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EQUILIBRIUM TEMPERATURE OF MOIST AIR PARCEL DUE TO COMPETITIVE GROWTH OF CLOUD DROPLET ON CLOUD CONDENSATION NUCLEI

by

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SYNOPSIS

Usually the equilibrium cloud droplet radius is estimated by the traditional Köhler model which is based on the chemical potential equilibrium and which assumes the vapor pressure and the temperature to be constant in the course of vapor condensation. However, the Köhler model fails to take account of the number density of cloud condensation nuclei (CCN) and is subjected to the limitation of the maximum vapor pressure (critical saturation ratio) in the estimation of the cloud droplet radius. In order to estimate the cloud droplet size, the temperature and the vapor pressure (saturation ratio) in the equilibrium state, a new model has been developed, taking account of the water vapor decrease and the latent heat release in the air parcel. This is done by coupling the mass and the heat conservation laws on the air parcel to the chemical potential equilibrium. The new model can be applied to the competitive growth of plural cloud droplets as well as to non-competitive growth of a single cloud droplet. Variations of size, temperature and saturation ratio with number density of CCN are simulated numerically by the new model. The results of simulations are as follows: (1) the reduction of the equilibrium size due to competitive growth is enhanced by the increase in the initial saturation ratio of vapor in the air parcel, and (2) the temperature variation of the air parcel including the cloud droplets is too small to change the values of physical properties, but large enough to make the air parcel statically unstable (or thermally convective) in the atmosphere.

INTRODUCTION

The temperature change of the earth's atmosphere has been the focus of the world's attention these days. Rising global temperatures due to global warming are expected to raise sea levels and to change local climate conditions which bring about increase in precipitation and evaporation, intense rainstorms and drier soils. Human activity accounts for these changes through mainly the release of greenhouse gases which trap the solar heat around the earth. However, the temperature change of the very local atmosphere is possibly caused by vapor condensation on cloud condensation nuclei (CCN) which releases the solar heat accumulated in the atmospheric water vapor.

When the atmospheric vapor condenses into liquid water to form droplets in an air parcel, latent heat is released by condensation of vapor and a new static state of the air parcel comes up. Condensation of the atmospheric vapor in an air parcel is considered to occur through the two mechanisms as: (1) reduction of the temperature of the air parcel by uplifting the air parcel, and (2) reduction of the surface vapor pressure of the CCN-containing droplets below the bulk vapor pressure without uplifting the air parcel. The first is the primary and ordinary mechanism of atmospheric vapor condensation, although it usually needs the aid of CCN. However, from the viewpoint of static stability of the air parcel, it should be noted that a small temperature increase due to the second condensation mechanism enables the stationary air parcel to be statically unstable. This is because, in the case of the stationary air parcel, the condensation makes the parcel temperature (strictly speaking, virtual potential temperature) always higher than the surrounding temperature and then the kinetic energy of the air parcel increases [see ,e.g., Asai (1)]. In this case the stationary air parcel always becomes absolutely unstable. Instability of the air parcel in lower part of an atmospheric layer brings about thermal convective flow. The thermal convective flow in the atmospheric layer is usually expected to cause cumuliform cloud with precipitation. Thus, anthropogenic CCN affects the climate change, especially rainfall. Local anthropogenic climate change is of great interest to advanced hydrologists as well as global one.

As mentioned previously, air pollution is closely related to rainfall through such phenomena as the cloud droplet formation on CCN with heat release, static instability of air parcel with proceeding to thermal, and growth of convective cloud with generating precipitation. Therefore, the hydrological study on water cycles, which consists of evaporation, precipitation, and both surface and subsurface runoff, and the ecological and meteorological study on air pollution, which treats anthropogenic air pollutant emitted to eco-system by human activity, are also intimately related with each other. A new field of study which is called "eco-meteorohydrology" is expected to solve new hydrological problems caused by human activities.

Evapo-transpiration of liquid water and condensation of water vapor play an important role in the transportation of the water and its dissolved substances between the atmosphere and the earth, even though the transported amount of the water (precipitation) is only a small part of the surface water over the earth (oceans, lakes, rivers and so forth). Polluted gaseous and particulate matter emitted from natural and anthropogenic sources into the atmosphere are scavenged by the cloud droplets and the rain drops. Therefore, rainfall contributes to the transportation of not only water itself but also the atmospheric pollutants to the earth. There are two steps in the pollutant mass transfer from the atmosphere to the rainwater, which control the rainwater quality. The first one is the in-cloud scavenging by cloud droplets and is called rainout. Rainout has not been made clear yet [Shiba et al. (18); Oishi et al. (11); Matsui et al. (7)]. The second one is the below-cloud scavenging by rain drops and is called washout. Washout has become quite evident thanks to the work of Shiba et al. (14, 15 and 16) and Oishi et al. (12).

