Modeling Study of Ice Formation in Warm-Based Precipitating Shallow Cumulus Clouds

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ABSTRACT

Observations of large concentrations of ice particles in the dissipating stage of warm-based precipitating shallow cumulus clouds point to the limitations of scientists’ understanding of the physics of such clouds and the possible role of cloud dynamics. The most commonly accepted mechanisms of ice splinter production in the riming process have limitations to properly explain the rapid production of ice bursts. A more detailed description of the temporal and spatial evolution of hydrometeors and their interaction with cloud condensation nuclei and ice nuclei is needed to understand this phenomenon. A cloud model with bin-resolved microphysics can describe the time-dependent evolution of liquid droplets and ice particles and provide insights into how the physics and dynamics and their interaction may result in ice initiation and ice multiplication. The authors developed a 1.5-dimensional non-hydrostatic convective cloud and aerosol interaction model with spectral (bin) microphysics. The number and mass concentrations of aerosols, including ice nuclei and cloud condensation nuclei, were explicitly followed. Since both in situ observations of bioaerosols and laboratory experiments pointed to efficient nucleation capabilities at relatively warm temperatures, it was assumed that ice-nucleating bioaerosols are involved in primary ice particle formation in condensation and immersion modes. Results show that bioaerosols can be the source of primary ice pellets, which in turn lead to high ice concentrations.

1. Introduction

Ice formation in both deep and shallow cumulus clouds impacts the atmospheric circulation through its impact on precipitation, on diabatic heating, and on the earth’s radiation budget because of the differences of the optical properties of water and ice particles. Although shallow cumulus clouds are widespread in the tropics and subtropics (Rangno and Hobbs 2005; Masunaga and Kummerow 2006; Warren et al. 2007), the ice-formation mechanism in warm-based precipitating shallow cumulus clouds (WPSCCs) is not well understood (Rangno and Hobbs 2005).

The rapid formation of exceptionally high ice particle concentrations in WPSCCs has often been observed in both maritime and continental regions (Koenig 1963; Mssop 1968; Hobbs and Rangno 1998; Baker 2001). However, concentrations of ice nuclei (IN) in the atmosphere that are active at relatively warm temperatures are very low (Mohler et al. 2007). Mechanisms
other than primary ice formation must play an important role in the ice formation of a WPSCC (Rangno and Hobbs 2005). To elucidate ice multiplication in WPSCCs, we should resolve two related scientific issues: 1) the evolution of cloud droplet spectra in cumulus clouds and 2) the possible candidates for IN and ice nucleation mechanisms triggered by them in WPSCCs.

Ice formation in WPSCCs is strongly related to their cloud droplet spectra (Hobbs and Rangno 1998). Observations show that cumulus cloud droplet spectra are broader than those predicted by condensational growth in an adiabatically ascending parcel that is only driven by buoyancy. However, both buoyancy and pressure perturbations determine the development of cumulus clouds (List and Lozowski 1970; Yau 1979; Schlesinger 1980; Zhao and Austin 2005). Therefore, it is necessary to simulate the impact of pressure perturbations on the droplet spectral evolution in cumulus clouds. Sun et al. (2010) found that the cloud droplet spectrum evolution of WPSCCs is favorable for ice multiplication through the Hallett–Mossop (H–M) mechanism in the simulations with perturbation pressures considered. However, little is known about the candidates of IN and ice nucleation mechanisms in WPSCCs. Even though Ariya et al. (2009) proposed that ice-nucleating bioaerosols may act as IN to trigger the ice multiplication in WPSCCs, there is much we still do not know about the minimum concentration of bioaerosols and the prevalent nucleation mode required to trigger ice multiplication in WPSCCs. Condensation freezing may play an important role in the primary ice formation of WPSCCs in addition to immersion freezing, which is considered only as a nucleation mode in the simulations for such clouds (Ariya et al. 2009; Sun et al. 2010). The sources and chemical compositions of IN in the atmosphere have been a puzzling issue for several decades, and there is still much to be learned in this area. Evidence keeps emerging in favor of both mineral and carbonaceous origins of ice nuclei. In situ observations further showed that biological particles dominate the carbonaceous fraction in some regions (Pratt et al. 2009; Prenni et al. 2009). Soil dust and biological particles are the predominant IN active at relatively warm temperatures (Phillips et al. 2009; Pratt et al. 2009).

The main components of mineral dusts have different ice-forming capabilities. The temperatures at which ice starts to be nucleated by silicate particles range between −10° and −20°C, and other particles, such as kaolinite and montmorillonite, have an ice initiation temperature as cold as −35°C via the deposition mode when ice supersaturation is greater than 10% (Welti et al. 2009). Connolly et al. (2009) found that mineral dusts from Asia, the Sahara Desert, and Arizona are not active as IN at temperatures warmer than −12°C. In recent years, airborne IN observed in the atmosphere have been mainly associated with mineral dusts (DeMott et al. 2003; Richardson et al. 2007; Klein et al. 2010; Chou et al. 2011). However, using an atmospheric lidar, Ansmann et al. (2008) suggested that dust aerosols may not be effective as IN at temperatures warmer than −18°C, since ice particles at warmer temperatures were not found at the top of altocumulus clouds at locations close to the Sahara. Even though a recent observation showed that Saharan dust aerosols transported to central Europe can initiate ice particles at temperatures as warm as −8°C (Klein et al. 2010), mineral particles may only be partial contributors and not the dominant sources of atmospheric IN at relatively warm temperatures (warmer than −10°C). Biological particles have been identified as a widespread source of IN in almost all climates (Szyrmer and Zawadzki 1997; Sun 2008) in both continental regions (Pratt et al. 2009; Prenni et al. 2009) and oceanic regions (Knopf et al. 2011). Bioaerosols collected in precipitation show high nucleation capacities at relatively warm temperatures (Christner et al. 2008). Bowers et al. (2009, 2010) further found airborne bacteria acting as IN at temperatures above −10°C. The ubiquity and the high ice-nucleating ability of bioaerosols may render them as an important player in cloud ice formation at relatively warm temperatures. DeMott and Prenni (2010) suggested that carbonaceous aerosols acting as IN in clouds warmer than −15°C are likely to be airborne biological particles. Note that there is a large variety of biological particle concentrations (Burrows et al. 2009). We still do not know the potential roles for biological particles in atmospheric processes (Morris et al. 2011). Recent studies found that biological ice nucleus concentrations are not important on the global scale in cloud formation (Hoose et al. 2010a; Sesartic et al. 2011). However, their roles in atmospheric processes may be important on a small scale and at warm temperatures.

Considering the direct and indirect evidence described above, we suggest that likely candidates for efficient IN at warm temperatures in the atmosphere might be bioaerosols and that they might be expected to play an important role in ice crystal formation in WPSCCs. However, little is known about ice initiation mechanisms of ice-nucleating bioaerosols in WPSCCs and the relationship between huge concentrations of ice particles in WPSCCs (Rangno and Hobbs 2005) and the low concentrations of biological IN in the atmosphere (Hoose et al. 2010b,a).
To accomplish this, it is necessary to numerically simulate how the ice nucleation by ice-nucleating bioaerosols occurs in WPSCCs microphysically and dynamically. Diehl and Wurzler (2010) used an air parcel model to simulate ice nucleation by bacteria with bin microphysics. Phillips et al. (2009) investigated the role of bioaerosols in ice formation with bulk microphysics. Parcel models are dynamically too simple to describe the cloud droplet spectrum evolution of cumulus clouds because the most important property of the air continuity cannot be considered in such models. Furthermore, bulk microphysics cannot microphysically describe the evolution of the IN-containing cloud droplet spectra properly. To overcome these problems within current computation limitations, we use a two-cylinder time-dependent cloud and aerosol interaction model that incorporates explicit microphysical processes for cloud condensation nuclei (CCN) and IN. Section 2 provides the dynamical framework of the model. Section 3 describes microphysical processes of the model. Section 4 presents the model setup. Section 5 describes simulations based on a parameterization that is constrained by observations. Section 6 presents a sensitivity study of the effects of aerosol concentrations on ice formation in WPSCCs. Section 7 presents results based on a parameterization constrained by results from laboratory experiments.

