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Broadening of cloud droplet size
distributions by condensation in turbulence

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Abstract

To consider the growth of cloud droplets by condensation in turbulence, the Fokker-Planck equation is derived for the droplet size distribution (droplet spectrum). This is an extension of the statistical theory proposed by Chandrakar and coauthors in 2016 for explaining the broadening of the droplet spectrum obtained from the ‘II-chamber’, a laboratory cloud chamber. In this Fokker-Planck equation, the diffusion term represents the broadening effect of the supersaturation fluctuation on the droplet spectrum. The aerosol (curvature and solute) effects are introduced into the Fokker-Planck equation as the zero flux boundary condition at $R^2 = 0$, where $R$ is the droplet radius, which is mathematically equivalent to the case of Brownian motion in the presence of a wall. The analytical expression for the droplet spectrum in the steady state is obtained and shown to be proportional to $R \exp(-cR^2)$, where $c$ is a constant. We conduct direct numerical simulations of cloud droplets in turbulence and show that the results agree closely with the theoretical predictions and, when the computational domain is large enough to be comparable to the II-chamber, agree with the results from the II-chamber as well. We also show that the diffusion coefficient in the Fokker-Planck equation should be expressed in terms of the Lagrangian autocorrelation time of the supersaturation fluctuation in turbulent flow.
Keywords cloud droplet size distribution, turbulence-cloud droplet interaction, Fokker-Planck equation, direct numerical simulation

1. Introduction

Clouds play a crucial role in Earth’s weather and climate system, yet our understanding of clouds remains limited. One of the challenges in improving understanding is the vast range of scales involved. If we focus on clouds without ice (so-called ‘warm clouds’, Lau and Wu 2003), the relevant processes range from the condensation-nucleation of aerosol particles at nanometer scales, to the condensation growth of droplets at scales of a few to several tens of micrometers, then to the collision-coalescence growth of hundreds of micrometers up to raindrops on scales of millimeters (Houze 2014). Furthermore, because flows inside clouds are inherently turbulent, they involve various sizes of eddies and associated fluctuations, ranging from the largest energy-containing scale of a few kilometers to the smallest scale of a few millimeters where the molecular viscosity is dominant. These turbulent eddies interact with each other, and interact with the droplets as well, making the modeling and understanding of cloud turbulence a highly complex problem (Bodenschatz et al. 2010).

The turbulence properties of clouds have been revealed by recent high-resolution in-situ measurements (Siebert et al. 2006, 2015; Bodenschatz
For example, Siebert and Shaw (2017) used helicopter-borne measurements and observed the fine-scale turbulence structures of cumulus clouds newly formed in the atmospheric boundary layer. From temperature and humidity measurements with resolutions as high as several tens of centimeters, they demonstrated that the supersaturation fluctuation has an amplitude on the order 1%.

Because of its potential importance for the growth of droplets in clouds, turbulence has been investigated as one of the key candidates for improving numerical weather models and cloud microphysical parameterizations (Vaillancourt and Yau 2000; Shaw 2003; Devenish et al. 2012; Grabowski and Wang 2013). Although clarifying the effect of turbulence in clouds is not an easy task as the turbulence itself is still not fully understood (Frisch 1995; Wyngaard 2010; Davidson et al. 2012), recent developments in high-performance supercomputers have made it possible to conduct direct numerical simulations (DNSs) of cloud turbulence, and numerous studies have been carried out since the pioneering work by Vaillancourt et al. (2001, 2002).

For condensation growth, turbulent mixing and entrainment in clouds excite strong spatial and temporal fluctuations in temperature and humidity. These turbulent fluctuations cause differences in the local supersaturation experienced by each droplet, which in turn cause differences in
the growth histories of droplets and a broader droplet size distribution.

This mechanism was examined since the early 1960s by a group of studies known as the “stochastic condensation theory” (mostly Russian, see Sedunov 1974; Clark and Hall 1979; Korolev and Mazin 2003), but the importance of the mechanism was later reinforced by Cooper (1989) and a sophisticated microphysical Lagrangian model by Lasher-Trapp et al. (2005).

DNSs on condensation growth of cloud droplets in a turbulent environment have been conducted and revealed various features, such as the essential role of large-scale turbulent motions, the fine-scale structures at the cloudy-clear air interface, and two-way coupling between the supersaturation fluctuation and cloud droplets (Celani et al. 2005, 2009; Lanotte et al. 2009; Sardina et al. 2015; Kumar et al. 2014, 2018).

For collision growth, vertically straight trajectories of gravitationally sedimenting droplets are perturbed by turbulent fluctuations in the air velocity field. Moreover, the acceleration of droplets due to the turbulent velocity field can be comparable to gravitational acceleration, causing complex droplet trajectories and phenomena such as the inertial clustering effect (Sundaram and Collins 1997) and the sling effect (Falkovich and Pumir 2007). This leads to the enhancement of the collision rate especially among droplets of similar sizes and to accelerated rain initiation (Franklin et al. 2005; Ayala et al. 2008b; Onishi et al. 2015; Chen et al. 2018a).
The knowledge accumulated from above studies has resulted in the development and implementation of new cloud microphysical parameterizations that account for the effect of turbulence on the condensation growth (Grabowski and Abade 2017; Sardina et al. 2018) or on the collision-coalescence growth of droplets (Franklin et al. 2007; Grabowski and Wang 2009; Ayala et al. 2008a; Seifert et al. 2010; Onishi and Seifert 2016). In addition, several studies have conducted DNSs including both the condensation and collision-coalescence processes in order to obtain a more comprehensive understanding of the effect of turbulence on the continuous growth of droplets (Chen et al. 2018b; Saito and Gotoh 2018; Kunishima and Onishi 2018).

On the other hand, an interesting laboratory experiment was recently conducted by Chandrakar et al. (2016) (hereinafter referred to as “C16”) which focused on the effect of turbulence on the condensation growth of cloud droplets. They used a laboratory chamber with a working volume of 3.14m$^3$, referred to as the “II-chamber” (Chang et al. 2016), and produced a steady state turbulent cloud in the chamber in the following way. Moist Rayleigh-Benard convection is excited in the chamber to produce turbulent fluctuations in the air velocity and supersaturation fields. Aerosol particles are injected into the system at a constant rate, removed from the system by sedimentation, and the steady state particle number density is determined.
through the balance between the source and sink. Aerosol particles initiate
the generation of cloud droplets in the moist air, and their sizes change in
the fluctuating supersaturation field. The droplet size distributions in sta-
tistically steady states were measured and demonstrated to be narrower for
experiments with greater number densities of cloud droplets. This tendency
is associated with the dispersion aerosol indirect effect (Chandrakar et al.
2018b). In C16, a statistical theory was also proposed to account for their
experimental results, and it was shown that this theory can explain the
statistical properties of the droplet size distribution fairly well for a certain
range of experimental parameters.

Although there are differences in the physical parameters and conditions
of atmospheric clouds and the laboratory clouds in C16, we believe that the
II-chamber experiment is highly useful for validating the results of numerical
simulations for the following reasons. First, in contrast to cloud observation
in the atmosphere, the II-chamber experiment provides detailed information
on the laboratory cloud which was obtained under closely controlled lab-
oratory conditions. This is useful for comparing between the laboratory
experiment and numerical simulations, and for the identifying important
factors for droplet growth in turbulent clouds. Second, the chamber’s vol-
ume of about $3m^3$ is achievable in DNSs. For example, resolving the smallest
turbulent motion requires a grid length of about 1-2 mm. With this grid
length, we need about 1000$^3$ grid points to fill a domain of 3m$^3$, which is feasible with recent high-performance computers. Thus, the inter-comparison of the laboratory experiment, DNSs, and statistical theory is possible, which is useful and important for a deeper understanding of cloud microphysical processes. It also should be noted that the domain size of 3 m$^3$ is close to the finest grid size used in the latest high-resolution large-eddy simulations (LESs) of clouds (Sato et al. 2018) ($\Delta x = \Delta y = 6.25$m, $\Delta z = 5$m). This suggests that useful knowledge on cloud microphysical processes can be provided from DNSs to LESs with little extrapolation.

With the above motivation, the main purpose of the present study is to conduct DNSs using our DNS model the “cloud microphysics simulator” (Gotoh et al. 2016), and compare the results with the statistical theory and the laboratory experiment by C16. First, we make several extensions to the statistical theory by C16. We obtain an analytical expression for the droplet size distribution at a steady state by deriving the Fokker-Planck equation for the droplet size distribution. Aerosol (curvature and solute) effects on droplet growth are introduced as the zero flux boundary condition, which is mathematically equivalent to the case of Brownian motion in the presence of a wall. Next, we compare this theory with the experimental results of a small-scale DNS (box length is $L_{\text{box}} = 12.8$cm). Finally, we conduct a large-scale DNS ($L_{\text{box}} = 102.4$cm) with a domain size comparable to the
size of the Π-chamber.

Readers should be aware that our target in this study is the Π-chamber experiment by C16 and the physical parameters and conditions are accordingly different from those in atmospheric clouds. Therefore the interpretation of the present results needs great care in the context of atmospheric clouds. We discuss these points in detail in Sec. 4.

The remainder of the present paper is organized as follows. In Sec. 2, we consider the statistical theory proposed in C16 and its extension. In Sec. 3, we conduct DNSs. A discussion and summary are provided in Sec. 4 and Sec. 5, respectively.

2. Statistical theory by Chandrakar et al. (2016) and its extension

Before introducing our extensions, we first briefly review the statistical theory proposed by C16. First, the size of the cloud droplet evolves in response to the ambient supersaturation as

\[
\frac{dR^2}{dt} = 2K_s S,
\]

where \( R \) is the droplet radius, \( S \) is the supersaturation at the droplet position, and \( K_s \) is assumed to be constant for simplicity. Second, the evolution of the supersaturation is assumed to be given by the following stochastic
differential equation:

\[ dS(t) = S(t + dt) - S(t) = \left[ \frac{S_{eq} - S}{\tau_t} - \frac{S}{\tau_c} \right] dt + \left( \frac{2\sigma_{S_0}^2 dt}{\tau_t} \right)^{1/2} \xi(t) \]  \tag{2}

where \( \sigma_{S_0} \) is the standard deviation of the supersaturation without cloud droplets, \( S_{eq} \) is the equilibrium value of the supersaturation, \( \xi(t) \) is a Gaussian white noise process with a zero mean:

\[ \langle \xi(t) \rangle = 0, \]  \tag{3}

\( \delta() \) is the Dirac delta, and angle brackets indicate the ensemble average.

In (2), the effects of the turbulent fluctuation of the supersaturation are represented in terms of \( \tau_t \) based on a Langevin model, where \( \tau_t \) is referred to as the turbulent mixing time [Here we simply refer to \( \tau_t \) as the turbulent mixing time and determine the appropriate value for \( \tau_t \) from the simulation results in Sec. 3]. On the other hand, the term with \( \tau_c \) in (2) represents the effect of cloud droplets on the supersaturation through condensation/evaporation. The parameter \( \tau_c \) is referred to as the phase relaxation time (Cooper 1989), and is inversely related to the number density \( n_d \) and mean radius of cloud droplets by

\[ \tau_c^{-1} \propto n_d \bar{R}, \]  \tag{4}

where the overbar denotes the volume average. See Appendix A for the precise form of \( \tau_c \).
Using (1) and (2), C16 derived estimates of the statistical properties of the supersaturation fluctuation and the droplet size distribution for a statistically steady state. Here, the number density \( n_d \) is assumed to be constant due to the balance between the injection and removal of cloud droplets, and cloud droplets are assumed to remain in the system with a residence time \( \tau_{\text{res}} \). The variance of the supersaturation is given by

\[
\sigma_S^2 = (S')^2 = \frac{\tau_s}{\tau_t} \sigma_{S_0}^2 \left( < \sigma_{S_0}^2 > \right),
\]

where \( S' = S - \overline{S} \) is the supersaturation fluctuation. The variance of the squared radius is given by

\[
\sigma_{R^2}^2 = \frac{8K_8^2 \sigma_{S_0}^2 \tau_s^2}{\tau_t} \tau_{\text{res}},
\]

where \( \tau_s \) is referred to as the system timescale and is given by the harmonic mean of \( \tau_t \) and \( \tau_c \) as

\[
\tau_s = \left( \frac{1}{\tau_t} + \frac{1}{\tau_c} \right)^{-1} = \frac{\tau_t \tau_c}{\tau_t + \tau_c}.
\]

See C16 for the detailed derivations of (5) and (6).

