \mu m in radius, 1 \mu m = 10^{-6} m), with mass doubling at the bin edges. Similar to Feingold et al. (1996), the concentration (N_i) and mass (M_i) of water drops and haze particles are calculated in every bin as well as the total mass of the embedded aerosol particles in the kth mass category (M_{a,k}). At the initial time of the simulation, the aerosol particles are assumed to be wet (haze particles) if the saturation ratio is larger than 95%. In this case, the mass of the condensed water is assumed to be equal to the mass of the aerosol particles and the aerosol particle is transferred to the next bin as a result of this additional mass. Diffusional growth of these wet aerosol particles is then calculated. Because these wet aerosol particles have a very rapid growth rate, the numerical solution of the diffusion equation would require an extremely short time step. To avoid this problem, the diffusion equation for haze particles was solved using a linear approximation method. The diffusional growth rate is (Pruppacher and Klett 1997):

\[
\frac{dm}{dt} = 4\pi r_d \left( \frac{s - \frac{2\sigma_{wa}}{R_a r_d f_d}}{T_a^2 - 1} + \frac{\epsilon D_w}{e_{sat,w}} \right) f_a, \tag{1}
\]

where \(m\) and \(r_d\) are the mass and the radius of the wet aerosol particle, respectively; \(M_a\) is the mass of the aerosol particle the drop formed on; \(M_a\) and \(M_i\) are the molecular weight of the water and the ammonium sulfate; \(s\) is the water-soluble fraction; \(p_w\) and \(p_r\) are the density of the water and the aerosol particles; the term \(\sigma_{wa}\) is the van’t Hoff factor; \(s, T, k_b, D_w\) are the supersaturation, temperature, thermal conductivity of air, and diffusivity of the vapor in air, respectively; \(f_a\) is the ventilation coefficient; \(R_a\) is the gas constant for water vapor; \(\alpha_{wa}\) is the surface tension between the water and air; \(e_{sat,w}\) is the saturation vapor pressure over flat water surface; and \(L_w\) is the latent heat of condensation.

It is assumed that the growth rate changes linearly inside the bin, that is,

\[
dm/dt = a_k m(t) + b_k, \tag{2}
\]

where the value of \(a_k\) and \(b_k\) could be calculated if the growth rates are known at the bin edges. The mass of the aerosol particles (\(m_a\)) within drops with mass of \(m_k\) is calculated by

\[
m_a = 0.5 \left( \frac{M_{a,k-1}}{N_{k-1}} + \frac{M_{a,k}}{N_k} \right), \tag{3}
\]

where \(M_{a,k}\) is the total mass of the aerosol particles inside the drops in the kth bin, and \(N_k\) is the number of the drops in the kth bin. From Eq. (2), the time necessary for the drops to grow from a mass of \(m_k\) to \(m_{k+1}\) could be calculated as

\[
\Delta t = \frac{1}{a_k} \ln \left[ \frac{(a_k m_k + b_k)}{(a_k m_k + b_k)} \right]. \tag{4}
\]

The time necessary to grow from \(m_{k+1}\) to \(m_{k+2}\), and so on, could be also given in a similar manner. If \(\Delta t\) is given for each bin, the new size of the drop with mass of \(m_k\) can be calculated in the following manner:

1) Search for the jth bin for which (Fig. 1)

\[
\Delta t = \sum_{i=k}^{j} \Delta t_i < \Delta t < \sum_{i=k}^{j+1} \Delta t_i, \tag{5}
\]

where \(\Delta t\) is the time step used in the model.
Compared with that calculated by using a fourth-order Runge–Kutta scheme, it is the tendency term due to advection and the subgrid-scale diffusion of temperature and vapor content, and \( \tau = 0.5(s(t) + s(t + \Delta t)) \). The second term accounts for the diffusional growth of haze particles, water drops, and pristine and rimed ice particles (sinks of vapor). To numerically solve the above equation, the so-called secant iteration method is applied.

The calculation of supersaturation is based on the implicit method suggested by Hall (1980):

\[
[s(t + \Delta t) - s(t)] / \Delta t = T + f(\tau),
\]

where \( T \) is the tendency term due to advection and subgrid-scale diffusion of temperature and vapor content, and \( \tau = 0.5(s(t) + s(t + \Delta t)) \). The second term accounts for the diffusional growth of haze particles, water drops, and pristine and rimed ice particles (sinks of vapor). To numerically solve the above equation, the so-called secant iteration method is applied.
In addition to the diffusional growth of the above-mentioned particles, the following microphysical processes are considered (see Part I for details):

1) Collision and coalescence of water drops. The moment-conserving technique (e.g., Reisin et al. 1996) is used for the calculation of the time evolution of the hydrometeor size distributions. The moment-conserving technique has been demonstrated to accurately model the evolution of hydrometeor distributions by Tzivion et al. (1987, 1999). The main achievement of the moment-conserving technique is the prevention of artificial broadening of the hydrometeor size distribution by numerical diffusion. In addition, the moment-conserving technique conserves mass and number concentration independent of time step and bin size.