2. Dynamical framework

To study ice nucleation and ice multiplication, not only is high resolution of the particle size bins necessary to adequately describe the distributions of aerosol particles (CCN and IN) and water droplets and ice particles, but also the mass of CCN or IN in hydrometeors should be followed. Since such requirements need an explicit treatment of the hygroscopic growth of aerosols from the nanometer size range of dry aerosols to the millimeter size range of hydrometeors, we had to choose a 1.5-dimensional (1.5D) Eulerian model due to the current limited computational resources. The model described here has been used to simulate ice formation in WPSCCs (Ariya et al. 2009; Sun et al. 2010). The more detailed description is needed to clarify the advantages and the disadvantages of this model. The first 1.5D cylindrical model was presented by Asai and Kasahara (1967) to investigate the influence of the compensating downward motions on cumulus cloud formation and evolution. In such a model, two circular concentric air columns describe the updraft/cloud region (inside column) and the compensating downward-motion region (outside column). A 1.5D model allows us to describe the exchange processes between the lateral sides of the two cylinders. Holton (1973) further developed a parameterization method to include perturbation pressure in a 1D model. Yau (1980) presented a method to include explicit computation of perturbation pressure. Including perturbation pressure resolved the modeling problem of unrealistically large gradients of vertical velocities at the tops of simulated cumulus clouds. We further assume that horizontally, the vertical wind velocities are distributed sinusoidally in order to allow for a vortexlike circulation at the cloud top (Zhao and Austin 2005). Vertical eddy fluxes were also calculated in order to consider the changes of the vertical distribution of heat, momentum, aerosols, and hydrometeors induced by wind velocity deformation and thermal instability. The ratio of the horizontal size of the updraft region and that of the compensating downward-motion region impacts the evolution of the simulated cumulus clouds (Yau 1980). The radii ratio of the two cylinders has been specified to be 0.16 (Ariya et al. 2009; Sun et al. 2010). We performed many sensitivity tests and compared model runs with observations of the Cooperative Convective Precipitation Experiment (CCOPE) case (19 July 1981) (Dye et al. 1986). The radii ratio of 0.25 can result in a good agreement between the observations and the simulations.

The dynamics of this model are shown in Fig. 1a. Lateral exchanges between the two cylinders are allowed through the horizontal velocity \( \tilde{u} \). The vertical wind velocities \( w_a \) and \( w_b \) are horizontally distributed sinusoidally. The equations in the inner cylinder are written as follows:

\[
\frac{\partial (\rho_0 \bar{w}_a)}{\partial t} = - \frac{\partial (\bar{w}_a \rho_0 \bar{w}_a)}{\partial z} - 2 \frac{\rho_0 \bar{w}_a \tilde{u}_a}{a} + 2 \frac{\rho_0}{a} \bar{w}_b - \bar{w}_a \frac{(\bar{w}_b - \bar{w}_a)}{\partial z} - \rho_0 g \frac{\partial K_M}{\partial z} \frac{\partial \bar{w}_a}{\partial z} + c_p \theta_0 \rho_0 \frac{\partial \bar{q}_a}{\partial z} + \left[ \frac{\partial \bar{q}_a}{\partial z} - \frac{\partial \bar{q}_{ab}}{\partial z} - (\bar{q}_{ca} + \bar{q}_{ia}) \right] \rho_0 g \tag{1}
\]
b. Thermodynamic equation

$$\frac{\partial (\rho_0 \bar{\theta})}{\partial t} = - \frac{\partial (\bar{\rho}_d \bar{\theta})}{\partial z} - 2 \rho_0 \bar{\theta} \bar{u} + \frac{2}{\alpha^2} \rho_0 (\bar{w}_b - \bar{w}) (\bar{\theta}_b - \bar{\theta})$$

$$- \rho_0 \frac{\partial (K_H \bar{\theta})}{\partial z} + \frac{L \theta_0}{c_p T_0} \frac{\partial \rho_0 \bar{\theta}_v}{\partial t} \mid_{\text{mic}}$$

(2)

c. Continuity of water vapor

$$\frac{\partial (\rho_0 \bar{q}_{\text{sat}})}{\partial t} = - \frac{\partial (\bar{\rho}_d \bar{q}_{\text{sat}})}{\partial z} - 2 \rho_0 \bar{q}_{\text{sat}} \bar{u} + \frac{2}{\alpha^2} \rho_0 (\bar{w}_b - \bar{w}) (\bar{q}_{\text{sat}} - \bar{q})$$

$$- \rho_0 \frac{\partial (K_M \bar{q}_{\text{sat}})}{\partial z} + \rho_0 \frac{\partial \bar{q}_{\text{sat}}}{\partial t} \mid_{\text{mic}}$$

(3)

d. Continuity of hydrometeors

$$\frac{\partial f_{\text{wat/ice},a}}{\partial t} = - \frac{\partial (\bar{w} - U_{\infty,\text{wat/ice}} f_{\text{wat/ice},a})}{\partial z} - 2 \frac{\partial f_{\text{wat/ice},a}}{\partial \bar{u}}$$

$$+ \frac{2}{\alpha^2} (\bar{w}_b - \bar{w}) (f_{\text{wat/ice},b} - f_{\text{wat/ice},a})$$

$$- \frac{\partial (K_M f_{\text{wat/ice},a})}{\partial z} + f_{\text{wat/ice},a} \mid_{\text{mic}}$$

(4)

where $a$ is the inside radius, $b$ is the outside radius, the ratio of $a$ and $b$ is 0.25, we assumed that $a$ is 1.5 km for warm-based shallow convection, variables with the subscript $a$ represent variables of the inner cylinder, variables with the subscript $b$ represent variables between the inner cylinder and outer cylinder, $\alpha^2$ is set to 0.1 (Asai and Kasahara 1967), and $f_{\text{wat/ice}}$ denotes the number density distribution function of water droplets or ice crystals. Also, $w$ denotes the vertical velocity, $U_{\infty,\text{wat/ice}}$ is the terminal velocity of hydrometeors, $\theta$ is the potential temperature, $\theta_0$ is the virtual potential temperature, and $q_w$ is the mixing ratio of water vapor. The bar above a symbol denotes the average values over the inner cylinder domain and over a domain between the inner cylinder and the outer cylinder. The variables with a tilde represent the effect of dynamic entrainment. The variable $\pi'$ denotes the deviation of nondimensional entrainment. The variable $\pi'$ represents the effect of dynamic entrainment. The variable $\pi'$ represents the effect of dynamic entrainment.

3. Microphysical processes

The microphysical processes of this model especially focus on the interactions between aerosols and clouds. The properties of the atmospheric aerosol are much more complicated than can be described faithfully in cloud models. It is plausible that a considerable portion of aerosols in the atmosphere are suitable candidates for CCN. IN that are active at temperatures warmer than $-35^\circ C$; however, can only come from a very small fraction of those aerosols that contain insoluble substances. It is impossible to track the proportions of CCN and IN in each hydrometeor. Instead, we define two functions, $f(m, m_{ap})$ and $g(m, m_{IN})$. The former is the concentration of hydrometeors (either water or ice) of mass $m$ that contain any particle (solid or dissolved) (e.g., ammonium sulfate and/or IN) of mass $m_{ap}$; for most hydrometeors $m_{ap}$ will be the CCN mass. The latter is the concentration of hydrometeors of $m$ that contains an ice nucleus of mass $m_{IN}$, regardless of the total particle mass in the hydrometeor. So, $g(m, m_{IN})$ is a subset of $f(m, m_{ap})$. The meaning of functions used in our model with a fixed-bin scheme is described in Table 1.