### 2.1 Physical interpretation and Fokker-Planck equation

The above statistical theory by C16 describes the interaction between cloud droplets and the supersaturation fluctuation in what is assumed to be the most straightforward way. First, the effect of cloud droplets on
the supersaturation fluctuation is described in (5). If the supersaturation is positive/negative, cloud droplets make the ambient air drier/moister by condensation/evaporation, respectively. The overall effect is that they reduce the amplitude of the supersaturation fluctuation and $\sigma_S < \sigma_{S_0}$. Second, the effect of the supersaturation fluctuation on cloud droplets is described in (6). As the amplitude of the supersaturation fluctuation ($\sigma_{S_0}$) increases, the droplet size distribution becomes broader and $\sigma_{R^2}$ increases.

We can also understand the above theory through an analogy with Brownian theory. From (1) and (2), we can regard $R^2$ and $2K_sS$ as the position and velocity of Brownian particles, respectively, and rewrite (6) as

$$\sigma_{R^2}^2 = 2D\tau_{\text{res}}.$$  

(8)

where $D$ is the diffusion coefficient for Brownian motion given by

$$D = \left[4K_s\sigma_S^2\right] \tau_1 = \left[4K_s^2\left(\sigma_{S_0}^2\frac{\tau_s}{\tau_1}\right)\right] \tau_1.$$  

(9)

Just like the spatial distribution of Brownian particles, the droplet size distribution diffuses due to the supersaturation fluctuation $\sigma_S$. For the time parameter $\tau_1$ in (9), the C16 result (6) is recovered if we substitute the system timescale $\tau_s$ for $\tau_1$ as

$$\tau_1 = \tau_s.$$  

(10)

Based on the analogy with Brownian motion, we can extend the above theory to consider the evolution of the size distribution. Since we can regard
the squared radius of cloud droplets \( (R^2) \) as the position of the Brownian particles with the diffusion coefficient \( D \) given by (9), the size distribution for \( R^2 \), or \( n(R^2, t) \), evolves according to the following Fokker-Planck equation:

\[
\frac{\partial n(R^2, t)}{\partial t} = D \frac{\partial^2 n(R^2, t)}{\partial (R^2)^2} - \frac{1}{\tau_{res}} n(R^2, t) + J_0 \delta(R^2 - R_0^2). \tag{11}
\]

The first term on the right side in (11) represents the broadening effect of the supersaturation fluctuation. For simplicity, the diffusion coefficient \( D \) is assumed to be constant. The term with \( \tau_{res} \) represents the sink, where cloud droplets are removed from the system with a residence time of \( \tau_{res} \). The term with \( J_0 \) represents the source, where cloud droplets are injected into the system at a constant rate \( J_0 \) and an injection radius \( R_0 \).

The boundary condition is essential to determine the distribution function. When the radius of a cloud droplet is small, the evolution equation (1) is no longer valid. We should consider the curvature and solute effects, which we hereinafter simply refer to as “aerosol effects”, and terms associated with the Köhler curve are introduced as explained later in (31). This is the case for the II-chamber, since aerosol particles are injected into the chamber with initial radii ranging from several tens to hundreds of nanometers.

As a simple way to include the aerosol effects in the theoretical framework, we adopt the idea by Siewert et al. (2017). To a first approximation,
the effect of the Köhler curve is to keep the droplet radius positive. More precisely, even when the supersaturation in the ambient air is negative, the droplet radius is kept at the corresponding equilibrium radius, which is small but positive definite. Siewert et al. (2017) suggested that this effect can be approximated with a reflecting wall boundary condition as in the case of Brownian motion, which is mathematically equivalent to the zero flux boundary condition at $R^2 = 0$ as

$$\frac{\partial n(R^2, t)}{\partial R^2} = 0, \quad \text{at} \quad R^2 = 0. \quad \text{(12)}$$

For $R^2 \to \infty$ it is natural to assume that

$$n(R^2, t) \to 0 \quad \text{for} \quad R^2 \to \infty, \quad \text{(13)}$$

and the initial condition is

$$n(R^2, 0) = n_0(R^2). \quad \text{(14)}$$

The analytical solution for the above model is given in Appendix E (Chandrasekhar 1943), but hereafter we examine only the steady state solution for two typical cases, case a) : $R_0$ is large such that $R_0^2 \gg R_s^2$, and case b) : $R_0$ is small such that $R_0^2 \sim R_s^2$, where $R_s$ is a characteristic radius at which the Köhler curve attains the maximum and the aerosol effects become appreciable.
a. Steady state size distribution when the injection radius is large

For the steady state ($\partial/\partial t = 0$) where all three fluxes (source, sink, and flux due to the diffusion of particles in $R^2$-space) balance each other, we can obtain an analytical solution of (11). In the present case, the probability density function (PDF) of $R^2$ for the solution is given as follows:

$$P(R^2) = \frac{1}{2\beta^2} \exp \left(-\beta^{-2}|R^2 - R_0^2|\right),$$

where $\beta$ has the dimension of length and is defined as

$$\beta = (D\tau_{\text{res}})^{1/4}. \quad (16)$$

From (15), the average of $R^2$ and the variance are given by

$$\overline{R^2} = \int_0^\infty R^2 P(R^2) dR^2 = R_0^2, \quad (17)$$
$$\sigma^2_{R^2} = \int_0^\infty (R^2 - \overline{R^2})^2 P(R^2) dR^2 = 2\beta^4 = 2D\tau_{\text{res}}, \quad (18)$$

respectively, where we additionally assumed that $R_0 \gg \beta$. Equation (18) agrees with (8).

b. Steady state size distribution when the injection radius is small

The solution of the Fokker-Planck equation in this case is found to be

$$P(R^2) = \frac{1}{2\beta^2} \exp \left(-\beta^{-2}|R^2 - R_0^2|\right) + \frac{1}{2\beta^2} \exp \left(-\beta^{-2}|R^2 + R_0^2|\right), \quad (R^2 > 0). \quad (19)$$
Taking the limit $R_0 \to 0$ for simplicity, the PDF of $R^2$ in the steady state is given by

$$P(R^2) = \frac{1}{\beta^2} \exp\left(-\beta^{-2}R^2\right).$$  \hspace{1cm} (20)

The average of $R^2$ is given by

$$\overline{R^2} = \int_0^\infty R^2 P(R^2) dR^2 = \beta^2,$$  \hspace{1cm} (21)

which is different from $R_0^2(\approx 0)$ for the source. This difference is due to the presence of the reflecting wall and differs from the result obtained without aerosol effects (17). The variance of $R^2$ is given by

$$\sigma_{R^2}^2 = \int_0^\infty (R^2 - \overline{R^2})^2 P(R^2) dR^2 = \beta^4 = D\tau_{res},$$  \hspace{1cm} (22)

which does not have the factor 2, in contrast to (18). The statistical properties of the distribution (20) are summarized in Appendix D.

Several studies on cloud turbulence used the Fokker-Planck equation to derive the droplet size distribution for the steady state, but with slightly different settings (McGraw and Liu 2006; Siewert et al. 2017). For example, McGraw and Liu (2006) obtained the same functional form as (20), but their Fokker-Planck equation did not include the source and sink terms and required a negative mean supersaturation to attain a steady state size distribution. We consider the effect of the mean supersaturation later in (53).
3. Numerical experiments

Here, we conduct DNSs of cloud droplets in turbulence and compare the results with the theory in the previous section. The numerical domain under consideration is a cubic box with sides of length $L_{\text{box}}$ which is assumed to be in the chamber interior far from the wall. We also assume that $L_{\text{box}}$ is sufficiently small. Accordingly, the turbulence is regarded as being homogeneous isotropic and periodic boundary conditions in three directions are applied.

3.1 Governing equations

a. Fluid

The velocity field of air is governed by the incompressible Navier-Stokes equations:

$$\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} = -\frac{1}{\rho_a} \nabla p + \nu_a \nabla^2 \mathbf{u} + \mathbf{f},$$

$$\nabla \cdot \mathbf{u} = 0,$$

where $p$ is the pressure fluctuation, $\rho_a$ and $\nu_a$ are the density and kinematic viscosity of dry air, respectively, and $\mathbf{f}$ represents the external force for the velocity field. The temperature $T$ and the water vapor mixing ratio $Q$ are expressed as the sum of the mean and fluctuation as

$$T = T + \theta,$$
respectively, where the overbar denotes the volume average over the box.

The evolution equations for $\theta$ and $q$ are given by

\[
\frac{\partial \theta}{\partial t} + \mathbf{u} \cdot \nabla \theta = \frac{L_v}{c_p} (C_d - \overline{C_d}) + \kappa_T \nabla^2 \theta + f_\theta, \tag{26}
\]

\[
\frac{\partial q}{\partial t} + \mathbf{u} \cdot \nabla q = - (C_d - \overline{C_d}) + \kappa_v \nabla^2 q + f_q, \tag{27}
\]

where $\kappa_T$ and $\kappa_v$ are the thermal diffusivity and diffusivity of water vapor, respectively, $L_v$ is the latent heat of vaporization, $c_p$ is the specific heat of air at constant pressure, and $C_d$ is the condensation rate defined later in (34). $f_\theta$ and $f_q$ represent the external forces on $\theta$ and $q$, respectively.

We next consider evolution equations for the mean values, $\overline{T}$ and $\overline{Q}$.

In the II-chamber, the moist Rayleigh-Benard convection and turbulence keep the chamber interior in a statistically steady state, and $\overline{T}$ and $\overline{Q}$ are accordingly kept at statistically steady values. As a simple model for this effect, we use nudging terms as follows:

\[
\frac{\partial \overline{T}}{\partial t} = \frac{L_v}{c_p} \overline{C_d} - \frac{\overline{T} - T_0}{\tau_0}, \tag{28}
\]

\[
\frac{\partial \overline{Q}}{\partial t} = - \overline{C_d} - \frac{\overline{Q} - Q_0}{\tau_0}, \tag{29}
\]

where terms with $\tau_0$ mean that the average values are nudged to $T_0$ and $Q_0$ with a timescale $\tau_0$. 

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b. Particles

Cloud droplets are assumed to be smaller than the grid size and are treated as point particles. The evolution equations for the \( j \)-th cloud droplet are given by

\[
\frac{d\mathbf{X}_j(t)}{dt} = \mathbf{u}(\mathbf{X}_j),
\]

\[
\frac{dR_j(t)^2}{dt} = 2K_s \left[ S(\mathbf{X}_j) - \frac{A}{R_j(t)} + \frac{B}{R_j(t)^3} \right],
\]

where \( \mathbf{X}_j(t) \) and \( R_j(t) \) are the position and radius, respectively, of the \( j \)-th cloud droplet, and \( \mathbf{u}(\mathbf{X}_j) \) and \( S(\mathbf{X}_j) \) are the fluid velocity and supersaturation, respectively, at the droplet position. The parameter \( K_s \) is a diffusion coefficient for droplet growth that depends on the pressure and temperature (Pruppacher and Klett 1997). \( K_s \) is set to a constant because its fluctuations are sufficiently small in the present DNS. Terms with \( A \) and \( B \) in (31) represent the curvature and solute effects, respectively, definitions of which are given in Appendix B.

The supersaturation at the droplet position, \( S(\mathbf{X}_j(t), t) \) in (31), is defined by

\[
S(\mathbf{X}_j(t), t) = \frac{Q(\mathbf{X}_j(t), t)}{Q_{vs}(\mathbf{X}_j(t), t)} - 1.
\]

The saturation mixing ratio for the water vapor \( Q_{vs} \) at the droplet position is determined by Tetens’ formula:

\[
Q_{vs}(\mathbf{X}_j(t), t) = \varepsilon_0 \frac{610.78}{P_0} \exp \left[ 17.629 \frac{T(\mathbf{X}_j(t), t) - 273.16}{T(\mathbf{X}_j(t), t) - 35.86} \right],
\]
where $\varepsilon_0 = R_d/R_v \approx 0.62$, $R_d$ and $R_v$ are the gas constants for dry air and water vapor, respectively, and $P_0$ is the environmental pressure.

