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**New cloud
microphysical model
and parameterization**

N. Kuba and Y. Fujiyoshi

Development of a cloud microphysical model and parameterizations to describe the effect of CCN on warm cloud

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First, a hybrid cloud microphysical model was developed that incorporates both Lagrangian and Eulerian frameworks to study quantitatively the effect of cloud condensation nuclei (CCN) on the precipitation of warm clouds. A parcel model and a grid model comprise the cloud model. The condensation growth of CCN in each parcel is estimated in a Lagrangian framework. Changes in cloud droplet size distribution arising from condensation and coalescence are calculated on grid points using a two-moment bin method in a semi-Lagrangian framework. Sedimentation and advection are estimated in the Eulerian framework between grid points. Results from the cloud model show that an increase in the number of CCN affects both the amount and the location of precipitation. Additionally, results from the hybrid microphysical model and Kessler's parameterization were compared.

Second, new parameterizations were developed that estimate the number and size distribution of cloud droplets given the updraft velocity and the number of CCN. The parameterizations were derived from the results of numerous numerical experiments that used the cloud microphysical parcel model. The input information of CCN for these parameterizations is only several values of CCN spectrum (they are given by CCN counter for example). It is more convenient than conventional parameterizations those need values concerned with CCN spectrum, C and k in the equation of $N=CS^k$, or, breadth, total number and median radius, for example. The new parameterizations' predictions of initial cloud droplet size distribution for the bin method were verified by using the aforesaid hybrid microphysical model. The newly developed parameterizations will save computing time, and can effectively approximate components of cloud microphysics in a non-hydrostatic cloud model. The parameterizations are useful not only in the bin method in the regional cloud-resolving model but also both for a two-moment bulk microphysical model and for a global model. The effects of sea salt, sulfate, and organic carbon particles were also studied with these parameterizations and global model.

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1. Introduction

Accurate representation of the effect of aerosols on precipitation and on the optical properties of clouds is a key to improving climate models. Previous studies have shown that variations in cloud nuclei are primarily responsible for variations in cloud droplet concentrations and colloidal stability (Twomey and Squires, 1959; Twomey and Warner, 1967; Harshvardhan, 2002). Differences in cloud microstructure cause differences in optical properties, precipitation efficiency, and cloud lifetime (Twomey, 1974; Albrecht, 1989). For example, an increase in the number of aerosol particles suppresses precipitation (Rosenfeld, 1999, 2000; Andreae et al., 2004; Givanti and Rosenfeld, 2004). It is therefore desirable to use cloud microphysical models that can simulate the effects of cloud condensation nuclei (CCN) on the cloud microstructure in climate or cloud models.

Many studies have used detailed microphysical models (Clark, 1973; Takahashi, 1976; Hall, 1980; Reisin et al., 1996; Khain et al., 1999). Vertical intervals between grid points in those studies exceeded 100 m, but grid point supersaturation values were used to estimate CCN nucleation. The number concentration of cloud droplets significantly influences the radiative properties and precipitation efficiency of clouds and depends on the CCN spectrum and maximum value of supersaturation near the cloud base. Supersaturation changes very rapidly near the cloud base, so its maximum value cannot be estimated from values on grid points with intervals exceeding tens of meters. Furthermore, supersaturation is affected by updraft velocity and by the number of activated nuclei. It is therefore advantageous to calculate CCN growth by condensation in a Lagrangian framework to determine if and when each nucleus is activated.

Chen and Lamb (1999) used a detailed microphysical and chemical cloud model (Chen and Lamb, 1994) to simulate orographic cloud formation. A Lagrangian approach using 15 parcels to represent 15 vertical layers was used to examine microphysical processes. The results agreed with observations, but it is difficult to apply the technique to different situations. Khairoutdinov and Kogan (1999) developed a large

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eddy simulation model that includes an explicit microphysics scheme. Their grid point vertical interval (25 m) is smaller than previous studies but not small enough to allow an accurate estimate of maximum supersaturation near the cloud base. Furthermore, their CCN classification cannot accurately estimate the number of activated CCN (number of cloud droplets).

Rainfall amounts and precipitation area are affected by raindrop fall velocity, which has a wide distribution (from 10 cm s^{-1} to 10 m s^{-1}). Differences in fall velocity cannot be neglected if precipitation is to be simulated precisely, yet conventional bulk models put all rainwater in only one category. Cotton et al. (2003) showed that the Colorado State University Regional Atmospheric Modeling System (RAMS), which has a bulk microphysical model but simulates sedimentation by emulating a full-bin model, can get the results agreeing with the results from a full bin resolving microphysics model in a large eddy simulation (LES). It shows that the difference in fall velocity caused by the difference in the size of droplets is important. A bin model can resolve differences not only in fall velocity but also in collection efficiency more accurately according to the resolution of the bin framework.

The microphysical model in this study is based on the two-moment bin method developed by Chen and Lamb (1994). In addition, a Lagrangian framework is used to estimate the CCN activation and to give the initial cloud droplet size distribution for the bin model. The model estimates both the condensational growth of each cloud condensation nucleus and the time change of supersaturation in a Lagrangian framework. Therefore, the number concentration of activated CCN and the size distribution of cloud droplets can be estimated accurately. As a result, the relationship between CCN and precipitation efficiency in warm clouds can be studied precisely.

The model developed in this study can also be used to verify and improve the microphysical bulk model. It is difficult, however, to install this hybrid cloud microphysical model with both Lagrangian and Eulerian frameworks into a non-hydrostatic 3-D cloud model. It is also difficult to input a full CCN spectrum into a simulation. Therefore, parameterizations were developed that predict the initial cloud droplet size distribution for

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a cloud microphysical model that uses the bin method. Input CCN data for the parameterizations can be simplified if the most influential factor can be found. The impacts on precipitation of small particle CCN, large particle CCN, and giant particle CCN were estimated systematically using the hybrid cloud microphysical model. The parameterizations developed were then verified using the hybrid cloud microphysical model. The parameterizations thus developed are useful not only for the bin method in regional cloud-resolving models but also for bulk microphysical models in global models.

2. Model description

The hybrid microphysical cloud model was developed to produce accurate estimates of the number concentration of cloud droplets and the effect of CCN on the microstructure of clouds. This hybrid cloud microphysical model estimates the maximum values of supersaturation and the number concentration of cloud droplets using a parcel model with a Lagrangian framework. This hybrid cloud microphysical model also uses a bin model in the grid point model with semi-Lagrangian or Eulerian frameworks to estimate condensation, coalescence, sedimentation, and advection of cloud droplets and raindrops. Table 1 shows the two schemes.

2.1. Cloud microphysical model (Lagrangian framework)

CCN activation for each grid point is estimated by a parcel model. When the relative humidity at a grid point reaches 100% for the first time, or when relative humidity at a grid point exceeds 100% but no cloud water exists on the windward side of the grid point, an air parcel that includes CCN and vapor starts to rise from the windward side of the grid point.

CCN activation and the initial growth of cloud droplets by condensation are computed in a Lagrangian framework that incorporates the solute effect on CCN. This method precludes numerical diffusion of droplet size distribution. CCN size distributions are

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approximated using discrete radii classes with prudent choices of initial values. The number of CCN included in one class should be small compared to the number of activated cloud droplets. In this study, CCN (up to $12\ \mu\text{m}$ in radius) are partitioned into 185 separate classes, of which more than 100 classes can be activated and evolve into cloud droplets. The minimum input CCN radius must be smaller than the radius of the smallest CCN that can be activated, because the simulated results would be invalid if all CCN were activated.

Considering the time constants at which CCN reach their equilibrium radii in the ambient humidity (100% at cloud base), CCN smaller than $0.1\ \mu\text{m}$ in radius, and CCN larger than $1\ \mu\text{m}$ in radius, are initially assumed to be in equilibrium at 99% and 90% RH , respectively. Intermediate CCN are initially assumed to be in equilibrium between 99 and 90% RH as a function of radius.

The time evolution of the representative droplet radius in each class is calculated as detailed in Takeda and Kuba (1982) and Kuba et al. (2003). The time step in this model is 0.05 s. The growth by condensation of each droplet during each step is limited so that the radius of each droplet does not exceed its equilibrium radius.

When droplets condensed on CCN grow large enough to be distinguished from non-activated nuclei, the cloud droplets' size distribution calculated by the parcel model is assigned to the grid point as the initial cloud droplet size distribution for the bin model. The mixing ratio of vapor and potential temperature in the parcel are also assigned to the grid point.

When relative humidity at a grid point exceeds 100% (cloud droplets exist already) but no cloud water exists on the windward side of the grid point, the inflow of cloud droplets from the windward side of the grid point is estimated by using the cloud droplet size distribution in the parcel model.