2) Collision between haze particles and water drops including Brownian, phoretic, and gravitational collection.

3) Ice formation via deposition and condensation freezing is simulated using a curve fit to in situ ice crystal measurements by Cooper (1986).

4) Freezing of supercooled drops by contact and immersion nucleation. The formula suggested by Meyers et al. (1992) is used for the number concentration of active contact nuclei. The immersion nucleation rate is given by Bigg’s freezing (Bigg 1953). If the diameter of the supercooled drop is <50 μm, a pristine ice crystal is formed, otherwise graupel is created.

5) Rimed ice formation through the collision of pristine ice crystals and small water drops. If the mass of the water drop is greater than that of the pristine ice crystal, then graupel forms.

6) Graupel formation through the collision of a rimed ice crystal with a water drop of larger mass.

7) Snowflake aggregate formation through the self-collisions of pristine ice crystals and rimed ice crystals.

8) Snowflake aggregate formation due to collisions between pristine and rimed ice crystals.

9) Growth of snowflake aggregates through the collection of pristine and rimed ice crystals.

10) Graupel formation through the riming of aggregates.

11) Diffusional growth for aggregates and graupel.

12) Secondary ice production during the riming of graupel using the formulas suggested by Harris-Hobbs and Cooper (1987).

3. One-dimensional parcel model calculations

Some preliminary one-dimensional parcel model calculations were made to test the numerical technique described above and to investigate the effect of weak updraft speeds (0.05 and 0.1 m s⁻¹) on the evolution of the drop size distribution. The formation and diffusional growth of water drops were calculated in a non-mixing, constant ascent rate rising air parcel (no coagulation between the drops was considered). The initial size distributions of the aerosol particles used in the calculation was given by the lognormal size distributions suggested by Jaenicke (1988). We assumed that all the aerosol particles have the same size-independent soluble fraction consisting of ammonium sulfate. Initially the aerosol particles were distributed in the first 27 bins, between radii of −0.0015 and −8.0 μm. Maritime (MARITIME), rural continental (CONT-A), urban (CONT-B), and extreme urban (CONT-C) type size distributions were used (Fig. 4). The continental distributions are consistent with the Hobbs et al. (1985) observations of giant and smaller particles at various
locations across the United States. The maritime distribution is consistent with previous aerosol particle measurements presented in Pruppacher and Klett (1997). The urban and extreme urban distributions are slightly modified from Jaenicke (1988) to avoid the extreme high numbers of haze particles generated by his urban distribution even in the case of a small water-soluble fraction. The total number concentrations of the aerosol particles are \(8.10 \times 10^9\), \(2.11 \times 10^9\), \(2.19 \times 10^9\), and \(2.27 \times 10^9\) m\(^{-3}\) for the cases of MARITIME, CONT-A, CONT-B, and CONT-C, respectively. Differences between the continental cases are small if the total concentrations are compared, but if the concentrations of particles larger than \(0.1\) \(\mu\)m (the typical size necessary for activation at low supersaturation) are compared the differences are larger. The concentrations of the aerosol particles greater than a radius of \(0.1\) \(\mu\)m were \(4.54 \times 10^7\), \(1.47 \times 10^8\), \(2.22 \times 10^8\), and \(3.12 \times 10^8\) m\(^{-3}\), respectively.

Figures 5–7 show the evolution of the droplet size distributions for the various cases. The size distributions are given at three different levels: (i) about 10 m above the condensation level, close to the height where maximum supersaturation is attained; (ii) 150 m above the condensation level, where the supersaturation is nearly steady state; and (iii) at the top of the 250-m-thick layer. The main characteristics of the size distributions at the end of the simulation are summarized in Table 1. The thin vertical lines denote the critical drop radii at maximum supersaturation. The critical drop radii, which depend on the water-soluble fraction, were calculated from the equations suggested by Pruppacher and Klett (1997). The position of these lines coincides well with the radii where the size distributions have their local minimum. In the case of the MARITIME aerosol distribution, the droplets formed on CCN and haze particles are well separated (Fig. 5). Increasing the soluble fraction, \(v\), by a factor of 10 (from 0.1 to 1.0) results in about a 10% increase in the CCN concentration independent of the updraft velocity. (The CCN concentrations were calculated from the calculated size distributions. The concentrations of the droplets in the bins larger than the local minimum were summed.) In the case of the CONT-A aerosol size distribution, the haze particles were well separated in radius space from the water droplets (Fig. 6). However, when the water-soluble fraction is equal to 1.0, the separation between the haze particles and water drops is not so pronounced as it was in the MARITIME cases (cf. Figs. 6a,b versus Figs. 5a,b). Note that when the updraft is very weak (0.05 m s\(^{-1}\)), the shape of the size distribution of the water drops for the CONT-A cases resembles that of the MARITIME cases (Figs. 5b,d and Figs. 6b,d). Although the concentration of water drops larger than the critical size was higher in the cases of larger water-soluble fraction and larger updraft velocity, the difference between the spectrums is mostly confined to sizes less than 5 \(\mu\)m. The lower solubility results in about a
10% increase of the mean drop radius and of the mean volume radius (see Table 1). While the higher water-soluble fraction results in about 10% increase in CCN concentration, the number concentration of the CCN shows significantly more sensitivity to updraft velocity. Specifically, as the updraft is increased from 0.05 to 0.1 m s\(^{-1}\) in the CONT-A case, the CCN number concentration increased from 75 to over 140 cm\(^{-3}\) (Fig. 6).