Note that there are several properties of the droplet dynamics that are neglected in the present DNS, such as gravity, droplet inertia, and collision-coalescence. The reason for these simplifications is to make the model in our DNS simpler and closer to that in the statistical theory in Sec. 2 to facilitate a comparison between the two. Possible effects of these simplifications are discussed in Sec. 5.

The droplet size changes according to (31). The condensation rate $C_d(x, t)$ in (26) and (27) is then calculated by

$$C_d(x, t) = \frac{1}{\rho_v(\Delta x)^3} \sum_{j=1}^{N_\Delta} \frac{dm_j(t)}{dt};$$  

where $N_\Delta$ is the number of cloud droplets within the grid cell $(\Delta x)^3$ and $m_j$ is the mass of the $j$-th cloud droplet defined as

$$m_j(t) = \frac{4\pi}{3} \rho_w R_j(t)^3;$$

where $\rho_w$ is the liquid water density.

### 3.2 Experimental setup

#### a. Nudging and thermodynamic parameters

We assumed that the environmental pressure $P_0$ was 1013.25 hPa (= 1 atm) and that the mean temperature in the domain was kept close to
Accordingly, we set the nudging parameter as $T_0 = 290$ K in (28).

Other parameters are summarized in Table 1. With these parameters, the diffusion coefficient $K_s$ in (31) was estimated as $K_s = 1.097 \times 10^{-6}$ cm$^2$s$^{-1}$ for sufficiently dilute drop (Pruppacher and Klett 1997).

For parameter $Q_0$ in (29), we set $Q_0$ so that the corresponding supersaturation $S_0(T_0, Q_0)$ was 0 % based on (32) and (33). The value of $Q_0$ used in the present DNS is shown in Table 1. The reason for this choice of $S_0$ is that the main focus of the present study is on the effect of the supersaturation fluctuation. We discuss the effect of this choice of $S_0$ in Sec. 3.4.

For the nudging timescale $\tau_0$, we assume that $\overline{T}$ and $\overline{Q}$ are mainly determined by the largest eddies. We set $\tau_0 = 0.9$ s and $\tau_0 = 2.0$ s for the small- and large-scale DNS, respectively. These values are roughly consistent with the large-eddy turnover time (see Table 2).

**b. Solute**

The solute dissolved in each cloud droplet is NaCl (sodium chloride). For simplicity, the amount of solute is fixed to $m_s = 1.13 \times 10^{-15}$ g, which corresponds to a sphere with an equivalent radius of 50 nm. The parameters for the aerosol effects are summarized in Appendix B. With these parameters, from the definitions in (59) and (60) we have $A = 1.089 \times 10^{-7}$ cm and $B = 1.665 \times 10^{-16}$ cm$^3$, respectively. Figure 1a shows the Köhler curve.
\[ f(R) = (A/R) - (B/R^2). \] The peak of the curve is at \( R_c = (3B/A)^{1/2} = 0.677 \, \mu m \) and \( S_c = 2A/(3R_c) = 0.107\%, \) where \( R_c \) is the critical radius and \( S_c \) is the critical supersaturation. The curve crosses the horizontal axis at \( R_{eq} = (B/A)^{1/2} = 0.391 \, \mu m \), which is the equilibrium radius for the supersaturation \( S = 0. \)

c. Injection and removal of particles

Cloud droplets are injected into the system at a constant rate \( J_0 \) with uniform random initial positions, and they are removed from the system randomly following a residence time \( \tau_{res}. \) The evolution equation for the number density of cloud droplets \( n_d \) is written as

\[
\frac{\partial n_d}{\partial t} = J_0 - \frac{1}{\tau_{res}} n_d. \tag{36}
\]

In the steady state, the two terms on the right side balance and we obtain

\[ n_d = J_0 \tau_{res}. \tag{37} \]

We used a fixed value of \( \tau_{res} = 580 \, s (=9.7 \, \text{min.}) \) based on the results from the \( \Pi \)-chamber in C16. We changed the injection rate \( J_0 \) and conducted experiments with different number densities. The values of \( J_0 \) and \( n_d \) are summarized in Table 3.
d. Random force

The force for the velocity field $f$ in (23) is a solenoidal, Gaussian random variable with a zero mean, representing white noise (Gotoh et al. 2002), and is applied to the low wavenumber band as

$$\langle f(k, t) \rangle = 0,$$
$$\langle f(k, t) f(-k, s) \rangle = \left( \frac{L_{\text{box}}}{2\pi} \right)^3 \left( \delta_{ij} - \frac{k_i k_j}{k^2} \right) \frac{F(k)}{4\pi k^2} \delta(t - s),$$

where $P_{ij}(k) = \delta_{ij} - \frac{k_i k_j}{k^2}$ and $F(k)$ is the spectrum of the random force defined as

$$F(k) = \begin{cases} c_f & (1 \leq k L_{\text{box}} \leq 4) \\ 0 & \text{(otherwise)} \end{cases}$$

The value of $c_f$ is set to 30 cm$^2$s$^{-3}$ for the small-scale DNS (Run1–Run10), and to 20 cm$^2$s$^{-3}$ for the large-scale DNS (Run11–Run14).

For forces on scalar fields $f_\theta$ and $f_q$ in (26) and (27), respectively, we used only $f_q$ and set $f_\theta = 0$. [As long as we consider the supersaturation, it does not matter whether fluctuations arise from the temperature or water vapor mixing ratio. We also conducted simulations with $f_q = 0$ and non-zero $f_\theta$ to excite the supersaturation fluctuation, and confirmed that the same result was obtained when the statistics of the supersaturation fluctuation were unchanged]. The force $f_q$ is a Gaussian random variable with zero mean, representing white noise, and applied to the low wavenumber band. The amplitude of $f_q$ is tuned so that the standard deviation of the
supersaturation fluctuation without cloud droplets is \( \sigma_{S_0} = 0.014 \) at a statistically steady state. This value is obtained from the measurements with the II-chamber in C16.

e. DNS

We first conduct small-scale DNS (Run1–Run10). The numerical domain is a triply periodic cubic box with sides of length \( L_{\text{box}} = 12.8 \) cm. We numerically integrate the evolution equations (23) (26)–(31) using the second-order Runge-Kutta scheme with time increment \( \Delta t = 1.0 \times 10^{-3} \) s. For the spatial discretization of the flow field, we use the pseudo spectral method with \( N = 128 \) grid points in each dimension and a grid length \( \Delta x = L_{\text{box}}/N = 1 \) mm. The velocity and scalar fields for the droplet position are linearly interpolated, and the same weighting is used to calculate the condensation rate in (34), i.e., the particle in the cell method (PIC).

We conduct two kinds of experiments: those with and without aerosol effects. For experiments without aerosol effects (Run1–Run5 in Table 3), we integrate (31) without the second and third terms on the right side. Cloud droplets are injected into the system with an initial radius \( R_0 = 20 \) \( \mu \)m, which is sufficiently large that they are removed from the system before their radius becomes smaller than zero. The purpose of these idealized experiments is to check the consistency between the DNS result and the sta-
tistical theory (15)–(17) as a first step. On the other hand, for experiments
with aerosol effects (Run6–Run10 in Table 3), we integrate all of equation
(31). Injected cloud droplets are initially unactivated and have a radius
\( R_0 = 0.39 \, \mu m \), which is the equilibrium radius for zero supersaturation as
shown in Fig. 1. The injection of unactivated cloud droplets is a model for
the injection of aerosol particles in the II-chamber experiment. When the
droplet radius is smaller than 200 nm, we integrate (31) with an implicit
scheme and use a Newton-Raphson type iterative scheme to determine the
radius in the next step (Shima et al. 2009).

Cloud droplets are initially distributed randomly in the domain with
radius \( R_0 \) and number density given by (37). Before each run, equations
(23), (26)–(31) are integrated for a few large-eddy turnover times without
condensation, injection or removal of cloud droplets. The period of numer-
ical integration is 4000 s (4 million steps) for the small-scale DNS to obtain
a statistically steady state. The turbulence parameters in the statistically
steady state are summarized in Table 2. Definitions of the turbulence pa-
rameters are given in Appendix C.

We also conduct large-scale DNS (Run11–Run14) to examine the effect
of the domain size. The details are provided in Sec. 3.4.

We use the cloud microphysics simulator, which is a DNS model for cloud
turbulence developed in previous studies (Gotoh et al. 2016; Saito and Go-
toh 2018; Saito et al. 2018). The supercomputers used in the present study are mostly the K-computer at the Research Organization for Information Science and Technology (RIST) in Kobe and the Fujitsu FX100 installed at Nagoya University. Parallelization of the computer program is described in Saito and Gotoh (2018).

3.3 Results of small-scale DNS

a. Supersaturation fluctuation

We first investigate the effect of cloud droplets on the supersaturation fluctuation. Figure 2a shows PDFs for the supersaturation fluctuation in statistically steady states for Run1–Run10, and Table 4 summarizes the standard deviation of the supersaturation ($\sigma_S$) and $\tau_c$ for each experiment. From (5), the theory predicts that $\sigma_S$ decreases with decreasing $\tau_c$. This tendency is confirmed by comparing Fig. 2a and Table 4.

Figure 2b is the same as Fig. 2a except that each distribution is normalized by its variance. The PDFs in Fig. 2b collapse onto a single curve (labeled “Theory”), which is a Gaussian distribution with a zero mean and unit variance, confirming that the supersaturation fluctuation is well approximated by the Gaussian distribution. This is consistent with the observations in the II-chamber by C16 and also with that in atmospheric clouds (Siebert and Shaw 2017).
We next consider the turbulent mixing time $\tau_l$. As explained in (2), the statistical theory developed by C16 uses a single timescale $\tau_l$ for turbulent mixing. $\tau_l$ is assumed to be the Lagrangian autocorrelation time for the turbulent velocity, which is effectively the same as the turbulence large-eddy turnover time (Desai et al. 2018; Pope 2000). This relies on the assumption that the mixing of scalar field is dominated by the large scale velocity fluctuations.

In order to confirm the validity of the above assumption, we conducted the following investigation. The turbulent velocity field mixes scalar fields through the advection terms in the fluctuation equations for temperature (26) and the water vapor mixing ratio (27). Since the equations for the velocity field (23) are not coupled with the scalar fields in the present DNS, the turbulence statistics for the velocity field are independent of the scalar fields. Then, if we can choose a single timescale for $\tau_l$, its value should be the same for Run1–Run10 because the turbulence statistics in the velocity field are the same for each of these runs. Substituting the definition $\tau_s^{-1} = \tau_t^{-1} + \tau_c^{-1}$ into (5) and solving for $\tau_t$, we have

$$\tau_t = \left( \frac{\sigma_{S_0}^2 - \sigma_S^2}{\sigma_S^2} \right) \tau_c,$$  \hspace{1cm} (41)

so that all of the parameters on the right side can be obtained from experimental results. By substituting $\sigma_{S_0} = 0.014$ and the results for Run1–Run10 into the right side of (41), we estimate the value of $\tau_t$ for each run.
Figure 3 shows the estimated $\tau_\ell$ as a function of the phase relaxation time $\tau_c$ for Run1–Run10. As shown in the figure, the estimates for $\tau_\ell$ for these runs, with or without the aerosol effects, are grouped closely around a single horizontal line. In view of the fact that $\tau_c$ varies by almost two orders of magnitude (from 0.14 s to 9.0 s) among Run1–Run10, this agreement is remarkable, strongly supporting the validity of using a single value for $\tau_\ell$ (By close observation of Fig. 3, we find that for Run1–Run5 the estimated $\tau_\ell$ value becomes larger for smaller $\tau_c$, but this trend is weak and we can regard it as a constant to a first approximation).