2.2. Cloud microphysical model (Semi-Lagrangian framework)

The cloud droplet size distribution at a grid point is formulated using bins of fixed masses. Representative masses for each bin are given by $x_i = x_1 2^{(i-1)/k}$, where k

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represents the fineness of classification. The time evolutions of the number and total mass (n_i and m_i , respectively) of droplets with masses between the two boundaries of the i -th bin, that is, between $b_i = x_i 2^{-1/2k}$ and $b_{i+1} = x_{i+1} 2^{-1/2k}$, are computed.

Time changes due to growth by condensation and coalescence are calculated in the semi-Lagrangian framework by using the two-moment bin method developed by Chen and Lamb (1994) as shown in the appendix to minimize numerical diffusion of cloud droplet size distribution. Additionally fine resolution ($k=2$ in this study) is used. There are 71 bins for radii between $1 \mu\text{m}$ and 3.25 mm .

The time steps for growth by condensation and coalescence are 0.5 s , but the time step for coalescence can be shortened to prevent multiple collisions in one time step (the shortest time step for coalescence is 0.005 s). Sedimentation and advection of droplets are estimated in the Eulerian framework among grid points.

The solute effect on cloud droplet growth by condensation is not considered here. This simplification introduces little error because almost all cloud droplets are sufficiently dilute at this stage, with the exception of those unusual droplets that form on rare, large CCN. However, these large drops grow mostly by coalescence and the condensational growth rate $(dr/dt)_{\text{cond}}$ is much smaller than r . Underestimating the growth rate due to condensation alone does not significantly affect estimates of their overall growth rate. Details on such underestimates of growth rate are in the appendix of Kuba et al. (2003).

3. Numerical experiments

The dynamical framework of this study was designed to test the warm rain microphysical model Case 1 of the fifth WMO Cloud Modeling Workshop (Szumowski et al., 1998). The dynamical cloud model predicts an evolving flow for 50 min and performs a two-dimensional advection of the temperature and water variables (domain: $9 \text{ km} \times 3 \text{ km}$, dx and dz : 50 m , dt : 3 s). The advection scheme is a modified version of that by Smolarkiewicz (1984). Figure 1 shows the wind field at 25 min , the time that corresponds

to the peak updraft.

This simple model cannot estimate the effect of rainfall-induced drag on the dynamics. The effect of the change in drag caused by CCN differences will be studied in subsequent work. However, the model can estimate the effects of CCN on cloud microstructure and rainwater generation.

3.1. Effect of small-particle CCN

We used the three CCN size distributions (A, B, and C) (Fig. 2) to study the effect of the number of small-particle CCN. A, B, and C are typical size distributions for clean maritime, lightly polluted maritime, and heavily polluted maritime CCN respectively. The difference between the three cases is the number of small CCN (radius <math>< 0.1 \mu\text{m}</math>). Their ratios are 1, 5, and 10, respectively. Table 2 lists the ratio of the number of CCN in each case to that in case A for the three size regimes (small, large, and giant particles). For simplicity, all CCN are assumed to be NaCl and all grid points have same CCN size distribution in this study.

Figure 3 shows cloud droplet size distributions at the cloud base, at the center of the cloud, at 5.5 min for the three cases. The concentrations of cloud droplets for cases A, B, and C are 103, 325, and 550 cm^{-3} , respectively. Larger numbers of small-particle CCN lead to a smaller mode radius for cloud droplets. Figure 4a shows the accumulated rainfall at 50 min for three cases. The figure also lists values averaged in the domain. Smaller rainfall amounts result from the case with larger numbers of CCN, in agreement with many studies (Twomey, 1974; Albrecht, 1989; Saleeby and Cotton, 2004). It is also shown in Fig. 4a that the more CCN lead to the more widespread rainfall area. This study shows that rainfall area is also affected by the number of CCN because increased numbers of small CCN significantly reduce not only the rainwater production rate but also the fall velocity of the raindrops as a result of their smaller size. Small rain droplets can not fall against the strong updraft at the center of this cloud, they need longer time to become large enough to fall even in no updraft area (outer area in this study).

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Kessler's parameterization was installed on the same kinematic framework for comparison. The parameterization is expressed as follows (Cotton and Anthes, 1989):

$$R = \alpha(Q_c - Q_{c0})H(Q_c - Q_{c0}) + \beta Q_c Q_r^{0.875} \quad (1)$$

where R is the production rate of rainwater, Q_c and Q_r are the mixing ratios of cloud water and rain water, and α , β and Q_{c0} are constants. $H(Q_c - Q_{c0})$ is the Heaviside function that is introduced to represent the threshold process. The following typical combination of values is used here, following the code distributed by Szumowski et al. (1989):

$$(\alpha, \beta, Q_{c0}) = (0.001, 2.2, 0.0005)$$

The terminal fall velocity V_t (cm s^{-1}) of rain water is assumed to be

$$V_t = V_0(1000\rho)^{-0.5}(\rho Q_r)^\gamma \quad (2)$$

where ρ is air density (g cm^{-3}). As in the code distributed by Szumowski et al. (1989), the following values are used:

$$(V_0, \gamma) = (36.34, 0.136).$$

Mean rainfall amount in the domain at 50 min calculated from simulations using these values is 1.37 mm. A situation similar to cases A, B, and C was simulated with different values of (α, β, Q_{c0}) as follows:

$$(\alpha, \beta, Q_{c0}) = (0.003, 2.2, 0.0005) \text{ for case } Ka$$

$$(\alpha, \beta, Q_{c0}) = (0.001, 1.1, 0.0005) \text{ for case } Kb$$

$$(\alpha, \beta, Q_{c0}) = (0.0005, 1.1, 0.001) \text{ for case } Kc.$$

Figure 4b shows the accumulated 50-min rainfall for these cases. The combination of coefficients produced a mean rainfall amount in the domain of 1.6 mm, 1.0 mm, or

0.5 mm, respectively. Adjusting the coefficients (α , β , Q_{c0}) controls the averaged accumulated rainfall. However the horizontal distributions of rainfall amounts in Fig. 4b are very different from those in Fig. 4a, because the bulk model cannot accurately express differences in fall velocity for raindrops with a wide range of radii ($40\ \mu\text{m}\sim 2\ \text{mm}$). On the other hand, the bin model can estimate the fall velocity of raindrops for each bin.

3.2. Effect of large-particle CCN

Cases D and E, described next, investigate the role of the large-particle CCN ($0.1\ \mu\text{m} < \text{radius} < 1\ \mu\text{m}$) in warm cloud. Case D adds large-particle CCN to case A (clean maritime case). Case E adds large-particle CCN to case C (small-particle rich case). In both cases large particle CCN are added mainly in the size range between 0.1 and $0.8\ \mu\text{m}$, moderately in the size ranges between 0.08 and $0.1\ \mu\text{m}$ and between 0.8 and $1.0\ \mu\text{m}$. Figure 5a shows the cloud droplet size distributions at the cloud base, at the center of the cloud, at 5.5 min for cases A and D. The cloud droplet concentrations for cases A and D are 103 and $209\ \text{cm}^{-3}$, respectively. Similarly, Fig. 5b shows cloud droplet size distributions for cases C and E; the cloud droplet concentrations are 550 and $583\ \text{cm}^{-3}$, respectively. Figure 5 shows that adding large-particle CCN reduces the mode radius of cloud droplets and that this effect is larger for cases with fewer CCN. If there are many CCN, adding large-particle CCN increases the number of large cloud droplets.

Figure 6a shows accumulated rainfall at 50 min for cases A and D. Adding large-particle CCN decreases the amount of rainfall for cases with small numbers of small-particle CCN. Figure 6b shows accumulated rainfall at 50 min for cases C and E. Adding large-particle CCN does not affect rainfall amounts when there are large numbers of small-particle CCN. Rainwater is produced mainly from water condensed on small-particle CCN for the cases with small numbers of CCN (cases A and D). In this case, adding large-particle CCN suppresses the growth rate of droplets condensed on small-particle CCN. Water condensed on large CCN does not produce a lot of rain in any case.

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3.3. Effect of giant-particle CCN

Cases F–I, described below, help define the role of giant-particle CCN (radius > 1 μm) in warm cloud. Case F adds giant CCN to case A (clean maritime case), and case G removes giant CCN from case A. Case H adds giant CCN to case C (small-particle rich case) and case I (continental case) removes giant CCN from case C.

