# Roles of Drop Size Distribution and Turbulence in Autoconversion Based on Lagrangian Cloud Model Simulations

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#### Abstract

The roles of the drop size distribution (DSD) and turbulence in the autoconversion rate A are investigated by analyzing Lagrangian cloud model (LCM) data for shallow cumulus clouds. The correlations of DSD and turbulence with other cloud parameters are estimated, and they are applied to parameterize their effects on A. A new parameterization of A is proposed based on it, as  $A = \alpha qc7/3Nc-1/3H(Rc-Rc0)$  with  $\alpha = aNc-X(Rc-Rc0)(1+b\epsilon)$ , where qc, Nc, and Rc are the mixing ratio, the number concentration, and the volume mean radius of cloud droplets, respectively.  $\epsilon$  is the dissipation rate, Rc0 is the threshold value of Rc, H is the Heaviside step function, and X, a, and b are constants. Here, Nc-X(Rc-Rc0) represents the effect of DSD, via its correlation with Nc and qc, while A [?] qc7/3Nc-1/3 represents the effect of the gravitational collisional growth for given DSD and turbulence. The correlation between turbulence and DSD makes b larger than expected based on turbulence-induced collision enhancement. The effects of DSD and turbulence and their correlations with qc and Nc explain a wide range of exponent values of qc and Nc in many existing parameterizations of A. The new parameterization is compared with the LCM data and applied to a bulk cloud model (BCM) while clarifying the difference between the cloud droplet mixing processes of the LCM and BCM. The importance of DSD and turbulence in the raindrop formation in shallow cumulus clouds are shown by comparing the results from A with and without these effects.

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6	
7	Key Points:
8	• Identification of the role of the drop size distribution and turbulence in autoconversion by
9	analyzing Lagrangian cloud model data
10	• Correlations of the drop size distribution and turbulence with other cloud parameters and
11	its application to the parameterization
12	• Development of a new parameterization of autoconversion and its application to a bulk
13	cloud model
14	

## 15 Abstract

The roles of the drop size distribution (DSD) and turbulence in the autoconversion rate A are 16 investigated by analyzing Lagrangian cloud model (LCM) data for shallow cumulus clouds. The 17 correlations of DSD and turbulence with other cloud parameters are estimated, and they are applied 18 to parameterize their effects on A. A new parameterization of A is proposed based on it, as 19  $A = \alpha q_c^{7/3} N_c^{-1/3} H(R_c - R_{c0}) \text{ with } \alpha = a N_c^{-X} (R_c - R_{c0})(1 + b\varepsilon) \text{, where } q_c \text{, } N_c \text{, and } R_c \text{ are the}$ 20 mixing ratio, the number concentration, and the volume mean radius of cloud droplets, respectively. 21  $\varepsilon$  is the dissipation rate,  $R_{c0}$  is the threshold value of  $R_c$ , H is the Heaviside step function, and X, 22 a, and b are constants. Here,  $N_c^{-X}(R_c - R_{c0})$  represents the effect of DSD, via its correlation with 23  $N_c$  and  $R_c$ , while  $A \propto q_c^{7/3} N_c^{-1/3}$  represents the effect of the gravitational collisional growth for 24 given DSD and turbulence. The correlation between turbulence and DSD makes b larger than 25 expected based on turbulence-induced collision enhancement. The effects of DSD and turbulence 26 and their correlations with  $q_c$  and  $N_c$  explain a wide range of exponent values of  $q_c$  and  $N_c$  in 27 many existing parameterizations of A. The new parameterization is compared with the LCM data 28 and applied to a bulk cloud model (BCM) while clarifying the difference between the cloud droplet 29 mixing processes of the LCM and BCM. The importance of DSD and turbulence in the raindrop 30 formation in shallow cumulus clouds are shown by comparing the results from A with and without 31 these effects. 32

#### 34 Plain Language Summary

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One of the most important aspects of numerical weather prediction is to predict how much 36 precipitation is generated from cloud droplets within a cloud; this process is referred to as 37 autoconversion. Although autoconversion is known to be determined by the mass and number 38 concentration of cloud droplets, it can also be affected by other factors, such as the drop size 39 distribution (DSD) and turbulence. We clarify the effects of DSD and turbulence on 40 autoconversion by analyzing the results of a realistic cloud field simulated by a new type of cloud 41 model, in which cloud droplets are simulated as Lagrangian particles, and propose a 42 parameterization to predict the autoconversion rate. It is found that the correlations of DSD and 43 turbulence with other cloud parameters must be considered for the parameterization of their effects 44 45 in autoconversion. We also apply the new parameterization to numerical weather prediction, and show that the DSD and turbulence play an important role in raindrop formation in shallow cumulus 46 47 clouds.