The difference between the CONT-A and CONT-C aerosol size distributions is the roughly 2 times higher

![Image](https://journals.ametsoc.org/jas/article-pdf/62/7/2037/3482580/jas3452_1.pdf)

**Fig. 5.** Evolution of the particle size distribution when the initial aerosol size distribution is MARITIME. The size distributions are given at three different levels above the condensation level. Here, \(w\) represents the ascending velocity of the air parcel, while \(\varepsilon\) represents the water-soluble fraction of the aerosol particles. The thin vertical lines are drawn at the critical drop radius belonging to the maximum supersaturation.
concentration of the aerosol particles larger than 0.1 μm in the CONT-C size distribution. The increased aerosol concentration did not result in a significant increase of the CCN concentration (Fig. 7) due to the lower supersaturation in the CONT-C case. The corresponding reduction of the maximum supersaturation (due to a higher aerosol concentration) significantly increases the minimum size of the activated aerosol particles (about 0.2 μm, see Fig. 4). Figure 7b shows that it is not easy to separate the haze particles from the water.
drops when the water-soluble fraction is high and the updraft is very weak. While the number concentration of activated aerosol particles is about 110 cm\(^{-3}\), the total concentration of haze particles and water drops larger than 1.0 \(\mu\)m was 170 cm\(^{-3}\). In this case, the haze particles depleted much more vapor than in other cases. The mass ratio of the haze particles and the water droplets was calculated at the level where the supersaturation reached its maximum value. This ratio was about 0.001 in the MARITIME cases, about 0.01 in the CONT-A cases, and about 0.2 in the CONT-C cases if the water-soluble fraction is equal to 1.0. The lower soluble fraction results in lower ratios in every case. This means that if the updraft is very weak (as in the case of the stably stratified layer cloud), the diffusional growth of haze particles could not be neglected in polluted air masses.

Using the four aerosol size distributions, the relation between the maximum supersaturation and the number concentration of the activated aerosol particles (CCN) was calculated. The discrete points in Fig. 8 (denoted by squares and circles, triangles and crosses) show how the number concentration of the CCN depends on the updraft velocity for the different aerosol size distributions and water-soluble fractions. In this figure, the two lines (solid and dashed) give the field-measured relation between the supersaturation and the CCN concentration used in the previous research reported in Part I. The dots, squares triangles, and crosses denote the CCN concentration calculated from aerosol size distributions of CONT-A, MARITIME, CONT-B, and CONT-C, respectively. The numbers next to each plotted symbol indicate the vertical velocity of the simulations in cm s\(^{-1}\).

Part I. In brief, a moist low-level flow impinging on an idealized two-dimensional barrier was simulated with the MM5 mesoscale model in order to reproduce conditions observed in stably stratified clouds producing freezing drizzle.

The concentration of aerosol particles was held constant from the ground to 3 km MSL. Above this height, the concentration decreased linearly. In regions where the humidity was larger than 95%, all of the aerosol particles were assumed to deliquesce and the particles from the 4th size category were transferred into the (k + 1)th size category. As done previously, the soluble fraction, \(c_s\), was preset to 0.1 or 1.0.

It is important to emphasize that this case study is not a simple constant updraft case as presented in the previous section. Supersaturation is not only determined by the vertical motions, but also by the horizontal distribution of water vapor mixing ratio, which often maximizes over the slope. As the particles move into the interior region of the cloud they deplete more vapor, and the supersaturation decreases. The supersaturation can again increase in the middle region of the hill where the updraft is stronger.

The main difference in the current simulation from the Part I simulations is that the humidity was reduced from 100% to 95% above the ground (Fig. 9). Because this reduction affects the results previously published in Part I, the previous simulation was repeated using the current sounding and both cases were compared in section 4d.

4. Two-dimensional model simulations

In this section, two-dimensional simulations are conducted using the size distributions shown in Fig. 4 to investigate the effect of giant nuclei and the aerosol size distribution and solubility on the drizzle formation in a stably stratified layer cloud. To investigate the effect of giant nuclei, the size distributions were truncated at 1.5 \(\mu\)m. This implies that the concentration of giant nuclei is about 0.3 cm\(^{-3}\) in the case of MARITIME and CONT-A aerosol size distributions.