Rainout and washout are also controlled by the air flow field where cloud droplets grow and rain drops fall. Takahashi (21) constructed a 1-dimensional unsteady drop growth model and applied the model to the cylindrical flow field with constant radius where cloud grows and rainfall occurs. With this model he calculated the salt content of drops with other various variables and showed that the number density of cloud droplets was the critical factor in initiation of warm rain and that giant nuclei increased the salt content of rain by almost one order higher than the case without giant nuclei. Takahashi's study is very noteworthy concerning the application of cloud dynamics to pollutant mass transfer between the air and precipitation. His research is a pioneering work in "eco-meteorohydrology" and it introduces a way by which we can deal with such new environmental problems as climatic change in precipitation.

Recently, the studies on rainfall hydrology in connection with the water circulation process between the atmosphere and the ground have considerably progressed especially in the microscopic treatment of rainfall process as is seen in the analysis of the vertical profile of the rain drop size distribution [Nakagawa et al. (8 and 9)] and in the rainfall prediction method utilizing the knowledge of convective cloud processes [Oishi et al. (10)]. Since cloud droplets and rain drops work as a cluster of micro-scale chemical reactors during rainout and washout, it is very important for prediction of rainwater quality to estimate the precise size of cloud droplets and rain drops. Such microscopic treatment in rainfall process is also an effective approach for estimating the pollutant mass transfer between the atmosphere and the rainwater. For rainfall volume prediction it is recognized that the cloud formation process is important as well as the rainfall process. The same is true for rainfall quality prediction, because the in-cloud scavenging are considerably accountable for the rainfall quality [Shiba et al. (18)]. Matsui et al. (7) combined a numerical simulation of cumuliform clouds with pollutant mass transfer between the air and water. Oishi et al. (11) performed the numerical simulation of acid rain formation by combining the chemical process with detailed cloud microphysical process considering the size distribution of precipitation by the bin method. These studies prove that a new study field "eco-meteorohydrology" demanded by anthropogenic environmental issue is steadily advancing in the traditional hydrology.

The size of cloud droplets is an important factor in controlling the rainout of atmospheric pollutants in clouds. Until now, the traditional Köhler model has been utilized to estimate the equilibrium droplet size. However, the traditional Köhler model takes no account of consumption of vapor and release of latent heat, and is dependent on CCN number density. In order to estimate the equilibrium cloud droplet size in cloud (air parcel), a new model was developed which takes into account the water vapor consumption and the latent heat release. The new model consists of the chemical potential equilibrium, the mass conservation and the heat energy conservation to overcome the deficits in the traditional Köhler model.

CLOUD DROPLET FORMATION CAUSED BY CCN

First, it should be noted that the cloud droplets treated here are those which are formed by condensation of the water vapor without the occurrence of the ice phase in clouds at all. Also cloud droplet interactions are not considered here, that is, the secondary processes of droplet growth by collision and coalescence and of collisional breakup are not treated here.

It is well known [Pruppacher and Klett (13)] that in the atmosphere the excess of water vapor over the saturated vapor pressure, which varies with the atmospheric temperature, condenses into liquid water and forms droplets. Once a very small water droplet is generated, the flux of water vapor flows into the droplet, since the vapor pressure of the atmosphere (supersaturated) is greater than the pressure over the droplet surface (saturated). This process is a homogeneous nucleation, and supersaturation may be achieved by cooling of air parcel. However, supersaturation of several hundred percent would be necessary for droplet formation in homogeneous water vapor, but typically supersaturation remains below 10% and most often even below 1% in the atmosphere. Therefore, it is hard to condense vapor to droplets by cooling alone and, furthermore, cooling needs to be aided by something which can drive the flux of water vapor into liquid. According to Raoult's Law [Asai et al. (1), Atkins (2)], aerosol particles are capable of initiating drop formation at the observed low supersaturation, because they can reduce the drop-phase vapor pressure lower than the atmospheric vapor pressure. This process is a heterogeneous nucleation and traditionally the equilibrium droplet size is estimated by adopting the Köhler model [Pruppacher and Klett (13)]. Such aerosol particles which enable vapor to condense into droplets are called cloud condensation nuclei (CCN). Ammonium sulfate $(NH_4)_2SO_4$ is a well known major CCN [Corbett et al. (4), and Mason (6)], which is formed by the gas phase chemical reaction of SO_2(g) with NH_3(g) [Kim et al. (5)] and is very soluble in liquid water.