#### 49 **1 Introduction**

The parameterization of autoconversion, which calculates the rate of raindrop formation by the coalescence of two cloud droplets, plays a critical role in cloud microphysics parameterization, as it triggers precipitation. Nonetheless, it still remains one of the most uncertain parts of cloud microphysics parameterization. In most parameterizations, the rate of autoconversion A is expressed in terms of the mixing ratio and the number concentration of cloud droplets,  $q_c$  and  $N_c$ , respectively, as

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$$\mathbf{A} = \alpha q_c^m N_c^{-n} \Phi \,. \tag{1}$$

57 Here,  $\alpha$  is a proportional constant and  $\Phi$  is an additional multiplicative function (Table 1). In many parameterizations,  $\Phi = 1$  (Beheng, 1994; Khairoutdinov & Kogan, 2000; Kogan, 2013), but 58 in some parameterizations,  $\Phi$  represents a threshold function such as  $H(R_c - R_{c0})$  (Liu & Daum, 59 60 2004; Tripoli & Cotton, 1980), the influence of the drop size distribution (DSD) (Berry & Reinhardt, 1974; Liu & Daum, 2004; Liu et al., 2007; Seifert & Beheng, 2001), or the influence 61 of turbulence (Seifert et al., 2010). Here,  $R_c$  is the volume mean radius of cloud droplets, which 62 is defined as  $q_c = (4\pi\rho_w/3\rho_a)R_c^3N_c$ ,  $R_{c0}$  is its threshold value,  $\rho_w$  and  $\rho_a$  are the density of 63 water and air, respectively, and H is the Heaviside step function. Wide value ranges are suggested 64 for m and n, i.e., m = 2.33-4.7 and n = 0.33-3.3. Accordingly, A varies greatly across different 65 schemes, and this often causes discrepancies of several orders of magnitude (Menon et al., 2003; 66 Wood, 2005; Hsieh et al., 2009). 67

A large number of previous works have attempted to clarify the roles of various processes 68 in autoconversion, such as broadening of DSD (Baker et al., 1980; Blyth, 1993; Cooper, 1989; 69 Lasher-Trapp et al., 2005) and turbulence-induced collision enhancement (TICE) (Franklin, 2008; 70 Pinsky & Khain, 2002; Wang & Grabowski, 2009). Attempts have been made to incorporate the 71 effects of these processes into the parameterization of autoconversion; for example, Seifert et al. 72 (2010), Franklin (2008), and Seifert and Onishi (2016) studied TICE, and Berry and Reinhardt 73 (1974), Liu and Daum (2004), and Milbrandt and Yau (2005a, b) examined the DSD. Wang and 74 Grabowski (2009) showed that TICE could help overcome the delayed initiation of precipitation. 75 Noh et al. (2018) showed that the effect of DSD could explain the so-called aging period problem 76 associated with rainwater production that occurs too early and too low in a cloud (Shipway & Hill, 77 2012; Straka & Rasmussen, 1997). 78

	$A = \alpha q_c^m N_c^{-n} \Phi$		
	т	п	Φ
Kessler (1969)	1	0	$H(q_c-q_{c0})$
Tripoli and Cotton (1980)	7/3	1/3	$H(R_c-R_{c0})$
Beheng (1994)	4.7	3.3	1
Khairoutdinov and Kogan (2000)	2.47	1.79	1
Kogan (2013)	4.22	3.01	1
Liu and Daum (2004)	3	1	$f(\sigma_c / R_c)H(R_6 - R_{60})$
Liu et al. (2007)	3	1	$f(\sigma_{c} / R_{c}, q_{c}^{-2} N_{c}^{3/2})$
Berry and Reinhardt (1974)	2	0	$(R_c^4 - c_1)(R_c - c_2)$
Seifert and Beheng (2001)	4	2	$ au^{0.68} (1 -  au^{0.68})^3$
Seifert et al. (2010)	4	2	$ au^{0.68}(1- au^{0.68})^3(1+barepsilon)$
			$b = \operatorname{Re}_{\lambda}^{1/4} f(\overline{r}_c)$

**Table 1** List of the parameterizations of A ( $R_6$  = the mean radius of the sixth moment of the droplets,  $\tau$  = internal time scale ( $q_r / q_l$ ), Re<sub> $\lambda$ </sub> = Taylor microscale Reynolds number,  $\overline{r_c}$  = mean radius of the cloud droplets, and  $c_1$ ,  $c_2$  = constants).

Considering the difficulty of obtaining reliable observation data, a widely used approach for evaluating cloud microphysics parameterizations is to analyze the results obtained from a box collision model, which calculates the evolution of the droplet spectrum induced by collision by solving the stochastic collection equation (SCE), used for a spectral bin model (SBM), based on a given initial DSD. For the initial DSD, observed DSD data (Hsieh et al., 2009; Wood, 2005) or an idealized DSD, such as a gamma or lognormal distribution (Berry & Rheinhardt, 1974; Franklin, 2008; Lee & Baik, 2017; Noh et al., 2018; Seifert & Beheng, 2001; Seifert et al., 2010) are used.

