

Theories of Competitive Cloud Droplet Growth and Their Application to Cloud Physics Studies

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ABSTRACT

In a rising cloud parcel, a small droplet establishes quasi-steady-state fields of vapor density ρ and temperature T around it, approximately satisfying the common relationship, $\nabla^2\rho = \nabla^2T = 0$, where ∇^2 is the Laplacian operator. The fields at this stage merely transfer vapor and heat between the droplet surface and the infinite environment without changing their distributions in space; lowering of ρ and T fields is primarily due to cloud parcel lifting and not by the droplet growth. As the growth continues, the fields begin to compete with others of adjacent droplets and limit their expanse within each territory referred to as "cell." The supersaturation reaches the maximum shortly thereafter, and the growth mode becomes totally competitive. Under this cell boundary-controlled competitive kinetics, the fields establish a new steady state and transfer properties between the droplet surface and the space inside the cell where $\nabla^2\rho$ and ∇^2T take nonzero constant values. Under this condition, the ρ and T fields are lowered due to both parcel lifting and droplet growth.

To evaluate the difference between the droplet growth of conventional theories and the competitive growth under steady-state conditions, equations describing the latter vapor and temperature fields as well as the mass growth rates are analytically obtained in closed forms under both Maxwellian and diffusion-kinetic conditions. A factor responsible for the competitive kinetics is identified as a function of only the ratio between the droplet radius and the distance between the droplet center and the boundary with the surrounding droplets. Deviation of droplet mass growth rate for the competitive kinetics from that for noncompetitive growth is found to be negligible under the ordinary range of cloud conditions. However, it is pointed out that under unusual situations, laboratory processes in particular, consideration of the deviation may become necessary.

1. Introduction

Clouds form when moist air parcels rise through the cloud base and nucleation of water vapor condensation on foreign particles takes place with subsequent growth of the formed droplets under relative humidity exceeding 100%. During the growth process, the water vapor in air is continually transported to the droplet surface and generates the latent heat of condensation. The heat generated is removed by conduction at the same rate as the condensation heating, thereby establishing a quasi-steady state between the two transport processes. Under the steady state, the vapor and temperature fields take a form that makes the transfer rates of both vapor and heat the same across any closed surface that envelopes the droplet.

At the inner boundary of the space for heat and vapor transfers, which is the droplet surface, the steady-state condition is violated due to the droplet radius increasing. Application of the steady-state assumption brings in an error of a few tenths of a percent (Fuchs 1959). Any dissolved solute in the droplet helps the nucleation

to occur by lowering the vapor pressure at the droplet surface. The surface tension of the droplet, on the other hand, increases the vapor pressure above that which is in equilibrium with a flat surface of pure water. However, both of these effects rapidly disappear as the droplet grows (Mason 1971).

Fields for both vapor and heat transfer are limited by inner and outer boundaries. The inner boundary is represented by the droplet surface, the outer one by the environment. Two analytic solutions exist for droplet growth kinetics depending on the assumption at the surface. Maxwell (1890) considered growth of the droplet in which the saturation vapor pressure of water with flat surface and the temperature were assumed just above the droplet surface. This Maxwellian condition allows both fields to be continuous right up to the droplet surface, whereas it has been noted that when mass, heat, or even momentum is to be exchanged between the gaseous and condensed phases, an inefficiency appears that is expressed by a condensation, thermal accommodation, or momentum accommodation coefficient, respectively. As a result, the condition of the gaseous phase just above the surface deviates from that of equilibrium. Under such a condition, an analytic solution of droplet growth kinetics can be used (Fukuta and Walter 1970), and the treat-

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ment is sometimes referred to as “diffusion kinetic” (Smirnov 1971). These two theories have been frequently used in cloud physics depending on the conditions and purpose.

The transfer of heat and vapor takes place through the respective fields established around the droplet in question. For the Maxwellian treatment, which causes the largest temperature difference between the droplet surface and the environment due to the lack of diffusion-kinetic resistance effect (Fukuta and Walter 1970), application of a 1% supersaturation at a temperature of 10°C and a pressure of 1000 mb causes a warming of 0.09°C at the droplet surface. The saturation vapor density, the latent heat of condensation at the droplet surface, the thermal conductivity of air, and the diffusivity of water vapor in air all depend on temperature. In this case, use of these constants throughout the system at the environmental temperature underestimates the growth rate by less than 0.4%.