Figures 7a and b show the cloud droplet size distribution at the cloud base, at the center of the cloud, at 5.5 min for cases A, F, G, C, H, and I; cloud droplet concentrations are 103, 102, 103 cm^{-3} , 550, 541, and 550 cm^{-3} , respectively. The addition of giant CCN does not affect the mode radius of cloud droplets, but it does affect the number of large cloud droplets. Figures 8a and b show accumulated rainfall at 50 min for cases A, F, G, C, H, and I. Figure 8a shows that the addition of giant-particle CCN increases the rainfall at the center of cloud. It is because that drops condensed on giant-particle CCN become large enough to fall against the strong updraft at the center of cloud. However many droplets condensed on CCN smaller than 1.0 μm , also grow enough to start coalescence growth, and become rain drops. Therefore addition of giant CCN does not dramatically affect rainfall for cases with small numbers of CCN (Fig. 8a). For cases with small numbers of small-particle CCN, rainwater mainly originates from water condensed on small-particle CCN. In contrast, if there are many small-particle CCN, the more giant-particle CCN leads to the higher amounts of rainfall (Fig. 8b). When there are many small-particle CCN, droplets condensed on small-particle CCN can not grow enough to start coalescence growth, and then rainwater originates mostly from water condensed on giant-particle CCN and droplets caught by droplet condensed on giant-particle CCN. These results agree with the results in Kuba and Takeda (1983), Cooper et al. (1997), Feingold et al. (1999), and Saleeby and Cotton (2004), in which they showed that the effect of giant-particle CCN on rainfall efficiency of clouds is most remarkable in cases with numerous small-particle CCN. Additionally this study shows clearer differences in rainfall amounts and in the area of precipitation that arise from differences in the CCN spectrum.

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4. Parameterizations

4.1. Parameterizations to predict the number of cloud droplets

Section 3 showed that the most influential CCN factor is the number of small-particle CCN. A parameterization is developed to relate cloud droplet number (N_d cm^{-3}) to the updraft velocity at the cloud base (V_{base} m s^{-1}) and the cumulative number of CCN that can be activated at $S\%$ supersaturation ($N_c(S)$ cm^{-3}) (Kuba et al., 2003; Kuba and Iwabuchi, 2003). In this study, those factors are extended for a wider range of updraft velocities, as in Kuba and Iwabuchi (2003). Figure 9 shows relationship between the number of cloud droplets and the number of CCN that can be activated at fixed supersaturation $S\%$. The results of the numerical simulations with many kinds of CCN size distributions using the cloud microphysical parcel model are shown by marks. Based on these fitting curves approximations are developed as follows;

$$N_d = AN_c(S)/(N_c(S) + B) \quad (3)$$

For $V_{\text{base}} \leq 0.24 \text{ ms}^{-1}$: $S = 0.2\%$
 $A = 4710 V_{\text{base}}^{1.19}$
 $B = 1090 V_{\text{base}} + 33.2$

For $0.24 \leq V_{\text{base}} \leq 0.5 \text{ ms}^{-1}$: $S = 0.4\%$
 $A = 11700 V_{\text{base}} - 1690$
 $B = 10600 V_{\text{base}} - 1480$

For $0.5 \leq V_{\text{base}} \leq 1.0 \text{ ms}^{-1}$: $S = 0.5\%$
 $A = 4300 V_{\text{base}}^{1.05}$
 $B = 2760 V_{\text{base}}^{0.755}$

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$$\begin{aligned} \text{For } 1.0 \leq V_{\text{base}} \leq 3.0 \text{ ms}^{-1} : S &= 1.0\% \\ A &= 7730 - 15\,800 \exp(-1.08 V_{\text{base}}) \\ B &= 6030 - 24\,100 \exp(-1.87 V_{\text{base}}) \\ \text{For } 3.0 \leq V_{\text{base}} \leq 10.0 \text{ ms}^{-1} : S &= 2.0\% \\ A &= 1140 V_{\text{base}} - 741 \\ B &= 909 V_{\text{base}} - 56.2 \end{aligned}$$

In case of the larger updraft, we need to count CCN number under the higher supersaturation to get the good correlation between CCN number and droplet number. Critical supersaturations of 0.2%, 0.4%, 0.5%, 1.0%, and 2.0% correspond to radii of $0.036 \mu\text{m}$ ($0.048 \mu\text{m}$), $0.023 \mu\text{m}$ ($0.031 \mu\text{m}$), $0.019 \mu\text{m}$ ($0.027 \mu\text{m}$), $0.012 \mu\text{m}$ ($0.017 \mu\text{m}$) and $0.0077 \mu\text{m}$ ($0.011 \mu\text{m}$) for dry nuclei of NaCl ($(\text{NH}_4)_2\text{SO}_4$), for example. $N_c(S)$ for $S=0.2\%$, 0.4% , 0.5% , 1.0% , or 2.0% reflects the number of small CCN and large CCN. The input information of CCN for these parameterizations is only several values of CCN spectrum (they are given by CCN counter for example). It is more convenient than conventional parameterizations those need values concerned with CCN spectrum, C and k in the equation of $N=CS^k$ (as in Twomey, 1959) or, breadth, total number and median radius, for example.

Typically, global models do not include cloud microphysical models; thus parameterizations will be useful in global models to approximate the effects of aerosol particles. This parameterizations was incorporated into the global model CCSR/NIES/FRCGC-AGCM, which also includes the aerosol transportation model SPRINTARS (Takemura et al., 2000, 2002), to test application of the parameterization. CCSR/NIES/FRCGC-AGCM is an atmospheric general circulation model that has been developed based on CCSR/NIES-AGCM (Numaguti, 1993; Numaguti et al., 1995). Particle radii that correspond to the critical supersaturations 0.2%, 0.4%, 0.5%, 1.0%, and 2.0%, and an assumed size distribution of CCN for each constituent are used to estimate the number of CCN $N_c(S)$, because SPRINTARS output is the global distribution of the masses of sea salt particles, sulfate particles, organic carbon particles, black carbon particles,

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and dust particles.

To calculate the particle radius that corresponds to each critical supersaturation, sea salt particles are assumed to be NaCl and sulfate particles are assumed to be $(\text{NH}_4)_2\text{SO}_4$. Many different chemical constituents comprise organic carbon particles. It is difficult to treat each chemical constituent separately, so an approximation is needed for the average nature of organic carbon particles in the real atmosphere. The first step is to adopt the approximation derived by Ghan et al. (2001): the material density and hygroscopicity of organic carbon particles are approximately 1 and 0.14, respectively (see their Table 1). These values allow a calculation of the dry radius for each supersaturation. Critical supersaturations of 0.2%, 0.4%, 0.5%, 1.0%, and 2.0% correspond to radii of $0.074 \mu\text{m}$, $0.047 \mu\text{m}$, $0.040 \mu\text{m}$, $0.025 \mu\text{m}$, and $0.016 \mu\text{m}$ for a dry nucleus of organic carbon particles. Black carbon and dust particles are assumed insoluble; they become CCN only when coated by water-soluble constituents. Black carbon and dust particles can therefore be excluded from the CCN if sea salt, sulfate, and organic carbon aerosol particles are estimated sufficiently without regard to whether they are mixed with insoluble matter.

Because CCSR/NIES/FRCGC-AGCM does not resolve the cloud updraft, turbulent kinetic energy is used to estimate updraft velocity as in Lohmann et al. (1999). Figure 10a shows the annual mean value (for 2000) of effective cloud droplet radii at cloud tops warmer than 273 K. These annual mean values were calculated by T. Takemura (personal communication) following the methods in Takemura et al. (2005). Figure 10a shows a land-ocean contrast of the effective radius of cloud droplets that is often retrieved from NOAA/AVHRR (Advanced Very High Resolution Radiometer) data (Nakajima and Nakajima, 1995; Kawamoto, 2001). To estimate the effect of organic carbon aerosol particles on the effective radius of cloud droplets, Fig. 10b shows the same calculations as in Fig. 10a, but organic carbon particles are neglected. Comparisons of Fig. 10a and b show that considering organic carbon particles leads to decrease in annual mean value of cloud droplet effective radius over South Africa, Australia, and South America. In these regions, concentration of organic carbon particles is not small

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compared with whole CCN concentration. Therefore, the effect of organic carbon particles on the cloud microstructure is not negligible over South Africa, Australia, and South America.