Many parameters used in cloud microphysics parameterizations, such as DSD and turbulence, are correlated with each other in real clouds. In a box collision model, however, it is difficult to consider these correlations, and thus these parameters are often regarded as independent. One way to cope with this problem is to use large eddy simulation (LES), in which the evolution of the DSD and the correlations between parameters are naturally reproduced during the evolution of clouds. Khairoutdinov and Kogan (2000) and Kogan (2013) analyzed data obtained from warm cloud fields simulated by LES coupled with an SBM for the parameterization of autoconversion,

- 96 but they did not consider the effects of DSD, turbulence, and correlations of parameters.
- Lagrangian cloud models (LCMs) have recently received much attention as a new type of 97 cloud model (e.g., Andrejczuk et al., 2008, 2010; Arabas et al., 2015; Dziekan et al., 2019; 98 Hoffmann et al., 2017; Riechelmann et al., 2012; Shima et al., 2009; Sölch & Kärcher, 2010). In 99 such a model, the flow field is simulated by LES, and droplets are treated as Lagrangian particles 100 that undergo cloud microphysics while interacting with the surrounding air. LCMs have been 101 shown to overcome many problems faced by existing Eulerian models, including numerical 102 diffusion (Grabowski et al., 2019; Hoffmann, 2016). LCMs have recently been used to successfully 103 investigate detailed microphysical processes such as aerosol-cloud interactions (Andrejczuk et al., 104 105 2010; Hoffmann et al., 2015) or precipitation processes (Arabas & Shima, 2013; Dziekan & Pawlowska, 2017; Hoffmann et al., 2017; Naumann & Seifert, 2016). 106
- 107 Recently, Noh et al. (2018) obtained information about *A* by analyzing a box collision 108 model, which used the collision scheme in the LCM. In this case, autoconversion was calculated 109 directly by capturing the moments of the conversions of individual Lagrangian droplets from cloud 110 droplets to raindrops. An analysis of the box collision model results, in which a lognormal DSD 111 was assumed, revealed the relation as
- 112

$$A = \alpha q_c^{7/3} N_c^{-1/3} H(R_c - R_{c0})$$
<sup>(2)</sup>

which agreed with the results of Tripoli and Cotton (1980). Contrary to Tripoli and Cotton (1980), however,  $\alpha$  increases linearly with the dissipation rate  $\varepsilon$  and the standard deviation of the DSD  $\sigma$ . Accordingly,  $\alpha$  was parameterized as

$$\alpha = a(\sigma - \sigma_0)(1 + b\varepsilon)H(\sigma - \sigma_0), \qquad (3)$$

with  $a = 1.0 \text{ cm}^{-1} \mu \text{m}^{-1} \text{ s}^{-1}$  and  $b = 8.8 \times 10^{-3} \text{ cm}^{-2} \text{ s}^{3}$ , where  $\sigma_{0}$  is the threshold value of  $\sigma$ . However, in this parameterization,  $\sigma$  and  $\varepsilon$  are regarded as independent of  $q_{c}$  and  $N_{c}$ .

In the present work, we investigate how the effects of DSD and turbulence on *A* appear in realistic field simulations of shallow cumulus clouds using the LCM, in which the parameters are correlated. We carry out an extensive analysis of the correlations between parameters (Sections 3.2, 3.3, and 3.5), and propose how the effects of DSD and turbulence can be parameterized based on it, which results in a new parameterization of *A* (Section 3.4). We also investigate how exponent values of  $q_c$  and  $N_c$  vary when *A* is parameterized only in terms of  $q_c$  and  $N_c$ , as is done in many existing parameterizations (Section 3.6). The new parameterization is compared with the LCM data (Section 4.1) and applied to a bulk cloud model (BCM) while clarifying the difference between the cloud droplet mixing processes of the LCM and BCM (Section 4.2). The roles of DSD and turbulence in the parameterization of *A* are investigated by comparing the results from *A* with and without these effects (Section 4.1 and 4.3).

#### 130 2 Model and Simulation

## 131 2.1 Lagrangian cloud models (LCM)

The employed LCM is the same as that used in Hoffmann et al. (2017) and Noh et al. 132 (2018). To handle an extremely large number of cloud droplets, the concept of a super-droplet, 133 134 which represents a large number of real droplets with identical features, is introduced. Each superdroplet moves under the influence of the turbulent flow field and the gravitational force while 135 experiencing condensational and collisional growth. The turbulent flow field is calculated by an 136 LES model, Parallelized Large-Eddy Simulation Model (PALM; Maronga et al., 2015; Raasch & 137 Schröter, 2001). The collision scheme follows the algorithm introduced by Shima et al. (2009), in 138 which collision occurs stochastically with the probability proportional to the collection kernel. The 139 collision scheme is known to reproduce the realistic evolution of a cloud droplet spectrum 140 (Unterstrasser et al., 2017). The collection kernel includes the effect of TICE (Ayala et al., 2008; 141 Wang & Grabowski, 2009). One can refer to Hoffmann et al. (2017) and Noh et al. (2018) for a 142 detailed explanation of the utilized LCM. 143

#### 144 **2.2 Bulk cloud model (BCM)**

A new parameterization of A is applied to a BCM. The BCM employs the two-moment scheme of Morrison et al. (2005), the extension of Seifert and Beheng (2001, 2006), which predicts  $q_c$ ,  $q_r$ ,  $N_c$ , and  $N_r$  based on the assumption that the DSD follows a gamma distribution. Here,  $q_r$  and  $N_r$  are the mixing ratio and number concentration of raindrops, respectively. However, our parameterizations of the activation and condensational growth processes are different from those of Seifert and Beheng (2006). The number of activated aerosols is calculated by following the parameterization developed by Khvorostyanov and Curry (2006) for the prediction of  $N_c$ , and the increase in  $N_c$  is calculated by the difference between the predicted and existing  $N_c$  values at each time step. Condensational growth is parameterized by following Khairoutdinov and Kogan (2000). The BCM is already included in the PALM code, so the LCM and the BCM share the same dynamical core of LES, which enables direct comparisons among the effects of different cloud microphysical processes of LCM and BCM.