The initial conditions of the 2D simulations were similar to that used in the previous research reported in
olution of the maximum cloud water mixing ratio (Figs. 10a,b) and the maximum drizzle mixing ratios (Figs. 10c,d) for the various cases. Drizzle is defined as water drops with diameters greater than 40 μm and cloud water drops less than 40 μm.

A significant difference between the GIANTON and GIANTOFF cases was found for the simulation CONT-A with a soluble fraction of 1.0. In the MARITIME cases, the presence of giant nuclei was negligible, as in the case of MARITIME aerosol size distribution and soluble fraction of 0.1 (open squares in Figs. 10b,d). The maximum drizzle mixing ratio reaches a value of 0.1 g kg\(^{-1}\) in every case except in the simulations where the initial aerosol size distribution was urban (CONT-B) and extreme urban (CONT-C); simulations CB10GOFF and CC10GOFF, respectively. When the soluble fraction is 0.1, drizzle starts to form at about 100 min in the MARITIME case and at least 20 min later in the rural continental cases (CONT-A). After the first drizzle forms, its mixing ratio increases quickly and reaches the maximum value of 0.12 g kg\(^{-1}\).

**Fig. 9.** Sounding used to initialize the cloud. Wind barbs are plotted at every \(\sigma\) level of the model. Full barb represents 5 m s\(^{-1}\).

**Table 2. Summary of the simulations.**

<table>
<thead>
<tr>
<th>Type of the size distribution</th>
<th>Soluble fraction ((\varepsilon))</th>
<th>Giant nuclei</th>
<th>Ice physics</th>
</tr>
</thead>
<tbody>
<tr>
<td>M01GOFF MARITIME</td>
<td>0.1</td>
<td>No</td>
<td>No</td>
</tr>
<tr>
<td>M10GOFF MARITIME</td>
<td>1.0</td>
<td>No</td>
<td>No</td>
</tr>
<tr>
<td>M01GON MARITIME</td>
<td>0.1</td>
<td>Yes</td>
<td>No</td>
</tr>
<tr>
<td>M10GON MARITIME</td>
<td>1.0</td>
<td>Yes</td>
<td>No</td>
</tr>
<tr>
<td>CA01GOFF CONT-A</td>
<td>0.1</td>
<td>No</td>
<td>No</td>
</tr>
<tr>
<td>CA10GOFF CONT-A</td>
<td>1.0</td>
<td>No</td>
<td>No</td>
</tr>
<tr>
<td>CA01GON CONT-A</td>
<td>0.1</td>
<td>Yes</td>
<td>No</td>
</tr>
<tr>
<td>CA10GON CONT-A</td>
<td>1.0</td>
<td>Yes</td>
<td>No</td>
</tr>
<tr>
<td>CB10GOFF CONT-B</td>
<td>1.0</td>
<td>No</td>
<td>No</td>
</tr>
<tr>
<td>CC10GOFF CONT-C</td>
<td>1.0</td>
<td>No</td>
<td>No</td>
</tr>
<tr>
<td>M01ICEON MARITIME</td>
<td>0.1</td>
<td>Yes</td>
<td>Yes</td>
</tr>
<tr>
<td>C10ICEON CONT-A</td>
<td>1.0</td>
<td>Yes</td>
<td>Yes</td>
</tr>
<tr>
<td>P1MICEOFF* Maritime</td>
<td>—</td>
<td>—</td>
<td>No</td>
</tr>
<tr>
<td>P1MICEON* Maritime</td>
<td>—</td>
<td>—</td>
<td>Yes</td>
</tr>
<tr>
<td>P1ICEOFF* Continental</td>
<td>—</td>
<td>—</td>
<td>No</td>
</tr>
<tr>
<td>P1ICEON* Continental</td>
<td>—</td>
<td>—</td>
<td>Yes</td>
</tr>
</tbody>
</table>

*P1 means that the case was presented in Rasmussen et al. (2002). In these cases CCN concentration as a function of the supersaturation is given instead of the size distribution type. The shape of the CCN function depends on the type of the air mass.
within an hour. When the soluble fraction is 1.0, drizzle forms later (from 0.5 to 1.0 h), and reaches a maximum value of 0.11 g kg\(^{-1}\) more slowly (about 2 h). The slower formation of drizzle in the latter case could be explained by the difference in the number concentration of the activated aerosol particles for the two soluble fractions. Higher solubility leads to more activated aerosol particles and a smaller critical radius for a given supersaturation. This is due to the larger amount of salt at higher solubility, allowing the cloud droplet to have a smaller radius for activation. Since the total number of activated particles consists of all the particles larger than the critical radius, the total number of particles activated will be higher in this case.