As is mentioned above, realistic condensation of the atmospheric vapor is supposed to occur by cooling of the air parcel with aid of small aerosol particles as liquid CCN or solid CCN. And yet condensation treated here is due to CCN alone. This is because our focus of attention is on the climate change caused by CCN as product of human activity from viewpoint of "eco-meteorohydrology", and because with existence of active CCN (regardless of liquid or solid) it is not necessary for droplet formation to cool the air parcel. In the case where there is no cooling, that is, when the vapor is condensed into liquid water to form droplets, the temperature of the air parcel always goes up due to the latent heat released in the air parcel.

Defect of traditional Köhler model

To estimate the equilibrium cloud droplet size, the Köhler model [Pruppacher and Klett (13)] is widely utilized. This model is developed by a simple single equation including the unknown droplet radius as the one and only variable, although it takes some numerical calculations to obtain the droplet radius. This model also assumes that condensation goes on in inexhaustible water vapor at the constant pressure and constant temperature. However, it is clear that the condensational growth of the cloud droplet consumes the ambient water vapor and releases the latent heat. Therefore, the ambient water vapor pressure and the temperature should be changed in the course of condensation.

The assumption causes two apparent deficiencies in the Köhler model [Shiba et al. (17 and 19)]. The first is the remarkable characteristics of the Köhler model, and is the serious deficiency that the equilibrium radius cannot be obtained in case the given saturation ratio is greater than so called critical value which corresponds to the maximum value of the Köhler curve. In this case the users of the Köhler model say that the droplet is made active to keep the growth everlasting, although it is evidently non-realistic. Also from logical point of view, this is very eccentric and even seems distorted, because such interpretation as everlasting growth is based on non-equilibrium idea which is contradictory to the scope of the Köhler model which describes equilibrium state exclusively. It should be noted that the

Köhler model cannot predict the equilibrium radius outside the critical saturation ratio, granting the non-realistic assumption to be acceptable to use the Köhler model. The second deficiency is that the variation of the size with the number density of CCN, which is brought about by the competitive growth of the cloud droplets, cannot be taken into account in the estimation of the size. In fact, the Köhler model yields the same cloud droplet size regardless of the number of CCN.

Distinction of new model

The governing equation of Köhler model is based on only the thermodynamic equilibrium conditions between the droplet and the atmosphere. This is the cause of the above-mentioned deficiencies. In this regard, the thermodynamic equilibrium conditions in the Köhler model are joined together to be a single chemical potential equilibrium, although they are composed of equations about temperature, vapor pressure and chemical potential.

In order to take account of the variation of the water vapor content and the variation of the temperature in the air parcel, the new model developed here [Shiba et al. (20)] is supplemented by the mass conservation on the water (vapor and liquid) and the heat conservation on the air parcel to the chemical potential equilibrium. Therefore, the new model is composed of three governing equations coupling with each other. The new model can estimate the droplet radius, the temperature and the vapor pressure in the equilibrium state after competitive growth on any number of CCN for any saturation ratio, including a higher saturation ratio than the so called critical value in the Köhler model.

Governing equations of new model

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 the chemical potential equilibrium. The mathematical model consists of three governing equations which correspond to water mass conservation, heat energy conservation and chemical potential equilibrium (implicitly including temperature and pressure equilibria). The chemical potential equilibrium between liquid and vapor, thermal energy conservation and water mass conservation in the control volume of air parcel containing uniformly dispersed mono-sized cloud droplets are written as follows:

$$\mu_{\mathrm{w}}(S_{\mathrm{e}},T_{\mathrm{e}},a_{\mathrm{e}})=\mu_{\mathrm{v}}(S_{\mathrm{e}},T_{\mathrm{e}},a_{\mathrm{e}}) \tag{1}$$

$$d(m_{\mathrm{w}}h_{\mathrm{w}}+m_{\mathrm{v}}h_{\mathrm{v}}+m_{\mathrm{a}}h_{\mathrm{a}})=0 \tag{2}$$

$$d(m_{\mathrm{w}}+m_{\mathrm{v}}+m_{\mathrm{a}})=0 \tag{3}$$

where m_{x} = mass of x-phase [x: (w,v,a)=(liquid water, water vapor, air)] (g); h_{x} = enthalpy per unit mass of x-phase (cal/g); S_{e} = saturation ratio in equilibrium state (–); T_{e} = temperature in equilibrium state (K); a_{e} = droplet radius in equilibrium state (cm); and μ_{x} = chemical potential of x-phase, which is given by:

$$\mu_{\mathrm{x}}(S,T,a_{\mathrm{x}})=\mu_{\mathrm{x0}}(S,T)+\mathcal{R}T\ln(a_{\mathrm{x}}) \tag{4}$$

where μ_{x0} = integration constant in x-phase; $\mathcal{R}$ = universal gas constant; and a_{x} = activity in x-phase. It should be noted that Eq(1) is merely the necessary condition for thermodynamic equilibrium (the maximum entropy) and does not describe any conservation.