#### 157 **2.3 Simulation**

We simulated shallow cumulus cloud fields based on the Barbados Oceanographic and 158 159 Meteorological Experiment (BOMEX; Holland & Rassmusson, 1973). The initial profiles followed the LES intercomparison study (Siebesma et al., 2003) (Figure 1). To obtain information 160 about autoconversion under various conditions, a large number of simulations were carried out 161 162 with different initial cloud droplet concentrations and surface heat and moisture fluxes. Four different initial droplet number concentrations,  $N_0 = 60, 80, 100, \text{ and } 120 \text{ cm}^{-3}$ , were simulated, 163 and these concentrations are referred to as N60, N80, N100, and N120, respectively. In addition 164 to simulations with the original surface heat and moisture fluxes used for the BOMEX simulation 165  $(\overline{w\theta}_0 = 8 \times 10^{-3} \text{ K m s}^{-1} \text{ and } \overline{wq}_0 = 5.2 \times 10^{-5} \text{ m s}^{-1}, \text{ respectively})$  (Siebesma et al., 2003), additional 166 simulations with surface heat and moisture fluxes enhanced by 1.5 times were also carried out 167  $(\overline{w\theta}_0 = 12 \times 10^{-3} \text{ K m s}^{-1}, \text{ and } \overline{wq}_0 = 7.8 \times 10^{-5} \text{ m s}^{-1}).$ 168





170 Figure 1 Initial environmental profiles for the BOMEX simulation.

In most simulations, the activation process, whose main contribution is to determine  $N_{c}$ , 171 was bypassed to facilitate a large number of simulations while focusing on the collision process. 172 The radii of all super-droplets were initially given by  $r = 0.01 \mu m$ , and the droplets were not 173 174 allowed to evaporate and become any smaller. A time step of  $\Delta t = 0.2$  s was used. Two simulations including the activation process with different aerosol concentrations ( $N_a = 120, 200 \text{ cm}^{-3}$ ) were 175 also carried out to examine its possible effect and to conduct a comparison with the bulk model 176 results. In this case, the initial distribution of aerosols followed the setting of Hoffmann et al. 177 (2015), which was based on Derksen et al. (2009), and condensational growth included the 178 179 contributions from curvature and solute effects with a much smaller time step.

The model grid spacing was set to 20 m in each direction, with a total of  $120 \times 120 \times 144$ grid points (2.4 km × 2.4 km × 2.88 km), to perform a large number of simulations of the computationally demanding LCM. We performed simulations with a larger domain size (4.8 km × 4.8 km × 2.88 km) and smaller grid size (10 m) for the N60 case to examine the model sensitivity (Appendix). This shows that the vertical profiles of variables and the general correlation patterns between parameters remained valid. Integration was carried out for 6 hours, and the data obtained over the last 3 hours were used for analysis. Initially, 100 super-droplets were assigned to a grid, as this value is known to be sufficient for properly representing collisional growth (Arabas & Shima, 2013; Riechelmann et al., 2012; Unterstrasser et al., 2017).

#### 190 **2.4 Autoconversion calculation**

We detected collision events between super-droplets at every time step. Every collision event was assigned to the autoconversion, accretion, or self-collection category, depending on the radii of the involved super-droplets before and after the collision (e.g., Noh et al., 2018). Only autoconversion was considered in the present work. The probability that autoconversion occurred at a single grid during one-time step ( $\Delta t = 0.2$  s) was very small ( $10^{-5} - 5 \times 10^{-4}$ ).

The mass increases in raindrops from every collision event in a grid were added to 196 calculate the average value of A under a given condition during the last 3 hours for all simulations 197 (8 without activation and 2 with activation). The total number of grid data was approximately 2 198 million. For the calculation of  $A(q_c, N_c, \sigma_c, \varepsilon)$ , from the total grid data obtained over the last 3 199 hours, we collected grid data in which the parameters  $q_c$ ,  $N_c$ ,  $\sigma_c$ , and  $\varepsilon$  belonged to the same 200 interval and regrouped them into a set  $\{q_c, N_c, \sigma_c, \varepsilon\}$ . Here, the standard deviation of the DSD of 201 cloud droplets  $\sigma_c$  was used instead of that of the total droplets  $\sigma$  because the DSD does not 202 follow an ideal distribution and raindrops do not affect autoconversion.  $A(q_c, N_c, \sigma_c, \varepsilon)$  was then 203 calculated by the average value of A over the grid data belonging to a given group  $\{q_c, N_c, \sigma_c, \varepsilon\}$ . 204 The numbers of intervals for  $q_c$ ,  $N_c$ ,  $\sigma_c$ , and  $\varepsilon$  for a group  $\{q_c, N_c, \sigma_c, \varepsilon\}$  were 50, 50, 50, and 205 50, respectively. 206

207 Regarding the separation radius between a cloud droplet and a raindrop,  $r^* = 25 \,\mu\text{m}$  was 208 used, as in Noh et al. (2018), but the case with  $r^* = 40 \,\mu\text{m}$  was also analyzed. In the present 209 simulations, autoconversion tended to be very small at  $r^* = 40 \,\mu\text{m}$  when  $N_0$  was large.