In this model, the sizes of cloud droplets growing from haze droplets and raindrops growing via the warm-rain process are determined by condensation, evaporation, and stochastic collision and coalescence (Leroy et al. 2006; Sun 2008). The ice-phase microphysics includes nucleation, deposition, sublimation, aggregation, riming, and melting. Since we focus on ice nucleation and ice multiplication, ice particles are characterized as small spherical ice particles, including frozen cloud droplets (diameters less than 20 $\mu m$) and newly produced spherical ice splinters (Choularton et al. 1978),
with an assumed lognormal size distribution (standard deviation $\sigma$ is 1.15; the mean radius $r_m$ is 5.0 $\mu$m); columnar ice particles (see Hobbs and Rangno 1990, their Fig. 6) (diameters from 20 to 100 $\mu$m of size bins); and spherical ice particles [see Hobbs and Rangno (1990), their Fig. 6] (diameters from 100 to 600 $\mu$m of size bins). For the size bins of ice particles between 600 and 5000 $\mu$m, ice pellets have been implemented into the model for the simulations of WPSCCs. The ice spectrum of these shapes and types is presented by a 1D spectral function in our model. The density of ice particles has been assumed to be 0.9 cm$^{-3}$. With these prescribed characteristics, the diffusional and rime growth rates of ice particles with different shapes can be used to determine the ice particle mass increase as a spherical body with a normalized bulk density of 1.0 g cm$^{-3}$ in each size bin. In the size range of columnar ice particles (20–100 $\mu$m), the axis ratio is assumed to change sinusoidally with the mass increase to keep the spectral distribution of ice particles continuous, with a maximum ratio of 10.

### a. Microphysical processes for CCN

The hygroscopic growth of CCN is described as a continuous process through which haze drops become activated to form cloud drops. In this study, we further assume that CCN are predominantly composed of ammonium sulfate and that the impurity effect on cloud droplet activation (Sun and Ariya 2006) is ignored if aerosols contain other soluble or insoluble substances. Cloud droplets grow by condensation and collision–coalescence; meanwhile, some supercooled water droplets can also transform into ice particles. Water droplets can freeze in three ways in our model: the nucleation of cloud and rain droplets by IN, the freezing of rain droplets by collision with ice splinters, and the collision of haze droplets and cloud droplets with ice particles with equivalent spherical water droplet diameters greater than 100 $\mu$m (riming process). The whole process for the evolution of the water droplet and ice particle distributions (appendix A) is described in Eqs. (5) and (6), respectively.

### Table 1. Number distribution functions used in the 1.5D model.

<table>
<thead>
<tr>
<th>Functions</th>
<th>Total bins of aerosols</th>
<th>Total bins of hydrometeors</th>
</tr>
</thead>
<tbody>
<tr>
<td>$f_{wate}(m,m_{ap})$</td>
<td>90 mass bins of ammonium sulfate</td>
<td>130 mass bins of water drops</td>
</tr>
<tr>
<td>$f_{ice}(m,m_{ap})$</td>
<td>90 mass bins of ammonium sulfate</td>
<td>130 mass bins of ice particles</td>
</tr>
<tr>
<td>$g_{wate}(m,m_{IN})$</td>
<td>90 mass bins of ice nuclei</td>
<td>130 mass bins of water drops</td>
</tr>
<tr>
<td>$g_{ice}(m,m_{IN})$</td>
<td>90 mass bins of ice nuclei</td>
<td>130 mass bins of ice particles</td>
</tr>
</tbody>
</table>
b. Microphysical processes for IN

Heterogeneous freezing of ice in laboratory experiments can occur in association with IN, via condensation, immersion, deposition, and contact mechanisms. It is not feasible to include all these nucleation mechanisms in our simulations. Since only a small fraction of aerosols in the atmosphere are IN and their aged surfaces are always covered with soluble substances (Chen et al. 1998; Zhang and Carmichael 1999; Clarke et al. 2004; Sun and Ariya 2006; Pratt et al. 2010) and heterogeneous ice nucleation rarely occurred below water saturation (Heymsfield and Miloshevich 1993; Mohler et al. 2005; Dymarska et al. 2006), we consider that IN catalyzing ice formation through immersion freezing and condensation freezing are a part of the CCN distribution and are only distinguished from CCN by their extra water saturation (Heymsfield and Miloshevich 1993; Sun and Ariya 2006; Pratt et al. 2010) and heterogeneous ice nucleation rarely occurred below water saturation (Heymsfield and Miloshevich 1993; Mohler et al. 2005; Dymarska et al. 2006), we consider that IN catalyzing ice formation through immersion freezing and condensation freezing are a part of the CCN distribution and are only distinguished from CCN by their extra water saturation (Heymsfield and Miloshevich 1993; Sun and Ariya 2006; Pratt et al. 2010) and heterogeneous ice nucleation rarely occurred below water saturation (Heymsfield and Miloshevich 1993; Sun and Ariya 2006; Pratt et al. 2010)

\[
\frac{df_{\text{wat}}(m,m_{ap})}{dt}_{\text{mic}} \begin{cases} \frac{df_{\text{wat}}}{dt} \text{ condensation/evaporation} \\ - \frac{df_{\text{ice}}}{dt} \text{ nucleation} - \frac{df_{\text{wat}}}{dt} \text{ riming} \\ + \frac{df_{\text{wat}}}{dt} \text{ coalescence} + \frac{df_{\text{wat}}}{dt} \text{ melting} \end{cases}.
\]

(5)

\[
\frac{df_{\text{ice}}(m,m_{ap})}{dt}_{\text{mic}} \begin{cases} \frac{df_{\text{ice}}}{dt} \text{ deposition/sublimation} \\ + \frac{df_{\text{ice}}}{dt} \text{ nucleation} + \frac{df_{\text{ice}}}{dt} \text{ riming} \\ + \frac{df_{\text{ice}}}{dt} \text{ aggregation} - \frac{df_{\text{ice}}}{dt} \text{ melting} \end{cases}.\]

(6)

The detailed description of the above expressions can be found in appendix B.

c. Parameterizations of heterogeneous ice nucleation and ice multiplication

We selected two specific parameterizations of heterogeneous ice nucleation for ice formation comparisons in WPSCCs. One parameterization that provides dependencies on the chemistry and surface area of IN has an observational constraint (Phillips et al. 2008). The second is based on laboratory observations (Diehl and Wurzler 2004).

1) OBSERVATIONAL CONSTRAINT PARAMETERIZATION

Phillips et al. (2008) proposed an empirical parameterization of heterogeneous ice nucleation through deposition, condensation, and immersion freezing for dust/metallic aerosols (DM), inorganic black carbon (BC), and insoluble organic particles (O) in which bacteria are assumed to be the ice-nucleating material. The number of active IN per kilogram of air for each kind of aerosol \(n_{\text{IN,X}}\) is determined by air temperature, supersaturation with respect to ice, nucleation threshold temperature, fractional contribution of this kind of IN to the measured total IN concentrations, concentrations, and size distributions of a particular species of insoluble aerosol. A detailed description of the parameterization is given by Phillips et al. (2008). In our simulations, we only consider ice nucleation at water supersaturation.

2) LABORATORY-BASED PARAMETERIZATION

The immersion freezing mode is one ice initiation mechanism that is relevant to this study. We simulated ice nucleation in the immersion freezing mode with the parameterization of Diehl and Wurzler (2004). The immersion freezing rate of IN is based on the semi-empirical method, defined as

\[
R(m,m_{\text{IN}}) = -aB_{h,j}V_{\text{drop}} \exp[a(T_0 - T)] \frac{dT}{dt},
\]

(9)

where \(a\) is equal to 0.82 °C\(^{-1}\); \(V_{\text{drop}}\) is the volume of the water droplet; \(T_0\) is equal to 273.16 K; \(B_{h,j}\) is a constant
representing the ice-nucleating capability of IN, which relates the median freezing temperature \( T_m \) (Diehl and Wurzler 2004) and the volume of the drops \( V_{\text{drop}} \) (Pruppacher and Klett 1997); and \( dT/dt \) is the rate of temperature change of the cloud parcel. The temperature changes of saturated air parcels follow the modified pseudomoiost adiabat (Taylor 1989).

d. Ice multiplication

Laboratory experiments have documented the dependence of secondary ice production rates on temperature, the water drop spectrum, and riming rate (Mossop 1976, 1978a; Heymsfield and Mossop 1984), and the results have been expressed mathematically in various ways (Mossop 1978b; Harris-Hobbs and Cooper 1987; Cardwell et al. 2003). We have implemented the parameterization of secondary ice production of Harris-Hobbs and Cooper (1987) in the present model (see appendix C).