Equilibrium radius a_{e}, temperature T_{e} and saturation ratio S_{e} are calculated by the simultaneous equations obtained from Eqs(1) – (3) as:

$$\ln(S_{\mathrm{e}})=\frac{A_1}{a_{\mathrm{e}}}-\frac{A_2}{a_{\mathrm{e}}^3} \tag{5}$$

$$T_{\mathrm{e}}=T_0+\frac{L_{\mathrm{e}}(T_0)m_{\mathrm{we}}}{\Delta C_{\mathrm{p}}m_{\mathrm{we}}+C_{\mathrm{pv}}m_{\mathrm{ve}}+C_{\mathrm{pa}}m_{\mathrm{ae}}} \tag{6}$$

$$S_{\mathrm{e}}=S_0(1-A_3a_{\mathrm{e}}^3))A_4 \tag{7}$$

where $L_e(T_0)$ = latent heat of vapor at T_0 ($\approx$ 597.3 cal/g); C_{px} = isobaric specific heat of x-phase [cal/(g K)]; and $\Delta C_p = C_{pw} - C_{pv}$ [cal/(g K)]. Eq(6) is obtained from Eqs(2) and (3), rewriting the enthalpy with the specific heat at constant pressure. Eq(7) is derived from Eq(3), applying the equation of state to the vapor in the air parcel.

Coefficients A_1, A_2, A_3 and A_4 are given as:

$$A_1 = \frac{2M_w\sigma}{\mathcal{R}_3 T_e \rho_{we}} \quad (8)$$

$$A_2 = \frac{3\nu m_s M_w}{4\pi M_s \rho_{we}} \quad (9)$$

$$A_3 = \frac{4\pi\rho_{we}\mathcal{R}_1 T_e}{3M_w e(T_0)} N \quad (10)$$

$$A_4 = \frac{n(T_0)}{n(T_e)}\frac{e_{sat}(T_0)}{e_{sat}(T_e)} \approx \frac{e_{sat}(T_0)}{e_{sat}(T_e)} \quad (11)$$

where σ = surface tension (dyn/cm); ρ_{we} = density of water (g/cm^3); ν = van't Hoff factor (–); M_w and M_s = molecular weight of water and CCN, respectively (g/mol); m_s = mass of CCN (g); e = vapor pressure (atm); e_{sat} = saturation vapor pressure (atm); $\mathcal{R}_1$ and $\mathcal{R}_3$ = universal gas constants [=82.0 atm cm^3/(K mol), and 8.31$\times$10^7 erg/(mol K), respectively]; n = total moles of water and air (mol); and N = number density of CCN (1/cm^3). Eq(5) is very similar to the governing equation of the Köhler model, but S_e on the left hand side of Eq(5) is not constant but the variable given by Eq(7).

As Clement (3) has pointed out, in the case of multi-sized CCN Eqs(2) and (3), i.e., Eqs(6) and (7) should be applied to the volume contains sufficient number of CCN which is able to characterize the CCN size distribution (composition). However, because CCN size is assumed to be a uniform size throughout the air parcel (CCN number density is N), the equilibrium droplet radius, a_e, is also uniform. In such cases the constant N can be included in the coefficient A_3 and thus the governing equations can be applied to the volume which contains only one CCN. Because the minimum volume which characterizes the composition of CCN (control volume) has to contain only one CCN, Eqs(2) and (3) [or Eqs(6) and (7)] can be applied to the volume of $1/N$ cm^3. However, the numerical results are not dependent on whether the control volume is set to 1 cm^3 of N CCN or $1/N$ cm^3 of one CCN, because m_{xe} per $1/N$ cm^3 is identical with $N \times m_{xe}$ per 1 cm^3.

NUMERICAL SIMULATION

In the numerical simulation CCN is assumed to be ammonium sulfate $(NH_4)_2SO_4$ (M_s = 132.0 g/mol) whose initial dry radius a_{s0} is 0.1 μm. van't Hoff factor ν varies with the chemical composition of CCN. It is assumed that $\nu \approx 3$, because one molecule of $(NH_4)_2SO_4$ dissociates almost perfectly into three ions in water, that is, two moles of NH_4^+ and one mole of SO_4^{2-} are produced from one mole of $(NH_4)_2SO_4$. Characterizing the condensational growth with use of the Köhler curve, the critical supersaturation $S_c - 1$ and the critical radius a_c are 5.94×10^{-4} and 1.35×10^{-4} cm, respectively. The physical properties used for the numerical simulations are tabulated in Table 1 (at 273.15 K). ρ_s is the density of $(NH_4)_2SO_4$ and it is used for the estimation of m_s from the initial CCN radius a_{s0}, or inversely a_{s0} from m_s.

