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NEW PARCEL MODEL WITH DETAILED CLOUD MICROPHYSICS

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1. INTRODUCTION

Interaction between aerosol and cloud particles is important in some of today's meteorological issues, such as the indirect effect of aerosol particles in global climate change (Lohmann and Diehl, 2006), the change in stratospheric water vapor (Karcher, 2004), and the artificial adjustment of precipitation.

Aerosol particles with soluble and insoluble materials produce ice crystals through homogeneous and heterogenous freezing nucleations in upper troposphere. Their physicochemical nature and number concentration affect the properties of ice cloud, such as albed, coverage and life time, which have an effect on the radiative forcing of ice cloud. Those factors also modify the sedimentation rate of produced ice crystals, which control the dehydration rate of stratosphere. The basic (incomplete but available) knowledges on heterogenous nucleation of ice particles and droplet has been applied to the glaciogenic and hygroscopic seeding techniques for the precipitation enhancement, hail suppression and others.

For detailed discussion on those issues, basic studies with modeling approach on elemental processes relevant to the hydrometeor forming nuclei is needed as well as laboratory experiment. Recently, numerical modeling studies have been made to address the effect of ice nucleation with a sophisticated parameterization scheme. Gierens (2003) investigated the meteorological conditions for a transition between homogeneous and heterogeneous freezing nucleation using bulk parameterization for internal mixture of aqueous solution and ice forming insoluble material (soot). Lohmann and Diehl (2006) addressed the change in radiative forcing dependent on the chemical composition of mineral dust which works as ice nuclei in global climate model. It is neccessary to develop the model with detailed microphysics to provide a theoritical basis for such the parameterizations. Monier et al. (2006) tried to simulate the observation results on the properties of ambient and residual aerosol particles using their model which assumes an aerosol particle as a mixture of pure water and soluble material, and represent that property with two-dimensional bin scheme. Chen and Lamb (1994) developed a detailed physicochemical cloud model with multi-dimensional bin scheme to investigate the redistribution of atmospheric trace substances through cloud processes. In this work, we are further developing the microphysical processes in their model to study the hydrometeor nucleation processes in detail.

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2. NUMERICAL MODEL

2.1 Parcel Model

The microphysical parcel model developed by Chen and Lamb (1994) divides the categories of droplet and ice particle into multi-dimensional bins to express a variety of properties of hydrometeors. For droplets, the masses of pure water and soluble material were the variables for two-dimensional bin space. For ice particles, they adopted three-dimensional bin space, which is composed of the aspect ratio of particle and the massess of pure water and soluble material. Based on their model, we have developed a new parcel model with detailed cloud microphysics. The mass of insoluble material (ice nuclei) is introduced as another variable of droplet and ice particle categories to investigate the sensitivity of nucleation and residual material to the chemical components of soluble material and the size of insoluble portion of a particle. For ice particle category, the volume of particle is also introduced to simulate the successive change in the bulk density of ice particle in its growth process (Misumi et al., 2005).

Since the heterogeneous freezing nucleation is addressed in this work, we apply the model version of three-dimensional bin space for each of droplet (the masses of pure water, soluble and insoluble materials) and ice particle (the masses of pure water and soluble material, and aspect ratio).

2.2 Formulation for heterogeneous ice nucleation

The formulation for nucleation number (m^{-3}) through deposition mode in Chen and Lamb (1994) is modified so as to include the dependency on the size of insoluble particle and its surface area partially covered by aqueous solution (Fig. 1), in addition to the original function of super-saturation for ice, as follows.

$$N_{dep} = 4 \times 10^5 \left(\frac{s_i}{0.474} \right)^{4.5} \times F(R) \times G(X) \qquad (1)$$

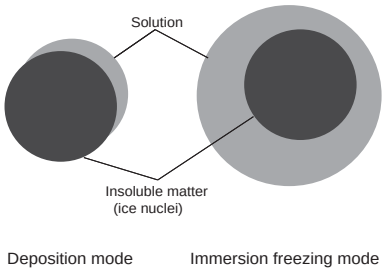


Fig. 1. Assumptions for the surface of deposition and immersion nuclei.