4. Model setup

a. Finite-difference scheme

The numerical transport algorithm adopted is the Multidimensional Positive Definite Advection Transport Algorithm (MPDATA) with the nonoscillatory option (Smolarkiewicz and Grabowski 1990; Schär and Smolarkiewicz 1996). Our model employs second-order-accurate MPDATA for both momentum and scalar variables involved in the dynamics and microphysics of hydrometeor growth. The number density distribution of aerosol particles, water droplets (including aerosol particles), and ice crystals are described by two number density distribution functions—\( f_{\text{water}}(\ln m) \) and \( f_{\text{ice}}(\ln m) \)—with respect to the natural logarithm of mass (Leroy et al. 2006). Aerosol particles have 90 bins with a radius from \( 8.0 \times 10^{-3} \) to \( 2.36 \times 10^{-2} \) \( \mu \text{m} \), and hydrometeors and aerosols together have 130 bins with a radius from \( 8.0 \times 10^{-3} \) to \( 2.4 \times 10^{4} \) \( \mu \text{m} \), so that the minimum mass of aerosols \( m_{\text{ap0}} \) is \( 2.0 \times 10^{-18} \) g.

The time step is 2 s, and a time-splitting procedure is applied in the numerical transport algorithm for both dynamical processes and microphysical processes in order to guarantee the Courant–Friedrich–Levy criterion.

b. Initial conditions

The initial thermodynamic conditions for the simulations of WPSCCs are the same as those used by Yau (1980) to represent an idealized sounding to simulate cumulus clouds. The temperatures at the sea surface and at the cloud base (800 m) are 24° and 17°C, respectively. The ambient temperature lapse rate is 6°C km\(^{-1}\) above the cloud base. To initiate convection we use a perturbation of vertical velocity. The initial vertical velocity impulse below cloud base is assumed to be

\[
w_\text{a} = \sin \left( \frac{\pi z}{H} \right), \quad z \leq H, \quad (10)
\]

where \( H \) is the height of the cloud base.

Figure 2 shows the size distribution of maritime aerosols (O’Dowd et al. 1997) together with multiples of the distributions. The concentration of maritime aerosol \( C_1 \) represents a clean ocean region; the threefold increase in the maritime aerosol concentration \( C_2 \) represents a clean continental region; the sixfold increase in the maritime aerosol concentration \( C_3 \) represents a slightly polluted continental environment; the twelfold increase in the maritime aerosol concentration \( C_4 \) represents a medium-polluted continental region; and the twenty-fourfold increase in the maritime aerosol concentration \( C_5 \) represents a heavily polluted continental region as may, for example, be found in response to biomass burning (Jayaratne and Verma 2001). A 6-times maritime aerosol concentration \( (C_3) \) is used as the control case study. The initial aerosol concentrations in all cases decreased exponentially with height above cloud base. Aerosols with diameters greater than 10 \( \mu \text{m} \) are not included in the simulations. The initial size of drops is determined by classical Kohler theory. The maximum size of haze drops is calculated by assuming equilibrium at 99% relative humidity at the cloud base.
5. Simulations based on the parameterization constrained by observations

In the parameterization scheme of Phillips et al. (2008), IN are dust particles, soot/black carbon, and bioaerosols represented by bacteria. The proportional contribution of each aerosol type to IN has been assumed to have a constant value that is independent of temperature and humidity. The relative contributions of dust and soot were set at $\frac{2}{3}$ and $\frac{1}{3} - 0.06$ [see Phillips et al. (2008), their Fig. 2], respectively. The estimate of the bacterial fraction, 0.06, was derived from the comparison of the total IN concentration predicted by the parameterization of Meyers et al. (1992) at $-30^\circ$C with the bacterial IN concentration that was estimated with a given concentration of bacterial cells, the fraction of the ice-nucleating bacteria of 1% (Lindemann et al. 1982), and the freezing fraction of cells belonging to the ice-nucleating bacteria of 0.1 at the same temperature (Vali et al. 1976; Gross et al. 1983).

Since we focus on the study of ice initiation and ice multiplication in WPSCCs in which cloud-top temperatures are around $-10^\circ$C, the concentration of IN at $-10^\circ$C in the atmosphere should be the upper bound on the ice nucleus concentrations and hence ice initiation at temperatures warmer than $-10^\circ$C. The concentration of IN active at temperatures warmer than $-10^\circ$C is normally less than 0.3 L$^{-1}$ [see Eidhammer et al. (2009), their Fig. 7]. Berezinski et al. (1988) observed an IN concentration of 0.27 L$^{-1}$ at $-10^\circ$C. Therefore, we assumed that the initial concentrations of bioaerosols, including non-ice-nucleating bioaerosols, are equal to different multiples of the concentration of IN at $-10^\circ$C, which vary in the in situ observation range (Burrows et al. 2009; Després et al. 2012). Furthermore, the impacts of concentrations of ammonium sulfate on ice formation should be also evaluated, since the size distribution and concentrations of cloud droplets impact the ice multiplication. Therefore, experiments listed in Table 2 include tests for concentrations of bacteria and concentrations of ammonium sulfate. We used the experiment $E_4$ as a control run for our study.

Note that both the initial size distribution of IN and their hygroscopicity influence their collision efficiencies with hydrometeors. The size distribution (Tong and Lightart 2000) and the activation of bacteria as CCN (Franc and DeMott 1998) are taken into account in our study. The size distribution of IN is selected to be a combination of two measured bacterial lognormal size distributions (Qian et al. 1995; Tong and Lightart 2000): for single cells, $\sigma = 1.35$ and $r_m = 0.78 \ \mu m$; for clusters, $\sigma = 1.35$ and $r_m = 1.97 \ \mu m$. This size distribution of bacteria results in the minimum bacterial mass $m_{\text{IN0}}$ to be $2.048 \times 10^{-15}$ g. The concentrations of single-cell particles and cluster-cell particles occupy 40% and 60% of the total bacterial particles, respectively.

### a. Time evolution of selected variables

The time evolution of vertical velocities and rainwater contents are shown in Fig. 3a. A significant feature of the results is the weak vertical gradient of vertical velocity compared with other studies using a similar dynamical framework (Asai and Kasahara 1967; Taylor 1989; Leroy et al. 2006). The strong gradient of vertical velocity found in previous studies resulted from inadequate treatment of the pressure perturbation (Sun 2008). In this study, we took account of the impact of rotation of the airflow on the perturbation pressure with an assumption that horizontal and vertical velocities are distributed sinusoidally. Furthermore, the gradient force of dynamic perturbation pressure (perturbation pressure induced by dynamic field) leads to acceleration at the cloud top and may speed up warm-rain formation (Sun 2008). A rainwater content greater than 0.1 g m$^{-3}$ appears within 35 min after the convection initiation with a multiplied maritime aerosol concentration ($6 \times 217$ cm$^{-3}$) (Fig. 3a). Liquid water converts into ice water in the cloud dissipating stage (Fig. 3b). The maximum ice water content reaches 0.5 m$^{-3}$. Figure 4 shows the spatial and temporal evolution of water drop concentrations. The maximum cloud droplet concentration reaches 600 cm$^{-3}$ (Fig. 4a). At the top of the cloud, there is a high concentration band of small raindrops (Fig. 4a). Correspondingly, the maximum concentration of bacteria-containing cloud droplets is 0.0025 cm$^{-3}$ (Fig. 4b). The spatial and temporal distribution of bacteria-containing raindrop concentrations is almost the same as that of the total raindrop concentrations, which means that all raindrops with a diameter greater than 2 mm contain bacteria. Bacteria are scavenged by raindrops through both nucleation and impaction scavenging. However, the maximum concentration of bacteria-containing small raindrops is less than that of total
small raindrops during most time periods. Therefore, some small raindrops do not contain bacteria in this simulation with an initial bacterial concentration of $13.5 \text{ L}^{-1}$ in which the concentration of ice-nucleating bacteria is $0.135 \text{ L}^{-1}$. Figure 5 shows the primary ice nucleation rate and the ice splinter production rate. The primary ice nucleation occurs both at the cloud-top interface of cloud-clear air through the condensation freezing mode and in the cloud through the immersion freezing mode. The maximum primary ice nucleation rate (MPINR) only reaches $1.038 \times 10^{-3} \text{ L}^{-1} \text{ s}^{-1}$. However, the maximum ice splinter production rate (MISPR) reaches $1.206 \text{ L}^{-1} \text{ s}^{-1}$. These results indicate that the ice multiplication dominates ice formation in WPSCCs.