Table 1 Values of Physical Properties at 273.15 K.

$\mathcal{R}_1$	$\mathcal{R}_3$	σ	e_{sat}	ρ_{we}	ρ_s	ν	$T(0)$
82.07	8.314$\times$10^7	75.67	6.108	1.001	1.769	3	273.15
atm cm^3/(mol K)	erg/(mol K)	dyn/cm	mb	g/cm^3	g/cm^3	—	K

Comparison of new model with Köhler model

Because N is virtually a unity in the Köhler model without the idea of CCN number density, a comparison between the two models must be made by putting $N = 1$ into the new model. In this case, growth is not competitive, and in the limited region below the critical saturation ratio, the estimated radius of the new model and that of the Köhler model are much alike.

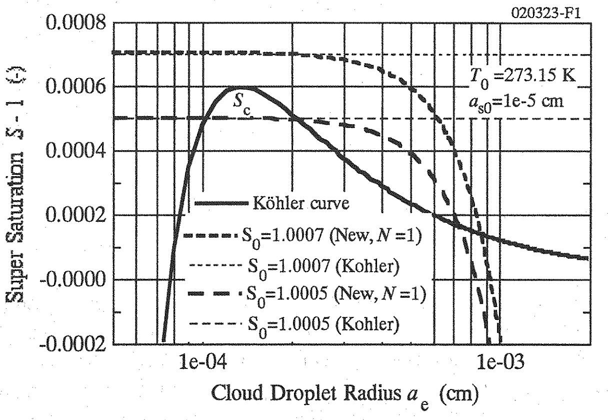

Fig. 1 Relationships between Super Saturation and Equilibrium Radius.

a) Köhler model

The Köhler model (equation) is obtained by replacing S_e of Eq(5) with S_0 as:

$$\ln (S_0) = \frac{A_1}{a_e} - \frac{A_2}{a_e^3} \tag{12}$$

This equation can be solved independently with respect to a_e to obtain the equilibrium cloud droplet radius. The procedure for obtaining the equilibrium droplet radius is illustrated by using Fig. 1. The equilibrium radius a_e is given as x-coordinate at the intersection of the Köhler curve with the horizontal line. The curve represented on the right hand side of Eq(12) is the Köhler curve (thick solid curve in Fig. 1) and the horizontal line (thin broken line in Fig. 1) approximately corresponds to the left hand side of Eq(12) is supersaturation line. If supersaturation $S_0 - 1$ is small, as usual atmospheric condition (below 10% and most often even below 1%), the left hand side of the Köhler equation $\ln(S_0)$ can be approximated by supersaturation $S_0 - 1$ as:

$$\ln(S_0) \approx S_0 - 1 \tag{13}$$

Therefore, Eq(12) is represented schematically as Fig. 1, where the abscissa is the droplet radius a_e in logarithmic scale and the ordinate is the supersaturation $S - 1$ in normal scale (the suffix 0 of S_0 is dropped because of dual use of Fig. 1 for the new model and the Köhler model). The upwardly-convex curve (opens downwards) is the Köhler Curve.

The Köhler model assumes the initial saturation ratio S_0 remains constant. Then, Supersaturation curve is represented by a horizontal line as shown by the thin dotted or the broken line. When the saturation ratio $S_0 = 1.0005$ (supersaturation: $S_0 - 1 = 5.0 \times 10^{-4}$, smaller than the critical supersaturation ratio 5.94×10^{-4}), there are two intersections. One is on the ascending branch ($a_e < a_c$) and the other is on the descending branch ($a_e > a_c$). Therefore, we can obtain the equilibrium radius by selecting the smaller one on the ascending branch, because the smaller one demands less energy. However, the Köhler curve has no intersection with the horizontal line, when $S_0 = 1.0007$, i.e., $S_0 - 1 = 7.0 \times 10^{-4}$ (greater than the critical supersaturation ratio 5.94×10^{-4}). In this case, the equilibrium radius cannot be obtained by using the Köhler model and the users of the Köhler model maintain that the droplet growth is everlasting.

b) New model

Such a singular case as the Köhler model fails to obtain the equilibrium droplet radius for the supersaturation above the critical value is never brought about in the new model. We can always obtain the equilibrium radius by means of the new model developed here, because the supersaturation curve given by Eq(7) in the new model is not a horizontal line but a descending curve as shown by the thick dotted curve ($S_0 = 1.0007$) and the thick broken curve ($S_0 = 1.0005$) in Fig. 1, which have at least one intersection for any initial saturation ratio (supersaturation). For example, in the case where $S_0 = 1.0005$ there are three intersections. However, in this case the first and the third radii are stable, but the second one is unstable. If there is more than one stable radius, the smallest radius is the right radius corresponding to the smallest energy consumption.

