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EFFECT OF CCN NUMBER DENSITY ON RADIUS AND TEMPERATURE OF CLOUD DROPLET GROWN UP COMPETITIVELY

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Abstract

Köhler model is utilized for the estimation of the equilibrium cloud droplet size, assuming constant vapor content and constant temperature. However, the condensational growth of the cloud droplet consumes water vapor and releases the latent heat in the air parcel. Therefore, the water vapor pressure and the temperature are changed from the initial state. Taking account of the variations of the water vapor content and the temperature, a new model for the estimation of the equilibrium cloud droplet size and temperature has been developed by modifying traditional Köhler model. The modification of Köhler model is based on the mass and the heat conservation laws for the water (vapor and liquid) in the air parcel occupied by cloud droplets. Variations of size and temperature with number density of cloud condensation nuclei (CCN) are simulated numerically.

KEYWORDS: *cloud droplet, competitive growth, equilibrium size and temperature, latent heat*

1. Introduction

Quality of rainwater is paid great attention in these days as is seen from various acid rain issues. The chemical substances contained in rainwater originate from such scavenging processes in the atmosphere as rainout by cloud droplets (in-cloud scavenging) and washout by raindrops (below-cloud scavenging). The size of cloud droplets is an important factor, which controls the rainout of atmospheric pollutants. Therefore, it is essential problem to estimate the size of cloud droplets as precisely as possible.

The formation of cloud droplets is commonly initiated by condensation of the atmospheric vapor on cloud condensation nuclei (CCN). The cloud droplet treated here is due to this kind of vapor condensation alone and not due to such secondary growth as coalescence. And further, the entrainment of the environment air, which may effect on mass and heat balances of water in the air parcel, is excluded.

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The equilibrium size of the cloud droplet condensed on cloud condensation nuclei (CCN) is estimated usually by traditional Köhler model. Köhler model is derived from the thermodynamic equilibrium which leads to thermal, mechanical and chemical potential equilibria between liquid and gas phases separated by a curved interface taking account of the effect of dissolved CCN (solute) on activity of a droplet (Pruppacher and Klett, 1980). However, Köhler model is based on the assumption that the cloud droplet grows in an infinitely large reservoir of water vapor at constant pressure and constant temperature. Therefore, the saturation ratio and the temperature are treated as invariable through the growth, although they are changed by the consumption of vapor and the release of latent heat due to the condensation.

The underlying imaginary assumption of Köhler model causes two apparent deficiencies in Köhler model, in case of treating the real world problems. The first is that the equilibrium size cannot be decided in case of the larger saturation ratio than the critical saturation ratio. This is attributed to the curious phenomenon that the droplet will grow ever larger. The second is that the variation of the size with the number density of CCN cannot be taken into account in the estimation of the droplet size. In order to improve these deficiencies and make a new model applicable to the competitive growth of droplets, the conventional Köhler model has been modified using the mass conservation and the heat conservation in the air parcel.

With use of this new model, variations of the cloud droplet size and the temperature with number density of CCN (particulate of ammonium sulfate) are simulated numerically. The results of numerical simulations show that the effect of CCN number density (i.e., the effect of competitive growth) is considerably great on the cloud droplet size but small on the temperature. However, from the meteorological point of view the temperature variations are large enough to make the air parcel unstable in the atmosphere. The temperature variations, which are caused by the latent heat stored in water vapor, drive atmospheric motions such as convective cloud generation.

2. Modeling of Heterogeneous Cloud Formation

Generation and growth of cloud droplets are realized by condensation of atmospheric water vapor. Water vapor condenses to liquid, if the vapor pressure over the cloud droplet is smaller than the vapor pressure in the ambient air. In this context, homogeneous droplet formation (absence of foreign substances) on basis of Kelvin law requires water vapor supersaturation as high as several hundred percent. Therefore, in the atmosphere, cloud droplet formation occurs via heterogeneous nucleation involving aerosol particles. It is because the droplet of an aqueous salt solution lowers the water vapor pressure over itself as far as the droplet can condense the ambient water vapor into itself. Such aerosol particles are capable of initiating droplet formation at low supersaturations observed in the atmosphere (below 10 % and most often below 1 %). They are called cloud condensation nuclei (CCN). In this paper heterogeneous droplet formation with CCN is treated exclusively.