In the subfreezing region, the maximum concentration of ice particles containing bacteria reaches $1.0 \text{ L}^{-1}$ in the dissipating stage (Fig. 6a). Bacteria-containing ice particles include bacteria-containing primary ice particles and bacteria-containing secondary ice particles. Bacteria-containing primary ice particles refer to ice particles nucleated by ice-nucleating bacteria and subsequent rimed ice particles resulting from collisions with cloud droplets and raindrops. Bacteria-containing secondary ice particles refer to ice particles nucleated by ice splinters that subsequently rimed with bacteria-containing water drops. Since in the parameterization scheme of Phillips et al. (2008) bacteria include ice-nucleating bacteria and nonice-nucleating bacteria, IN-containing ice particles refer to
the ice particles containing ice-nucleating bacteria that may initiate ice formation or be scavenged by secondary ice particles. Concentrations of ice particles as large as 155 L$^{-1}$ appeared in the dissipating stage (Fig. 6a), implying that a low concentration of IN can trigger ice multiplication. The primary nucleation of ascending small raindrops produces rimers (Fig. 6b) that result in ice crystal bursts via the H–M mechanism (Sun et al. 2010). The ice crystal multiplication process is directly influenced by the concentration of ice pellets. The initiation time and concentration of primary ice pellets are two determining factors that affect the production of ice bursts. These two factors are determined by both the production of primary ice particles and the evolution of raindrops. The concentration of primary ice pellets at the beginning of ice formation may be approximately estimated from the comparison between the total ice pellet concentration and the IN-containing ice pellet concentration. If the difference between them is small, then we can conclude that they represent the concentration of primary ice pellets that include ice pellets from small raindrop nucleation by ice-nucleating bacteria and from small raindrop nucleation by IN-containing ice particles. If the concentration of total ice pellets is highly greater than that of IN-containing ice pellets, then the production of the secondary ice pellets nucleated by the ejection of ice splinters becomes pronounced. Since bacteria-containing secondary ice pellets only occur when ice splinters collide with bacteria-containing raindrops or in a collision of secondary ice pellets with interstitial bacteria and bacteria-containing particles, bacteria-containing ice pellets may be regarded as bacteria-containing primary ice pellets or IN-containing ice pellets when the total concentration of ice pellets is close to that of bacteria-containing ice pellets. Hence, we can use the concentration of bacteria-containing ice pellets instead of IN-containing ice pellets to determine the primary ice pellets in the ice initiation stage. Figure 6b shows the evolution of the concentration of the bacteria-containing ice pellets and the total concentration of ice pellets. In the dominant stage of the primary ice production, there is little difference between them and their concentrations are far less than the concentration of ice-nucleating bacteria. However, in the dominant stage of secondary ice production, the concentration of the total ice pellets is much greater than that of the bacteria-containing ice pellets. The ice pellet band is finally formed through the nucleation of the small raindrop band and then the secondary ice production rate reaches its maximum value. These results indicate that a high concentration of ice particles can occur in WPSCCs when primary ice pellets appear even though their concentrations may be far less than those of ice-nucleating bacteria.

b. The impact of ice nucleus concentration on ice crystal multiplication

The production of primary ice pellets is influenced by the concentration of IN active at temperatures above $-10^\circ$C. There are large uncertainties in the measurement of IN in this temperature range (Mohler et al. 2007), and the concentration of IN may be lower than the value we used in the simulation described above. To show the impact of ice nucleus concentration on ice bursts, we simulated the ice formation process with different ice nucleus concentrations (Table 2). The highest bacterial concentration is 27 L$^{-1}$, among which the concentration of ice-nucleating bacteria is 0.27 L$^{-1}$; and the lowest bacterial concentration is 0.27 L$^{-1}$, among which the concentration of ice-nucleating bacteria is 0.0027 L$^{-1}$ at $-10^\circ$C. Table 3 shows the maximum ice particle concentrations, MPINR and MISPR values. The ice multiplication process occurred in all the simulations. The MPINR values increase with an increase of bacterial concentration. However, the MISPR values change slightly with an increase of bacterial concentration. These results indicate that ice multiplication in WPSCCs is insensitive to the concentration of IN. Figures 6c and 6d show the maximum concentration of ice particles and ice pellets in the simulation with a bacterial concentration of 0.27 L$^{-1}$. The maximum concentration of bacteria-containing ice particles is 0.02 L$^{-1}$ (Fig. 6c), which includes both ice particles that contain and do not contain ice-nucleating bacteria. We also simulated this case with the riming process omitted. The results show that the concentration of ice particles and the concentration of bacteria-containing
ice particles are the same and only reach $3.0 \times 10^{-4}$ L$^{-1}$ because there is no secondary ice particle formation. These results indicate that out of the initial ice-nucleating bacteria concentration of $2.7 \times 10^{-3}$ L$^{-1}$, a concentration of $3.0 \times 10^{-4}$ L$^{-1}$ was activated. The maximum concentration of activated ice-nucleating bacteria in experiment E1 may be approximately one-tenth of their initial concentration or even less due to the scavenging of secondary ice particles. The small raindrop band is also nucleated even though the MPINR value is as low as $1.7 \times 10^{-5}$ L$^{-1}$ s$^{-1}$ (Fig. 6d).

Table 3 in the experiments from E1 to E5 demonstrates that there is little difference in the maximum concentration of ice crystals for different bacterial concentrations if ice multiplication occurs. Our simulations also show that the impact of ice nucleus concentration on the ice concentration in WPSCCs is little if the small raindrop band reaches the ice multiplication temperature zone.

6. Sensitivity study of the effects of aerosol concentrations on ice formation in WPSCCs

The impacts of aerosols on the microphysics of deep convective clouds have been studied in recent years (Khain and Pokrovsky 2004; Khain et al. 2004, 2005; Cui et al. 2006; van den Heever et al. 2006; Lee et al. 2009).

<table>
<thead>
<tr>
<th>Case</th>
<th>Maximum ice concentration (L$^{-1}$)</th>
<th>MPINR (L$^{-1}$ s$^{-1}$)</th>
<th>MISPR (L$^{-1}$ s$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>E1</td>
<td>139.243</td>
<td>$1.741 \times 10^{-3}$</td>
<td>1.239</td>
</tr>
<tr>
<td>E2</td>
<td>146.904</td>
<td>$8.661 \times 10^{-3}$</td>
<td>1.249</td>
</tr>
<tr>
<td>E3</td>
<td>153.175</td>
<td>$5.419 \times 10^{-3}$</td>
<td>1.219</td>
</tr>
<tr>
<td>E4</td>
<td>155.337</td>
<td>$1.038 \times 10^{-3}$</td>
<td>1.206</td>
</tr>
<tr>
<td>E5</td>
<td>157.316</td>
<td>$1.954 \times 10^{-3}$</td>
<td>1.198</td>
</tr>
<tr>
<td>E6</td>
<td>0.098</td>
<td>$8.901 \times 10^{-4}$</td>
<td>$8.853 \times 10^{-4}$</td>
</tr>
<tr>
<td>E7</td>
<td>30.011</td>
<td>$9.347 \times 10^{-4}$</td>
<td>0.300</td>
</tr>
<tr>
<td>E8</td>
<td>261.551</td>
<td>$1.138 \times 10^{-3}$</td>
<td>1.654</td>
</tr>
<tr>
<td>E9</td>
<td>549.649</td>
<td>$1.063 \times 10^{-3}$</td>
<td>5.099</td>
</tr>
</tbody>
</table>
However, high concentrations of ice crystals have been observed in the dissipating stage of WPSCCs (Mossop 1985; Hobbs and Rangno 1985; Rangno and Hobbs 1991; Blyth and Latham 1993; Rangno and Hobbs 2005). The role of aerosols in these clouds will be quite different than that in deep convective clouds. For example, the H–M mechanism is not a dominant source of ice particles in deep clouds, but it may be a dominant process in WPSCCs.