## 210 **3** Analysis of the LCM Results and a New parameterization

#### 211 **3.1 Evolution of clouds**

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Figure 2 shows the evolutions of the vertical profiles of horizontally averaged A,  $q_c$ , 212  $N_c / N_0$ ,  $\varepsilon$ ,  $\sigma_c$ , and  $R_c$  at N60, helping us to understand how various parameters contribute to 213 autoconversion and how they are related to each other. Here, A and  $q_c$  are calculated by horizontal 214 averaging as in previous results (e.g., Wyszogrodzki et al., 2013), but  $\sigma_c$ ,  $\varepsilon$ ,  $N_c/N_0$ , and  $R_c$  are 215 averaged only within clouds with  $q_c > 10^{-5}$  kg kg<sup>-1</sup>, as they are more relevant to autoconversion 216 generation.  $\varepsilon$  is calculated directly from the LES (e.g., Riechelmann et al., 2012). Hereafter, all 217 figures represent results obtained with the original surface heat and moisture fluxes unless 218 otherwise specified.  $q_c$  appears at the cloud base and increases in magnitude while moving upward. 219 Intermittently large values of A,  $q_c$ , and  $\varepsilon$  appear near the cloud top (e.g., t = 3, 4, 5.2 hours), 220 which suggests that raindrops are formed at the top of a strong thermal with undiluted or slightly 221 222 diluted cores (Khain et al. 2013).



Figure 2 Time series of the horizontally averaged variables at N60: (a) A, (b)  $q_c$ , (c)  $N_c / N_0$ , (d) 225  $\mathcal{E}$ , (e)  $\sigma_c$ , (f)  $R_c$  (Here  $N_c / N_0$ ,  $\mathcal{E}$ ,  $\sigma_c$ , and  $R_c$  are averaged only within clouds.)

226	Figure 3 shows the horizontal mean vertical profiles of the corresponding parameters (A,
227	$q_c$ , $N_c / N_0$ , $\varepsilon$ , $\sigma_c$ , and $R_c$ ), averaged over the last 3 hours of each simulation with different
228	values of $N_0$ . The profiles agree with previously obtained simulation results in general (Siebesma
229	et al., 2003; Wyszogrodzki et al., 2013). All $\sigma_c$ , $R_c$ and $\varepsilon$ increase with the height <i>z</i> within clouds,
230	and $\varepsilon$ becomes very large near the cloud top, as expected from Figure 2.
231	The stronger condensational growth at smaller $N_0$ values causes larger $\sigma_c$ and $R_c$
232	(Chandrakar et al., 2016; Grabowski & Wang, 2013). It is also found that the small increase in $\sigma_c$
233	and $R_c$ increases A greatly. On the other hand, $q_c$ , $\varepsilon$ and $N_c / N_0$ are less sensitive to $N_0$ , as they
234	are mainly controlled by cloud dynamics such as entrainment, mixing, and convection. Profiles of
235	the cloud fraction, which will be shown in Figure 13, is also insensitive to $N_0$ either. The increases
236	in $q_c$ , $\varepsilon$ , $\sigma_c$ , and $R_c$ with z and the decreases in $\sigma_c$ , $R_c$ , and A with $N_0$ suggest the presence of
237	correlations between these parameters.





Figure 3 Vertical profiles of the horizontal mean variables averaged over the last 3 hours of 240 simulations (blue: N60, green: N80, orange: N100, red: N120): (a) A, (b)  $q_c$ , (c)  $N_c / N_0$ , 241 (d)  $\mathcal{E}$ , (e)  $\sigma_c$ , (f)  $R_c$  (Here  $N_c / N_0$ ,  $\mathcal{E}$ ,  $\sigma_c$ , and  $R_c$  are averaged only within clouds.) 242

## 245 **3.2 Relation between** $\sigma_c$ and $R_c$

It is well known that the width of DSD tends to increase with the mean size of cloud 246 droplets (Chandrakar et al., 2016; Cooper, 1989; Lasher-Trapp et al., 2005; Martin et al., 1994; 247 Politovich, 1993; Shaw, 2003). Figures 2 and 3 also show that  $R_c$  and  $\sigma_c$  both increase with 248 height. The broadening of the DSD may be caused by the different condensational growth histories 249 of various droplets in the turbulent environment of a cloud affected by entrainment (Cooper, 1989; 250 Lasher-Trapp et al., 2005; Luo et al., 2020; Shaw, 2003) or by the presence of collisional growth 251 (Lu & Seinfeld, 2006). 252 Figure 4 shows the variations in the average standard deviations of the DSD for a given 253 volume mean radius both for whole droplets ( $R_v$ ,  $\sigma$ ) and for cloud droplets smaller than  $r^* = 25$ 254  $\mu m (R_c, \sigma_c)$ . When  $R_{\nu}$  is small ( $R_{\nu} < 12 \mu m$ ),  $\sigma$  increases linearly with  $R_{\nu}$ , while  $\sigma / R_{\nu}$  ( $\simeq 0.3$ ) 255 is insensitive to  $N_0$  (Figure 4a). Here,  $\sigma/R_v \approx 0.3$  is consistent with the values obtained from 256 observation data (0.25 - 0.45), although the integral radius is used instead of  $R_{\nu}$  in those cases 257 (Martin et al., 1994; Pawlowska et al., 2006; Politovich, 1993; Zhao et al., 2006). However,  $\sigma$ 258 starts to grow more slowly with  $R_{\nu}$  at larger values of  $R_{\nu}$  ( $R_{\nu} > 12$  µm), and ultimately,  $\sigma$ 259 decreases with  $R_{\nu}$ . In this case, when  $N_0$  is larger, the decrease in  $\sigma/R_{\nu}$  starts earlier, and  $\sigma$ 260