2.1 Equilibrium Radius of Cloud Droplet

Non-steady growth process of cloud droplets on CCN can be described by a set of differential equations derived from the mass and heat conservation which considers the chemical potential equilibrium (Pruppacher and Klett, 1980; Takeda and Kuba, 1982; Kuba and Takeda, 1983; and Shiba et al., 2000). However, it is not so easy to integrate the differential equations numerically with respect to time and to obtain precise solutions, especially in the very early stage of the growth, because of the instability of the numerical calculation (poor convergence). On the other hand, the equilibrium radius is obtained easily by Köhler model described by an algebraic equation based on the thermodynamic equilibrium (between liquid and gas) alone, on the assumption that both water vapor pressure and temperature are not changed with the growth (Pruppacher and Klett, 1980). However, as mentioned earlier, Köhler model has two big deficiencies. The first is that the equilibrium size cannot be decided, if the vapor has the larger saturation ratio than the critical saturation ratio. The second is that Köhler model gives the same radius regardless of the number of cloud droplets, in other words, the variation of the size with the number density of CCN cannot be taken into account in the estimation of the size. These deficiencies are caused by the fact that Köhler model does not consider both the water vapor reduction and the release of latent heat, which are due to consumption by condensation into droplet.

2.2 Construction of New Model

The system of the cloud droplet growth on CCN is a heterogeneous system consists of two phases (gas and liquid; in very early stage it consists of three phases containing solid CCN) which are separated from each other by a surface of discontinuity (droplet surface). The new model developed here allows the reduction of water vapor pressure and the increase of temperature in the air parcel by coupling two conservation equations with Köhler model. This model takes CCN number density into account, but does not CCN size distribution. This is because, firstly this study aims at constructing a new model instead of traditional Köhler model and investigating the effect of CCN number density on the equilibrium size of droplets, and secondly consideration of CCN size distribution makes the numerical computation complicated with increase in size-class. The mathematical model consists of three governing equations (Shiba et al., 2002), which are derived from the chemical potential equilibrium (including the temperature and pressure equilibrium between droplet and vapor), the water mass conservation and the heat energy conservation. These three governing equations give the working equations for computing the cloud droplet radius, the saturation ratio and the temperature in the air parcel as follows as:

$$\ln(S_e) = \frac{A_1}{a_e} - \frac{A_2}{a_e^3} \tag{1}$$

$$S_e = S_0(1 - A_3 a_e^3)A_4 \tag{2}$$

$$T_e = T_0 + \frac{L_e(T_0)m_{we}}{(C_{pw} - C_{pv})m_{we} + C_{pv}m_{ve} + C_{pa}m_{ae}} \tag{3}$$

$$A_1 = \frac{2M_w\sigma}{R_3 T_e \rho_{we}} \tag{4}$$

$$A_2 = \frac{3\nu m_s M_w}{4\pi M_s \rho_{we}} \tag{5}$$

$$A_3 = \frac{4\pi\rho_{we} R_1 T_e}{3M_w e(T_0)} N \tag{6}$$

$$A_4 = \frac{n(T_0)}{n(T_e)}\frac{e_{sat}(T_0)}{e_{sat}(T_e)} \approx \frac{e_{sat}(T)}{e_{sat}(T_e)} \tag{7}$$

where a_e is equilibrium radius; S_e and S_0 are equilibrium and initial saturation ratio, respectively; T_e and T_0 are equilibrium and initial temperature, respectively; C_{pw}, C_{pv} and C_{pa} are specific heat capacity of liquid water, vapor water and air at constant pressure, respectively; m_{we}, m_{ve} and m_{ae} are mass of liquid water, vapor water and air, respectively; L_e is latent heat of evaporation of water; M_w and M_s are molecular weight of water and CCN, respectively; m_s is mass of CCN; R_1 and R_3 are universal gas constants; ρ_{we} is density of water; ν is van't Hoff factor; σ is surface tension of water against the air; e is water vapor pressure; e_{sat} is saturation vapor pressure; n is total moles of vapor and air in the air parcel; and N is CCN number density. The value and the unit of these physical properties are tabulated in Tables 1 and 2.