Ice crystal bursts occur in both maritime and continental WPSCCs even though the aerosol concentrations in them are different (Hallett et al. 1978; Hobbs and Rangno 1985; Rangno and Hobbs 1991; Blyth and Latham 1993; Rangno and Hobbs 2005). The role of aerosols in these clouds will be quite different than that in deep convective clouds. For example, the H–M mechanism is not a dominant source of ice particles in deep clouds, but it may be a dominant process in WPSCCs.

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The sensitivity simulations were run for five different scenarios of aerosol concentrations. These five cases are characterized as different air pollution situations.

**a. Effect on cloud drop spectra and precipitation**

The spatial and temporal evolution of cloud drop spectra depends on both cloud microphysics and dynamics and the interaction between them. For the same meteorological conditions, since water vapor supplies will be basically unchanged, an increase in cloud condensation nucleus concentration will result in a decrease in cloud drop sizes. Consequently, the height of the centers of maximum concentration of large cloud drops (diameters greater than 24 μm) increases with increasing aerosol concentration (Fig. 7). However, the concentration of large droplets increases with an increase of aerosol concentration and then decreases with

![FIG. 7. Spatial and temporal evolution of water drop concentrations in the inner cylinder for the cases of (a) E6, (b) E7, (c) E8, and (d) E9 in different size categories. Cloud drops with diameters > 24.0 μm (cm⁻³, shaded area), small raindrops with diameters > 600 μm (L⁻¹, dotted lines), and raindrops with diameters > 2 mm (m⁻³, solid lines).](image-url)
a further increase of aerosol concentration. Both the lowest concentration and highest concentration of aerosols result in low concentrations of large cloud drops. The decrease in size of cloud droplets leads to a delayed appearance of drizzle drops (diameters greater than 100 μm) and decreased concentrations of them (Fig. 7). As a result, both concentrations of raindrops and the rainwater content decrease with increasing concentrations of CCN beyond some optimum value, and precipitation is delayed (Figs. 3a, 4a, 7, and 8).

An increase of aerosol concentration with multiples of the maritime aerosol concentration can suppress precipitation in WPSCCs (Figs. 3a and 8). This result is consistent with that of 3D simulations for such clouds (Feingold et al. 2005; Khain et al. 2008).

**b. Effect on ice crystal spectra**

With an increase in aerosol concentration, the concentration of ice pellets generated by the freezing of raindrops may decrease due to the decrease in the concentrations of raindrops (diameters greater than 600 μm) (Fig. 7). However, ice multiplication is also highly related to the concentration of cloud droplets available to be rimed by ice pellets. Table 3 in the experiments from E6 to E9 shows that the maximum concentration of ice particles increases with an increase in concentration of CCN with little changes in MPINR and big changes in MISPR. The total concentration of large ice particles (diameters greater than 150 μm) increases through the riming process when high concentrations of cloud drops are present in the clouds (Fig. 9). The concentrations of ice crystals are not proportional to those of ice pellets and increase with increasing concentrations of CCN. This is because ice splinter production rates increase with the increasing concentrations of cloud droplets if the cloud droplet spectra still meet the requirements of the mechanism, even though the ice pellet concentrations decrease. Figure 8 also shows the concentration of ice crystals with diameters less than 20 μm, which includes primary ice crystals from condensation.
freezing at the cloud top, ice splinters produced in the zone, and small melted ice particles near the cloud base. The maximum values of ice splinter concentrations increase from $1.0 \times 10^{-4}$ to $1.0$ L$^{-1}$. Since high aerosol concentrations result in high concentrations of cloud drops, the ice splinter production rates for the cases with higher aerosol concentrations are larger than those for the cases with smaller ones if the cloud drop-size distribution meets the requirements for the mechanism to be effective. However, it should be noted that there must be an aerosol concentration limit beyond which riming efficiencies are reduced, so that the mechanism becomes ineffective.

**c. Effect on the ice water content and dynamics**

With the concentration of the riming-dominant ice particles increasing (Fig. 9), the ice water contents increase with an increase of the aerosol concentration. The maximum ice water content increases from 0.07 to more than 1.7 g m$^{-3}$ in continental polluted conditions. The resulting increase in latent heat release leads to strengthened updrafts in the upper part of clouds (Fig. 8). In addition, there are small increases in updraft at lower levels in the cloud due to reduced precipitation drag in the simulations with higher aerosol concentrations. However, with a low concentration of aerosols, more precipitation drag leads to a strong downdraft, which in turn triggers a relatively strong secondary convection (Fig. 8). The interaction between aerosols and cumulus clouds is a process involving both microphysics and dynamics. The impact of aerosols on the microphysics and precipitation has been understood in combination with the dynamical fields of cumulus clouds (Khain et al. 2005; Tao et al. 2007; Khain et al. 2008). It should be noted that ambient wind shear has not been considered in our 1.5D model. It should also be noted that the temperature of cloud base is more than 15°C in our case study. If the height of cloud base increases, then the concentration of raindrops is expected to decrease, the precipitation efficiency of such WPSCCs will be lower than those of the thicker WPSCCs we simulated, and the ice concentration will also decrease.

![Fig. 9. Spatial and temporal evolution of ice particle concentrations in the inner cylinder for the cases of (a) E6, (b) E7, (c) E8, and (d) E9 in different size categories. Small ice concentration (diameter < 20 μm) (L$^{-1}$, shaded area) and large ice concentration (diameter > 150 μm) (L$^{-1}$, solid lines).](image-url)
7. Simulations based on a laboratory constraint parameterization

Diehl and Wurzler (2004) proposed a parameterization constrained by results from laboratory experiments for different IN acting via the immersion freezing mode. The freezing of a water drop containing a specific ice nucleus is independent of the size of the IN and is based on a median freezing temperature that was derived from laboratory experiments. The Diehl and Wurzler parameterization is a function of temperature, ice nucleus composition, droplet volume, the cooling rate of clouds, and the concentration of soluble material in cloud droplets. It should be noted that insoluble aerosols implemented into the model only refer to ice-nucleating aerosols and exclude non-ice-nucleating aerosols in this parameterization scheme. We repeated the experiment of E4 with a concentration of ice-nucleating bacteria of $0.135 \text{L}^{-1}$ and the IN only acting through the immersion freezing mode.

Different nucleation modes can lead to different concentrations of primary ice particles in different regions of cumulus clouds. Figure 10 shows ice particle concentrations from this simulation. The maximum concentration of total ice particles and bacteria-containing ice particles reaches 140 and 0.002 $\text{L}^{-1}$ (Fig. 10a), respectively. Note that ice formation at the cloud top is delayed due to the absence of condensation nucleation. The maximum concentrations of both ice particles and bacteria-containing ice particles are less than those from the simulation that includes both condensation nucleation and immersion nucleation in the experiment E4 with the observational-constraint parameterization approach. This result is consistent with the simulation with a parcel model [see Eidhammer et al. (2009), their Fig. 7]. The parameterization of Diehl and Wurzler (2004) might underestimate ice concentrations at relatively warm temperatures. This result further indicates that condensation freezing at cloud top is also an important pathway for ice formation in cumulus clouds.

The above-mentioned modeling results imply that ice-nucleating bioaerosols can trigger the ice multiplication process in WPSCCs under different assumed ice nucleation parameterizations. However, ice multiplication in WPSCCs is also determined by many other factors (Sun et al. 2010), among which a small raindrop band plays a very important role. The concentration of small raindrops and the location of the small raindrop band are determined by both dynamical and microphysical processes. The initial temperature and humidity profiles and the environmental wind field all impact raindrop formation. Therefore, further in situ observations are needed in future.