261 becomes smaller.

The decrease in  $\sigma/R_{\nu}$  with  $N_0$  is consistent with the findings of many previous reports (Chandrakar et al., 2016; Lu & Seinfeld, 2006; Martins & Silva Dias, 2009; Miles et al., 2000). The mass density distribution of droplets shows that the enhanced broadening of the DSD at lower  $N_0$  is mainly attributed to the presence of stronger collisional growth (Figure 5), which is induced by stronger condensational growth.





Figure 4 Variations in the mean standard deviation of the DSD with the volume mean radius (blue: N60, green: N80, orange: N100, and red: N120): (a)  $\sigma$  with  $R_{\nu}$  (whole droplets), (b)  $\sigma_c$  with  $R_c$  (only cloud droplets)

The decrease of  $\sigma$  with  $R_{\nu}$  at a large  $R_{\nu}$  value is related to the fact that droplets in the cloud core that are unaffected by entrainment tend to have larger  $R_{\nu}$  values but smaller  $\sigma$  values, while droplets near the cloud edge that are affected by entrainment tend to have smaller  $R_{\nu}$  values but larger  $\sigma$  values (Burnet & Brenguier, 2007; Hoffmann et al., 2017; Lasher-Trapp et al., 2005; Lu et al., 2013; Pardo et al., 2020; Tölle & Krueger, 2014).

When only cloud droplets are considered (Figure 4b), the variation pattern of  $\sigma_c$  with  $R_c$ is similar, but the decrease in  $\sigma_c$  with  $R_c$  starts earlier, thus decreasing the maximum  $\sigma_c$ . This phenomenon is due to the limiting effect induced by the threshold radius. That is,  $\sigma_c$  must approach zero as  $R_c$  approaches  $r^*$  because only droplets smaller than  $r^*$  are considered for the calculation of  $\sigma_c$ .





Figure 5 Mass density distributions of droplets (blue: N60, green: N80, orange: N100, and red: N120). A dashed vertical line represents  $r^* = 25 \,\mu\text{m}$ . The distributions are obtained from the data within clouds during the last 3 hours of simulations.

## 287 **3.3 Variations in** $\alpha$ with $\sigma_c$ , $R_c$ , $\varepsilon$ , and $N_c$

Noh et al. (2018) showed that the relation  $A \propto q_c^{7/3} N_c^{-1/3}$  is observed for given DSD and 288 turbulence from an analysis of the box collision model results, as given by (2) (Figure 7 in Noh et 289 al. (2018)). This relation implies the collisional growth of larger cloud droplets with  $R_c$  colliding 290 gravitationally with smaller cloud droplets with no terminal velocity ( $V_T \simeq 0 \text{ m s}^{-1}$ ). The relation 291  $A \propto q_c^{7/3} N_c^{-1/3}$  is obtained from  $A \sim K N_c q_c$ ,  $K \sim E R_c^2 V_T(R_c)$ , and  $V_T(R_c) \propto R_c^2$  following Stokes' 292 law, if the collection efficiency E can be represented by the mean value (e.g., Liu & Daum, 2004; 293 Tripoli & Cotton, 1980). Accordingly, we assume that the relation (2) remains valid for given DSD 294 and turbulence, while  $\alpha$  varies with DSD and turbulence, as in Noh et al. (2018). However, the 295 correlations of DSD and turbulence with cloud parameters  $q_c$  and  $N_c$  can modify (3). Figure 5 296 also shows that the general pattern of the mass density distribution, which can be idealized by the 297 lognormal or gamma distribution, is maintained regardless of the value of  $N_0$  for cloud droplets. 298

Figure 6 shows the variations of  $\alpha$  in (2) with  $\sigma_c$  and  $\varepsilon$  at different values of  $N_0$  (= 60, 100 cm<sup>-3</sup>), which is calculated by  $A/(q_c^{7/3}N_c^{-1/3})$ . The results show that  $\alpha$  increases with  $\sigma_c$  and  $\varepsilon$ , as expected from (3). However,  $\alpha$  is much larger at smaller  $N_0$  values, as expected from Figure 3. The dependence on  $N_0$  also represents the dependence on  $N_c$  since  $N_c/N_0$  is almost independent of  $N_0$  (Figure 3e). Figure 4 shows that  $R_c$  is larger for a given  $\sigma_c$  at  $R_c > 12 \ \mu m$ when  $N_0$  is smaller. Therefore, a larger  $\alpha$  at a smaller  $N_0$  may suggest that  $\alpha$  becomes larger at larger  $R_c$ , even if  $\sigma_c$  is the same.