Note that the time evolution of drizzle and total water content in the CA01GON case agrees almost exactly with that of the M01GON case. This, and the similarity between the CA10GON and M10GON cases, suggests that in stably stratified layer clouds, drizzle formation depends only weakly on the aerosol size distribution if the air mass is not extremely polluted. The extreme urban case (CC10GON) has the highest cloud water content (near 0.7 g kg\(^{-1}\)) and the lowest drizzle water mixing ratio at the end of the simulation (−0.035 g kg\(^{-1}\)), suggesting that the high number concentration of droplets prevents the efficient onset of the collision–coalescence process in this case.

Figure 11 shows the dependence of time necessary to reach the drizzle mixing ratio of 0.01 g kg\(^{-1}\) as a function of the mean number concentration of particles (haze and drops) with radii larger than 1.0 \(\mu\)m at the time that drizzle formation is initiated. The best-fit line shows that there is a very good correlation between the number concentration of particles larger than 1.0 \(\mu\)m and the time period necessary for initiating drizzle.

Feingold et al. (1999) found similar results for the case of stratocumulus clouds using a box model with zero mean updraft and various liquid water contents and droplet number concentrations. The Feingold (1999) results are shown in Fig. 11 for a liquid water content of 0.5 g m\(^{-3}\). The average liquid water during the current simulations is closer to 0.1 to 0.2 g m\(^{-3}\), and thus the longer times for drizzle formation onset in our data is to be expected and consistent with the results of Feingold et al. (1999). Since Feingold et al. (1999) performed most of their simulations for the higher liquid water contents, we were not able to plot their data for the lower liquid water contents. However, they do have one data point at a liquid water content of 0.15 g m\(^{-3}\) and a concentration of 25 cm\(^{-3}\), which indicates a time
of 120 min for the onset of drizzle, similar to the value of 130 min found for the same conditions in the current study (Fig. 11).

Both datasets show that the width of the cloud droplet spectra plays an important role in the onset of drizzle, with wider spectra producing drizzle faster than narrower spectra, as expected. It should also be noted that there is a supersaturation difference between the continental and maritime distributions that helps determine this result as well.

Despite the larger difference between the CONT-A and CONT-B size distributions (Fig. 4), the onset of drizzle formation and maximum drizzle mixing ratios is nearly the same (Fig. 10c). This is due to the small difference of only 50 cm$^{-3}$ in the cloud droplet concentration for these two cases (cf. CA10 and CB10 in Fig. 11). The relatively small difference in size distribution between CONT-B and CONT-C (Fig. 4), however, results in a large difference in drizzle onset and mixing ratio (Fig. 10c). In this case the cloud droplet concentration increased to over 250 cm$^{-3}$. This is sufficient to suppress the formation of drizzle significantly. This result suggests that a number concentration of particles larger than 1.0 μm of over 250 cm$^{-3}$ mitigates drizzle formation significantly.

Figures 12–15 show the time evolution of the water drop size distribution at different grid points in the cases of CA01GOFF, CA10GOFF, CA10GON, and CC10GOFF, respectively. The size distributions are drawn at four different horizontal positions: near the cloud edge (400 km), in the updraft region (500 and 550 km), and at top of the hill (600 km). The main characteristics of the size distributions are summarized in Tables 3, 4, 5, and 6. To avoid the possible effect caused by the fluctuation of the haze particle concentration, only water drops larger than 5 μm in radius were taken into consideration when the characteristic parameters (concentration, mean radius, dispersion, volume mean radius, and drizzle mixing ratio) were calculated. A comparison of Fig. 12 (CA01GOFF case) and Fig. 13 (CA10GOFF case) shows that the lower soluble fraction not only results in earlier formation of drizzle, but a larger horizontal region over which the size distribution is wider. While there is no significant difference between the size distributions at a horizontal distance of 450 km (near to the cloud edge), the lower solubility results in earlier and more significant broadening of the spectra in the next 50 km. By the end of the simulation the characteristics of the spectra become very similar in these two cases at a horizontal distance of 600 km (see Tables 3 and 4). A more significant difference remained at the horizontal distance of 550 and 500 km near to the surface. In this region—where the updraft velocity is larger—the lower solubility results in significantly more drizzle.

The broadening of the spectra is far slower in the CC10GON case (Fig. 15). The spectra remains narrow except near the ground at a horizontal distance of 600 km (see Table 6). Integrating the drizzle mixing ratios over the two-dimensional domain resulted in an order of magnitude lower mass for this case compared with the CB10GOFF case.

b. The effect of giant nuclei

The effect of giant nuclei on drizzle formation was tested using the untruncated aerosol size distribution shown in Fig. 4 (the previous simulations were truncated at 1.5-μm radius). A significant effect of giant nuclei was found for the CONT-A size distribution with a soluble fraction of 1.0 (Figs. 10a,c). The unique aspect of this simulation was the relatively high concentration of activated CCN. The addition of giant nuclei added collection centers that would otherwise not have formed because of the relatively small sizes of the droplets as a result of their high concentration. This result agrees with that of Feingold et al. (1999), who also found that giant nuclei only impacted precipitation formation in stratocumulus clouds when the CCN concentration was high.