8. Conclusions

We simulated ice initiation and ice multiplication in WPSCCs. Our results indicate that ice multiplication in these kinds of clouds can be explained by the H–M mechanism. The impacts of various microphysical processes on this mechanism have been evaluated. Different ice initiation mechanisms, ice nucleus concentrations, and concentrations of CCN all affect the ice multiplication process. However, a supercooled small raindrop band plays a central role in ice multiplication (Sun et al. 2010). The formation of raindrops and their subsequent freezing is a precondition for ice crystal bursts in WPSCCs. The idea of nucleation of supercooled raindrops to accelerate ice multiplication was proposed by Hallett et al. (1978).
several decades ago. Sun (2008) further proposed that there should be an upward-moving small raindrop band within cumulus clouds. Other factors affect the timing and the magnitude of ice multiplication.

The involvement of the ice multiplication mechanism leads to an additional way of ice formation in WPSCCs. An increase in the concentration of CCN in these kinds of clouds does not necessarily delay ice formation, even though the sizes of cloud drops become smaller. Moreover, the undelayed onset of ice crystal formation results in the formation of a greater ice water content through the riming process because more cloud drops are available at subfreezing heights. As a result, the development of clouds will be strengthened because of more latent heat release compared to clouds forming in environments with smaller concentrations of CCN. Therefore, the thermodynamic effect (Hegerl et al. 2007; Romps and Kuang 2010) that impacts the development of deep cumulus clouds is still meaningful for WPSCCs, but the mechanisms are different. Furthermore, an increase in the concentration of CCN can weaken the secondary convection, since the compensating downdraft of the initial convection will become less strong due to a reduced warm-rain precipitation drag.

Ice concentrations in WPSCCs are related to the concentrations of both CCN and IN. Ice-nucleating bacteria can trigger ice multiplication even though their concentrations are very small. Other ice-nucleating bioaerosols, such as fungi, pollen, and leaf debris, may also trigger ice multiplication (Sun 2008) even though there are no reports of in situ observations of their concentrations. Ice-nucleating bioaerosols dominate ice initiation at temperatures warmer than −8°C. Therefore, any parameterization of ice formation should consider bioaerosols’ effects on ice formation.

Our findings concerning the evolution of cumulus cloud spectra and the ice initiation and multiplication processes are based on a 1.5D nonhydrostatic convective cloud and aerosol interaction model. It should be emphasized that the 1D framework imposes some constraints on the generality of our results. A 1D framework cannot consider the impacts of environmental wind shear on the evolution of cloud drop spectra. Environmental wind shear can strengthen the lateral entrainment. Depending on whether the entrained air is dry or wet may affect the microphysics in different ways. Furthermore, there is evidence that a thin subsiding shell surrounds the cumulus cloud boundary (Rodts et al. 2003). Our model has only a simplified vortex circulation at cloud top and lateral mixing, which impacts the cloud physics. These deficiencies should be addressed in future studies, including numerical experiments in the framework of a 3D cloud model with bin-resolved microphysics.

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APPENDIX A

Explicit Expression for Microphysical Processes of CCN

With specific expressions for the microphysical processes of CCN, the number density distributions of hydrometeors with water mass \( m \) and CCN mass \( m_{ap} \), \( f_{wat}(m, m_{ap}) \) and \( f_{ice}(m, m_{ap}) \) are calculated by numerical integration of the following two equations:

\[
\frac{\partial f_{wat}(m, m_{ap})}{\partial t} = - \frac{\partial}{\partial m} \left[ f_{wat}(m, m_{ap}) \frac{dm}{dt}_{wat} \right]_{mic} - f_{wat}(m, m_{ap}) R_{hetero}(m) \\
- \int_{m_{ap}}^{\infty} f_{wat}(m, m_{ap}) \int_{m_{ap}}^{\infty} K(m', m) f_{ice}(m, m_{ap}) dm'_{ap} dm' \\
- \int_{m_{drop}}^{m^2/2} f_{wat}(m, m_{ap}) \int_{m_{ap}}^{m/m'} K(m, m') f_{wat}(m', m_{ap}) dm'_{ap} dm' \\
+ \int_{m_{drop}}^{m^2/2} f_{wat}(m - m', m_{ap} - m'_{ap}) K(m - m', m') f_{wat}(m', m'_{ap}) dm'_{ap} dm' \\
+ \frac{\partial}{\partial m} \left[ f_{ice}(m, m_{ap}) \frac{dm}{dt}_{melting} \right]_{mic} 
\]  

(A1)
\[
\frac{\partial f_{\text{ice}}(m, m_{\text{ap}})}{\partial t} \bigg|_{\text{mic}} = - \left[ \frac{\partial}{\partial m} \left[ f_{\text{ice}}(m, m_{\text{ap}}) \frac{dm}{dt} \right]_{\text{ice}} \right]_{\text{mic}} f_{\text{wat}}(m, m_{\text{ap}}) R_{\text{hetero}}(m)
\]

\[
- \int_{m_{\text{drop}}}^{\infty} f_{\text{ice}}(m, m_{\text{ap}}) K(m, m_{\text{drop}}) f_{\text{wat}}(m_{\text{drop}}, m_{\text{ap}}') dm_{\text{ap}}' dm_{\text{drop}}
\]

\[
+ \int_{m_{\text{drop}}}^{m_{\text{ice}}} m_{\text{ice}}^{m_{\text{drop}} - m_{\text{ap}}} f_{\text{wat}}(m_{\text{drop}}, m_{\text{ap}}') K(m - m_{\text{drop}}, m_{\text{ap}}) f_{\text{ice}}(m - m_{\text{drop}}, m_{\text{ap}} - m_{\text{ap}}') dm_{\text{ap}}' dm_{\text{drop}}
\]

\[
- \int_{m_{\text{ice}}}^{\infty} f_{\text{ice}}(m, m_{\text{ap}}) K(m, m_{\text{ap}}') Es_{\text{ice}}(m', m_{\text{ap}}') dm_{\text{ap}}' dm'
\]

\[
+ \int_{m_{\text{ice}}}^{m/2} m_{\text{ap}}^{m_{\text{ice}} - m_{\text{ap}}/2} f_{\text{ice}}(m - m_{\text{ap}} - m_{\text{ap}}) K(m - m_{\text{ap}}, m_{\text{ap}}') Es_{\text{ice}}(m', m_{\text{ap}}') dm_{\text{ap}}' dm'
\]

\[
- \left[ \frac{\partial}{\partial m} \left[ f_{\text{ice}}(m, m_{\text{ap}}) \frac{dm}{dt} \right]_{\text{melting}} \right]
\]

(A2)

where \( m_{\text{ice}} \) is the minimum mass of ice crystals, \( m_{\text{drop}} \) is the minimum mass of water drops, and \( m_{\text{ap}} \) is the minimum mass of CCN in all ice and water particles; \( R(m) \) is the drop freezing rate for a drop with water mass \( m \); \( k \) is the collection kernel between the two particles (Sun 2008). \( Es \) is the sticking efficiency between the two ice particles; we assumed that \( Es \) is 0.3 given by Latham and Saunders (1970). The flux method for finding the numerical solution of the stochastic collection process of the masses of both aerosol and water is presented by Bott (2000). This method is based on the assumption that the probability for the collision of two hydrometeors (ice or water) only depends on the water mass of them. However, in our simulations, the collection kernel between two hydrometeors depends on both their water mass and aerosol mass, which may slightly impact the aerosol redistribution after a collision. We assumed that the ice nucleation capabilities for drops with identical water mass but with different masses of CCN are the same. Under such an assumption, we can derive the freezing rates of drops containing CCN through that of drops containing IN as follows:

\[
R_{\text{hetero}}(m) = \int_{0}^{\infty} R_{\text{hetero}}(m, m_{\text{IN}}) g_{\text{wat}}(m, m_{\text{IN}}) dm_{\text{IN}}
\]

\[
\int_{0}^{\infty} f(m, m_{\text{ap}}) dm_{\text{ap}}
\]

(A3)