$$F(R) = 1 - \exp\left(-\frac{R^2}{R_n^2}\right),\ R_n = 1 \times 10^{-7}\ \mathrm{m} \qquad (2)$$

$$G(X) = \frac{1}{2}\left(1 - \cos\left(\pi X\right)\right), \qquad X = \frac{A_{nw}}{A_n} \qquad (3)$$

where $F(R)$ is the function for the size dependency, $G(X)$ is for the coverage of aqueous solution over insoluble particle. s_i is the super-saturation for ice, R is the radius of insoluble particle. A_n and A_{nw} are the total area of insoluble particle and the area of surface which is not covered by aqueous solution, respectively. Their ratio X is in the range of $0 < X < 1$. A_{nw} can be calculated using the volume of aqueous solution, the radius of insoluble particle and the contact angle of 12 degrees, with a help of the theory in Pruppacher and Klett (1978, Eq. 9-19). Figure 2a and 2b show the functions $F(R)$ and $G(X)$, respectively. The scaling parameter R_n is determined as 1×10^{-7} m, since the dependency of nucleation rate on the size of insoluble particle is significant below this value (Pruppacher and Klett, 1978). When the size of insoluble particle is larger and the coverage of aqueous solution is smaller (X is larger), these functions get larger. This means that the nucleation rate becomes larger, as shown in Fig. 3a.

For the nucleation number (per single droplet) through immersion freezing mode, $F(R)(R_n = 3 \times 10^{-8}$ m) is multiplied to the original function of temperature in Chen and Lamb (1994).

$$N_{im} = m(R_d)\exp\left[-0.68(7 + T_c)\right] \times F(R) \qquad (4)$$

where $m(R_d)$ is the mass of droplet with radius of R_d, T_c is the temperature in Celsius and R is the radius of insoluble particle. Here, the scaling parameter R_n is determined as 3×10^{-8} m, since the dependency of immersion freezing nucleation rate on the size of insoluble particle is significant below this value (Pruppacher and Klett, 1978). The $F(R)$ is shown in Fig. 2a (broken line), which means that when the size of insoluble particle is larger, these function get larger, namely, the nucleation rate becomes larger (Fig. 3b).

3. NUMERICAL EXPERIMENT

Two-dimensional log-normal distribution is assumed as initial size distribution of two-component aerosol particles. The number concentration is 500 cm^{-3}. The mode radius is 0.01 μm and dispersion is 0.22, for both of soluble and insoluble (ice nuclei) components (EXP-I). In order to investigate the sensitivity of simulation result to the size of insoluble particle, another case (EXP-II) in which the mode radius of 0.1 μm for insoluble particle is also examined. Initial atmospheric condition is given as 520 hPa, -20^{o}C and 75 % for air pressure, temperature and humidity, respectively. Those values are summarized in Table 1. Ascending speed of 2 ms^{-1} is assumed.

Table 1. Initial conditions for numerical experiment.

Experiment	EXP-I	EXP-II	EXP-III
Height	5200	5200	5200 m
Pressure	520	520	520 hPa
Temperature	-20	-20	-20 ^{o}C
Relative humidity	75	75	75 %
Aerosol particle number	500	500	500 cm^{-3}
Dispersion of soluble material	0.22	0.22	0.22
Mode radius of soluble material	0.01	0.01	0.01 μm
Dispersion of insoluble material	0.22	0.22	**0.42**
Mode radius of insoluble material	0.01	**0.1**	0.01 μm

4. RESULTS

Figure 4a shows the vertical profiles of meteorological and microphysical variables from the EXP-I. Air parcel which started to ascend from 5200 m reached water-saturation at about 5600 m and kept it until around 6900 m, and then departed from water-saturation but still kept super-saturation of several % for ice. The ice particle number concentration (solid line; Nice) exceeded 1 kg^{-1} at about 5500 m and 10^3 kg^{-1} at about 5800 m. Finally, it reached 7.17×10^6 kg^{-1}. Before the air parcel reached water-saturation, as shown in Fig. 4b, ice particles were mainly produced by deposition nucleation. After that, immersion freezing nucleation became dominant. In this stage, deposition nucleation continued in small magnitude, because the particles which were not able to go over their critical super-saturation still remained and produced ice particles through deposition nucleation.