The effect of the giant nuclei on drizzle formation in the current case is shown in Fig. 16, which shows the time evolution of the relative difference in the domain maximum drizzle mixing ratios between the GIANTON and GIANTOFF cases. The presence of giant nuclei results in earlier formation of drizzle drops, but this effect remains relatively large for only very small drizzle mixing ratios and decreases very rapidly as drizzle mixing ratio increases (see Fig. 10). The effect of giant nuclei lasts for a longer time period (about 1 h) for the CONT-A case aerosol size distributions when the soluble fraction of 1.0 (the case with the highest number of activated aerosol particles for CONT-A size...
Fig. 12. The time evolution of the particle size distribution in the cases of CA01G0FF at 12 different grid points. The position of the grid points is given in each panel. The vertical coordinate $z$ means the height above the ground.
Fig. 13. Same as Fig. 12, but for the CA10GOFF case.
Fig. 14. Same as Fig. 12, but for the CA10GON case.
Fig. 15. Same as Fig. 12, but for the CC10OFF case.
Table 3. Main characteristics of the size distributions at the end of the simulation in Fig. 12: x, z, and y are the coordinates of the grid points. The number concentrations, \( n \), are given in \( \text{cm}^{-3} \); the mean drop sizes \( \langle r \rangle \), the dispersion of the size distributions \( \sigma \), and the volume mean radii \( \langle r \rangle \) are given in \( \mu \text{m} \). The drizzle mixing ratios \( q_{\text{dr}} \) are given in \( \text{g} \, \text{kg}^{-1} \). The parameters were calculated for the water drops larger than 5 \( \mu \text{m} \) in radius.

<table>
<thead>
<tr>
<th>CA10GOF</th>
<th>CA10GON</th>
</tr>
</thead>
<tbody>
<tr>
<td>( x = 450 )</td>
<td>( x = 450 )</td>
</tr>
<tr>
<td>( z = 600 )</td>
<td>( z = 600 )</td>
</tr>
<tr>
<td>( n = 39.0 )</td>
<td>( n = 39.0 )</td>
</tr>
<tr>
<td>( r_{\text{d}} = 9.0 )</td>
<td>( r_{\text{d}} = 9.0 )</td>
</tr>
<tr>
<td>( \sigma = 2.7 )</td>
<td>( \sigma = 2.7 )</td>
</tr>
<tr>
<td>( q_{\text{dr}} = 0.002 )</td>
<td>( q_{\text{dr}} = 0.002 )</td>
</tr>
</tbody>
</table>

Table 4. Same as Table 3, but characteristics of the size distribution are from Fig. 13.

<table>
<thead>
<tr>
<th>CA10GOF</th>
</tr>
</thead>
<tbody>
<tr>
<td>( x = 450 )</td>
</tr>
<tr>
<td>( z = 600 )</td>
</tr>
<tr>
<td>( n = 37.7 )</td>
</tr>
<tr>
<td>( r_{\text{d}} = 7.7 )</td>
</tr>
<tr>
<td>( \sigma = 2.5 )</td>
</tr>
<tr>
<td>( q_{\text{dr}} = 0.001 )</td>
</tr>
</tbody>
</table>

Table 5. Same as Table 3, but characteristics of the size distribution are from Fig. 14.

<table>
<thead>
<tr>
<th>CA10GON</th>
</tr>
</thead>
<tbody>
<tr>
<td>( x = 450 )</td>
</tr>
<tr>
<td>( z = 600 )</td>
</tr>
<tr>
<td>( n = 33.5 )</td>
</tr>
<tr>
<td>( r_{\text{d}} = 8.1 )</td>
</tr>
<tr>
<td>( \sigma = 2.6 )</td>
</tr>
<tr>
<td>( q_{\text{dr}} = 0.001 )</td>
</tr>
</tbody>
</table>

Table 6. Same as Table 3, but characteristics of the size distribution are from Fig. 15.