Effect of CCN number density on equilibrium radius, temperature and saturation ratio

If the air parcel contains more than one CCN, the growth of the cloud droplets is competitive and the competition is supposed to get keen with increase in the CCN number density. In order to investigate the effects of CCN number density, the normalized variables for a_e, T_e and S_e are introduced as follows:

$$\hat{a}_e = \frac{a_e - a_p}{a_p} \tag{14}$$

$$\hat{T}_e = \frac{T_e - T_0}{\Delta T_{as}} \tag{15}$$

$$\hat{S}_e = \frac{S_e - S_0}{S_0} \tag{16}$$

where a_p = potential radius (cm), which is obtained by Köhler model assuming $S_0 = 1$; and ΔT_{as} = condensational temperature variation (K) of the atmosphere due to small vertical perturbation ΔH (m) in altitude. Assuming $\Delta H = 1$ m, in the atmosphere of 1000 hPa and 0 K, ΔT_{as} is given by:

$$\Delta T_{as} = \Delta \Gamma_{as} \times \Delta H = 4.0 \times 10^{-3} \text{ K} \tag{17}$$

$$\Delta \Gamma_{as} = \Gamma_a - \Gamma_s \approx 4.0 \times 10^{-3} \text{ K/m} \tag{18}$$

where Γ_a and Γ_s = dry adiabatic lapse rate ($\approx 9.8 \times 10^{-3}$ K/m) and saturation adiabatic lapse rate ($\approx 5.8 \times 10^{-3}$ K/m), respectively.

When the moist air parcel is uplifted, latent heat is produced due to vapor condensation by cooling of air parcel. Because the lapse rate is depressed by the latent heat due to condensation, the temperature of the moist air parcel becomes higher than that of the dry ambient atmosphere. However, the temperature difference ΔT between the uplifted air parcel and the ambient atmosphere varies depending on the degree of the both lapse rates (dry, supersaturation and so on) and ΔT is not necessarily positive. If $\Delta T > 0$, the air parcel is statically unstable and is capable for buoyant convection. As the maximum lapse rate depression of the air parcel without CCN is $\Delta \Gamma_{as}$ given by Eq(18), the maximum temperature increase due to the uplift of 1 m is ΔT_{as} which corresponds to the most unstable state. It is thought that ΔT_{as} is a kind of scale to measure the static instability of the air parcel in which vapor condensation occurs.

On the other hand, in the air parcel containing CCN the vapor can condense on CCN without the cooling by uplifting of the air parcel. Therefore, the temperature (T_e) of the stationary air parcel with CCN becomes always higher than that (T_0) of the ambient atmosphere (without CCN). Because the temperature difference ($\Delta T = T_e - T_0$) is always positive, the stationary air parcel with CCN becomes always statically unstable. Eq(15) means that the larger the normalized temperature ($\hat{T}_e$) is, the more unstable the air parcel with CCN becomes.

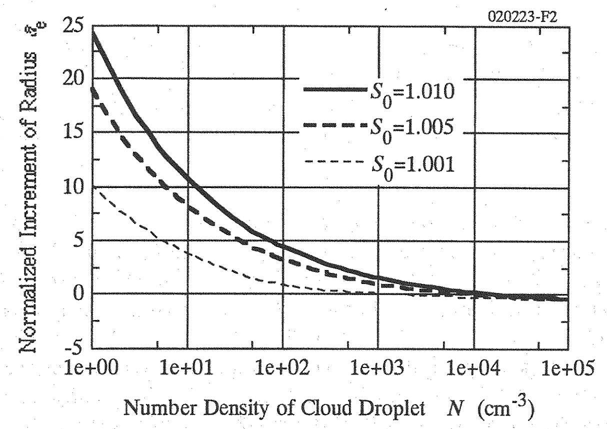

Fig. 2 Relationships between Equilibrium Radius and Number Density of CCN.

a) Equilibrium cloud droplet radius

In Fig. 2 the relationships between normalized equilibrium radius $\hat{a}_e$ and CCN number density N are plotted parametrically in the initial saturation ratio S_0. As is supposed, the larger N becomes, the smaller $\hat{a}_e$ gets due to competitive growth. This competitive growth cannot be estimated by the Köhler model. However, as is seen in Fig. 1, in the case that $N = 1$ for the supersaturation less than critical value (5.94×10^{-4}) the radius estimated by the new model is almost the same as the radius obtained by the Köhler model. This is because the supersaturation curve by the new model can be approximated by the horizontal line with which the ascending branch of the Köhler curve intersects.