When the ice nuclei included in aerosol particles were larger, as shown in Fig. 5a, the period of water-saturation became shorter. The water-saturation in EXP-II was kept from 5600 to 6500 m. The timing of ice initiation became earlier. The ice particle number concentration (solid line; Nice) in the EXP-II exceeded 1 kg^{-1} or 10^3 kg^{-1} at the heights 200 to 300 m lower than in the EXP-I. The ice particle number concentration finally reached 3.16×10^6 kg^{-1} which was smaller than in the EXP-I. As seen in Fig. 5, deposition nucleation was major process to produce ice particles before water-saturation was reached, and after that, immersion freezing nucleation became dominant, like in the EXP-I. In the former stage (before water-saturation was reached), the magnitude of nucleation rate through deposition mode was much larger in the EXP-II than in the EXP-I. In contrast, in the latter stage, deposition nucleation had no

effect, which means that the deposition nuclei had been used up.

5. DISCUSSION

The nucleation rate through deposition mode was dependent on the size of insoluble portion in aerosol particle. For larger insoluble particle, the coverage of aqueous solution over its surface tends to be smaller so that the deposition mode is enhanced in the EXP-II. The larger surface area provides more activation sites for immersion freezing mode so as to make the nucleation rate larger. Enhanced nucleation through both modes produces more ice particles in the EXP-II. This escalates the consumption of water vapor due to deposition growth of produced ice particles. That is why the period of water-saturation becomes shorter in the EXP-II. In the EXP-I, the water-saturation is kept longer so that more droplets can be stored until air parcel ascends up to higher altitudes (lower temperature) at which larger nucleation rate through immersion freezing mode is expected.

In the EXP-I and EXP-II, we adopted the same value (0.22) to the dispersion of insoluble particle size distribution. In order to examine the effect of dispersion on simulation results, we conducted another experiment (EXP-III) in which the dispersion was set as 0.42. As a result (Fig. 6), the initiation of ice particle was slightly earlier (a few tens meters lower in altitude) in the EXP-III, because the nucleation rate through deposition mode became slightly larger than in the EXP-I. This comes from the wider size distribution of insoluble particles which provides larger particles, that is to say, more activation sites for ice nucleation. The final number concentration of ice particle (5.56×10^6 kg^{-1}) was slightly smaller than in the EXP-I. A small effect on model result was seen in the EXP-III. However, it is revealed that the effect of larger dispersion has the same tendency as in the EXP-II where the larger mode radius of insoluble particle was applied.

The formulation for nucleation rate used in this work sufficiently shows the effect of insoluble particle size and coverage of aqueous solution over it. DeMott et al. (1997) and Gorbunov et al. (2001) have provided the empirical formulas which express the size effect of insoluble portion (soot) in aerosol particle. However, the effect of surface condition (coverage of aqueous solution) and chemical component is still open to question. It is necessary to improve the functional forms and parameters for detailed discussion on heterogenous ice nucleation. Recently, a cloud simulation chamber (Tajiri et al., 2006) has been developed by Meteorological Research Institute, Tsukuba, Japan, to simulate cloud-aerosol processes in adiabatically ascending air parcel. We plan to further improve the formulation through a comparison with chamber experiments.

6. SUMMARY

A new parcel model with detailed microphysics has been developed on the basis of Chen's microphysical model, in order to study the effect of aerosol particles on the initiation of water and ice clouds. This model has the hydrometeor categories of droplet and ice particle, each of which is divided into multi-dimensional bins to

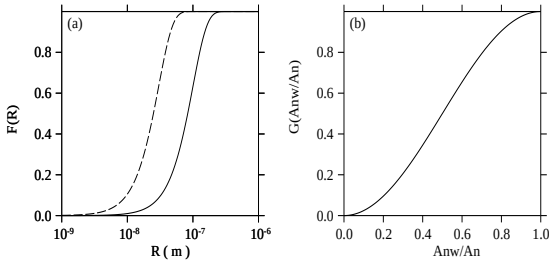
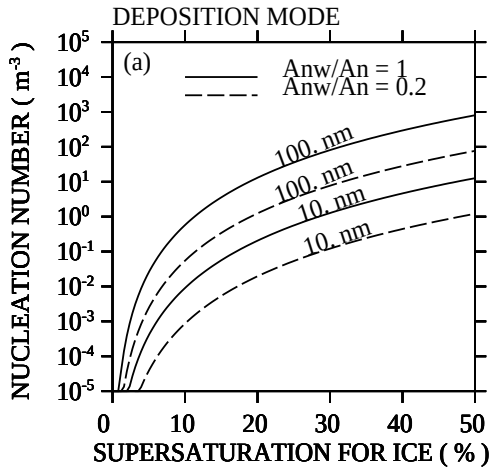