Figure 6 Scatter plots of  $\alpha$  vs.  $\sigma_c$  (violet:  $0 \text{ cm}^2 \text{ s}^{-3} \leq \varepsilon < 20 \text{ cm}^2 \text{ s}^{-3}$ , blue:  $20 \text{ cm}^2 \text{ s}^{-3} \leq \varepsilon < 40 \text{ cm}^2 \text{ s}^{-3}$ , green:  $40 \text{ cm}^2 \text{ s}^{-3} \leq \varepsilon < 60 \text{ cm}^2 \text{ s}^{-3}$ , orange:  $60 \text{ cm}^2 \text{ s}^{-3} \leq \varepsilon < 80 \text{ cm}^2 \text{ s}^{-3}$ , red:  $80 \text{ cm}^2 \text{ s}^{-3} \leq \varepsilon < 100 \text{ cm}^2 \text{ s}^{-3}$ ). The means (circles) and standard deviations (crosses) of  $\alpha$  vs.  $\sigma_c$  are included: (a) N60, (b) N100.

The present result reveals that  $\alpha$  depends on both  $R_c$  and  $\sigma_c$ , although the dependence on  $R_c$  was not considered in Noh et al. (2018). It implies that the variation of  $q_c$  and  $N_c$  with constant  $R_c$  gives  $A \propto q_c^{7/3} N_c^{-1/3}$ , but the tendency of increasing *E* with  $R_c$  (e.g., Long, 1974) can cause the increase of  $\alpha$  with  $R_c$ . The effect of DSD thus means the effects of both  $R_c$  and  $\sigma_c$ hereafter in the present paper. In this case, the effect of DSD can also be represented in terms of  $R_c$  and  $N_c$  since  $\sigma_c / R_c$  varies with  $N_c$  (Figure 4). A parameterization in terms of  $\sigma_c$  appears somewhat unnatural, considering that two different groups of data with small and large values of  $R_c$  represent the same  $\sigma_c$  in Figure 4. More importantly, information about  $\sigma_c$  is not available from the atmospheric models. Therefore, it is more desirable to parameterize the effect of DSD in terms of  $R_c$  and  $N_c$ .

Figure 7 shows the variations in  $\alpha$  with  $R_c$  and  $\varepsilon$  at different values of  $N_0$ corresponding to Figure 6.  $\alpha$  increases with  $R_c$ , starting at a certain value of  $R_c$  (=  $R_{c0}$ ), and  $\alpha$ is larger at smaller values of  $N_0$ , similar to Figure 6. This result also reflects the fact that  $\sigma_c$  is larger for the same  $R_c$  value ( $R_c > 12 \mu$ m) when  $N_0$  is smaller (Figure 4). The values of  $R_{c0}$  are consistent with those used in Tripoli and Cotton (1980) ( $R_{c0} = 7 - 10 \mu$ m).



327

Figure 7 Scatter plots of  $\alpha$  vs.  $R_c$  (violet:  $0 \text{ cm}^2 \text{ s}^{-3} \le \varepsilon < 20 \text{ cm}^2 \text{ s}^{-3}$ , blue:  $20 \text{ cm}^2 \text{ s}^{-3} \le \varepsilon < 40 \text{ cm}^2 \text{ s}^{-3}$ , green:  $40 \text{ cm}^2 \text{ s}^{-3} \le \varepsilon < 60 \text{ cm}^2 \text{ s}^{-3}$ , orange:  $60 \text{ cm}^2 \text{ s}^{-3} \le \varepsilon < 80 \text{ cm}^2 \text{ s}^{-3}$ , red:  $80 \text{ cm}^2 \text{ s}^{-3} \le \varepsilon < 100 \text{ cm}^2 \text{ s}^{-3}$ ). The means (circles) and standard deviations (crosses) of  $\alpha$  vs.  $R_c$  are included. A dashed violet line represents the variation in the mean  $\alpha$  with  $R_c$  for  $0 \text{ cm}^2 \text{ s}^{-3} \le \varepsilon < 20 \text{ cm}^2 \text{ s}^{-3}$ : (a) N60, (b) N100.

It is important to mention that each data point in Figure 6 and 7 represents the average value of a set  $\{q_c, N_c, \sigma_c, \varepsilon\}$ , not the value at every grid, as explained in Section 2.4. On the other hand, the probability distribution functions of data show that the data with the radius near the mean value of  $R_c$  dominate for a given  $\varepsilon$  range (Figure 8). It means that most data are located within a

range represented by the mean and standard deviation of  $R_c$  in Figure 7. Furthermore, large 337 variations of  $\alpha$  in Figure 6 and 7 reflect the fact that the characteristic of DSD is different 338 depending on the location; that is,  $\sigma_c / R_c$  is large in the cloud core but small near the cloud edge 339 affected by entrainment, as discussed in the previous section. For example, a small portion of data 340 that shows large  $\alpha$  at small  $R_c$  may represent the data from the cloud edge with large  $\sigma_c / R_c$ . 341 However, Figure 8 shows that the contribution of scattered data away from the mean  $R_c$  to A is 342 small. It should also be mentioned that  $\alpha$  varies with  $\varepsilon$  significantly even within the same color 343 range of  $\varepsilon$  in Figure 7. 344