In the region of N less than about 1000 cm^{-3}, $\hat{a}_e$ decreases drastically with increase in N. On the other hand, for N larger than about 1000 cm^{-3}, the decrease is moderate. A comparison of Fig. 2 with Figs. 3 and 4, suggest these results may be attributed to the difference of the vapor consumption mode between the two regions of N. The effect of N on the decrease in the equilibrium size, that is, the sensitivity of the size reduction to N, can be estimated by the gradient of the $\hat{a}_e$ curves against N. The effect proves to be enhanced with decrease in N and also with increase in S_0. Anyway, Fig. 2 suggests that the Köhler model causes a considerable prediction error in the equilibrium radius, because this model does not consider N.

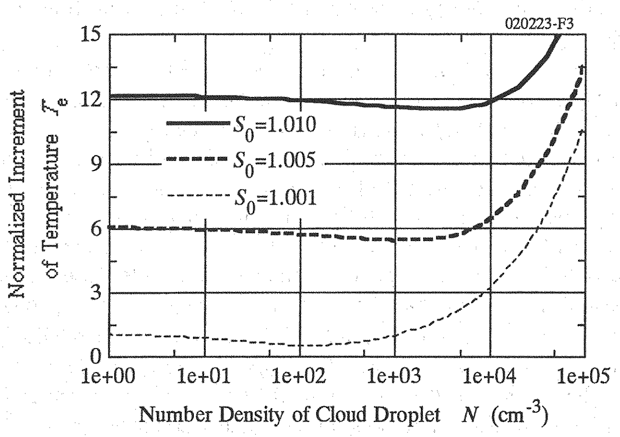

Fig. 3 Relationships between Temperature and Number Density of CCN.

b) Equilibrium temperature

In Fig. 3 the relationships between $\hat{T}_e$ and N are shown parametrically in S_0. They reveal that $\hat{T}_e$ increases with increase in S_0. Under usual atmospheric conditions the temperature increase $T_e - T_0$ is less than about 10^{-2} K for $N < 10^4$ and it does not change the physical properties effectively. However, with increase in N the temperature increase $T_e - T_0$ becomes considerably larger than ΔT_{as} which is the maximum temperature increase due to condensation and brought about by uplifting of the air parcel in the atmosphere as mentioned before. From a meteorological point of view, such a temperature increase is noteworthy, because it causes the thermal convection by generating the statical instability in the atmosphere. Thus a temperature increase has the potential to trigger off a cumuliform cloud which brings about local precipitation.

$\hat{T}_e$ curves have the minimum. The descending branch of $\hat{T}_e$ curves (low N side) correspond to the decrease in heat generation, i.e., the decrease in condensation. This means that the amount of condensation per one droplet decreases considerably with each increase in N to bring about sharp reduction in droplet size with increase in N. In fact $\hat{a}_e$ curves in Fig. 2 sharply decrease in small N region. The sharp ascending branch of $\hat{T}_e$ curves (high N side) in Fig. 3 show that the heat generation increases considerably with increase in N (high N side). This suggests that the vapor consumption sharply increases, that is, $\hat{S}_e$ decreases sharply in this region.

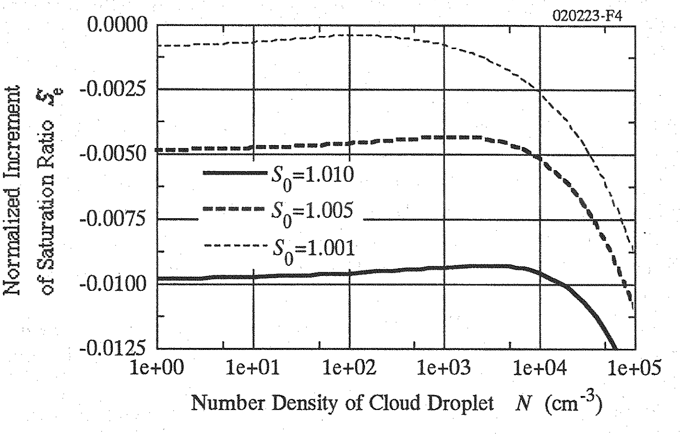

Fig. 4 Relationships between Saturation Ratio and Number Density of CCN.

c) Equilibrium saturation ratio

Fig. 4 represents the relationships between $\hat{S}_e$ and N parametrically in S_0. Because the vapor in the air parcel is diminished to change into droplets (i.e., $S_e < S_0$), it is expected that $\hat{S}_e$ defined by Eq(16) becomes negative. $\hat{S}_e$ curves shown in Fig. 4 agree well with such an expectation. It can be seen that the reduction of vapor in the air parcel is enhanced with increase in S_0. Downwards-open $\hat{S}_e$ curves have the maximum. Therefore, $\hat{S}_e$ curves and $\hat{T}_e$ curves organize a symmetry of the shape with respect to x-axis. This can be said a matter of course, because the latent heat release to raise $\hat{T}_e$ is done by vapor condensation to lower $\hat{S}_e$.