<table>
<thead>
<tr>
<th>CC10GOF</th>
</tr>
</thead>
<tbody>
<tr>
<td>( x = 450 )</td>
</tr>
<tr>
<td>( z = 600 )</td>
</tr>
<tr>
<td>( n = 145.2 )</td>
</tr>
<tr>
<td>( r_{\text{d}} = 5.2 )</td>
</tr>
<tr>
<td>( \sigma = 0.7 )</td>
</tr>
<tr>
<td>( q_{\text{dr}} = 0.000 )</td>
</tr>
</tbody>
</table>

The effect of giant nuclei could be further examined by the comparison of Fig. 13 (CA10GOF case) and Fig. 14 (CA10GON case). Although the spectra are broader in the GIANTON case at 3-h simulation time at every grid point, the difference becomes negligible by the end of the simulation. The presence of the giant nuclei results in a broader size distribution during the first half of the simulation, resulting in the formation of drizzle from 100 min onwards. After 180 min, the drop-

...
let spectra become broader as a result of collision-coalescence of cloud droplets formed on the aerosol particles less than 1.5 μm. At 360 min, the droplet spectra are nearly identical (see Tables 4 and 5). Thus, although the broadening of the spectra started earlier in the case with giant nuclei, after 2 h, the rate of broadening decreased as a result of cloud water depletion by the giant nuclei themselves, thus there is a slight negative feedback in the presence of giant nuclei. As noted, the spectra became near steady state and identical only after 5 h of simulation time.

Comparison of Fig. 12 (CA01GOFF case), Fig. 13 (CA10GOFF), and Fig. 14 (CA10GON case) shows that lower solubility aerosol particles can more efficiently increase drizzle formation than the presence of giant nuclei for a rural continental air mass. Although the spectra were wider at every grid point at 3-h simulation time in the CA10GON case, later the spectral broadening became faster in the CA01GOFF case. In CA01GOFF, the size distribution of the drizzle size drops become steady state after 4 hr of simulation time near the ground at distances of 550 and 600 km. Except at the level of 600 m, by the end of the simulation the concentration of water drops larger than 50 μm was higher at every grid point in the case of CA01GOFF.

The effect of giant nuclei on the areal extent of drizzle is shown in Figs. 17–19 for simulations CA10GOFF, CA10GON, and CA01GOFF. The outer contours with dashed lines in the figures give the cloud region where cloud water mixing ratio is larger than 0.01 g kg⁻¹. The solid lines denote the drizzle mixing ratios. The value of the first isoline is 10⁻⁴ g kg⁻¹ and the increment is 0.01 g kg⁻¹. The model time is given in the upper right corner of each panel.

Fig. 16. The relative difference between the domain maximum drizzle mixing ratio of the GIANTON and the GIANTOFF maritime and continental simulations as a function of model time.

Fig. 17. Two-dimensional cross section of cloud and drizzle water mixing ratio for simulation CA10GOFF at 2, 3, 4, 5, and 6 h. The outer contours with dashed lines give the cloud region where cloud water mixing ratio is larger than 0.01 g kg⁻¹. The solid lines denote the drizzle mixing ratios. The value of the first isoline is 10⁻⁴ g kg⁻¹ and the increment is 0.01 g kg⁻¹. The model time is given in the upper right corner of each panel.

areas bounded by the 0.01 g kg⁻¹ drizzle mixing ratio isopleths for the two cases. Comparison of the simulations CA10GON (Fig. 18) and CA01GOFF (Fig. 19) shows that the lower concentration of the activated aerosol particles in the low solubility run produces more drizzle than the run with giant nuclei and higher concentration of aerosol (CA10GON). Although the
drizzle starts to form later in the CA01GOFF case, by 3 h, the drizzle content is larger in CA01GOFF than in CA10GON.

c. Regeneration of aerosol particles

Although the regeneration of aerosol particles after evaporation of the droplets is not an important issue for this study, we show in the following that the current numerical technique allows for the determination of the size distribution of the aerosol particles at the lee side of the hill where the droplets evaporate. Because of the application of a one-dimensional size distribution for water drops and haze particles, the discrete bin width limits the size information about the mass of the aerosol particles inside the drops. As the size of the water drops increase, they are transferred into a broader bin where the uncertainty about the mass of the aerosol particles further increases.

Figure 20 shows the size distribution of the aerosol particles in the lee of the barrier at a horizontal distance of 700 km for two different altitudes: 1) close to the ground and 2) about 600 m above ground level. Results are shown from case CA01GOFF at the end of the
simulation. Because of the above-mentioned uncertainty in aerosol mass for larger bin sizes, strong fluctuations in the number concentration of aerosols at the submicron size are evident. To reduce this effect, the data were smoothed by substituting the aerosol concentration in each bin with a mean value calculated by the following equation:

\[
N_k = \frac{N_{k-1} + N_k + N_{k+1}}{3}.
\]

The total concentration of the aerosol particles hardly changed over the domain. Initially it was about 2100 cm\(^{-3}\) over the domain, and by the end of the simulation it was about 1950 cm\(^{-3}\) at both downslope points. This small difference is due to the fact that the concentration of the smaller aerosol particles less than 0.1 \(\mu m\) was hardly reduced by the collision processes, although Brownian, phoretic, and gravitational collision processes were taken into consideration in the model. Sensitivity tests show that the role of the phoretic forces has no significant effect on the final aerosol size distribution even in the downslope region where the saturation decreases below water saturation.