4.2. Parameterizations to predict the droplet size distribution

5 Both the number of cloud droplets and the shape of the distribution are needed to derive the initial cloud droplet size distribution in the bin model. Gamma distributions have been used in many studies to express the cloud droplet size distribution, and they are written as follows:

$$n(r) = C r^\beta \exp(-Dr) dr \quad (4)$$

$$10 \quad C = \frac{N_d}{\beta!} \left(\frac{4\pi}{3Q} (\beta + 3)(\beta + 2)(\beta + 1) N_d \right)^{\frac{\beta+1}{3}}$$

$$D = \left(\frac{4\pi}{3Q} (\beta + 3)(\beta + 2)(\beta + 1) N_d \right)^{\frac{1}{3}} .$$

Here, $n(r)$ is the number density of cloud droplets (cm^{-4}), N_d is the number concentration of cloud droplets (cm^{-3}), and Q is the cloud water (g cm^{-3}). β is an integer (2 or 4 in this study). When we can clearly distinguish cloud droplets from non-activated wet CCN and adjusted cloud water reaches the critical value, the gamma distribution is assigned to the bin as the initial cloud droplet size distribution.

5. Verification of parameterizations

20 The hybrid cloud microphysical model described in Sect. 2 is used as truth to verify the parameterizations in Sect. 4. The new model uses the parameterizations instead of the parcel model. Parameterization verification is achieved by comparing the results from

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the new model using both the parameterizations and bin method, with the results from the model using the hybrid cloud microphysical model that uses both a parcel model and bin method.

The procedure for using parameterizations to predict initial cloud droplet size distribution for the bin model instead of the parcel model is as follows:

1. When the condensed cloud water determined by adjusting the supersaturation at a grid point reaches 1.5×10^{-5} (g g^{-1}), N_d is derived by Eq. (3). This threshold was derived from many trials that tested whether cloud droplets could be clearly distinguished from non-activated CCN.
2. Using N_d and the adjusted condensed cloud water Q , the cloud droplet size distribution $n(r)$ (Eq. 4) is derived.
3. The number and total mass of droplets included in each bin are calculated by integrating Eq. (4).

Figure 11a shows the cloud droplet size distributions derived explicitly by the parcel model and approximated by Eqs. (3) and (4) for case A. Cloud droplet size distributions derived from the parameterization with a Gamma distribution are wider than those derived using the parcel model, and the large cloud droplets that condense on giant-particle CCN cannot be described. Differences in the cloud droplet size distribution between the parcel model and the parameterization are smaller at later stages and at higher levels in the cloud (Fig. 11b), except for large droplets condensed on giant-particle CCN. Because the numbers of cloud droplets in both the parcel model and the parameterization are same and the smaller droplets grow faster, the differences become smaller. However the lack of large cloud droplets condensed on giant-particle CCN in the cloud droplet size distribution derived from parameterization is not solved even in this stage. It is the restriction of this parameterization.

Figure 12a–c shows the accumulated surface rainfall at 50 min for cases A, B, and C, respectively. Differences between the results from the parcel model and those from the

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parameterizations are not large (mean values in the domain are also shown in Fig. 12). Parameterizations can express differences in rainfall amounts caused by differences in the number of small CCN. Using 4 as β offers little improvement over 2, so 2 is used from now on.

5 The effect of adding large or giant CCN was tested in cases for which large CCN or giant CCN were increased five-fold over case A (clean maritime case) or C (small-particle rich case). Figure 13a shows accumulated surface rainfall at 50 min for case A and the case with high concentrations of large CCN (case D) or giant CCN (case F) from the hybrid cloud model that uses the parcel method and two-moment bin method. Figure 13b is the same as Fig. 13a but shows the results from the cloud model that uses parameterization and the two-moment bin method. Adding large CCN reduces the rainfall amounts; adding giant CCN has little effect on the rainfall amounts. Comparisons of Figs. 13a and b show that the parameterization predicts the effects of adding large or giant CCN.

15 Figures 13c and d are similar to Figs. 13a and b, but large CCN (case E) or giant CCN (case H) are added to case C (small-particle rich case). As noted in Sect. 3.2, adding large CCN does not affect the rainfall amounts for this case. Adding giant CCN causes an increase in rainfall over the small-particle rich case (Fig. 13c for the hybrid cloud microphysical model) as shown in Sect. 3.3. In contrast, the method using parameterizations and the two-moment bin cannot model the effect of adding giant CCN (Fig. 13d). The parameterizations use the number of CCN, so differences in the number of giant CCN cannot be reflected in the initial cloud droplet size distribution of the bin model. However, the effect of giant CCN on warm rain is not so large for usual cases (cases A, B, and C, for example), as discussed in Sect. 3.3. Therefore these parameterizations may be useful for estimating the effect of CCN on warm rain in non-hydrostatic 3-D cloud models except for cases in which giant-particle CCN has large effect on precipitation. In these cases the hybrid cloud microphysical model that uses both parcel model and bin method is useful.

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6. Conclusions

A hybrid cloud microphysical model was developed that combines Lagrangian and Eulerian frameworks. The model can estimate the effect of CCN on cloud microstructure. The effect of CCN on warm cloud rainfall was studied using this microphysical model and a simple two-dimensional cloud model, and the following conclusions were reached:

1. The number of small-particle CCN has a large impact on the amount of rainfall. Larger numbers of small-particle CCN cause a smaller mode radius of cloud droplets at the cloud base and lead to lighter rainfall. Adding small-particle CCN reduces both the amount of rainfall and the fall velocity of raindrops, which affects the rainfall area.
2. Adding large-particle CCN leads to a decrease in the mode radius of cloud droplets at the cloud base when there are small numbers of small CCN. On the other hand, when there are large numbers of small-particle CCN, adding large-particle CCN leads to an increase in large cloud droplets at the cloud base. Adding large-particle CCN leads to a decrease in the amount of rainfall when there are small numbers of small-particle CCN; it does not affect the amount of rainfall when there are large numbers of small CCN. When there are few CCN, rainwater is produced from condensation onto small-particle CCN but not from condensation onto large-particle CCN.
3. When there are small numbers of small-particle CCN, adding giant-particle CCN leads to a slight decrease in rainfall, which suggests that almost all rain water is produced by condensation onto small-particle CCN. On the other hand, when there are large numbers of small-particle CCN, adding giant-particle CCN leads to a modest increase in the amount of rainfall, which suggests that rain water is produced mainly from condensation onto giant-particle CCN and small cloud droplets caught by large droplets condensed on giant-particle CCN.

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4. Rainfall calculated by a bulk microphysical model using Kessler's parameterization can be tuned by changing the coefficients. However, the relationship between the coefficients and CCN properties is unclear. Furthermore, the bulk model does not express the proper fall velocities of raindrops that are distributed over a wide velocity range. A high-resolution cloud model therefore needs a cloud microphysical model that uses a bin method.

The kinematic framework in this study is so simple that a change in cloud microstructure cannot affect cloud dynamics. Therefore, a quantitatively accurate estimate of the effect of CCN on warm rain cannot be studied here. However, the basic role of CCN in warm rain production can be estimated using the hybrid cloud microphysical model. The effect of CCN on warm rain can be estimated quantitatively when the hybrid cloud microphysical model is incorporated into a three-dimensional cloud dynamical model. In addition, the model can be used to verify and improve the microphysical bulk model.

Parameterizations to predict the initial cloud droplet size distribution for the bin model were developed. The results from a cloud model using these parameterizations and a two-moment bin model were compared to results from a hybrid cloud microphysical model that combines Lagrangian and Eulerian frameworks. Replacing the parcel model with the parameterizations developed in this study saves computing time at the cost of a small degree of error. A cloud model that includes the parameterizations and a two-moment bin model estimates the effects of CCN on cloud microstructure with sufficient accuracy. These parameterizations use only the number of CCN that can be activated under a certain supersaturation (0.2–2.0%). Thus, the effect of giant-particle CCN on warm rain precipitation cannot be estimated. However, the effect of giant-particle CCN is not large compared to the effects of small and large-particle CCN. If the effect of seeding giant-particle CCN must be estimated, a combination of two cloud-droplet size distributions can be used as the initial cloud droplet size distribution for the bin model, although only one gamma distribution was used in this study, or the hybrid cloud microphysical model developed in Sect. 2 in this study is useful. The developed parameterizations and the two-moment bin method are now running in

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a three-dimensional non-hydrostatic cloud model, CReSS (Tsuboki and Sakakibara, 2002). The parameterizations to predict the number of cloud droplets can be also applied to a GCM. GCM simulations that include an aerosol transport model and the developed parameterizations show that the effect of organic carbon particles on the cloud microstructure is not negligible over South Africa, Australia, and South America.

Appendix A

Two-moment bin method

The two-moment bin method was developed by Chen and Lamb (1994) and described in detail in their paper. Only highlights are described here.

A.1. Growth by condensation

1) The growth of the boundary of a bin is calculated by

$$b'_i = b_i + \Delta t \left(\frac{dx}{dt} \right)_{x=b_i}^{\text{cond}} \quad (\text{A1})$$

where b_i ($i=1, 2, \dots, im$) is the fixed boundary of the i -th bin and $\left(\frac{dx}{dt} \right)^{\text{cond}}$ is the condensational growth rate of mass.