In the presence of a temperature gradient, the mass flux of water vapor diffusion toward the growing droplet surface is influenced not only by the gradient of vapor density but also by the temperature gradient (Fleagle and Businger 1963). Similarly, the heat energy flux is affected not only by the temperature gradient but also by the vapor density gradient. The errors produced by ignoring the temperature gradient effect on the vapor diffusion and the vapor density effect on heat conduction are 2.8% at 20°C for water droplets and 2.0% at -30°C for ice crystals (see appendix A).

Fall of a cloud droplet makes the temperatures and vapor fields deform. However, in the Stokes regime of fall, where cloud droplets belong, the increase in the rate of growth on the front face of the droplet equals the decrease on its rear face. Hence, the overall growth rate is equal to that of a stationary droplet (Fuchs 1959).

The extent of the effects just described is thus either known or accessible by known methods. Accordingly, improvement of the two theories given for steady-state droplet growth, that is, the Maxwellian and the diffusion kinetic for these effects, may be carried out.

2. Fundamental equations for vapor density and temperature fields around a growing droplet

For the temperature and vapor density fields around the growing droplet without their advective transportation, we can express their changes with respect to time at an arbitrary position in an elementary volume of isotropic space based on the diffusional transportation and the source-sink effect as

$$\frac{\partial \rho}{\partial t} = D \nabla^2 \rho + F_1(x, y, z, t) \quad (1)$$

and

$$\frac{\partial T}{\partial t} = \kappa \nabla^2 T + F_2(x, y, z, t), \quad (2)$$

where ρ is the vapor density, t the time, D the diffusivity of water vapor in air, F_1 the source-sink term for vapor density, T the temperature, F_2 the source-sink term for the temperature that is attributed to the original source-sink term of heat energy, respectively; and

$$\nabla^2 = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2} \quad (3)$$

is the Laplacian operator for Cartesian coordinates of x , y , and z ; and

$$\kappa = \frac{K}{c_p \rho_a} \quad (4)$$

is the thermal diffusivity with K , c_p , and ρ_a being the thermal conductivity, specific heat at constant pressure, and density of the air, respectively. The first terms on the right-hand sides of Eqs. (1) and (2) describe the property transportation in an elementary volume by diffusion mechanism where the difference between the incoming and the outgoing fluxes decides the increase of the property level.

In the customary treatment (e.g., Fletcher 1962; Pruppacher and Klett 1978; Rogers and Yau 1989), steady-state temperature and vapor density fields around a growing or evaporating droplet are taken with $\partial \rho / \partial t = 0$ and $F_1 = 0$ as well as $\partial T / \partial t = 0$ and $F_2 = 0$, so that

$$\nabla^2 \rho = \nabla^2 T = 0 \quad (5)$$

is assumed. The ρ and T fields in this equation merely permit their transfer between the droplet surface and an infinite environment without changing the fields in between them.

Figure 1 describes the ρ and T fields around a droplet in the adiabatic cooling cloud air in which both fields keep lowering. Therefore, at the beginning of the process, changes of ρ and T exist, contrary to the aforementioned treatment, and $\partial \rho / \partial t - F_1 = \text{const}$ and $\partial T / \partial t - F_2 = \text{const}$ hold. Nevertheless, at this stage, they are influenced only by the cloud process and not yet by the droplet growth. By introducing the above conditions into Eqs. (1) and (2), one can show that condition (5) is also valid in the early stage of cloud droplet growth (see noncompetitive growth part of Fig. 1). In this stage, both fields at the would-be cell boundary, R , are totally dry adiabatic, showing no effect of droplet growth.

In the early droplet growth under a given supersaturation, the radius changes rapidly causing a deviation from the steady-state assumption of ρ and T fields. The problem was explicitly treated by Reiss and LaMer (1950) for the ρ field alone under $F_1 = 0$, leaving the ρ and T at the droplet surface unsolved. They noted the existence of impermeable shells (cell boundaries) between droplets and included zero gradient of ρ and T there. Their treatment, solved only for the ρ field, is hence regarded as nonsteady-state, cell boundary-controlled kinetics without source-sink.