It should be noted that in the region of small N the $\hat{S}_e$ (negative value of $\hat{S}_e$ means vapor reduction) increases with increase in N, in other words, the reduction of vapor in the air parcel decreases with increase in N, although reduction of vapor is expected to increase with increase in N. This may be explained by the fact that such a sharp reduction of individual droplet size due to competitive growth (see Fig. 2) lowers the total amount of vapor changed into droplets until N gives the maximum of $\hat{S}_e$ (the minimum of vapor consumption).

CONCLUSIONS

From the results of numerical simulations made by the new mathematical model developed here, it is concluded that:

1. The reduction of the equilibrium cloud droplet radius ($\hat{a}_e$) due to competitive growth becomes more remarkable with increase in the initial saturation ratio (S_0).
2. The increase of the normalized equilibrium temperature ($\hat{T}_e$) with increase of CCN number density (N) is not monotonous but $\hat{T}_e$ curves have the minimum value, that is, the curves open upwards with the descending and the ascending branches.
3. A temperature increase by competitive growth of cloud droplet ($T_e - T_0$) is too small to have an effect on the equilibrium cloud droplet radius, but large enough to make the air parcel statically unstable and to trigger off the thermal convection in the atmosphere, comparing with the temperature increase due to adiabatic uplift of moist air parcel (ΔT_{as}).
4. The variation of vapor consumption ($\hat{S}_e$) with CCN number density (N) coincides well with the variation of temperature ($\hat{T}_e$) due to latent heat release which is estimated from both CCN number density (N) and the droplet radius reduction ($\hat{a}_e$).

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APPENDIX–NOTATION

The following symbols are used in this paper;

a_{c} = critical radius of cloud droplet given by Köhler model;
a_{e} = equilibrium radius of cloud droplet;
a_{p} = potential radius of cloud droplet given by Köhler model;
a_{s0} = initial dry radius of cloud condensation nucleus;
a_{x} = activity in x-phase ("x" is "a" for air, "v" for water vapor and "w" for liquid water);
$\hat{a}_{\mathrm{e}}$ = normalized equilibrium radius of cloud droplet;
A_1 = coefficients in Eq(5);
A_2 = coefficients in Eq(5);
A_3 = coefficients in Eq(7);
A_4 = coefficients in Eq(7);
C_{p} = difference of isobaric specific heat given by $C_{\mathrm{pw}} - C_{\mathrm{pv}}$;
C_{px} = isobaric specific heat of x-phase;
e = water vapor pressure;
e_{sat} = saturation water vapor pressure;
h_x = enthalpy per unit mass of x-phase;
L_{e} = latent heat of water vapor;
m_{x} = mass of x-phase;
m_{xe} = equilibrium mass of x-phase;
m_{s} = mass of CCN;
M_{s} = molecular weight of CCN;
M_{w} = molecular weight of water;
n = total moles of air, water vapor and liquid water;
N = number density of CCN;

$\mathcal{R}$ = universal gas constant;
$\mathcal{R}_1$ = universal gas constant [= 82.06 atm cm^3/(mol K)];
$\mathcal{R}_3$ = universal gas constant [= 8.314 erg/(mol K)];
S = saturation ratio of moist air;
S_0 = initial saturation ratio of moist air;
S_e = equilibrium saturation ratio of moist air;
$\hat{S}_\mathrm{e}$ = normalized equilibrium saturation ratio of moist air;
T = temperature;
T_0 = initial temperature;
T_e = equilibrium temperature;
$\hat{T}_\mathrm{e}$ = normalized equilibrium temperature;
ΔT_as = temperature increment due to condensation of saturation vapor in the air parcel;
Γ_a = dry adiabatic lapse rate (= 9.8×10^{-3} K/m);
Γ_s = saturation adiabatic lapse rate (= 5.8×10^{-3} K/m);
$\Delta\Gamma_\mathrm{as}$ = difference of adiabatic lapse rate given by $\Gamma_\mathrm{a} - \Gamma_\mathrm{s}$;
μ_x = chemical potential of x-phase;
μ_x0 = integration constant for chemical potential of x-phase;
ν = van't Hoff factor;
π = circular constant;
ρ_s = density of CCN;
ρ_we = density of water; and
σ = surface tension of cloud droplet.

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