From the estimation of $\tau_\ell$ in Fig. 3, the average and standard deviation are given by $\tau_\ell = 0.846 \pm 0.021$ s, respectively, hence we have $\tau_\ell \approx 0.85$ s which is indicated by the horizontal line in Fig. 3. Since the large-eddy turnover time $T_E$ is 0.58 s (Table 2), the estimated $\tau_\ell(=0.85$ s) is greater than $T_E$ by about 50%. However, the estimation at least confirms that the appropriate choice of $\tau_\ell$ is closer to the timescales for the largest velocity fluctuations. This is consistent with the findings of previous DNS studies of cloud turbulence, where it was shown that the turbulence condensation dynamics are mainly determined by the large flow scales (Sardina et al. 2015; Götzfried et al. 2017). In the following analysis, we use $\tau_\ell = 0.85$ s. Using $\tau_\ell = 0.85$ s to calculate the right side of (5), we obtain the theoretical values for $\sigma_S$ shown in the second column in Table 4.
b. Droplet size distribution: cases without aerosol effects

We next consider the effect of the supersaturation fluctuation on the droplet size distribution. We begin with cases without aerosol effects, namely, Run1–Run5.

From (15), the theory predicts that the PDF of the squared radius \( R^2 \) is a maximum at the size of the injected particles \( R_0^2 \) and has exponentially decaying tails in a statistically steady state. This is confirmed by Fig. 4a, which shows PDFs for \( R^2 \) in statistically steady states for Run1–Run5 with semi-log scaling. Each distribution has its maximum at \( R_0^2 = 400 \) μm² and linearly decreasing tails in the semi-log plot. The width of the PDF becomes smaller with increasing run number, or for runs with higher number densities of cloud droplets and smaller supersaturation fluctuations (Tables 3 and 4). Figure 4b is the same as Fig. 4a but the PDFs are normalized by

\[
\hat{P}(\hat{R}^2) = \beta_1^2 P(\hat{R}^2), \quad \hat{R}^2 = \beta_1^2 |R^2 - R_0^2|,
\]

where \( \beta_1^2 \) is estimated from the DNS results as

\[
\beta_1^2 = \frac{\sigma_{R^2}}{\sqrt{2}}
\]

based on (18). After normalization, all distributions in Fig. 4b collapse onto the theoretical curve labeled “Theory”: \( \frac{1}{2} \exp(-|\hat{R}^2|) \) from (15).

The collapse of the DNS results onto the theoretical curve can also be obtained from the time evolution. Figure 5a shows the time evolution of
the variance of the squared radius ($\sigma_{R^2}^2$) for Run1–Run5. As predicted from
the Langevin theory, and also as shown in previous DNS studies (Sardina
et al. 2015; Siewert et al. 2017), $\sigma_{R^2}^2$ grows linearly with time. With the
effect of the sink in (11), the evolution of $\sigma_{R^2}^2$ is (see appendix E)

$$
\sigma_{R^2}^2 = 2D\tau_{\text{res}}(1 - e^{-t/\tau_{\text{res}}}) = 2\beta^4(1 - e^{-t/\tau_{\text{res}}}).
$$

(44)

Figure 5b is the same as Fig. 5a but the results are normalized by $\beta_1^4$ in
(43). All results collapse onto the theoretical curve: $2(1 - e^{-t/\tau_{\text{res}}})$.

We next consider the time parameter included in the diffusion coefficient.
In the statistical theory, the diffusion coefficient $D$ defined in (9) plays the
primary role in the broadening effect of the supersaturation fluctuation on
the droplet size distribution. The coefficient $D$ includes the time parameter
$\tau_1$. As explained in (8)–(10), the C16 result is recovered if we substitute
the system timescale $\tau_s$ for $\tau_1$. However, from the analogy with Brownian
motion, we can infer that $\tau_1$ should be the timescale of the supersaturation
fluctuation felt by each cloud droplet, or the Lagrangian autocorrelation
time for the supersaturation fluctuation.

In order to check the importance of the Lagrangian autocorrelation time,
we conducted an additional experiment. This experiment is the same as
Run1 except that each cloud droplet is fixed at the initial position after
it is introduced into the numerical domain. As defined in (7), the system
timescale $\tau_s$ is calculated from $\tau_l$ and $\tau_c$. The turbulent mixing time $\tau_t$ is
independent of the droplet motion and is solely determined by the turbulent velocity statistics in the present DNS as shown in Fig. 3. The phase relaxation time $\tau_c$ in (4) is calculated from the number density and mean radius of the cloud droplets, and is not affected by the droplet motion. Thus, $\tau_s$ is also independent of the droplet motion. If we rely on $\tau_1 = \tau_s$ in (9), the droplet size distribution should be the same for Run1 and in the additional experiment in a statistically steady state.

Results for Run1 and the additional experiment are compared in Fig. 6. First, PDFs for the supersaturation fluctuation for the two experiments in statistically steady states are compared in Fig. 6a, which shows no significant differences between the two. This result is consistent with the theory (5), because parameters $\tau_s$ and $\tau_l$ are unchanged. On the other hand, PDFs of $R^2$ for the two experiments in statistically steady states are compared in Fig. 6b, which shows significantly different results between the two experiments, in particular, the PDF for the additional experiment is narrower than that for Run1.

It is due to the change of the fluctuation timescale felt by cloud droplets that we obtained the narrower size distribution in the additional experiment. When the positions of cloud droplets are fixed, the fluctuation timescale experienced by cloud droplets is Eulerian. Since the scalar is basically Lagrangian invariant for zero diffusivity, the fluctuation timescale for scalars
is usually longer for the Lagrangian timescale than for the Eulerian one. Hence the smaller $\tau_1$ in (9) for the additional experiment and the narrower size distribution. From the above argument, we conclude that the time parameter $\tau_1$ in (9) should be the Lagrangian autocorrelation time for the supersaturation fluctuation, denoted hereinafter as $\tau_{\text{cor}}$:

\[
\tau_1 = \tau_{\text{cor}}. \tag{45}
\]

Table 5 compares $\sigma_{R^2}$ for Run1–Run5 with the theory (T0) calculated from (18), where $\tau_{\text{cor}}$ is used for the timescale $\tau_1$. T0 shows almost perfect agreement with the DNS results, which supports the validity of the statistical theory.

Even if we use the system timescale $\tau_s$ for $\tau_1$ in (18), the theory provides a good estimate for $\sigma_{R^2}$ in the present DNS, since $\tau_s$ is fairly close to $\tau_{\text{cor}}$ as shown in Table 5. However, it should be noted that the Lagrangian autocorrelation time $\tau_{\text{cor}}$ is greatly affected by the droplet motion in fluid. For example, $\tau_{\text{cor}}$ may change if we introduce the droplet inertia and gravity, which are known to cause substantial changes in the Lagrangian motion of relatively large cloud droplets. We discuss this point in Sec. 5.

c. Droplet size distribution: cases with aerosol effects

We next consider the results of Run6–Run10 which include aerosol effects. Figure 7 shows the time evolution of the radius of one droplet over 80
s along with the supersaturation experienced by that droplet in Run1. Note that these results are obtained after the droplet size distribution attains a statistically steady state (after 4000 s), and “t’ = 0” in the horizontal axis corresponds to t = 4000 s. The supersaturation is relatively negative from t’ = 0s to 40s, and the droplet radius is smaller than 3μm during this period. On the other hand, the supersaturation is relatively positive from t’ = 40s to 80s, and the droplet radius keeps growing, until exceeding 6μm. The wall effect originating from the aerosol effects can be clearly seen in Fig. 7 from t’ = 0s to 40 s. Around t’ = 20 s, for example, the droplet radius does not become smaller than zero but remains close to the equilibrium radius even when the supersaturation is negative.

We next examine the PDF of $R^2$ in a statistically steady state. From (20), the theory predicts that the PDF has its maximum around $R^2 = 0$ and an exponentially decaying right tail. This is confirmed by Fig. 8a, which shows the semi-log plot of the PDFs of $R^2$ for Run6–Run10 in statistically steady states. The PDFs have linearly decreasing right tails on a semi-log scale, and their widths decrease with increasing run number, or for runs with greater number densities of cloud droplets. Figure 8b is the same as Fig. 8a but each distribution is normalized by

$$\hat{P}(\hat{R}^2) = \beta_2^2 P(\hat{R}^2), \quad \hat{R}^2 = \beta_2^{-2} R^2,$$  \hspace{1cm} (46)
where $\beta^2_2$ is estimated from the experimental results as

$$\beta^2_2 = \sigma^2 R$$

(47)

based on (22). The curve labeled “Theory” in Fig. 8b indicates the theoretical curve $\exp(-\hat{R}^2)$, onto which all distributions collapse after normalization.

In cloud physics, PDFs of the droplet radius $R$ (or the diameter $d$) are often used to investigate cloud properties. Figure 9a shows PDFs of the droplet diameters for Run6–Run10 in statistically steady states. From the relationship $\int P(R^2)dR^2 = \int \mathcal{P}(R)dR$, where $P(R^2)$ and $\mathcal{P}(R)$ are PDFs for $R^2$ and $R$, respectively, $P(R^2)$ in (20) can be converted to $\mathcal{P}(R)$ as

$$\mathcal{P}(R) = 2\beta^{-2} R \exp \left( -\beta^{-2} R^2 \right),$$

(48)

which is the Rayleigh distribution. For the droplet diameter ($d = 2R$), the PDF is

$$P(d) = (2\beta^2)^{-1} d \exp \left[ -\left( 4\beta^2 \right)^{-1} d^2 \right].$$

(49)

Using this form, we normalize $P(d)$ as follows:

$$\hat{P}(\hat{d}) = \beta_2 P(\hat{d}), \quad \hat{d} = \beta^{-1}_2 d.$$  

(50)

Figure 9b shows the results for this normalization along with the theoretical curve: $(\hat{d}/2) \exp(-\hat{d}^2/4)$. Although the normalized distributions collapse
well onto the theoretical curve for \( \hat{d} > 0.5 \), they deviate significantly from
the theory around \( \hat{d} = 0 \), and have large peaks. These peaks correspond
to unactivated cloud droplets with a radius smaller than the critical radius.
Deviations are mainly due to differences in the settings used in the theory
and the DNS. In the theory, the radius of injected cloud droplets is assumed
to be \( R_0 = 0 \) for simplicity, whereas \( R_0 = 0.39 \, \mu \text{m} \) in the present DNS.

Because the mass of the solute dissolved in each cloud droplet is fixed
in the present DNS, the size distributions in Fig. 9 suggest that some of
these cloud droplets are activated and larger than the critical radius whereas
others are not, depending on the local supersaturation experienced by these
cloud droplets in the turbulent environment. This is qualitatively consistent
with the results of the \( \Pi \)-chamber experiment (see Fig. 3 in Chandrakar
et al. 2017).

Table 6 compares \( \sigma_{R^2} \) in statistically steady states for Run6–Run10 with
the theory (22), which is indicated by “T0”. T0 significantly deviates from
the DNS results, with deviations greater for experiments with smaller \( \sigma_{R^2} \).
For Run 10, T0 is almost twice as large as the DNS result. Such significant
deviations are contrary to the almost perfect agreement obtained for Run1–
Run5 in Table 5.