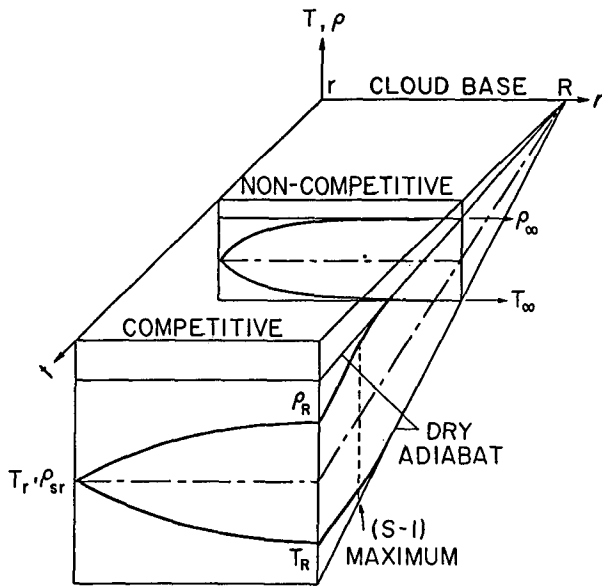


FIG. 1. Variation of vapor density ρ and temperature T fields around a droplet and associated transition of growth mode from noncompetitive to competitive while a cloud parcel is rising from the cloud base. The droplet growth condition is Maxwellian. The levels of ρ and T fields at the cell boundary R initially shift linearly with time along the respective dry adiabats. As the droplet grows, the fields spread and begin to interact with adjacent ones and the initial mode of transportation between the droplet surface and the infinite environment gradually shifts to a new steady-state mode in which the transportation proceeds between the droplet surface and the space inside the cell by changing the field style. This mode becomes totally dominant when the system passes through the zone of maximum supersaturation and the time rates of change of ρ and T fields become approximately controlled by the pseudoadiabatic process of the cloud. For notations, see the text.

As the droplet grows, the fields reach the cell boundary and begin to compete with those of other surrounding droplets. This process becomes totally dominant after the cloud system has passed through the zone of maximum supersaturation above the cloud base and ceased to nucleate new droplets. The supersaturation maximum depends on the nucleation-growth-cloud dynamics interaction above the cloud base (Fukuta 1990). In this limited space around the droplet, the excess vapor is made available and the generated heat by vapor condensation onto the droplet is distributed. The process is under the influence not only of the average but also of fluctuations of the updraft velocity and the droplet spacing (Srivastava 1989). The present paper deals only with an idealized system without such effects.

When the supersaturation maximum is passed and the droplet growth becomes competitive, the system establishes a new quasi-steady state: The cell space now receives uniformly the heating effect of droplet growth and loses the corresponding amount of vapor to the droplet surface. As a result, $\partial\rho/\partial t - F_1$ and $\partial T/\partial t - F_2$ become nonzero constants approximately correspond-

ing to the pseudoadiabatic rate of change, and the governing condition for both fields changes from Eq. (5) to

$$\nabla^2 \rho = \frac{1}{D} \frac{\partial \rho}{\partial t} - \frac{F_1}{D} = \text{const} \neq 0 \quad (6)$$

and

$$\nabla^2 T = \frac{1}{\kappa} \frac{\partial T}{\partial t} - \frac{F_2}{\kappa} = \text{const} \neq 0. \quad (6')$$

This is to say that, in this stage of competitive cloud droplet growth, the droplet growth and the adiabatic cloud process both contribute to changes of ρ and T , whereas in the early growth stage of cloud droplets, the former did not.

In Fig. 1, the competitive growth portion describes the condition of a growing cloud droplet after the supersaturation maximum. Both ρ and T fields have reached the cell boundary with ρ reduced from the dry adiabatic line to ρ_R and T raised from that to T_R . Note that dry adiabats for both ρ and T are lowering with time due to the adiabatic expansion. In addition, the gradients of ρ and T are now zero at the cell boundary, permitting no transfer of these properties across it.

Conditions (6) and (6') with the cell boundary restriction thus govern the droplet growth kinetics after the supersaturation maximum, and this mode of growth prevails for the remaining growth period much longer than that before the maximum. The growth process takes a fundamentally different mechanism compared with the conventional one. To treat the cell boundary-controlled growth of droplet, Carstens (1979) described a way in which a spherical shell, impermeable to vapor, is circumscribed about each droplet and in it the mass balance is considered. The treatment deals with the ρ field alone without the source-sink term and leaves ρ and T of the droplet surface unsolved. The ρ field treated is that which was originally obtained for condition (5) and has an infinite expanse. The field, therefore, fails to satisfy the necessary vapor impermeability condition at the cell boundary, causing inconsistency in the treatment. The same inconsistency also appears in the conventional approach to this competitive droplet growth kinetics where the solution of the noncompetitive, steady-state kinetics under condition (5) is applied with consideration of mass and energy balance. Evaluation of any error that arises from the use of noncompetitive kinetics in place of competitive ones based on an independent, analytic solution is necessary. It is, therefore, the purpose of this paper to obtain the analytic solution for the cell boundary-controlled (competitive) kinetics of the cloud droplet growth under conditions where ρ and T fields adjust to attain quasi-steady-state profiles with source-sink terms of the respective properties uniformly distributed in the cell space and to identify the error associated with the use of the conventional, non-

cell boundary-controlled (noncompetitive), steady-state kinetics.