Table 1. Value of Physical Properties (1)

R_1	R_3	σ	e_{sat}	ρ_{we}	ρ_s	ν
82.0	8.31×10^7	75.67	6.108	1.001	1.769	3
atm cm^3/(mol K)	erg/(mol K)	dyn/cm	mb	g/cm^3	g/cm^3	–

Table 2. Value of Physical Properties (2)

L_e	T_0	M_w	M_s	C_{pw}	C_{pv}	C_{pa}
597.3	273.15	18.0	132.0	1.00	0.45	0.24
Cal/mol	K	g/mol	g/mol	cal/(g C)	cal/(g C)	cal/(g C)

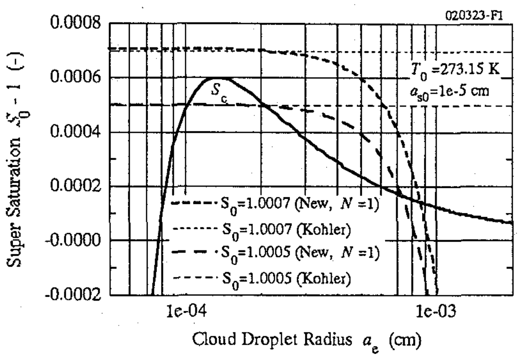

Figure 1. Difference between new model and Köhler model.

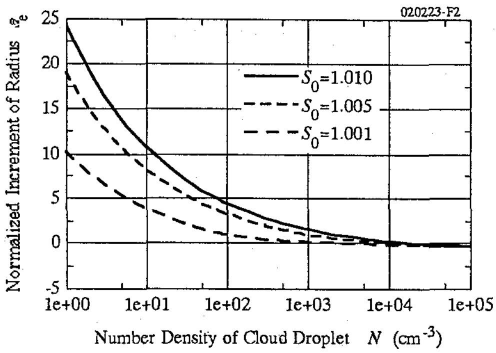

Figure 2. Relationship between equilibrium droplet radius and CCN number density.

3. Numerical Simulations

Using the new model developed here for competitive growth of cloud droplets, the equilibrium values of the cloud droplet radius, the temperature and the saturation ratio have been estimated numerically to investigate the effects of CCN number density (i.e., competitive growth of cloud droplets) on these equilibrium quantities. CCN is solid $(NH_4)_2SO_4$ particle, which is one of the major atmospheric aerosol made from gaseous pollutant SO_2(g) and NH_3(g). One molecule of $(NH_4)_2SO_4$ dissociates into three ions almost perfectly in liquid water. Therefore, van't Hoff factor can be assumed to be 3. The physical properties used for the numerical simulations are tabulated in Tables 1 and 2.

The algorithm of the numerical calculation of the equilibrium radius, the temperature and the saturation ratio is as follows as:

(A) Assuming equilibrium radius (e.g., use potential radius obtained by Equation (1) with $S_e = 1$), estimate the saturation ratio by Equation (2) and the temperature by Equation (3), respectively, to obtain the first approximated equilibrium values of a_e, S_e and T_e;

(B) Applying Newton-Raphson method to a set of Equations (1), (2) and (3), and using the estimated values in step (A) as the starting values, calculate the improved new values of a_e, S_e and T_e; and

(C) If the values of a_e, S_e and T_e can be regarded as convergent, stop the calculation (if not, repeat the above steps (B) and (C) with using the obtained approximated values as the starting values of the Newton-Raphson routine).