No significant difference could be observed between the size distributions if the radius of the particles is smaller than 0.5 \(\mu m\). While the lower concentration partly could be explained by the nucleation and the fallout of the water drops, the fluctuation about the initial size distribution could be the consequence of the problem caused by the finite bin width. Because of nucleation and aerosol scavenging by precipitation, the aerosol particles larger than 0.5 \(\mu m\) in radius are completely missing in the downslope region at 600 m above the ground. The concentration of these particles is about 0.5 cm\(^{-3}\). Initially the concentration of the micron-size particles was about 5 cm\(^{-3}\). A 2 times reduction could be observed near the ground. In this case only particles larger than 2.0 \(\mu m\) are missing, which is the last bin in the initial aerosol size distribution. This difference could be explained by the size dependence of the trajectory of water droplets. The smaller drops containing smaller aerosol particles follow the trajectory of the air and can leave the cloud at higher elevations. The larger drops probably form on larger nuclei or they contain larger nuclei because of collision–coalescence and fallout and their concentration will be larger at lower elevations. The size distribution of the aerosol particles at the lee side is also affected by the downdraft occurring on this side of the hill.

d. Comparison with results presented in Part I

The main difference between the microphysical model used in the current study from our earlier research (Rasmussen et al. 2002) relates to the formation of cloud drops directly from aerosols rather than by a CCN supersaturation curve. To investigate the effect this difference may have on the calculations, the Part I simulations were repeated using the initial condition applied in this research. One of the most important conclusions made in Part I was that the drizzle formation strongly depends on CCN concentration. The current research supports this. The results of M01GOFF and M01GON experiments agree very well with simulation P1MICEOFF (Fig. 10), and we obtained very similar results between the M01IEON and P1MICEON experiments (Fig. 21). It was also found that increasing the number concentration of activated aerosol particles by either changing the size distribution or their solubility delayed the onset and reduced the rate of drizzle formation. Good agreement was also found with the current urban (CONT-B) size distributions and the continental simulation in Part I. In the P1ICEOFF and P1ICEON simulations, the maximum CCN concentration varied between 200 and 300 cm\(^{-3}\), which we reproduced herein with the urban and extreme urban aerosol size distributions (CONT-B and CONT-C in Fig. 4).

5. Summary

This study investigated how the characteristics of aerosol particles (size distribution and solubility) as well as the presence of giant nuclei affects drizzle formation in stably stratified layer clouds. A new technique was developed to simulate the evolution of water drops from wet aerosol particles and implemented into the detailed microphysical model presented in Part I of this study. The detailed microphysical model was incorporated into a one-dimensional parcel model and a two-dimensional version of the MM5 mesoscale model. Experiments were performed with the parcel model using a constant updraft speed, and with the two-dimen-
sional model by simulating flow over a bell-shaped mountain. The simulation results showed:

1) Stably stratified clouds with weak updrafts (<10 cm s\(^{-1}\)) can form drizzle relatively rapidly for maritime size distributions with any aerosol particle solubility, and for continental size distributions with highly insoluble particles due to the low number of activated CCN (<100 cm\(^{-3}\)) as a result of low supersaturation achieved in these types of clouds (<0.1% typically). The presence of giant nuclei in these maritime clouds had little impact on drizzle formation.

2) Drizzle is suppressed in stably stratified clouds with weak updrafts (<10 cm s\(^{-1}\)) for highly soluble urban and extreme urban size distributions. These distributions had sufficiently increased numbers of relatively small water drops to mitigate drizzle formation and are characterized by significant increases of the aerosol concentration for particles larger than 0.1 \(\mu m\). Thus, drizzle formation may be suppressed near urban areas. Since biomass burning size distributions are similar to those found near urban areas (Pruppacher and Klett 1997), drizzle formation in stably stratified clouds in the vicinity of biomass burning is also likely to be suppressed.

It was found that giant nuclei only enhance drizzle formation when the CCN concentration is relatively high and only during the early stages of the cloud formation when the collision–coalescence process has not yet been established. Our simulations show that the CCN concentration should be greater than 100 cm\(^{-3}\) for giant nuclei to have an effect. This result agrees well with that of Feingold et al. (1999), who found a significant effect on drizzle formation in stratocumulus clouds only when the CCN concentration was above 150 cm\(^{-3}\), and no effect when the CCN concentration was less than 50 cm\(^{-3}\). The effect of the giant nuclei was only important in the first half of the simulation prior to the onset of collision–coalescence. It was shown that the reduction of CCN concentration due to the lower solubility of the aerosol particles can have a much stronger effect on drizzle formation than giant nuclei.

Acknowledgments. The research was supported by the Hungarian Scientific Research Fund (T043010). This research is also in response to requirements and funding by the Federal Aviation Administration (FAA). The view expressed are those of the authors and do not necessarily represent the official policy of the FAA. Comments and remarks given by the reviewers significantly improved this manuscript. The authors are grateful to Greg Thompson and Bill Hall for their insightful and thorough reviews of this paper.

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