3. Cell-boundary-controlled, steady-state kinetics

Suppose a droplet of radius r is located in a cloudy air parcel rising from the cloud base past the zone of maximum supersaturation. Due to the existence of other droplets, its ρ and T fields are now restricted within a cell space of radius R . The continuous lifting of the air parcel generates an additional supersaturation within the cell that is entirely removed by the growth process of the droplet, thereby maintaining a nearly constant level of supersaturation. The droplet has grown already for some period of time so that the Kelvin effect of surface tension to raise the droplet vapor pressure and the solute effect to reduce the vapor pressure, both of which are large for a small droplet, can now be ignored. Within the space between the droplet surface (inner boundary) and the cell boundary (outer boundary) transportation of the vapor takes place according to Fickian diffusion and that of heat based on Fourier's law of heat conduction. The vapor is removed uniformly from the limited space, and the heat generated at the droplet surface is similarly returned to it.

a. Maxwellian theory

In the process of exchanging vapor and heat between the air and the droplet surface, factors describing the efficiencies of their exchanges are known to exist. They are condensation and thermal accommodation coefficients and cause discrete profiles of the fields at the surface (Fukuta and Walter 1970). While this effect decreases with increasing droplet radius to give continuous field profiles at the surface, one can assume the continuous profiles regardless of the coefficients and the radius. Such a condition is called Maxwellian (Maxwell 1890) and gives an upper limit for the growth rate (Fukuta and Walter 1970). At the surface of the droplet, a relationship exists between the saturated vapor density, ρ_{sr} , and the temperature, T_r , which may be expressed by the Clausius-Clapeyron equation.

The problem of droplet growth with the Maxwellian condition under the cell boundary-controlled, steady-state kinetics considering conditions (6) and (6') involves two unknowns, ρ_{sr} and T_r , described by the following two equations: a continuity equation of latent and sensible heat fluxes given by the two fundamental transport equations of vapor and heat within the cell boundary and the Clausius-Clapeyron equation that connects ρ_{sr} and T_r at the droplet surface. Therefore, if suitably simplified in linear forms, in particular, it is analytically soluble provided that the field style is identified.

Under the steady-state condition of the system in question, the rate of supersaturation generation in the air space surrounding the droplet is nearly constant.

To maintain this state, removal of the generated supersaturation is required at the same rate. The removal takes place through automatic adjustment of vapor density and temperature gradients, and it is the process to determine the field style.

Consider an arbitrary spherical surface of radius r in between r and R . Within a given period of time, the total amount of supersaturation newly generated in the space between spherical surfaces of radius r and R must completely be compensated by the transfer of the vapor toward the droplet with the help of the gradient established, or

$$\frac{dm}{dt} = C_1 \left(\frac{4}{3} \pi R^3 - \frac{4}{3} \pi r^3 \right) = 4\pi r^2 D \frac{d\rho}{dr}, \quad (7)$$

where, m , t , C_1 , D , and ρ are, respectively, the droplet mass, the time, the proportional constant, the vapor diffusivity, and the vapor density. The volume change due to an expansion by lifting and additional heating by water vapor condensation are ignored. After integration with respect to r and inserting the boundary condition at the droplet surface, $r = r$, $\rho = \rho_{sr}$ and that at the cell boundary, $r = R$, $\rho = \rho_R$, Eq. (7) yields

$$\rho = - \frac{(\rho_R - \rho_{sr})r}{(R - r)[2R^2 - r(R + r)]} \times \left[\frac{2R^3}{r} + r^2 - 3R^2 \right] + \rho_R. \quad (8)$$

A different treatment (Carslaw and Jaeger 1959) gives the same result. The main difference between this equation and the Maxwellian vapor field without the cell boundary restriction (Fukuta and Walter 1970) is the term r^2 .