There are two reasons for the above deviations. The first is that the
theory (22) does not include the effect of the mean supersaturation. In the
present DNS, cloud droplets are injected into the system with an initial radius \( R_0 = 0.39 \text{ \mu m} \), attain the size distribution shown in Figs. 8a and 9a, and are removed from the system over a timescale \( \tau_{\text{res}} \). Thus, cloud droplets are growing in size on average and the mean condensation rate is positive \((\overline{C_d} > 0)\). From (28) and (29) for the steady state, we have

\[
\overline{T} = T_0 + \frac{L_v}{c_p} \tau_0 \overline{C_d}, \quad (51) \\
\overline{Q} = Q_0 - \tau_0 \overline{C_d}. \quad (52)
\]

When \( \overline{C_d} > 0 \), \( \overline{T} > T_0 \) and \( \overline{Q} < Q_0 \), leading to the mean supersaturation \( \overline{S} \) which is smaller than \( S_0 \) \((S_0 = 0\% \text{ for the present DNS})\). The mean supersaturations for Run6–Run10 are shown in the sixth column of Table 6. When the effect of the mean supersaturation \( \overline{S} \) is taken into account, the Fokker-Planck equation (11) has the following additional drift term:

\[
\frac{\partial n(R^2, t)}{\partial t} + 2K_s \overline{S} \frac{\partial n(R^2, t)}{\partial R^2} = D \frac{\partial^2 n(R^2, t)}{\partial (R^2)^2} - \frac{1}{\tau_{\text{res}}} n(R^2, t) + J_0 \delta(R^2 - R_0^2). \quad (53)
\]

Assuming \( R_0 = 0 \) and under the reflecting wall boundary condition (12), we obtain the same steady state solution as (20), except that \( \beta \) is replaced by \( \beta_- \), defined as follows:

\[
\beta_- = \frac{\beta}{(\sqrt{1 + \alpha^2} - \alpha)^{1/2}}, \quad (54)
\]
where $\beta^2$ is the root of a quadratic equation and

$$\alpha = \left(2K_s\bar{S}\right)\sqrt{\tau_{res}/(4D)}.$$  \hfill (55)

Using $\beta_-$, the variance of the squared radius is

$$\sigma^2_{R^2} = \beta^4 \left[\sqrt{1 + \alpha^2} - \alpha\right]^2.$$  \hfill (56)

Since $\alpha < 0$ for $\bar{S} < 0$, the effect of the negative mean supersaturation is to make $\sigma^2_{R^2}$ smaller. The theoretical values calculated from (56) are shown in the fourth column (T1) in Table 6. Although T1 is closer to the DNS results than T0, there are still significant differences between the theory (T1) and the DNS results.

The second reason for the above deviation is the over-simplification of the aerosol effects in the statistical theory. As explained in (12), one effect of the Köhler equation with solute is to make the droplet radius greater than zero. In addition, another effect is to make the effective supersaturation for droplet growth smaller than $S$. Figure 1a is reproduced in Fig. 1b on the semi-log scale, and shows that the Köhler curve for the present study is positive and typically has a magnitude of $10^{-2}\%$ for $R$ smaller than 15$\mu$m.

From (31), the positive value of the Köhler curve effectively acts as negative supersaturation on the cloud droplets. In order to include this effect in a simple way, we approximate the Köhler curve with a single value of $10^{-2}\%$, and replace $\bar{S}$ in (53) with $\left(\bar{S} - S_K\right)$, where $S_K = 10^{-4}$. The choice of
\( S_K = 10^{-4} \) is based on Fig. 9a, which shows that most of the cloud droplets in the present DNS are smaller than \( R = 15 \mu m \) (\( d = 30 \mu m \)). Theoretical values with the above correction are shown in the fifth column of Table 6 (T2). Although this approximation is crude, with it we obtain fairly close agreement between the theory (T2) and the DNS results.

### 3.4 Results of large-scale DNS

The results of the small-scale DNS described above show good agreement with the theory. However, the droplet size distributions shown in Fig. 9a are significantly narrower than the results of the II-chamber experiment. For example, for Run 6 in the present study, the droplets have a number density of 80 cm\(^{-3}\) (Table 3), which is close to the cloud droplet number density for the experiment “\( \dot{n}_a = 2/cm^3/min \)” by C16 as shown in their Table 1. Comparing our size distribution in Fig. 9a with that by C16 in their Fig. 2, the latter is much broader, reaching its tail well beyond a diameter of 30 \( \mu m \).

Based on the theory, (20) and (54), we can infer two possible reasons for the above discrepancy: the mean supersaturation (\( \overline{S} \)) and the Lagrangian autocorrelation time for the supersaturation fluctuation (\( \tau_{cor} \)). First, if \( \overline{S} \) is greater than that in the present DNS, the denominator in (54) becomes smaller, resulting in a larger \( \beta_- \) and a broader size distribution. Second, if
\( \tau_{\text{cor}} \) in (45) is greater than that in the present DNS, the diffusion coefficient \( D \) in (9) becomes larger and \( \alpha \) in (54) becomes smaller, both resulting in a larger \( \beta_- \) and a broader size distribution.

However, it is not easy to determine the relative importance of \( \overline{S} \) and \( \tau_{\text{cor}} \). For the mean supersaturation \( \overline{S} \), the measurements from the \( \Pi \)-chamber by Niedermeier et al. (2018) demonstrate that \( \overline{S} \) decreases from \( \sim 2\% \) to almost \( 0\% \) after the injection of aerosol particles, suggesting that it is difficult to keep \( \overline{S} \) positive in the chamber interior. However, this measurement is from an experiment with a relatively high number density of particles \( (\sim 1200 \text{ cm}^{-3}) \), and values of \( \overline{S} \) for experiments with lower number densities of particles are unknown. For the Lagrangian autocorrelation time \( \tau_{\text{cor}} \), \( \tau_{\text{cor}} \) is expected to be closely related to the turbulence statistics and the spatio-temporal scale of the largest eddies (Tennekes and Lumley 1972). Turbulence statistics for the \( \Pi \)-chamber experiment are given in Chang et al. (2016) (see their Table 3), who estimated that \( \eta=0.11 \text{cm}, \lambda=1.6 \text{cm}, \mathcal{L}=8.8 \text{cm}, \) and \( R_\lambda = 55 \). These estimates are fairly close to the corresponding turbulence statistics for the small-scale DNS in the present study (See Table 2). However, we should keep in mind the difficulty in measuring the turbulence statistics in the \( \Pi \)-chamber where, unlike a wind-tunnel experiment, there is no mean flow and hence Taylor’s hypothesis of frozen turbulence does not necessarily hold.
Given the uncertainties in the measurements of the mean supersaturation $\overline{S}$ and the turbulence statistics described above, we have to simplify the question so that the problem can be addressed by the present DNS. Because the effect of $\overline{S}$ for the droplet growth is well known, and also, as described in the previous subsection, $\overline{S}$ in the present DNS is affected by the nudging parameter $\tau_0$, the choice of which is largely arbitrary, here we focus on the effect of turbulence and consider the following question: if we make the domain and corresponding largest possible eddy size comparable to those for the II-chamber, can the broadening effect of the supersaturation fluctuation in the present DNS explain the droplet size distributions in the II-chamber?

In order to answer this question, we conducted the following large-scale DNS: Run11-Run14. The setups for these experiments are the same as those for Run6-Run10 except the box length is $L_{\text{box}} = 102.4$ cm, the grid number is $N = 512$ in each dimension, the grid length is $\Delta x = 2$ mm, the time increment is $\Delta t = 0.8 \times 10^{-3}$ s, the nudging time is $\tau_0 = 2$ s in (28) and (29), and the parameters shown in Table 3 are used. The turbulence parameters for Run11-Run14 are shown in Table 2. The period of numerical integration is 3600 s (4.5 million steps) in order for the large-scale DNS to obtain a statistically steady state.

Figure 10 shows PDFs of the droplet diameter in statistically steady...
states for Run11–Run14. In comparison to the results of the small-scale DNS in Fig. 9a, the distributions in Fig. 10 are broader (compare Run7 and Run12, which have almost the same number density of \( \sim 80 \text{ cm}^{-3} \)). Furthermore, the number densities for Run11–Run13 approximately correspond to the cloud droplet number densities for the \( \bar{n}_a = 1, 2, 4/\text{cm}^3/\text{min} \) experiments in C16, respectively. The results shown in their Fig. 2 and our Fig. 10, seem to agree quantitatively.

Figure 11 shows the spectra for the kinetic energy and the variance of the supersaturation for the large-scale DNS. Since the Taylor microscale Reynolds number \( R_\lambda \) is 207, a wavenumber range with a slope close to \(-5/3\) can be seen in the middle of these spectra. These results are consistent with the power spectral density for the vertical velocity fluctuations measured in the II-chamber (see Fig. 8 in Chang et al. 2016), but again the difficulties in measuring the turbulence statistics in the II-chamber should be noted.

Table 7 shows the standard deviation of the supersaturation (\( \sigma_S \)) for the large-scale DNS. By substituting the results for Run11–Run14 into the right side of (41), the turbulent mixing time for the large-scale DNS is estimated as \( \tau_l = 3.824 \pm 0.173 \approx 3.82 \text{ s} \), which is about 70% greater than the large-eddy turnover time (\( T_E = 2.2 \text{ s} \), see Table 2). This again confirms that the correct value of \( \tau_l \) is of the same order as \( T_E \), but also indicates the possibility that the ratio \( \tau_l/T_E \) weakly depends on the domain size and
other turbulence parameters such as the Reynolds number.

Table 8 shows the standard deviation of the squared radius ($\sigma_{R^2}$) for the large-scale DNS. Values of $\tau_{\text{cor}}$ are over 1 s, which are at least twice as large as $\tau_{\text{cor}}$ for the small-scale DNS (see Table 6). Accordingly, the diffusion coefficients ($D$) for the large-scale DNS are larger in (9), resulting in the broader size distributions as shown in Fig. 10. The experimental results for $\sigma_{R^2}$ seem to agree fairly well with the theoretical results (T2) in Table 8.

4. Discussion

The results in the present study are very satisfactory and encouraging for the future research of cloud and turbulence using the cloud microphysics simulator. At the same time, however, we should note several important differences between the present study and atmospheric clouds and turbulence. As described in Sec. 1, the main purpose of the present study was to compare our DNS results with the statistical theory and the laboratory experiment by C16. Since there are differences in the physical parameters and conditions of the atmospheric clouds and the laboratory clouds in the II-chamber, the interpretation of the present results in the context of the atmospheric clouds needs great care. Among those points, important ones are as follows:

1. **Time scale of turbulence.** The interaction between the supersatura-
tion fluctuation and cloud droplets substantially depends on the ratio between the phase relaxation time $\tau_c$ and the turbulence large-eddy turnover time $T_E$, that is, the Damköhler number: $Da = T_E/\tau_c$. The large eddy turnover time $T_E$ tends to increase with the Reynolds number. For example, $T_E$ is of the order of several tens of seconds in atmospheric clouds at $R_\lambda \approx 5000$ (Siebert and Shaw 2017), while it is shorter than several seconds in the present DNS at $R_\lambda \approx 200$ and the II-chamber experiment. The dumping effect of cloud droplets on the supersaturation fluctuation increases with the Reynolds number and thus more significant in atmospheric clouds [see Eq. (5)], which in turn affects the droplet growth by condensation/evaporation.

2. **Mean supersaturation.** In the present DNS, the mean supersaturation is kept close to a statistically steady value $[=S_0(T_0, Q_0)]$ by nudging terms in (28) and (29), and we set $S_0(T_0, Q_0) = 0$ for simplicity. In atmospheric clouds, however, droplet size distributions form in updrafts and associated mean supersaturation due to adiabatic cooling, when diffusion growth of droplet takes place. Because the positive mean supersaturation has the effect of narrowing the PDF of the droplet radius (not the squared radius) in ascending adiabatic volume (see Grabowski and Wang 2013, and references therein), the effect of the mean supersaturation should be taken into account when investigating the formation mechanism of the droplet size distribution.
in atmospheric clouds.

Note that the cloud microphysics simulator can incorporate the mean supersaturation effects, for example, by ascending the cubic box with updraft velocity that is self-consistently determined through the buoyancy force. Indeed the narrowing of the PDF of the droplet radius has successfully been simulated, showing that the cloud microphysics simulator is able to compute complex nonlinear interaction between mean and fluctuations (Saito and Gotoh, 2018). For the future studies of the cloud microphysics, however, it is necessary to verify the code and indispensable to obtain deeper understanding of the effects of the supersaturation fluctuation. As a simple but most effective way for this purpose, we compared the results of the cloud microphysics simulator with the experimental results by C16. The good agreement obtained in the present study is very encouraging for the future application of the cloud microphysics simulator.