2) The number and total mass of droplets with masses between b'_i and b'_{i+1} at time $t+\Delta t$ are expressed as N_i and M_i , as follows:

$$N_i = n_i \quad (\text{A2})$$

$$M_i = m_i + n_i \Delta t \left(\frac{dx}{dt} \right)_{x=m_i/n_i}^{\text{cond}} \quad (\text{A3})$$

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where n_i and m_i are the number and the total mass of cloud droplets included in the i -th bin at time t .

3) The number and total mass of droplets with masses between b'_i and b'_{i+1} at time $t + \Delta t$ can be expressed as the integral of the density function $f(x)$ and $f(x)x$ between b'_i and b'_{i+1} . $f(x)$ is approximated as

$$\begin{aligned} f(x) &= A + B(x - x_0) \\ x_0 &= (b'_i + b'_{i+1})/2 \end{aligned} \quad (\text{A4})$$

$$\int_{b'_i}^{b'_{i+1}} f(x) dx = N_i \quad (\text{A5})$$

$$\int_{b'_i}^{b'_{i+1}} f(x) x dx = M_i \quad (\text{A6})$$

Then A and B are

$$A = N_i / (b'_{i+1} - b'_i) \quad (\text{A7})$$

$$B = 12(M_i - x_0 N_i) / (b'_{i+1} - b'_i)^3 \quad (\text{A8})$$

4) New values n'_i or m'_i for the fixed bin are calculated by integrating the function $f(x)$ or $f(x)x$ for each suitable region.

A.2. Growth by coalescence

5) New particles produced by collision of particles in the i -th and j -th bins are included in the range between x_1 and x_2 .

$$x_1 = b_i + b_j \quad (\text{A9})$$

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$$x_2 = b_{i+1} + b_{j+1} \quad (\text{A10})$$

6) The number and total mass of new droplets with masses between x_1 and x_2 are expressed as N and M :

$$N = n_i n_j K \left(\frac{m_i}{n_i}, \frac{m_j}{n_j} \right) \Delta t \quad (\text{A11})$$

$$M = \left(\frac{m_i}{n_i} + \frac{m_j}{n_j} \right) N \quad (\text{A12})$$

where K is the particle interaction kernel.

7) The density function of the new droplets is approximated as

$$f(x) = A + B(x - x_0) \quad (\text{A13})$$

$$x_0 = (x_1 + x_2)/2$$

$$\int_{x_1}^{x_2} f(x) dx = N \quad (\text{A14})$$

$$\int_{x_1}^{x_2} f(x) x dx = M \quad (\text{A15})$$

Then A and B are

$$A = N / (x_2 - x_1) \quad (\text{A16})$$

$$B = 12(M - x_0 N) / (x_2 - x_1)^3 \quad (\text{A17})$$

8) The values n_i and m_i that should be added to the original value of the fixed bin are calculated by integrating the function $f(x)$ or $f(x)x$ for each suitable region. The concentration N is subtracted from the original values of the i -th and j -th bins. The mass of Nm_i/n_i or Nm_j/n_j is also subtracted from the original total mass of the i -th or j -th bins.

A.3. Incomplete distribution (for both condensation or coalescence processes)

When the function $f(x)$ is a negative value at the boundary of the region ($b'_i < x < b'_{i+1}$) or ($x_1 < x < x_2$), it is modified as follows:

if $f(b'_{i+1}) < 0$ or $f(x_2) < 0$;

$$f(x) = B'(x - x^*) \text{ for } b'_i < x < x^* \text{ or } x_1 < x < x^*$$

$$f(x^*) = 0$$

$$f(x) = 0 \text{ for } x^* < x < b'_{i+1} \text{ or } x^* < x < x_2 \quad (\text{A18})$$

where $B' = -2N_i(b'_i - x^*)$ or $B' = -2N(x_1 - x^*)$

$$x^* = 3M_i/N_i - 2b_i \text{ or } x^* = 3M/N - 2x_1.$$

if $f(b'_i) < 0$ or $f(x_1) < 0$; (A19)

$f(x) = B'(x - x^*)$ for $x^* < x < b'_{i+1}$ or $x^* < x < x_2$

$$f(x^*) = 0$$

$$f(x) = 0 \text{ for } b'_i < x < x^* \text{ or } x_1 < x < x^* \quad (\text{A20})$$

where $B' = 2N_i(b'_{i+1} - x^*)$ or $B' = 2N(x_2 - x^*)$ (A21)

$$x^* = 3M_i/N_i - 2b_{i+1} \text{ or } x^* = 3M/N - 2x_2. \quad (\text{A22})$$

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Table 1. The two computational schemes for cloud microphysical model.

	Parcel model	Bin model
Framework	Lagrangian	semi-Lagrangian
Fixed values	n_j Concentration of CCN included in each class. ($j=1, \dots, 185$)	$x_i = x_1 2^{(i-1)/k}$ Representative mass of droplets included in each bin. ($k=2, i=1, \dots, 71$)
Variable values	$x_j(t)$ Mass of droplets forming on CCN included in each class.	$n_i(t)$ Concentration of droplets included in each bin.
Activation	Takeda and Kuba (1982)	not considered
Condensation	Takeda and Kuba (1982)	Chen and Lamb (1994)
Coalescence	not considered	Chen and Lamb (1994)
Δt	0.05 s	0.5 s (condensation), <0.5 s (coalescence)

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Table 2. The ratio of the number of CCN in each case to that in case A.

Case	small particle CCN ($0.1 \mu\text{m} < \text{radius}$)	large particle CCN ($0.1 < \text{radius} < 1 \mu\text{m}$)	giant particle CCN ($1 \mu\text{m} < \text{radius}$)
A	1	1	1
B	5	1	1
C	10	1	1
D	1	5	1
E	10	5	1
F	1	1	5
G	1	1	0
H	10	1	5
I	10	1	0

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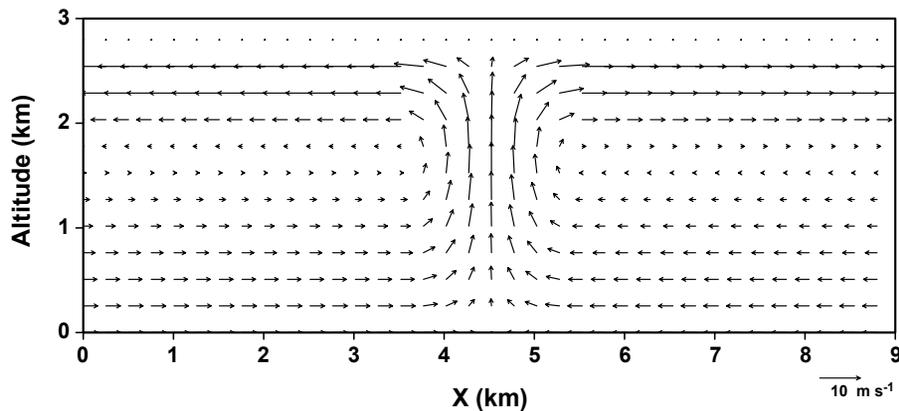


Fig. 1. Wind field at 25 min, corresponding to the peak updraft.

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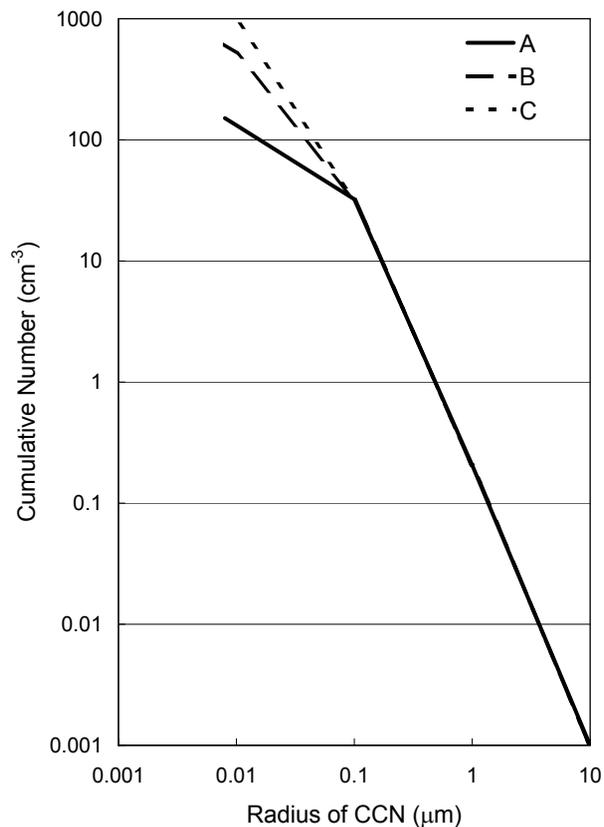


Fig. 2. Three CCN size distributions (A, B, and C) used in this study. The ratios of the number of small-particle CCN (radius <math> < 0.1 \mu\text{m}</math>) in B and C to A are 5 and 10, respectively.