3.1 New model and Köhler model

For the growth of a single cloud droplet, the difference between new model and Köhler model is illustrated in Figure 1. The initial dry CCN radius, a_{s0}, is 1×10^{-5} cm. In this case, the growth is not competitive. Therefore, the situation of new model and that of Köhler model are very much alike. Equilibrium radius is given as x-coordinate at the intersection of Köhler curve with a super saturation curve. Köhler curve is a thick upwardly-convex curve of 3-rd order hyperbola given by Equation (1). Köhler model assumes saturation ratio remains constant. Then, the super saturation curve in Köhler model represented by a horizontal line as shown by thin dotted or broken line. When the initial saturation ratio equals 1.0005, there are two intersections, and we can obtain the equilibrium radius. However, Köhler curve has no intersection with the line, when the initial saturation ratio equals 1.0007. Therefore, the equilibrium radius cannot be obtained by Köhler model for 1.0007 of the initial saturation ratio. Even in such a case, we can obtain the equilibrium radius by new model developed here. The super saturation curve in the new model is a descending curve as shown by the thick dotted or broken curve of a 3-rd order polynomial given by Equation (2), which has at least one intersection for any value of the initial saturation ratio.

3.2 Effects of CCN number density

In order to investigate the effects of CCN number density (i.e., effects of competitive growth) on the cloud droplet radius, the temperature and the saturation ratio in equilibrium state, the normalized variables $\hat{a}_e$, $\hat{T}_e$ and $\hat{S}_e$ are introduced for the cloud droplet radius, for the temperature and for the saturation ratio, respectively. These normalized variables are defined as follows as:

$$\hat{a}_e = \frac{a_e - a_p}{a_p} \tag{8}$$

$$\hat{T}_e = \frac{T_e - T_0}{\Delta T_{as}} \tag{9}$$

$$\hat{S}_e = \frac{S_e - S_0}{S_0} \tag{10}$$

where a_p is potential radius (given by equalizing the initial saturation ratio to unity in Köhler model); and ΔT_{as} is the temperature increment per 1 m uplift of the air parcel in the atmosphere, which is calculated from the difference between the saturated adiabatic lapse rate and the dry one. The temperature increment due to the growth of cloud droplets is too small to cause the detective variation in the equilibrium cloud droplet size. However, the temperature increment is considerably great, compared with the temperature rise due to vapor condensation by adiabatic uplift of vapor-saturated air parcel. To demonstrate this fact, the temperature incremnt is normalized as Equation (9).

Figure 2 represents effects of CCN number density (i.e., effect of competitive grwoth) on the equilibrium droplet radius defined by Equation (8). Continental air masses are generally richer in CCN than are maritime air masses. The median CCN number density ranges from a few tens to a few hundred cm^{-3} in air over oceans, and from a few hundred to a few thousand cm^{-3} in air over the continents. According to Pruppacher and Klett (1980), in relatively pure air CCN number density is very small (e.g., at Yellowstone Park an average of 15 cm^{-3}) and is large in air over urban area (e.g., at Washington D.C. from 2000 to 7000 cm^{-3}). As is supposed, the larger CCN number density is, the smaller the equilibrium radius becomes. That is, the competitive growth brings about the reduction of cloud droplet size. This effect of CCN number density on the equilibrium radius is strengthened with increase of the initial saturation ratio. Because the radii by Köhler model are approximately estimated from the intersections of the curves with y-axis (at N =1), the error in Köhler predictions can be roughly estimated by the curves in Figure 2. We can see that Köhler predictions bring about considerably great errors in the prediction of the competitive growth. It should be noted that even in the case of one CCN (non-competitive growth) Köhler prediction has an error due to the neglect of the vapor consumption, as is illustrated in Figure 1.