3. *Generation mechanism for supersaturation fluctuation.* Fluctuations of supersaturation in atmospheric clouds are considered to be correlated with fluctuations of vertical velocity. On the other hand, in the present DNS, we used the external scalar injections that are not correlated with the corresponding forces of the velocity field (see Sec. 3.2). One way to model the excitation of the supersaturation fluctuation which is correlated with the vertical velocity field is to use the scalar injection $\Gamma u_z$, propor-
tional to the vertical velocity $u_z$ times the prescribed vertical scalar mean 
gradient $\Gamma$ (Gotoh and Watanabe 2015). Such kind of scalar injections are 
often used in DNS studies on cloud turbulence (Celani et al. 2005; Lan-
otte et al. 2009; Sardina et al. 2015; Saito and Gotoh 2018), while several 
other studies use the same kind of forces as the present study (Siewert et al. 
2017). It is easily found that the dimensional estimate for the scalar injec-
tion in the wavenumber space yields $F_\theta(k) \propto \Gamma^2 \tau(k) E(k) \propto k^{-7/3}$, where 
$\tau(k) (\propto k^{-2/3})$ and $E(k) (\propto k^{-5/3})$ are the characteristic time and the kinetic 
energy spectrum of turbulence in the inertial range, respectively. Therefore 
it follows that the scalar injection proportional to the vertical velocity has 
most inputs at very low wavenumbers, which means that there are no dif-
fERENCE in effects between the low wavenumber injection and the injection 
proportional to the vertical velocity. Indeed the low order moments such 
as the spectrum of the scalar variances and Yaglom’s 4/3 laws obtained by 
simulations using two different injections are the same (Gotoh and Yeung 
2013; Saito and Gotoh 2018). The above arguments and facts support the 
suggestion by Siewert et al. (2017) that the properties of the supersatu-
ration field without droplets are expected to be insensitive to the specific 
form of the scalar injection.
5. Summary

The purpose of the present study was to conduct direct numerical simulations (DNSs) of the growth of cloud droplets by condensation in turbulence using our DNS model “cloud microphysics simulator”, and to compare the results with the statistical theory proposed by Chandrakar et al. (2016) (referred to as “C16”) and the experimental results from their laboratory cloud chamber, the “II-chamber”. For this purpose, we first made several extensions to the statistical theory by C16. We derived the Fokker-Planck equation for the droplet size distribution and introduced aerosol (curvature and solute) effects into the equation with a zero flux boundary condition, which is mathematically equivalent to the case of Brownian motion in the presence of a wall. We obtained an analytical expression for the size distribution in a steady state which is proportional to $R\exp(-cR^2)$, where $c$ is a constant. Next, we conducted a small-scale DNS ($L_{\text{box}} = 12.8\text{cm}$) and showed that the results agree closely with the theory. In addition, by careful analysis of the DNS data, we found that the proper value for the turbulent mixing time ($\tau_t$) in the theory is greater than the large-eddy turnover time for turbulence by about 50%. We also found that the diffusion coefficient in the Fokker-Planck equation should be expressed in terms of the Lagrangian autocorrelation time for the supersaturation fluctuation. Finally, we conducted a large-scale DNS ($L_{\text{box}} = 102.4\text{cm}$) with a domain size comparable
to the size of the II-chamber, and obtained steady state size distributions
which are quantitatively consistent with the results of the II-chamber ex-
periment.

As described in Sec. 3, there are many processes that are not included
in the present DNS for simplicity, such as particle inertia, gravity, collision-
coalescence and buoyancy. Nevertheless, it is still remarkable that quan-
titatively consistent results are obtained among the large-scale DNS, the
statistical theory, and the II-chamber experiment, suggesting that these
share the essence of the formation mechanism of the droplet size distribu-
tion by condensation in turbulence. This agreement provides validation for
our DNS and encourages further studies in this direction.

For future studies, we should clarify the effects of the simplifications in
the present DNS. First, while the II-chamber has solid boundaries and ex-
cites turbulence by moist Rayleigh-Benard convection, the present DNS as-
sumed a periodic domain and excited turbulence by random external forces.
The effects of these simplifications can be investigated by DNSs or large-
eddy simulations of moist Rayleigh-Benard convection with more realistic
boundary conditions.

Second, the present DNS neglected particle inertia and gravity. Because
these processes are known to cause substantial changes in particle trajecto-
tories especially for larger particles (Sundaram and Collins 1997; Falkovich
and Pumir 2007), these changes can in turn affect the Lagrangian autocorrelation time for the supersaturation fluctuation $\tau_{cor}$. In fact, Vaillancourt et al. (2002) reported that the gravitational sedimentation reduces the broadening effect of the supersaturation fluctuation on the droplet size distribution. Clarifying the effects of particle inertia and gravity on the correlation time could also be useful for informing cloud microphysical parameterizations proposed in recent studies (Grabowski and Abade 2017; Sardina et al. 2018).

Third, to facilitate the comparison with the statistical theory, the present DNS assumed that the particle removal process is independent of the particle size so that the residence timescale $\tau_{res}$ is constant. However, since particles in the P-chamber are removed by gravitational sedimentation, $\tau_{res}$ is expected to be shorter for larger particles. A possible effect of a shorter $\tau_{res}$ for larger particles was reported in the P-chamber experiment by C16. The effect of gravity could also change the functional form of the steady state size distribution obtained from the Fokker-Planck equation in Sec. 2, thereby changing the statistical properties of the distribution. Full scale simulation including those effects is really challenging and will be reported somewhere.

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Appendix A: Phase relaxation time $\tau_c$ 

The phase relaxation time $\tau_c$ is introduced by Cooper (1989). The derivation of $\tau_c$ is explained in Lanotte et al. (2009) (see also Sardina
\( \tau_c^{-1} = 4\pi K_s \frac{\rho_w}{\rho_a} \left( \frac{1}{Q_{vs}} + \frac{\varepsilon_0 L_v^2}{c_p R_d T^2} \right) n_d \overline{R} \left( \propto n_d \overline{R} \right). \) (57)

Here, the saturation mixing ratio \( Q_{vs} \) is given by (33). With \( T = 290 \) K and the parameters taking the values shown in Table 1, Eq. (57) gives

\[ \tau_c^{-1} = 2.776 \cdots [\text{cm}^2\text{s}^{-1}] \times n_d \overline{R}. \] (58)

This equation is used to estimate \( \tau_c \) in the present study.

**Appendix B: Parameters for Köhler curve**

For a drop of a sufficiently dilute aqueous solution, \( A \) and \( B \) in (31) are given as follows (Pruppacher and Klett 1997):

\[
A = \frac{2M_w \sigma_w}{R_g \rho_w T_A}, \quad \quad \quad (59)
\]

\[
B = \frac{3\nu A \Phi m_s M_w}{4\pi M_s \rho_w}, \quad \quad \quad (60)
\]

where \( T_A \) is the temperature around a drop. We set \( T_A \) as a constant because the temperature change in the present DNS is sufficiently small. Definitions of the other parameters in (59) and (60) are given in Tables 1 and 9.

We assume that the solute dissolved in each drop is NaCl (sodium chloride) with a fixed mass of \( m_s = 1.13 \times 10^{-15} \) g. This value of \( m_s \) corresponds to a sphere with a 50 nm radius and a density \( \rho_s = 2.16 \) g cm\(^{-3}\).
Substituting the parameters in Tables 1 and 9 into (59) and (60), we obtain

\[ A = 1.089 \times 10^{-7} \text{ cm and } B = 1.665 \times 10^{-16} \text{ cm}^3, \text{ respectively.} \]

Appendix C: Definitions of turbulence parameters

The kinetic energy is defined by

\[ E = \frac{1}{2} \langle u_i^2 \rangle = \int_0^\infty E(k)dk, \]  \hspace{1cm} (61)

where \( u_i \ (i = 1, 2, 3) \) are components of the velocity vector \( \mathbf{u} \) (repeated indices are summed), the angle brackets \( \langle \cdots \rangle \) represent the spatial and temporal averages, and \( E(k) \) is the kinetic energy spectrum. The mean energy dissipation rate is defined by

\[ \epsilon = \frac{\nu_a}{2} \langle (\partial_i u_j + \partial_j u_i)^2 \rangle. \]  \hspace{1cm} (62)

The integral scale, Taylor microscale, and Kolmogorov scale are respectively defined by

\[ L = \frac{3\pi}{4E} \int_0^\infty k^{-1}E(k)dk \]  \hspace{1cm} (63)

\[ \lambda = \sqrt{\langle u_1^2 \rangle / \langle (\partial_1 u_1)^2 \rangle} \]  \hspace{1cm} (64)

\[ \eta_K = \left( \frac{\nu_a^3}{\epsilon} \right)^{1/4}. \]  \hspace{1cm} (65)

The large-eddy turnover time and Kolmogorov time are respectively defined by

\[ T_E = L/u_{rms}. \]  \hspace{1cm} (66)
\[ \tau_K = \left( \nu_a / \epsilon \right)^{1/2}, \quad (67) \]

where \( u_{\text{rms}} = \sqrt{2E/3} \) is the root-mean-square velocity. The Taylor microscale Reynolds number is defined by

\[ R_\lambda = u_{\text{rms}} \lambda / \nu_a. \quad (68) \]

**Appendix D: Statistical properties of the size distribution**

The functional forms of the PDFs of \( R \) and \( R^2 \) are given in (20) and (48), respectively. The statistical properties of the size distribution are as follows. The mean radius:

\[ \overline{R} = \int_0^\infty R \mathcal{P}(R)dR = \frac{\sqrt{\pi}}{2} \beta. \quad (69) \]

The mean squared radius is given in (21) and \( \overline{R^2} = \beta^2 \). The mean cubic radius:

\[ \overline{R^3} = \int_0^\infty R^3 \mathcal{P}(R)dR = \frac{3\sqrt{\pi}}{4} \beta^3. \quad (70) \]

The variance of \( R \):

\[ \sigma_R^2 = \overline{R^2} - \left( \overline{R} \right)^2 = \frac{(4 - \pi)}{4} \beta^2. \quad (71) \]

The variance of \( R^2 \) is given in (22) and \( \sigma_{R^2}^2 = \beta^4 \). The relative dispersion:

\[ \frac{\sigma_R}{\overline{R}} = \sqrt{\frac{4 - \pi}{\pi}} = 0.5227 \ldots \approx 0.52. \quad (72) \]
The relative dispersion measured in the Π-chamber is between 0.2 and 0.4 (Desai et al. 2018; Chandrakar et al. 2018b). The mean volume radius:

\[ R_v = \left( \frac{R^3}{4} \right)^{1/3} = \left( \frac{3\sqrt{\pi}}{4} \right)^{1/3} \beta \]  

(73)

The effective radius:

\[ R_e = \frac{R^3}{R^2} = \frac{3\sqrt{\pi}}{4} \beta. \]  

(74)

The parameter \( k \) used in cloud physics is given by

\[ k = \frac{R^3_v}{R^3_e} = \left( \frac{3\sqrt{\pi}}{4} \right)^{-2} = 0.5658... \approx 0.57 \]  

(75)

(For details of the parameter \( k \), readers are referred to Martin et al. 1994; Chandrakar et al. 2018a) \( k = 0.80 \pm 0.07 \) for maritime airmass clouds and \( k = 0.67 \pm 0.07 \) for continental airmass clouds (Martin et al. 1994). \( k = 0.66 \pm 0.01 \) for the Π-chamber (Chandrakar et al. 2018a).