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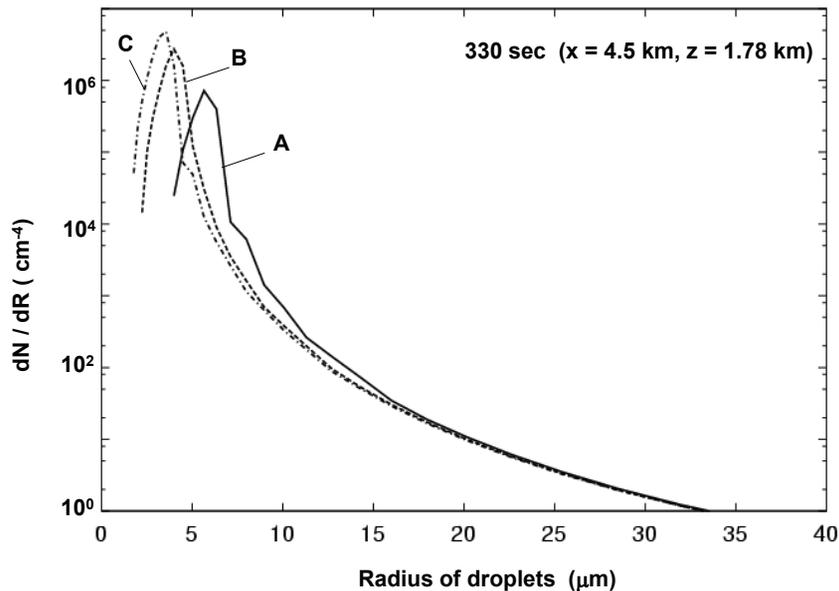


Fig. 3. Cloud droplet size distributions at the cloud base, at the center of the cloud, at 5.5 min for cases A, B, and C. The concentrations of cloud droplets in cases A, B, and C are 103 , 325 , and 550 cm^{-3} , respectively.

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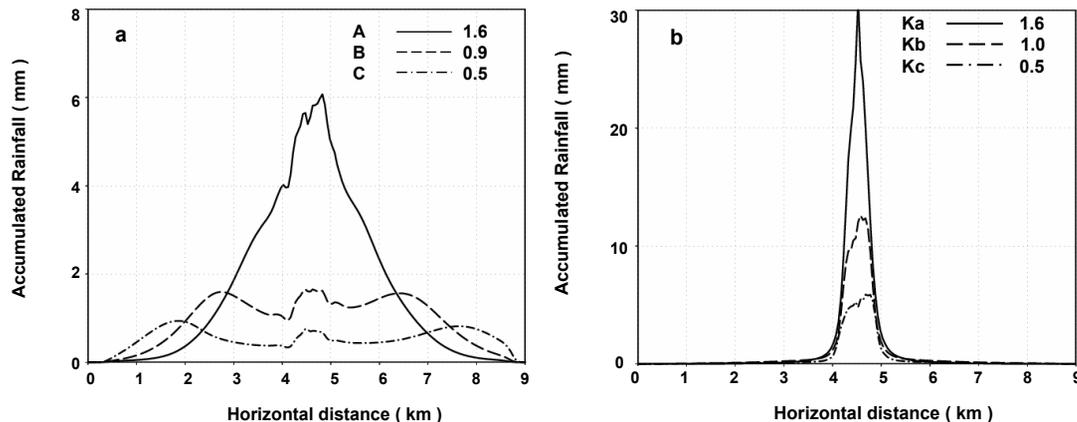


Fig. 4. Accumulated rainfall at 50 min. **(a)** Cases A, B, and C. Values averaged in the domain for cases A, B, and C are 1.6 mm, 0.9 mm, and 0.5 mm respectively. **(b)** Three cases using Kessler's parameterization. Values averaged in the domain for cases Ka, Kb, and Kc are 1.6 mm, 1.0 mm, and 0.5 mm, respectively.

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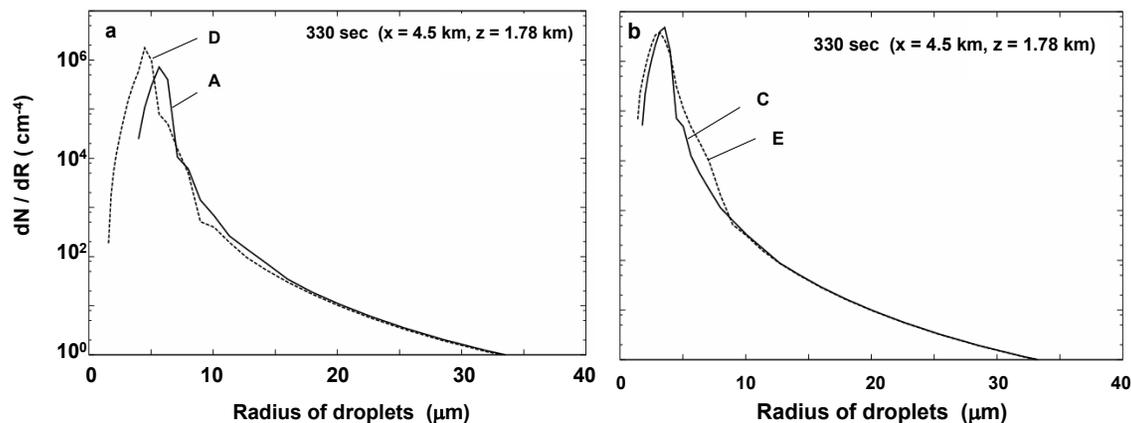


Fig. 5. Cloud droplet size distributions at the cloud base, at the center of the cloud, at 5.5 min. **(a)** Cases A and D (in case D, large-particle CCN are added to case A). The concentrations of cloud droplets in cases A and D are 103 and 209 cm^{-3} , respectively. **(b)** Cases C and E (in case E, large-particle CCN are added to case C). The concentrations of cloud droplets in cases C and E are 550 and 583 cm^{-3} , respectively.

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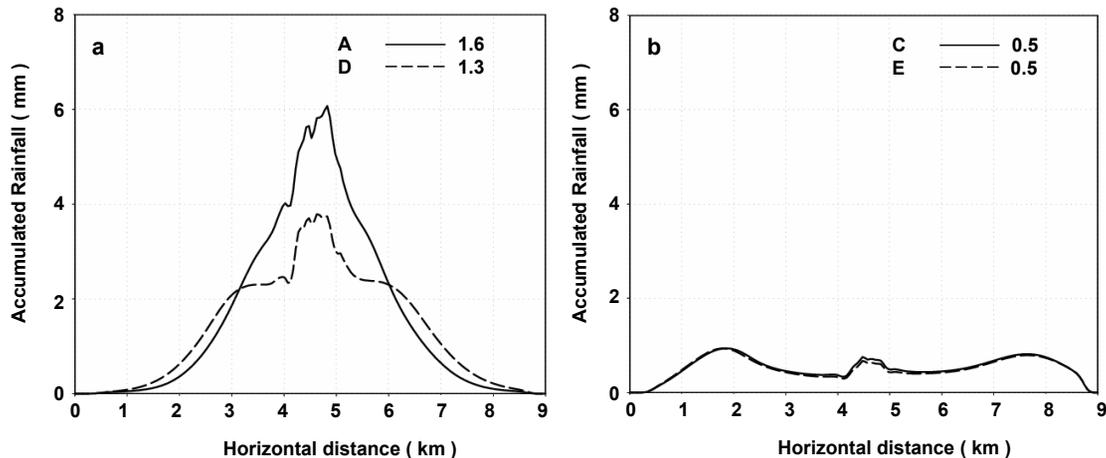


Fig. 6. Accumulated rainfall at 50 min. **(a)** Cases A and D. Values averaged in the domain for cases A and D are 1.6 mm and 1.3 mm, respectively. **(b)** Cases C and E. Values averaged in the domain for cases C and E are 0.5 mm and 0.5 mm, respectively.