**APPENDIX B**

Explicit Expression for Microphysical Processes of IN

To determine changes in \( g_{\text{wat}}(m, m_{\text{IN}}) \) and \( g_{\text{ice}}(m, m_{\text{IN}}) \) due to riming and coalescence in which IN and IN-containing hydrometeors may collide with IN-free hydrometeors, we assumed that \( m_{\text{IN}} \) in \( g_{\text{wat}}(m, m_{\text{IN}}) \) and \( g_{\text{ice}}(m, m_{\text{IN}}) \) is equal to zero if water droplets/ice particles contain no IN as shown:

\[
g_{\text{wat}}(m, m_{\text{IN}} = 0) = \int_{m_{\text{ap}}}^{\infty} f_{\text{wat}}(m, m_{\text{ap}}') dm_{\text{ap}}'
\]

\[
- \int_{m_{\text{IN}}}^{\infty} g_{\text{wat}}(m, m_{\text{IN}}') dm_{\text{IN}}'
\]

(B1)

\[
g_{\text{ice}}(m, m_{\text{IN}} = 0) = \int_{m_{\text{ap}}}^{\infty} f_{\text{ice}}(m, m_{\text{ap}}') dm_{\text{ap}}'
\]

\[
- \int_{m_{\text{IN}}}^{\infty} g_{\text{ice}}(m, m_{\text{IN}}') dm_{\text{IN}}', \quad (B2)
\]

where \( m_{\text{IN}} \) is the mass of the smallest ice nucleus.

For the hydrometeors containing IN with mass \( m_{\text{IN}} \neq 0 \), \( g_{\text{wat}}(m, m_{\text{IN}}) \) and \( g_{\text{ice}}(m, m_{\text{IN}}) \) are calculated by numerical integration of the following equations:
\[
\frac{\partial g_{\text{wat}}(m, m_{IN})}{\partial t}_{\text{mic}} = - \frac{\partial}{\partial m} \left[ \frac{g_{\text{wat}}(m, m_{IN})}{\partial t} \right]_{\text{wat}} - g_{\text{wat}}(m, m_{IN})R_{\text{hetero}}(m, m_{IN})
\]
\[
- \int_{m_{\text{mic}}}^{\infty} g_{\text{wat}}(m, m_{IN}) \int_{0}^{\infty} K(m_{\text{ice}}, m) g_{\text{ice}}(m_{\text{ice}}, m) \int_{IN}^{m_{\text{IN}}} dm_{\text{IN}} \frac{dm_{\text{IN}}}{dm_{\text{ice}}}
\]
\[
- \int_{m_{\text{drop}}}^{\infty} g_{\text{wat}}(m, m_{IN}) \int_{0}^{\infty} K(m, m_{\text{drop}}) g_{\text{wat}}(m_{\text{drop}}, m_{\text{drop}}') \int_{IN}^{m_{\text{IN}}} dm_{\text{IN}} \frac{dm_{\text{IN}}}{dm_{\text{drop}}}
\]
\[
+ \int_{m_{\text{drop}}}^{m/2} m_{\text{drop}} \int_{0}^{m_{\text{IN}}} g_{\text{wat}}(m_{\text{drop}}, m_{\text{IN}}) K(m - m_{\text{drop}}, m_{\text{drop}}') g_{\text{wat}}(m - m_{\text{drop}}, m_{\text{IN}} - m_{\text{IN}}') \int_{IN}^{m_{\text{IN}}} dm_{\text{IN}} \frac{dm_{\text{IN}}}{dm_{\text{drop}}}
\]
\[
+ \frac{\partial}{\partial m} \left[ \frac{g_{\text{ice}}(m, m_{IN})}{\partial t} \right]_{\text{melting}}
\]
\[
\text{(B3)}
\]

\[
\frac{\partial g_{\text{ice}}(m, m_{IN})}{\partial t}_{\text{mic}} = - \frac{\partial}{\partial m} \left[ \frac{g_{\text{ice}}(m, m_{IN})}{\partial t} \right]_{\text{ice}} + g_{\text{wat}}(m)R_{\text{hetero}}(m, m_{IN})
\]
\[
- \int_{m_{\text{drop}}}^{\infty} g_{\text{ice}}(m, m_{IN}) \int_{0}^{\infty} K(m_{\text{drop}}, m) g_{\text{wat}}(m_{\text{drop}}, m_{\text{drop}}') \int_{IN}^{m_{\text{IN}}} dm_{\text{IN}} \frac{dm_{\text{IN}}}{dm_{\text{drop}}}
\]
\[
+ \int_{m_{\text{drop}}}^{m/2} m_{\text{drop}} \int_{0}^{m_{\text{IN}}} g_{\text{ice}}(m_{\text{drop}}, m_{\text{IN}}) K(m - m_{\text{drop}}, m_{\text{drop}}') g_{\text{ice}}(m - m_{\text{drop}}, m_{\text{IN}} - m_{\text{IN}}') \int_{IN}^{m_{\text{IN}}} dm_{\text{IN}} \frac{dm_{\text{IN}}}{dm_{\text{drop}}}
\]
\[
- \int_{m_{\text{ice}}}^{\infty} g_{\text{ice}}(m, m_{IN}) \int_{0}^{\infty} K(m_{\text{ice}}, m) g_{\text{ice}}(m_{\text{ice}}, m_{\text{drop}}') \int_{IN}^{m_{\text{IN}}} dm_{\text{IN}} \frac{dm_{\text{IN}}}{dm_{\text{ice}}}
\]
\[
+ \int_{m_{\text{ice}}}^{m/2} m_{\text{ice}} \int_{0}^{m_{\text{IN}}} g_{\text{ice}}(m_{\text{ice}}, m_{\text{drop}}') K(m - m_{\text{ice}}, m_{\text{drop}}') g_{\text{ice}}(m - m_{\text{ice}}, m_{\text{IN}} - m_{\text{IN}}') \int_{IN}^{m_{\text{IN}}} dm_{\text{IN}} \frac{dm_{\text{IN}}}{dm_{\text{ice}}}
\]
\[
+ \frac{\partial}{\partial m} \left[ \frac{g_{\text{ice}}(m, m_{IN})}{\partial t} \right]_{\text{melting}}
\]
\[
\text{(B4)}
\]

**APPENDIX C**

**Parameterization Scheme for the Secondary Ice Production Rate**

The secondary ice production rate parameterized by Harris-Hobbs and Cooper (1987) is determined as follows:

\[
P = Cf(T) \int_{r_{13}}^{R_{a}} g(R) \pi(R + r)^2 |V(R)| \left( -\nu(r)N(R)n(r)E(R, r) dR dr \right)
\]
\[
\text{(C1)}
\]
\[
f(T) = \begin{cases} 
-(T + 3)/2, & -35 > T \geq -55, \\
(T + 8)/3, & -55 \geq T > -85, \\
0, & \text{otherwise},
\end{cases}
\]
\[
\text{(C2)}
\]
\[
G_x = \int n(r)^2 E(R, r) dr,
\]
\[
\text{(C6)}
\]
\[
h(R) = G_{c13}/G_{all}
\]
\[
\text{(C5)}
\]

where $E(R, r)$ is the collision efficiency between large ice particles and cloud drops; $R$, $V(R)$, and $N(R)$ are respectively the equivalent radius, terminal velocity, and size spectrum for large ice particles; $\nu(r)$, and $n(r)$ are respectively the cloud droplet radius, terminal velocity, and size spectrum; and $g(R)$ represents the importance of small cloud particles ($5 < \text{diameter} < 13 \mu m$) in the ice multiplication process, shown as

where the limits of integration cover radii from 2.5 to 6.5 $\mu m$ for $G_x = G_{c13}$ and cover all droplet sizes for $G_x = G_{all}$, $C$ is a constant, and $C = 0.21$. 

The above-mentioned production rate of secondary ice particles $P$ is determined according to the assumption that ice fragments are produced when large droplets ($>24.0 \mu m$ in diameter) strike a portion of the rimed ice pellet’s surface where small droplets ($<13.0 \mu m$) have accreted (Mossop and Wishart 1978). We applied this method in our modeling study.

REFERENCES