The plots in Figure 3 demonstrate the effects of CCN number density on the equilibrium temperature defined by Equation (9). The curves are plotted parametrically in the initial saturation ratio. As is supposed, the temperature increment increases with increase of the initial saturation ratio. These curves have the minimum values. The CCN number density for the minimum value increases with increase of the initial saturation ratio. Because the temperature increment of the air parcel containing the cloud droplets is due to the latent heat of vapor condensation into liquid water, the descending part of the curves implies that the increment of the condensed vapor in the air parcel (i.e., consumption of vapor) is diminished, although the growth of many cloud droplets is highly competitive. As is seen in Figure 2, this compels the individual droplet to decrease drastically in its radius in the region where CCN number density is less than about 1000. The increasing part of the curves indicates that the competitive growth is allowed to consume greater amount of the vapor with increase of CCN number density. This results in getting smaller decrement of droplet radius for large CCN number density in Figure 2, nevertheless the growth in large CCN number density region is more competitive than in small density region.

Figure 4 shows the effect of CCN number density on the normalized saturation ratio defined by Equation (10). As is seen from Figure 3 the temperature increment is positive, i.e., the latent heat is released, it can be supposed that the vapor is consumed by the condensation. Therefore, the increment of saturation ratio defined by Equation (10) should become negative, in other words, the saturation ratio decreases from the initial saturation ratio due to growth of cloud droplets. The plots of Figure 4 coinside with this guess and verifies the validity of the numerical results. From the relation between

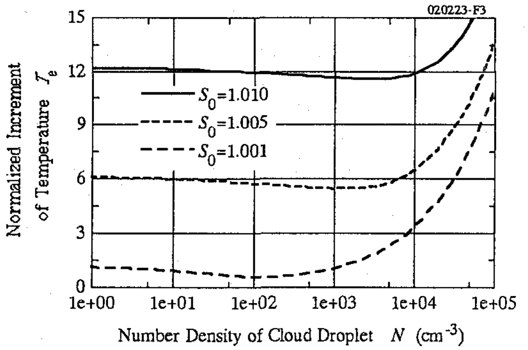

Figure 3. Relationship between temperature increment and CCN number density.

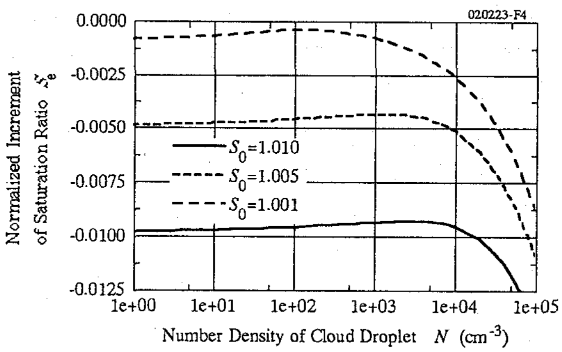

Figure 4. Relationship between saturation ratio decrement and CCN number density.

the temperature rise and the vapor consumption, we can also expect that the saturation-ratio decrement curves and the temperature increment curves should appear to be as if symmetrical about x-axis, comparing the appearance of the curves in Figure 3 with that of Figure 4. The saturation-ratio decrement (vapor consumption) curves have the maximum values, as the temperature increment curves have the minimum values. However, the reason why the saturation-ratio decrement (vapor consumption) curve does not monotonously decrease with increase of CCN number density is not clear to the authors at the moment.

4 Conclusions

From the results of the numerical simulation with the new model developed here for the competitive growth, which takes account of the consumption of water vapor and the release of latent heat, it is concluded that:

1. The larger the saturation ratio is, the more remarkable the droplet-size reduction is brought about by competitive growth;
2. Curve of temperature increment versus CCN-number density has the minimum value of temperature increment;
3. Increase in CCN causes considerably great temperature increment, compared with temperature decrement caused by adiabatic uplift of the saturated air parcel, although the temperature increment is too small to have sensible effect on the cloud droplet radius; and
4. Variation of saturation-ratio decrement seems to be in a reverse manner to variation of temperature increment.

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