By differentiating Eq. (8) with respect to r , we obtain the vapor density gradient

$$\frac{d\rho}{dr} = - \frac{(\rho_R - \rho_{sr})r}{(R - r)[2R^2 - r(R + r)]} \left(- \frac{2R^3}{r^2} + 2r \right). \quad (9)$$

Equation (9) $\rightarrow 0$ when $r \rightarrow R$, satisfying the necessary condition that no vapor transport takes place across the cell boundary. At $r = r$, Eq. (9) becomes

$$\frac{d\rho}{dr} = \frac{\rho_R - \rho_{sr}}{r} f_c, \quad (10)$$

where

$$f_c = \frac{2(1 + g + g^2)}{(2 - g - g^2)}, \quad (11)$$

and

$$g = \frac{r}{R}. \quad (12)$$

Here $f_c \rightarrow 1$, without cell boundary restriction or when $g = r/R \rightarrow 0$. Even under continuous generation of

supersaturation due to the lifting of a cloud parcel, which is not assumed in the conventional theory, the form of the vapor field remains the same as that without the cell boundary restriction if surrounding droplets are separated sufficiently. This is because the droplet growth effect is still negligible compared with the available space that ρ and T changes in Eqs. (1) and (2) are entirely controlled by respective source-sink terms satisfying condition (5) of the conventional theory.

Similarly, the corresponding temperature field may be obtained as

$$T = - \frac{(T_R - T_r)r}{(R - r)[2R^2 - r(R + r)]} \times \left[\frac{2R^3}{r} + r^2 - 3R^2 \right] + T_R, \quad (13)$$

where T_R and T_r are the air temperature at the cell boundary and that at the droplet surface, respectively (see Fig. 2).

Equation (13) gives

$$\frac{dT}{dr} = \frac{T_R - T_r}{r} f_c. \quad (14)$$

Fourier's law states that the sensible heat flux at the surface of a sphere of radius r is

$$\frac{dQ}{dt} = -4\pi r^2 K \frac{dT}{dr}, \quad (15)$$

where K is the thermal conductivity of air. The continuity condition of latent and sensible heat fluxes at the surface may be described as

$$\frac{dQ}{dt} = L \frac{dm}{dt}, \quad (16)$$

where L is the latent heat of condensation. Substitution of Eqs. (10) and (14) in Eqs. (7) and (15) and using Eq. (16) results in

$$\frac{LD}{K} = - \frac{T_R - T_r}{\rho_R - \rho_{sr}}. \quad (17)$$

Now the second condition, the Clausius-Clapeyron equation integrated from (T_r, ρ_{sr}) to (T_R, ρ_{sR}) and expressed in its linear form, becomes

$$\frac{L(T_r - T_R)}{R_v T_R^2} = \frac{\rho_{sr}}{\rho_{sR}} - 1, \quad (18)$$

where ρ_{sR} is the saturation vapor density of water at $T = T_R$. This assumes $e_s \propto \rho_s$ is the saturation vapor pressure of water, and it is valid only when T_s is constant. As a result, the $L/R_v T$ term in Eq. (18) is larger than the correct one that considers the effect of T on ρ_s , by less than 5% (Mason 1971). Equations (17) and (18) yield

$$\rho_{sr} - \rho_R = (S_R - 1) \left/ \left[\frac{DL^2}{KR_v T_R^2} + \frac{1}{\rho_{sR}} \right] \right., \quad (19)$$

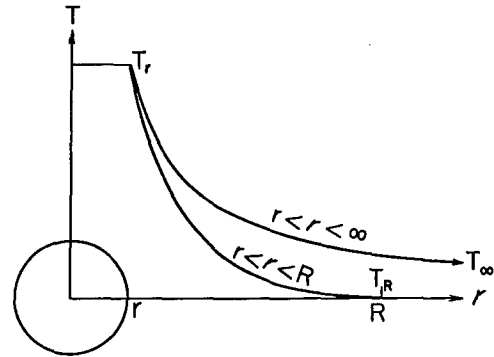


FIG. 2. Comparison of the temperature field between the noncompetitive ($r < r < \infty$) and the competitive ($r < r < R$) cases for growing cloud droplet. Note the gradient of the latter goes to zero at the cell boundary ($r = R$), allowing no transfer of heat across it, whereas the former permits a finite gradient at the boundary for heat transfer.

where $S_R = \rho_R/\rho_{sR}$ is the saturation ratio at $r = R$. Equation (19) is the solution for T_r and ρ_{sr} although under the condition of $T_r \approx T_R$, T_r is hidden.

Replacing Eqs. (19) and (10) into Eq. (7), we finally obtain the droplet mass growth rate equation

$$\frac{dm}{dt} = \left\{ 4\pi r (S_R - 1) \left/ \left[\frac{L^2}{KR_v T_R^2} + \frac{1}{D\rho_{sR}} \right] \right. \right\} f_c. \quad (20)$$

Equation (20) is identical to the commonly used Maxwellian growth rate except for f_c , the cell boundary-controlled (competitive) kinetics factor, which is a function of the ratio r/R .