**Appendix E: Solution of the Fokker-Planck equation**

Here we consider the solution to the Fokker-Planck equation (11) with the boundary conditions (12) and (13), and the initial conditions (14) (Chandrasekhar 1943). The analytical solution is expressed in terms of Green’s function as

\[
n(\rho, t) = e^{-t/\tau_{\text{res}}} \int_0^\infty G(\rho, \rho')n_0(\rho')d\rho' + \int_0^t e^{-s/\tau_{\text{res}}} G(\rho, \rho_0, s)ds, \quad (76)
\]

\[
G(\rho, \rho', t) = \frac{1}{\sqrt{4\pi D_t}} \left[ \exp \left( -\frac{(\rho - \rho')^2}{4D_t} \right) + \exp \left( -\frac{(\rho + \rho')^2}{4D_t} \right) \right] H(t), \quad (77)
\]

52
where \( \rho = R^2 \geq 0 \) for ease of writing and \( H(t) \) is the Heaviside function. It is readily found that \( G \) satisfies the boundary condition \( \partial G(\rho, \rho', t)/\partial \rho = 0 \) at \( \rho = 0 \) and the normalization

\[
\int_0^\infty G(\rho, \rho', t)d\rho = H(t). \tag{78}
\]

The first term of the right hand side of Eq.(76) arises from the initial condition of \( n \) which decays in time. Integration of \( n(\rho, t) \) over \( \rho \) yields the number density \( n_d(t) \) as function of time as

\[
n_d(t) = n_{d0}e^{-t/\tau_{\text{res}}} + J_0\tau_{\text{res}} \left( 1 - e^{-t/\tau_{\text{res}}} \right). \tag{79}
\]

where \( n_{d0} \) is the initial number density. The probability density function \( P(\rho, t) \) is given by

\[
P(\rho, t)d\rho = \frac{n(\rho, t)d\rho}{n_d(t)}. \tag{80}
\]

In order to make the arguments simpler we put

\[
n(\rho, 0) = n_{d0}\delta(\rho - \rho_0), \tag{81}
\]

so that the probability density function (PDF) \( P(\rho, t) \) at latter times is given by

\[
P(\rho, t) = \frac{1}{n_d(t)} \left[ n_{d0}e^{-t/\tau_{\text{res}}}G(\rho, \rho_0, t) + J_0 \int_0^t e^{-s/\tau_{\text{res}}}G(\rho, \rho_0, s)ds \right]. \tag{82}
\]

Now we consider two cases. First let \( \rho_* = R^2 \) be the square of the droplet radius at which the Köhler curve attains the maximum.
Case 1 \( \rho_* \ll |\rho - \rho_0| \sim \sqrt{D\tau_{\text{res}}} \ll \rho_0 \).

This corresponds to the domain of droplet radius in which the aerosol effects are negligible. Writing as \( \rho_0 - \rho = \xi \) we have the estimate \((\rho + \rho_0)^2 = 4\rho_0^2(1 - (\xi/\rho_0) + (\xi/\rho)^2/4) \sim 4\rho_0^2\). Then the second term of the right hand side of Eq.(77) becomes negligible compared to the first term, and thus the PDF is approximately

\[
P(\rho, t) = \frac{1}{n_d(t)\sqrt{4\pi D}} \left[ n_{d0} \frac{1}{\sqrt{t}} \exp \left( -\frac{t}{\tau_{\text{res}}} - \frac{(\rho - \rho_0)^2}{4Dt} \right) \right. \\
\left. + J_0 \int_0^t \frac{1}{\sqrt{s}} \exp \left( -\frac{s}{\tau_{\text{res}}} - \frac{(\rho - \rho_0)^2}{4Ds} \right) ds \right]. \quad (83)
\]

At latter times as \( t \gg \tau_{\text{res}} \) the contribution from the initial condition (the first term) vanishes and the second integral reduces to

\[
P_\infty(\rho) = \frac{1}{\sqrt{4D\tau_{\text{res}}}} \exp \left( -\frac{|\rho - \rho_0|}{\sqrt{D\tau_{\text{res}}}} \right), \quad (84)
\]

which is Eq.(15). Here we used the formula

\[
\int_0^\infty \exp \left( -ax^2 - \frac{b}{x^2} \right) dx = \frac{1}{2\sqrt{\pi/a}} \exp \left( -2\sqrt{ab} \right). \quad (85)
\]

The moments \( M_n(t) = \langle [\rho(t)]^n \rangle \) are computed by the standard way as

\[
M_n(t) = \int_0^\infty \rho^n P(\rho, t) d\rho \\
= \frac{1}{n_d(t)\sqrt{4\pi D}} \left[ n_{d0} \frac{1}{\sqrt{t}} e^{-t/\tau_{\text{res}}} \int_{-\rho_0}^\infty (\rho_0 + \xi)^n \exp \left( -\frac{\xi^2}{4Dt} \right) d\xi \right. \\
\left. + J_0 \int_0^t \frac{1}{\sqrt{s}} e^{-s/\tau_{\text{res}}} \int_{-\rho_0}^\infty (\rho_0 + \xi)^n \exp \left( -\frac{\xi^2}{4Ds} \right) dx ds \right]. \quad (86)
\]
The lower boundary $-\rho_0$ of the integral can effectively be replaced by $-\infty$ because of $\sqrt{Dt} \ll \rho_0$ in the present case. We obtain the mean and the variance as

$$\overline{\rho(t)} = \rho_0, \quad \sigma^2(t) = |\rho(t)|^2 - \rho(t)^2$$

$$= 2D\tau_{\text{res}} \frac{J_0\tau_{\text{res}}}{n_d(t)} \left[ 1 - \left( 1 + \left( 1 - \frac{n_{d0}}{J_0\tau_{\text{res}}} \right) \frac{t}{\tau_{\text{res}}} \right) e^{-t/\tau_{\text{res}}} \right]$$

$$\rightarrow 2D\tau_{\text{res}} \text{ as } t \rightarrow \infty.$$  

The mean value is independent of time and the variance approaches the constant $2D\tau_{\text{res}}$ at large $t$. When $\frac{n_{d0}}{J_0\tau_{\text{res}}} = 1$, Eq.(88) reduces to Eq.(44).

**Case 2** $\rho_s \sim \rho_0$.

This corresponds to the domain of small droplets so that the aerosols affect the distribution function at very small radius. Simple analytical expression of the PDF for all time is not available, and the precise functional form may be obtained by numerical integration of Eq.(82). However, when we focus on the asymptotic state at large time, the asymptotic PDF can be obtained. The first term (the initial contribution) of the right hand side of Eq.(82) vanishes and the second term is

$$P(\rho) = \frac{1}{\sqrt{4\pi D\tau_{\text{res}}}} \int_0^\infty \frac{1}{\sqrt{s}} e^{-s/\tau_{\text{res}}} \left[ \exp \left( -\frac{(\rho - \rho_0)^2}{4Ds} \right) + \exp \left( -\frac{(\rho + \rho_0)^2}{4Ds} \right) \right] ds$$

$$= \frac{1}{\sqrt{4D\tau_{\text{res}}}} \left[ \exp \left( -\frac{\rho - \rho_0}{\sqrt{D\tau_{\text{res}}}} \right) + \exp \left( -\frac{\rho + \rho_0}{\sqrt{D\tau_{\text{res}}}} \right) \right].$$  

(89)
At $\rho \ll \rho_*$ the PDF is

$$P(\rho = 0) = \frac{1}{\sqrt{D_{\tau_{\text{res}}}}} \exp \left( -\frac{\rho_0}{\sqrt{D_{\tau_{\text{res}}}}} \right), \quad (90)$$

and for large $\rho \gg \rho_* \sim \rho_0$

$$P(\rho) = \frac{1}{\sqrt{D_{\tau_{\text{res}}}}} \exp \left( -\frac{\rho}{\sqrt{D_{\tau_{\text{res}}}}} \right). \quad (91)$$

The mean and variance at large time are

$$\bar{\rho} = \rho_0 + \sqrt{D_{\tau_{\text{res}}}e^{-\rho_0/\tau_{\text{res}}}} \quad (92)$$

$$\sigma^2(t) = D_{\tau_{\text{res}}} \left( 2 - e^{-2\rho_0/\sqrt{D_{\tau_{\text{res}}}}} - 2 \frac{\rho_0}{\sqrt{D_{\tau_{\text{res}}}}} e^{-\rho_0/\sqrt{D_{\tau_{\text{res}}}}} \right) \quad (93)$$

$$\rightarrow \sqrt{D_{\tau_{\text{res}}}} \quad \text{for} \quad \rho_0 \to 0$$

$$\rightarrow D_{\tau_{\text{res}}} \quad \text{for} \quad \rho_0 \to 0$$

respectively, and they are Eqs. (21) and (22), respectively.

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1 (a) The Köhler curve $f(R) = (A/R) - (B/R^3)$, where $R$ is the droplet radius of $\mu$m, $A = 1.089 \times 10^{-7}$ cm$^{-1}$ and $B = 1.665 \times 10^{-16}$ cm$^{-3}$. The horizontal line indicates zero. (b) Same as (a) except that the vertical axis is on a logarithmic scale and the range of $R$ is extended to 20 $\mu$m.

2 (a) PDFs for the supersaturation fluctuation in the statistically steady state for Run1–Run10. (b) Same as (a) but the results of each experiment are normalized by the variance. The cyan curve indicates the Gaussian distribution with a zero mean and unit variance: $(2\pi)^{-1/2} \exp(-S^2/2)$.

3 Estimates of $\tau_i$ obtained by substituting the result of each experiment into the right side of (41). The horizontal axis is the phase relaxation time $\tau_c$ for each experiment. The horizontal line indicates the average 0.85 (See the text for details).

4 (a) PDFs of the squared radius ($R^2$) in statistically steady states for Run1–Run5. (b) Same as (a) but the results of each experiment are normalized by (42). The cyan curve indicates the theoretical curve: $\frac{1}{2} \exp(-|R^2|)$ (See the text for details).

5 (a) Time evolution of the variance of the squared radius ($\sigma^2_{R^2}$) for Run1–Run5. (b) Same as (a) but the results of each experiment are normalized by $D\tau_{res}$ (See the text for details). The cyan curve indicates the theoretical curve: $2(1 - e^{-t/\tau_{res}})$. The short line indicates a slope of 1.

6 Comparison of the results between Run1 and the additional experiment which is the same as Run1 except that the particles are stopped (indicated by “stop”). (a) PDFs for the supersaturation fluctuation in statistically steady states. (b) PDFs for the squared radius ($R^2$) of cloud droplets in statistically steady states. “No drop” in the left panel indicates results for simulations without particles.
Time evolution of the droplet radius \( R \) and supersaturation at the droplet position \( S \) for a single particle in Run6. The horizontal line indicates zero. Note that the time \( t' \) for the horizontal axis starts from the moment after the system has attained a statistically steady state (4000 s after the beginning of the simulation).

(a) PDFs of the squared radius \( R^2 \) in statistically steady states for Run6–Run10. (b) Same as (a) but the results of each experiment are normalized by (46). The cyan curve indicates the theoretical curve: \( \exp(-\bar{R}^2) \) (See the text for details).

(a) PDFs for the diameter \( d \) of cloud droplets in statistically steady states for Run6–Run10. (b) Same as (a) but the results of each experiment are normalized by (50). The cyan curve indicates the theoretical curve: \( (\bar{d}/2) \exp(-\bar{d}^2/4) \) (See the text for details).

PDFs for the diameter \( d \) of the cloud droplets in statistically steady states for Run11–Run14.