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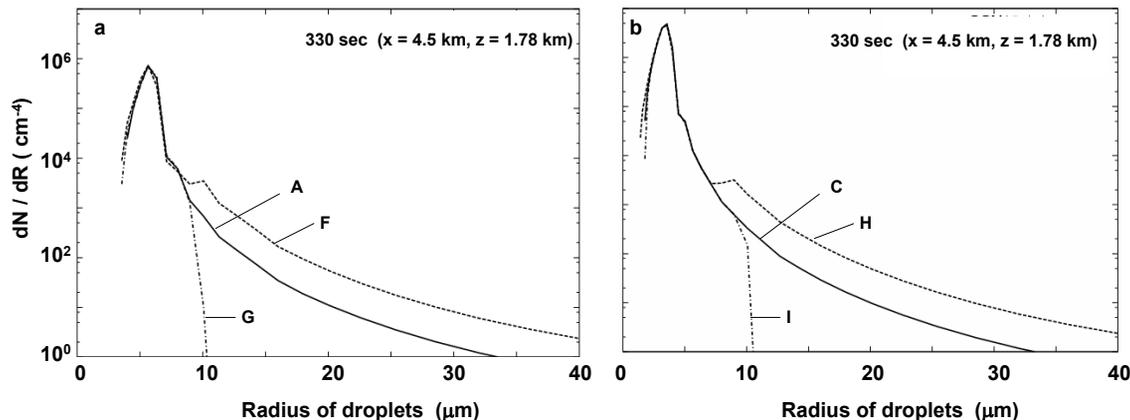


Fig. 7. Cloud droplet size distributions at the cloud base, at the center of the cloud, at 5.5 min. **(a)** Cases A, F, and G (Case F: giant CCN are added to case A; Case G: giant CCN are removed from case A). The concentrations of cloud droplets for cases A, F, and G are 103, 102, and 103 cm^{-3} , respectively. **(b)** Cases C, H, and I (Case H: giant CCN are added to case C; Case I: giant CCN are removed from case C). The concentrations of cloud droplets for cases C, H, and I are 550, 541, and 550 cm^{-3} , respectively.

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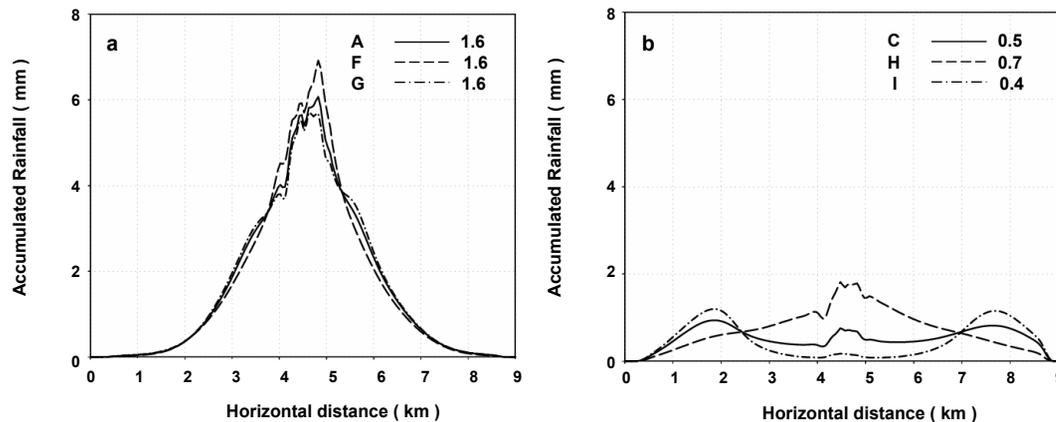


Fig. 8. Accumulated rainfall at 50 min. **(a)** Cases A, F, and G. Values averaged in the domain for cases A, F, and G are 1.6 mm, 1.6 mm, and 1.6 mm, respectively. **(b)** Cases C, H, and I. Values averaged in the domain for cases C, H, and I are 0.5 mm, 0.7 mm, and 0.4 mm, respectively.

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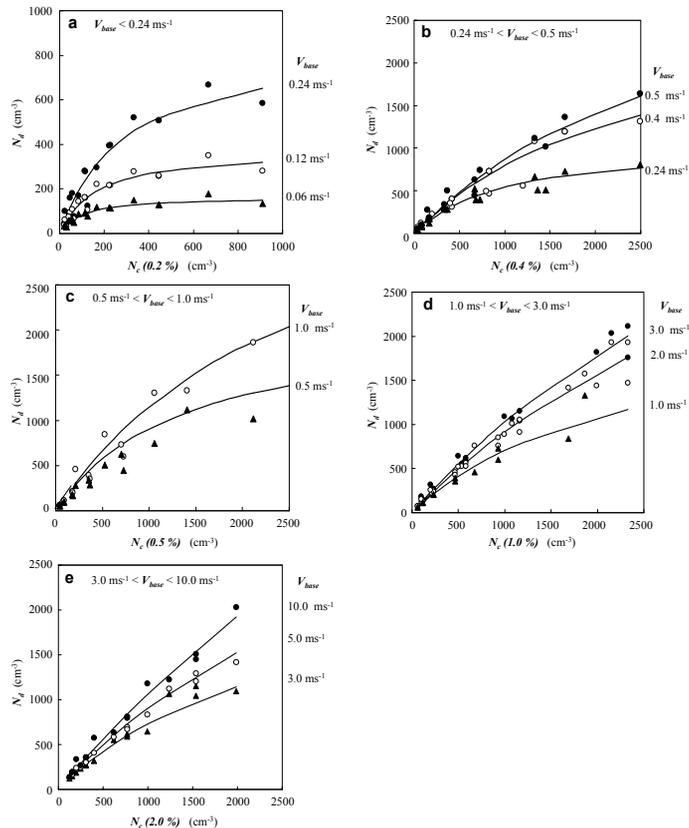


Fig. 9. Relationship between the number of cloud droplets (N_d) and the number of CCN $N_c(S)$ that can be activated at fixed supersaturation $S\%$. The results of the numerical simulations using the cloud microphysical parcel model are shown by marks; approximations using Eq. (1) are expressed by lines. The updraft velocity at the cloud base V_{base} varies by panel: **(a)** $V_{\text{base}} \leq 0.24 \text{ m s}^{-1}$ **(b)** $0.24 \leq V_{\text{base}} < 0.5 \text{ m s}^{-1}$ **(c)** $0.5 \leq V_{\text{base}} < 1.0 \text{ m s}^{-1}$ **(d)** $1.0 \leq V_{\text{base}} < 3.0 \text{ m s}^{-1}$ **(e)** $3.0 \leq V_{\text{base}} < 10.0 \text{ m s}^{-1}$.

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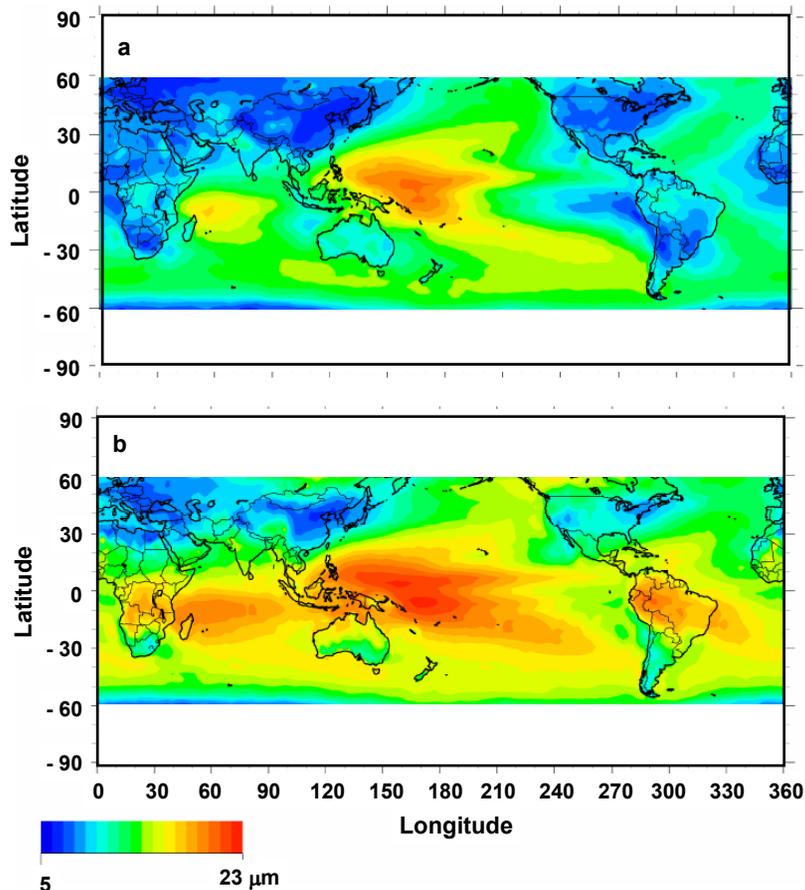


Fig. 10. Calculated annual mean (year 2000) of the effective radius of cloud droplets at the top of clouds that were warmer than 273 K. **(a)** Organic carbon aerosol particles are considered. **(b)** Organic carbon aerosol particles are ignored.