Figure 3 shows the relationship between f_c and r/R . It can be seen that $f_c > 1$, implying the dm/dt of cell boundary-controlled kinetics, which is closer to reality, is faster than the ordinary Maxwellian rate. As described in Fig. 2, the steady-state, competitive T field (lower curve) terminates with $T = T_R = T_\infty$ at $r = R$. This makes the growth rate-controlling gradient at the droplet surface ($r = r$) larger, even if the field takes the hyperbolic form of a conventional field (upper curve, see also Fukuta and Walter 1970). The hyperbolic form always develops as long as a property is transferred in an isotropic space to or from a spherical surface. In the competitive field, due to the necessity for additional transportation from the cell space to the droplet surface, the field becomes doubly steep and increases the growth rate. However, under the typical cloud condition of $r/R \approx 0.01$ or the average cloud liquid water content of about 0.5 g m^{-3} , $f_c \approx 1.015$ and the difference between the two treatments is negligible.

b. Diffusion-kinetic theory

The system of a droplet growing under the influence of a condensation coefficient, β , and a thermal accommodation coefficient, α , has four unknowns: ρ_{sr} , ρ_Δ ,

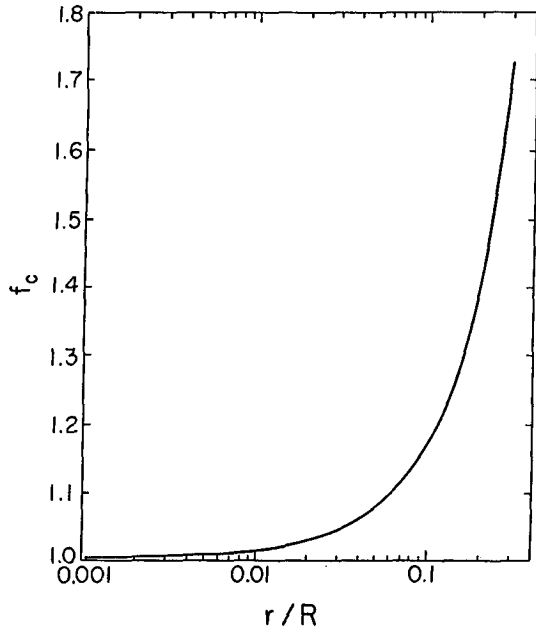


FIG. 3. The factor for cell boundary-controlled kinetics, f_c , logarithmically plotted as a function of the ratio between the droplet radius and the cell boundary distance, r/R .

T_r , and T_Δ , where ρ_Δ and T_Δ are the vapor density and the temperature of the droplet and those in air right above the droplet surface, respectively. To solve this system, four equations are necessary, preferably all in linear form. There are four different transport processes in the system: 1) the continuous vapor diffusion from the environment to the gaseous boundary right above the droplet surface (Fick's law), 2) the free molecular (Knudsen) flow of water vapor from this boundary to the droplet surface, 3) the free molecular flow of heat from the surface to the boundary layer, and 4) the continuous conduction of heat from the layer to the environment under the steady-state growth condition. Equality of these four processes leads to three equations. The three equations and the Clausius-Clapeyron equation at the droplet surface make up the four necessary equations to solve the problem. The present cell boundary-controlled condition affects only the diffusional fields of vapor and heat outside the boundary layer.

To treat the kinetic process of droplet growth in question, we take an approach similar to that of Fukuta and Walter (1970), by defining two normalization factors:

$$f'_\beta = \frac{\rho_R - \rho_\Delta}{\rho_R - \rho_{sr}} \tag{21}$$

and

$$f'_\alpha = \frac{T_R - T_\Delta}{T_R - T_r} \tag{22}$$

It should be noted that $f'_\beta \rightarrow f_\beta$ when $\rho_R \rightarrow \rho_\infty$, and $f'_\alpha \rightarrow f_\alpha$ when $T_R \rightarrow T_\infty$, where f_β and f_α are the factors free from cell boundary restriction. For the present system, Eqs. (10) and (14) become

$$\frac{d\rho}{dr} = \frac{\rho_R - \rho_\Delta}{r} f_c \tag{23}$$

and

$$\frac{dT}{dr} = \frac{(T_R - T_\Delta)}{r} f_c \tag{24}$$

Since the diffusional vapor and heat fluxes match the free molecular fluxes, respectively, at the outside of the boundary layer,

$$\left(\frac{R_v T_r}{2\pi}\right)^{1/2} \frac{2\beta}{(2-\beta)} (\rho_\Delta - \rho_{sr}) = D \frac{(\rho_R - \rho_\Delta)}{r} f_c, \tag{25}$$