(a) Time-averaged kinetic energy spectrum for Run11. Other runs (Run12–Run14) have similar kinetic energy spectra. (b) Time-averaged variance spectra of the supersaturation for Run11–Run14. The short line in each panel indicates a slope of -5/3.
Fig. 1. (a) The Köhler curve $f(R) = (A/R) - (B/R^3)$, where $R$ is the droplet radius of $\mu m$, $A = 1.089 \times 10^{-7}$ cm$^{-1}$ and $B = 1.665 \times 10^{-16}$ cm$^{-3}$. The horizontal line indicates zero. (b) Same as (a) except that the vertical axis is on a logarithmic scale and the range of $R$ is extended to 20 $\mu m$. 
Fig. 2. (a) PDFs for the supersaturation fluctuation in the statistically steady state for Run1–Run10. (b) Same as (a) but the results of each experiment are normalized by the variance. The cyan curve indicates the Gaussian distribution with a zero mean and unit variance: $(2\pi)^{-1/2} \exp(-S^2/2)$. 
Fig. 3. Estimates of $\tau_t$ obtained by substituting the result of each experiment into the right side of (41). The horizontal axis is the phase relaxation time $\tau_c$ for each experiment. The horizontal line indicates the average 0.85 (See the text for details).
Fig. 4. (a) PDFs of the squared radius ($R^2$) in statistically steady states for Run1–Run5. (b) Same as (a) but the results of each experiment are normalized by (42). The cyan curve indicates the theoretical curve: $\frac{1}{2} \exp(-|\hat{R}^2|)$ (See the text for details).
Fig. 5. (a) Time evolution of the variance of the squared radius \( \sigma_{R^2}^2 \) for Run1–Run5. (b) Same as (a) but the results of each experiment are normalized by \( D \tau_{\text{res}} \) (See the text for details). The cyan curve indicates the theoretical curve: \( 2(1 - e^{-t/\tau_{\text{res}}}) \). The short line indicates a slope of 1.
Fig. 6. Comparison of the results between Run1 and the additional experiment which is the same as Run1 except that the particles are stopped (indicated by “stop”). (a) PDFs for the supersaturation fluctuation in statistically steady states. (b) PDFs for the squared radius ($R^2$) of cloud droplets in statistically steady states. “No drop” in the left panel indicates results for simulations without particles.
Fig. 7. Time evolution of the droplet radius ($R$, red curve) and supersaturation at the droplet position ($S$, blue curve) for a single particle in Run6. The horizontal line indicates zero. Note that the time $t'$ for the horizontal axis starts from the moment after the system has attained a statistically steady state (4000 s after the beginning of the simulation).
Fig. 8. (a) PDFs of the squared radius ($R^2$) in statistically steady states for Run6–Run10. (b) Same as (a) but the results of each experiment are normalized by (46). The cyan curve indicates the theoretical curve: $\exp(-\hat{R}^2)$ (See the text for details).
Fig. 9. (a) PDFs for the diameter \(d\) of cloud droplets in statistically steady states for Run6–Run10. (b) Same as (a) but the results of each experiment are normalized by (50). The cyan curve indicates the theoretical curve: \((\bar{d}/2) \exp(-\bar{d}^2/4)\) (See the text for details).
Fig. 10. PDFs for the diameter ($d$) of the cloud droplets in statistically steady states for Run11–Run14.
Fig. 11. (a) Time-averaged kinetic energy spectrum for Run11. Other runs (Run12–Run14) have similar kinetic energy spectra. (b) Time-averaged variance spectra of the supersaturation for Run11–Run14. The short line in each panel indicates a slope of -5/3.
<table>
<thead>
<tr>
<th>Table</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Definitions of parameters and their corresponding values in the present DNS.</td>
</tr>
<tr>
<td>2</td>
<td>Mean turbulence parameters. $R_{\lambda}$ is the Taylor microscale Reynolds number, $u_{rms}$ is the root-mean-square velocity, $\epsilon$ is the mean energy dissipation rate per unit mass, $L$ is the integral scale, $\lambda$ is the Taylor microscale, $\eta_K$ is the Kolmogorov length, $k_{\text{max}}\eta$ is the cut off wavenumber normalized by the Kolmogorov length, $T_E$ is the large-eddy turnover time, and $\tau_K$ is the Kolmogorov time. Top: small-scale DNS. Bottom: large-scale DNS.</td>
</tr>
<tr>
<td>3</td>
<td>Experimental setups for cloud droplets and their statistics at steady states for the small-scale DNS (Run1–Run10) and the large-scale DNS (Run11–Run14). $R_0$ is the initial radius of cloud droplets, $\bar{R}$ is the mean droplet radius in the statistically steady state, $J_0$ is the injection rate of cloud droplets, $\bar{n}_d$ is the mean number density of cloud droplets in the statistically steady state, and $\tau_c$ is the phase relaxation time estimated from (57). “Yes”/“No” in the second column “Köhler” indicates that the growth equation (31) is integrated with/without terms including $A$ and $B$, respectively.</td>
</tr>
<tr>
<td>4</td>
<td>Statistics for the supersaturation fluctuation for the small-scale DNS. $\sigma_S$ is the standard deviation for the supersaturation, $\sigma_{S_0}(\tau_s/\bar{c})^{1/2}$ is the theoretical value from (5), $\tau_s$ is the system timescale (7), $\bar{c}$ is the phase relaxation timescale, and $\tau_t = 0.85$ s is used (See the text for details).</td>
</tr>
<tr>
<td>5</td>
<td>Statistics for the droplet size distributions in steady states for Run 1–5. $\sigma_{R^2}$ is the standard deviation of the squared radius $R^2$, “T0” is the theoretical value for $\sigma_{R^2}$ from (18), $\tau_{\text{corr}}$ is the Lagrangian autocorrelation time of the supersaturation fluctuation, $\tau_s$ is the system timescale, and $\tau_c$ is the phase relaxation timescale. $\tau_t = 0.85$ s is used for $\tau_s$.</td>
</tr>
</tbody>
</table>
Statistics for the droplet size distributions in steady states for Run 6–10. $\sigma_{R^2}$ is the standard deviation of the squared radius $R^2$. “T0” is the theoretical value for $\sigma_{R^2}$ from (22), “T1” is the theoretical value from (56) with the mean supersaturation $\mathcal{S}$ in the 6th column, “T2” is the same as T1 except that $\mathcal{S}$ is replaced by $\mathcal{S} - S_K$ where $S_K = 10^{-2}\%$. $\mathcal{S}$ is the mean supersaturation, $\tau_{\text{cor}}$ is the Lagrangian autocorrelation time for the supersaturation fluctuation, $\tau_s$ is the system timescale, and $\tau_c$ is the phase relaxation timescale. $\tau_t = 0.85$ s is used for $\tau_s$. See the text for details of T1 and T2.

Same as Table 4 but for the large-scale DNS (Run11–Run14). $\tau_t = 3.82$ s is used (See the text for details).

Same as Table 6 but for the large-scale DNS.

Parameters for the Köhler curve. The solute is NaCl (sodium chloride). $m_s = 1.13 \times 10^{-15}$ g of NaCl corresponds to a sphere with a 50 nm radius and a density $\rho_s = 2.16$ g cm$^{-3}$.
Table 1. Definitions of parameters and their corresponding values in the present DNS.

<table>
<thead>
<tr>
<th>Description</th>
<th>Symbol</th>
<th>Value</th>
<th>Unit</th>
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<tbody>
<tr>
<td>Density of liquid water</td>
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<td>g cm(^{-3})</td>
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<td>Density of air</td>
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<td>g cm(^{-3})</td>
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<td>Kinematic viscosity of dry air</td>
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<td>Thermal diffusivity</td>
<td>( \kappa_T )</td>
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<td>Diffusivity of water vapor</td>
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<td>( 2.49 \times 10^{-1} )</td>
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<td>hPa</td>
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<td>Latent heat of vapor</td>
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<td>Gas constant for water vapor</td>
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<td>( 462 \times 10^{4} )</td>
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<td>Specific heat at constant pressure</td>
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<td>( 1005 \times 10^{4} )</td>
<td>cm(^2)s(^{-2})K(^{-1})</td>
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<tr>
<td>Diffusion coefficient in (31)</td>
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<td>( 1.097 \times 10^{-6} )</td>
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<tr>
<td>Temperature for nudging</td>
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<td>K</td>
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<tr>
<td>Vapor mixing ratio for nudging</td>
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<td>( 1.18 \times 10^{-2} )</td>
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Table 2. Mean turbulence parameters. $R_\lambda$ is the Taylor microscale Reynolds number, $u_{\text{rms}}$ is the root-mean-square velocity, $\epsilon$ is the mean energy dissipation rate per unit mass, $L$ is the integral scale, $\lambda$ is the Taylor microscale, $\eta_K$ is the Kolmogorov length, $k_{\text{max} \eta_K}$ is the cut off wavenumber normalized by the Kolmogorov length, $T_E$ is the large-eddy turnover time, and $\tau_K$ is the Kolmogorov time. Top: small-scale DNS. Bottom: large-scale DNS.

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<thead>
<tr>
<th>$R_\lambda$</th>
<th>$u_{\text{rms}}$</th>
<th>$\epsilon$</th>
<th>$L$</th>
<th>$\lambda$</th>
<th>$\eta_K$</th>
<th>$k_{\text{max} \eta_K}$</th>
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Table 3. Experimental setups for cloud droplets and their statistics at steady states for the small-scale DNS (Run1–Run10) and the large-scale DNS (Run11–Run14). $R_0$ is the initial radius of cloud droplets, $\bar{R}$ is the mean droplet radius in the statistically steady state, $J_0$ is the injection rate of cloud droplets, $n_d$ is the mean number density of cloud droplets in the statistically steady state, and $\tau_c$ is the phase relaxation time estimated from (57). “Yes”/“No” in the second column “Köhler” indicates that the growth equation (31) is integrated with/without terms including $A$ and $B$, respectively.
<table>
<thead>
<tr>
<th>Run</th>
<th>$\sigma_s$ [%]</th>
<th>$\sigma_{S_0}(\tau_s/\tau_c)^{1/2}$ [%]</th>
<th>$\tau_s$ [s]</th>
<th>$\tau_c$ [s]</th>
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<td>1.0</td>
<td>0.44</td>
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Table 4. Statistics for the supersaturation fluctuation for the small-scale DNS. $\sigma_s$ is the standard deviation for the supersaturation, $\sigma_{S_0}(\tau_s/\tau_c)^{1/2}$ is the theoretical value from (5), $\tau_s$ is the system timescale (7), $\tau_c$ is the phase relaxation timescale, and $\tau_t = 0.85$ s is used (See the text for details).
Table 5. Statistics for the droplet size distributions in steady states for Run 1–5. \( \sigma_{R^2} \) is the standard deviation of the squared radius \( R^2 \), “T0” is the theoretical value for \( \sigma_{R^2} \) from (18), \( \tau_{\text{cor}} \) is the Lagrangian autocorrelation time of the supersaturation fluctuation, \( \tau_s \) is the system timescale, and \( \tau_c \) is the phase relaxation timescale. \( \tau_t = 0.85 \text{ s} \) is used for \( \tau_s \).
Table 6. Statistics for the droplet size distributions in steady states for Run 6–10. $\sigma_{R^2}$ is the standard deviation of the squared radius $R^2$, “T0” is the theoretical value for $\sigma_{R^2}$ from (22), “T1” is the theoretical value from (56) with the mean supersaturation $\overline{S}$ in the 6th column, “T2” is the same as T1 except that $\overline{S}$ is replaced by $\overline{S} - S_K$ where $S_K = 10^{-2}\%$, $\overline{S}$ is the mean supersaturation, $\tau_{cor}$ is the Lagrangian autocorrelation time for the supersaturation fluctuation, $\tau_s$ is the system timescale, and $\tau_c$ is the phase relaxation timescale. $\tau_t = 0.85$ s is used for $\tau_s$. See the text for details of T1 and T2.

<table>
<thead>
<tr>
<th>Run</th>
<th>$\sigma_{R^2}$ [(\mu m^2)]</th>
<th>T0 [(\mu m^2)]</th>
<th>T1 [(\mu m^2)]</th>
<th>T2 [(\mu m^2)]</th>
<th>$\overline{S}$ [10^{-2} %]</th>
<th>$\tau_{cor}$ [s]</th>
<th>$\tau_s$ [s]</th>
<th>$\tau_c$ [s]</th>
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<td>32</td>
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<td>-1.5</td>
<td>0.33</td>
<td>0.44</td>
<td>0.90</td>
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</table>
Table 7. Same as Table 4 but for the large-scale DNS (Run11–Run14). $\tau_t = 3.82$ s is used (See the text for details).

<table>
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<tr>
<th>Run</th>
<th>$\sigma_S$</th>
<th>$\sigma_{S0}(\tau_s/\tau_t)^{1/2}$</th>
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<td>0.97</td>
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<td>14</td>
<td>0.79</td>
<td>0.81</td>
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<tr>
<td>Run</td>
<td>$\sigma_{R^2}$ [(\mu\text{m}^2)]</td>
<td>T0 [(\mu\text{m}^2)]</td>
<td>T1 [(\mu\text{m}^2)]</td>
<td>T2 [(\mu\text{m}^2)]</td>
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Table 8. Same as Table 6 but for the large-scale DNS.
<table>
<thead>
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<th>Parameter</th>
<th>Value</th>
<th>Unit</th>
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<tbody>
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<tr>
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<td>J K$^{-1}$ mol$^{-1}$</td>
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<td>K</td>
</tr>
<tr>
<td>Mass of solute $m_s$</td>
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<tr>
<td>Total number of ions $\nu_s$</td>
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<td></td>
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Table 9. Parameters for the Köhler curve. The solute is NaCl (sodium chloride). $m_s = 1.13 \times 10^{-15}$ g of NaCl corresponds to a sphere with a 50 nm radius and a density $\rho_s = 2.16$ g cm$^{-3}$. 