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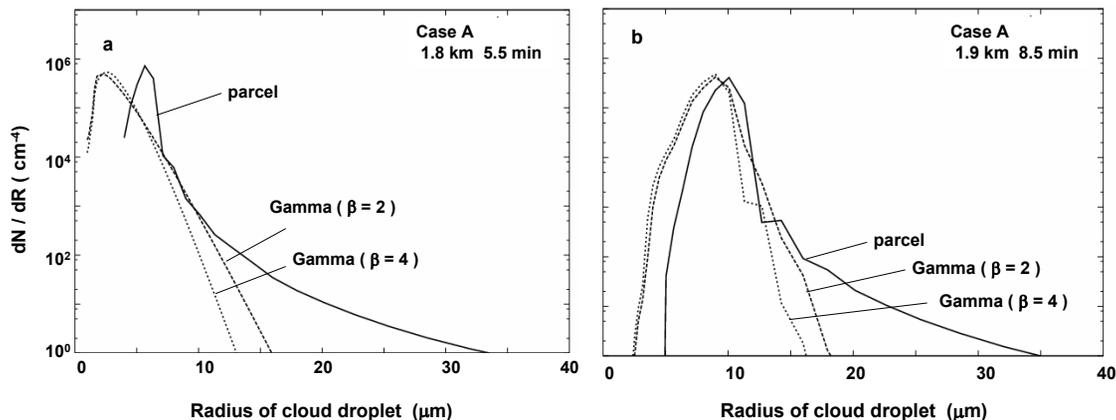


Fig. 11. Cloud droplet size distributions estimated using the parcel model and parameterized using Eqs. (1) and (2). CCN of case A in Fig. 2 are used here. **(a)** At the cloud base (1.775 km), at the cloud center, at 5.5 min. Concentrations of cloud droplets in the parcel model case and the parameterization using Eq. (1) case are 103 cm^{-3} and 116 cm^{-3} , respectively. **(b)** At a higher level (1.925 km), at the center of the cloud, at 8.5 min.

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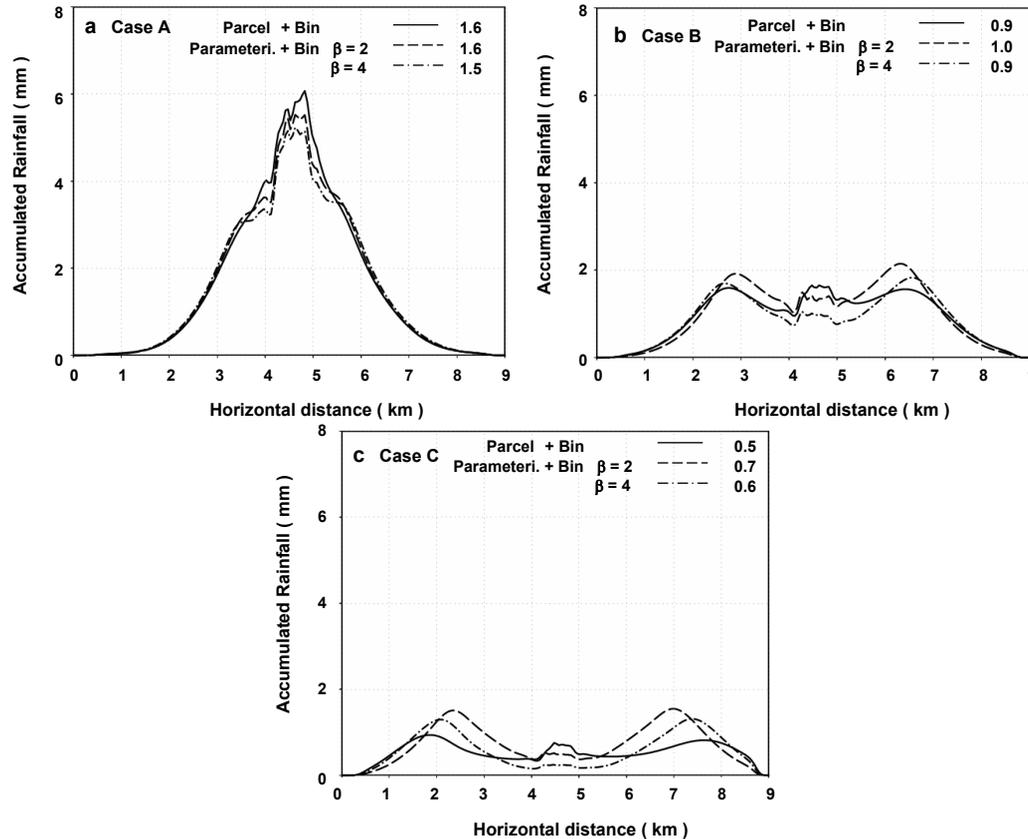


Fig. 12. Accumulated rainfall at 50 min. **(a)** CCN from case A in Fig. 2. Values averaged in the domain for cases using the parcel model and cases using parameterizations with $\beta=2$ and $\beta=4$ are 1.6 mm, 1.6 mm, and 1.5 mm, respectively. **(b)** As in (a), but for CCN from case B in Fig. 2. Averaged values are 0.9 mm, 1.0 mm, and 0.9 mm, respectively. **(c)** As in (a), but for CCN from case C in Fig. 2. Averaged values are 0.5 mm, 0.7 mm, and 0.6 mm, respectively.

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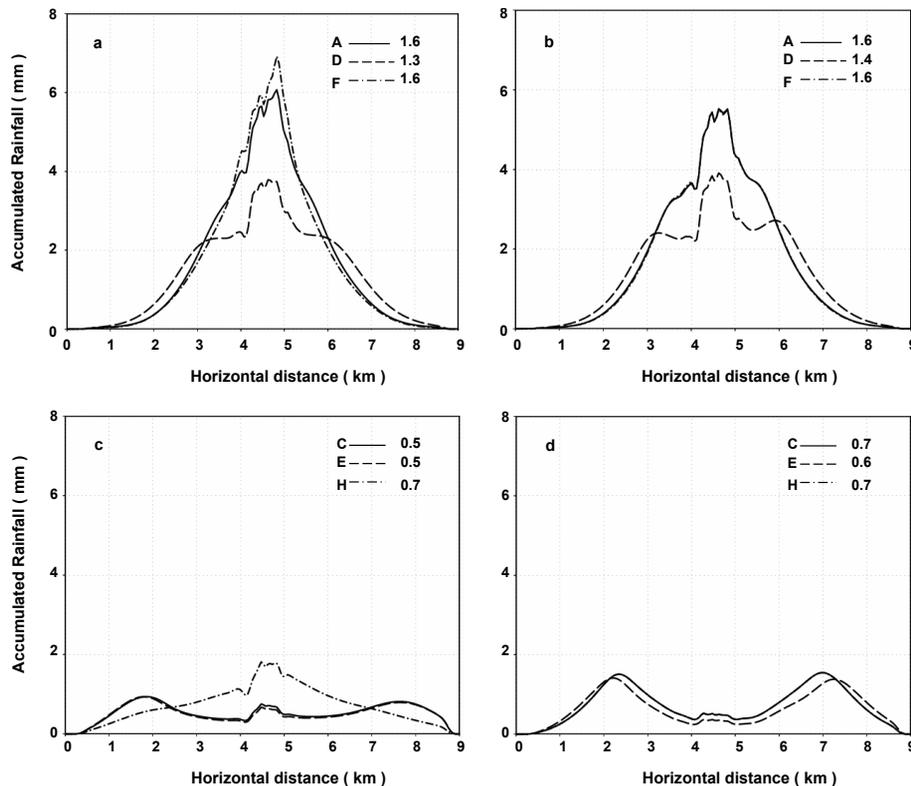


Fig. 13. Accumulated rainfall at 50 min. **(a)** Results using parcel model for cases A, D, and F. Values averaged in the domain are 1.6 mm, 1.3 mm, and 1.6 mm, respectively. **(b)** Results using parameterizations in cases A, D, and F. Values averaged in the domain are 1.6 mm, 1.4 mm, and 1.6 mm, respectively. **(c)** Results using parcel model in cases C, E, and H. Averaged values in the domain are 0.5 mm, 0.5 mm, and 0.7 mm, respectively. **(d)** Results using parameterizations in cases C, E, and H. Averaged values in the domain are 0.7 mm, 0.6 mm, and 0.7 mm, respectively.

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