and

$$\frac{p(c_v + 1/2 R_a)}{(2\pi R_a T_r)^{1/2}} \frac{2\alpha}{(2-\alpha)} (T_\Delta - T_r) = K \frac{(T_R - T_\Delta)}{r} f_c, \tag{26}$$

which, under the conditions of Eqs. (21) and (22), yield

$$f'_\beta = \frac{r}{r + l'_\beta}, \tag{27}$$

where

$$l'_\beta = \frac{(2-\beta)}{2\beta} \left(\frac{2\pi}{R_v T_r}\right)^{1/2} D f_c, \quad (T_r \approx T_R) \tag{28}$$

and

$$f'_\alpha = \frac{r}{r + l'_\alpha}, \tag{29}$$

with

$$l'_\alpha = \frac{(2-\alpha)}{2\alpha} \frac{K(2\pi R_a T_r)^{1/2}}{p(c_v + 1/2 R_a)} f_c. \tag{30}$$

Further, the continuity condition of the latent and the sensible heat fluxes gives, using Eqs. (16), (15), (23), and (24),

$$\frac{LD}{K} = - \frac{(T_R - T_\Delta)}{(\rho_R - \rho_\Delta)}. \tag{31}$$

Combination of this and Eq. (18), with the definitions given by Eqs. (21) and (22), yields a solution,

$$\rho_R - \rho_{sr} = (S_R - 1) \left/ \left[\frac{L^2 D}{KR_v T_r^2} \frac{f'_\beta}{f'_\alpha} + \frac{1}{\rho_{sR}} \right] \right. \tag{32}$$

Observing that at $r = r$, $d\rho/dr$ in Eq. (7) corresponds to Eq. (23) in the present system and substituting Eqs. (21) and (32) into Eq. (7), we obtain the diffusion-

kinetic equation of droplet mass growth under cell boundary restriction:

$$\frac{dm}{dt} = \left\{ 4\pi r (S_R - 1) \left/ \left[\frac{L^2}{KR_v T_R^2 f'_\alpha} + \frac{1}{\rho_{sR} D f'_\beta} \right] \right. \right\} f_c. \quad (33)$$

Equation (33) converges into the customary diffusion kinetic equation

$$\frac{dm}{dt} = 4\pi r (S - 1) \left/ \left[\frac{L^2}{KR_v T_\infty^2 f'_\alpha} + \frac{1}{\rho_{s\infty} D f'_\beta} \right] \right., \quad (34)$$

under the noncompetitive condition, as $S_R \rightarrow S$, $T_R \rightarrow T_\infty$, $\rho_{sR} \rightarrow \rho_{s\infty}$, $f'_\beta \rightarrow f'_\beta$, $f'_\alpha \rightarrow f'_\alpha$, and $f_c \rightarrow 1$. Also, by examining f'_β and f'_α terms in Eqs. (27) through (30), it is clear that β and α , the factors responsible for the diffusion-kinetic effects, are connected in such a manner that f_c increase is equivalent to β and α approach to unity, or reduction of the effects.

4. Discussion and concluding remarks

In the cloud parcel rising from the base, it has been pointed out that the water vapor and temperature fields around a droplet take approximately the commonly cited condition of $\nabla^2 \rho = \nabla^2 T = 0$ in its early stage of growth before reaching the supersaturation maximum, although lowering of ρ and T already exists for which cloud parcel lifting is solely responsible. There, the rapidly increasing droplet radius makes the condition deviate somewhat from the steady state. The steady-state fields under the condition of $\nabla^2 \rho = \nabla^2 T = 0$ simply indicate transfer of the vapor and heat between the droplet surface and the infinite environment without changing their distributions in space. As the droplet continues to grow in the cloudy expanding air, its ρ and T fields eventually come to compete with those of adjacent droplets and their expanse becomes limited within their own territory, called "cell." Soon the supersaturation maximum is reached and the droplet growth mode changes completely. Instead of transferring the vapor and heat between the droplet surface and the environment, the fields change to transfer the two properties between the droplet and the space inside the cell. Due to this shift of transportation mode, ρ and T keep changing at constant rates while maintaining their new steady-state profiles. Hence, the governing condition changes to $\nabla^2 \rho = \text{nonzero const}$ and $\nabla^2 T = \text{nonzero const}$ with the cell boundary restriction, from $\nabla^2 \rho = \nabla^2 T = 0$ of the conventional theories without boundary restriction. The nonzero constants thus occur due to ρ and T transportation in and out of the cell space.