Fig. 2. Functions for (a) the size dependency $F(R)$ (solid line for deposition mode and broken line for immersion freezing mode) and (b) the coverage of aqueous solution over insoluble particle $G(X)$



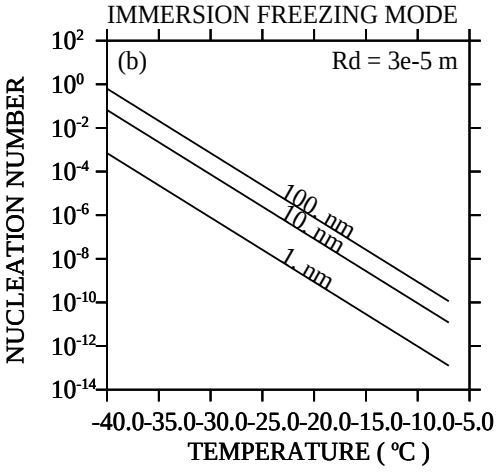


Fig. 3. Nucleation number through (a) deposition mode (m^{-3}) and (b) immersion freezing mode (per single particle of 30μm radius). Number is radius of ice particle.

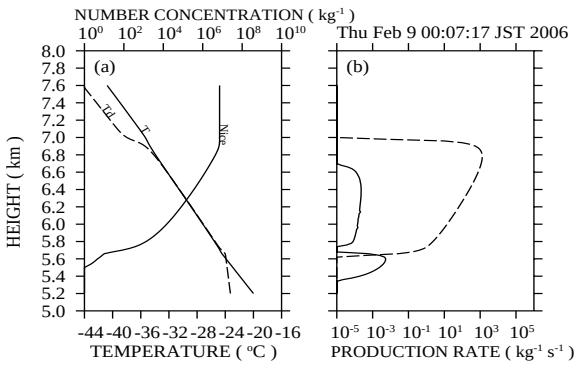


Fig. 4. Calculated profiles of (a) temperature (solid line; T), dew point temperature (broken line), ice particle number concentration (solid line; Nice), (b) ice nucleation rate through deposition mode (solid line) and immersion freezing mod (broken line), from the EXP-I.

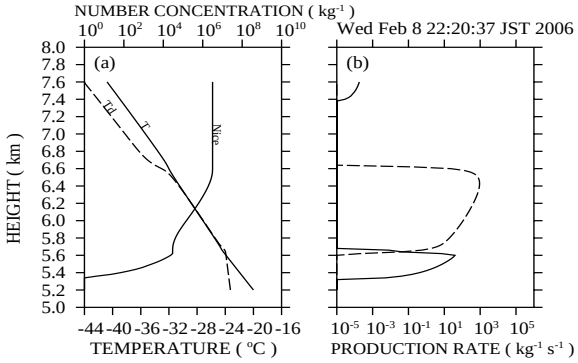


Fig. 5. Same as Fig. 4, but for the EXP-II.

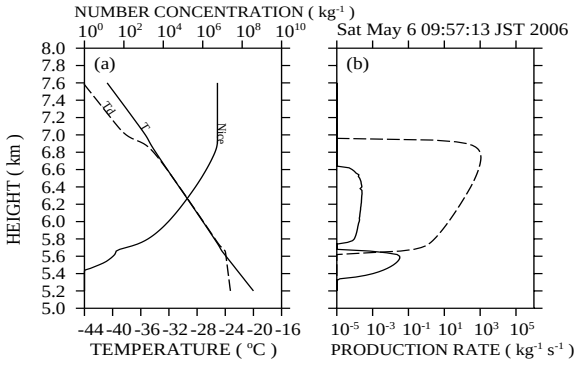


Fig. 6. Same as Fig. 4, but for the EXP-III.

express a variety of properties of hydrometeors.

Using this model, new formulation for heterogeneous ice nucleation are tested. For deposition mode, the nucleation rate is formulated so as to depend on the size of insoluble particle and its surface area partially covered by aqueous solution, as well as super-saturation for ice. For immersion freezing mode, the effect of insoluble particle size is introduced in addition to the temperature dependency so that the probability of active site occurrence on the surface of ice nuclei can be formulated.

As a result of numerical experiments, larger ice nuclei initiate ice particles earlier through deposition nucleation, and enhance nucleation through immersion freezing nucleation. Finally, consumption of water vapor becomes larger in the case of larger ice nuclei. It is revealed that the size of ice nuclei affects the timing of ice initiation, number concentration of ice particles, and amount of water vapor.

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