This cell-boundary-controlled (competitive), steady-state kinetics of cloud droplet growth under constant generation of supersaturation in the cell space due to cloudy parcel lifting and the corresponding removal by the droplet growth has been explicitly treated

for the fields of ρ and T and closed analytic solutions have been obtained. In the solution, the fields are found to deform relative to those of the conventional theories and to change their levels at constant rates while sustaining their steady-state profiles.

The field deformation is caused by an r^2 term in addition to the normal r^{-1} term (hyperbolic function) of noncell boundary-controlled (noncompetitive) kinetics. This quadratic term in r makes the gradient vanish at the cell boundary, satisfying the requirement of no vapor or heat transfer across the boundary. The derived fields of vapor density and temperature as well as the mass growth rate all converge to the noncell boundary-controlled, steady-state kinetics when the droplet radius r becomes negligible compared with the cell radius R .

The term responsible for this restrictive kinetics (f_c) is a function only of the radius ratio r/R . Increase of f_c in the restrictive diffusion kinetics is found to be equivalent to an increase in the condensation or the thermal accommodation coefficient or both in the conventional diffusion kinetics. The f_c term contributes to an increase in the mass growth rate for both Maxwellian and diffusion-kinetic theories. Use of analytic solutions obtained for the conventional droplet growth theories with mass and energy balance requires f_c correction, as discussed in section 2, because the solutions are valid only for the fields of infinite expanse. Carsten's approach is essentially the same as this, dealing with the fields of infinite expanse, and needs an identical correction, but the correction is negligible under the typical cloud condition (see section 3a). Nevertheless, for unusual natural and some artificial processes, such as condensation in the supersonic nozzle gas (Fukuta and Paik 1976) and in adiabatically expanding air in the cloud chamber with high mixing ratio of condensable gas, consideration of cell boundary-controlled kinetics under continuing generation of supersaturation may become necessary.

It should also be pointed out that application of the adiabatic expansion method for studying accommodation coefficients requires an extra caution since a link exists between f_c and accommodation coefficients, as discussed previously. There is, therefore, a possibility of obtaining a deceiving result for the coefficients due to the f_c factor, if the latter is not considered properly.

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APPENDIX A

Correction for Vapor Flux under Temperature Gradient

The water vapor mass flux density across a unit area of air space is given by

$$F_m = -\frac{1}{2} l \nabla(nm\bar{v}), \quad (\text{A1})$$

where \mathbf{i} , \mathbf{j} , \mathbf{k} are the respective unit vectors. A factor $1/3$ is sometimes used erroneously (Fleagle and Businger 1963) instead of $1/2$, and it has to be corrected (Moelwyn-Hughes 1957). Since

$$\nabla = \mathbf{i} \frac{\partial}{\partial x} + \mathbf{j} \frac{\partial}{\partial y} + \mathbf{k} \frac{\partial}{\partial z}, \quad (\text{A2})$$

\mathbf{i} , \mathbf{j} , \mathbf{k} being the respective unit vectors. A factor $1/3$ is sometimes used erroneously (Fleagle and Businger 1963) instead of $1/2$, and it has to be corrected (Moelwyn-Hughes 1957). Since

$$\rho = nm, \quad (\text{A3})$$

and

$$\bar{v} = \left(\frac{8kT}{\pi m} \right)^{1/2}, \quad (\text{A4})$$

k being the Boltzmann constant, Eq. (A1) reduces to

$$F_m = -\frac{1}{2} l \bar{v} \left(\nabla \rho + \frac{1}{2} \rho \nabla n T \right). \quad (\text{A5})$$

Application of the ideal gas law for the water vapor

$$\rho = \frac{e}{R_v T} \quad (\text{A6})$$

in Eq. (A5) yields

$$F_m = -\frac{1}{2} l \bar{v} \left(\nabla e - \frac{1}{2} e \nabla \ln T \right). \quad (\text{A7})$$

The second term in the bracket on the right-hand side of Eq. (A7) is the correction ΔF_m for the vapor flux due to the temperature gradient. Then

$$\frac{\Delta F_m}{F_m} \approx -\frac{1}{2} e \nabla \ln T / \nabla e. \quad (\text{A8})$$

For the Maxwellian field at the droplet surface under a low supersaturation, the Clausius-Clapeyron equation holds:

$$\frac{de_s}{dT} = \frac{e_s L}{R_v T^2}. \quad (\text{A9})$$

Multiplying $2T/e_s$ to both sides of Eq. (A9) and using it in Eq. (A8), we have

$$\frac{\Delta F_m}{F_m} \approx -\frac{R_v T}{2L}. \quad (\text{A10})$$

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