![](_page_19_Figure_4.jpeg)

Figure 8 Probability distribution function of the data with  $R_c$  and  $\varepsilon$  at N60 (violet: 0 cm<sup>2</sup> s<sup>-3</sup>  $\leq \varepsilon < 20$  cm<sup>2</sup> s<sup>-3</sup>, blue: 20 cm<sup>2</sup> s<sup>-3</sup>  $\leq \varepsilon < 40$  cm<sup>2</sup> s<sup>-3</sup>, green: 40 cm<sup>2</sup> s<sup>-3</sup>  $\leq \varepsilon < 60$  cm<sup>2</sup> s<sup>-3</sup>,

349 orange: 60 cm<sup>2</sup> s<sup>-3</sup>  $\leq \varepsilon$  < 80 cm<sup>2</sup> s<sup>-3</sup>, red: 80 cm<sup>2</sup> s<sup>-3</sup>  $\leq \varepsilon$  < 100 cm<sup>2</sup> s<sup>-3</sup>).

350

#### 351 **3.4 A new parameterization of** *A*

Figure 7 shows the tendency of  $\alpha$  to increase with  $R_c$  and  $\varepsilon$  but decrease with  $N_0$ . In particular,  $\alpha$  increases linearly with  $R_c - R_{c0}$  when the turbulence is weak (0 cm<sup>2</sup> s<sup>-3</sup>  $\leq \varepsilon < 20$   $cm^2 s^{-3}$ ), while its slope decreases with  $N_0$ . Furthermore, Figure 4 suggests that  $\sigma_c / R_c$  becomes smaller at larger  $N_0$  values in the range  $R_c > 12 \mu m$ , and this ratio dominates the contribution of the parameters to autoconversion. Based on these results, we can express the effect of DSD on  $\alpha$ as  $N_c^{-X}(R_c - R_{c0})$ , instead of  $(\sigma - \sigma_0)$  in (3), by using the proportionality of  $N_c$  to  $N_0$  (Figure 3e).

359

The relation in (3) can then be rewritten as

$$\alpha = a N_c^{-X} (R_c - R_{c0}) (1 + b\varepsilon) H(R_c - R_{c0}) .$$
(5)

The best fit of the data, shown in Figure 7, provides X = 1.0, a = 8.0, b = 0.22 cm<sup>-2</sup> s<sup>3</sup> and  $R_{c0} = 9$ 361  $\mu$ m. Table 2 shows the optimal values of constants used in the parameterization (6) for both  $r^*$  = 362 25 and 40  $\mu$ m. To obtain optimal coefficients, we first calculate  $\alpha(q_c, N_c, \varepsilon)$  from the dataset 363  $\{A, q_c, N_c, \varepsilon\}$  from all simulations, and regression analysis is carried out for  $R_c - R_{c0}$  and 364  $(R_c - R_{c0})\varepsilon$  with varying X and  $R_{c0}$ . The correlation of  $\varepsilon$  with  $q_c$  and  $N_c$  (or  $R_c$ ), which will be 365 discussed in the next section, is not parameterized here, because the information of  $\varepsilon$  can be 366 obtained from the atmospheric models; especially, it can be obtained directly in LES (e.g., Heath 367 et al., 2017; Huang et al., 2019). 368

369

Combining (5) with (2) results in the following:

370  $A = aq_c^{7/3} N_c^{-(1/3+X)} (R_c - R_{c0}) (1 + b\varepsilon) H(R_c - R_{c0}).$ 

Noh et al. (2018) proposed the parameterization of the threshold radius  $R_{c0}$  in (2) as a function of  $\sigma$  based on the result of a box collision model. This parameterization cannot be applied anymore since  $\sigma_c$  and  $R_c$  are closely related, as shown in Figure 4. Instead, the threshold

(6)

374  $R_{c0}$  is included naturally in (6).

One important implication of the present result is that the dependence of the parameterization of A on  $q_c$  and  $N_c$  is generated via two different mechanisms. One derives from the collisional process based on gravitational collision for a given DSD, and the other is from the effects of DSD and turbulence and their correlations with  $q_c$  and  $N_c$  in real clouds. The box collision model results cannot provide information about the latter in general. Furthermore, one can also expect that exponential coefficients m and n in (1), as shown in Table 1, can differ depending on whether the effects of DSD and turbulence are considered explicitly or not.

<i>r</i> <sup>*</sup> (µm)	X	а	b	<i>R</i> <sub>c0</sub> (µm)
25	1.0	8.0	0.220	9
40	1.15	1.95	0.514	10

Table 2 Values of *X*, *a*, *b* and  $R_{c0}$  obtained via multiple regression analyses at  $r^* = 25$  and 40 µm.

## 385 **3.5 Relation between** $\varepsilon$ and $\sigma_c$

Another important feature in (5) is that *b* is substantially larger than previous estimations based on box collision model results (Franklin, 2008; Noh et al., 2018; Onishi et al., 2015; Seifert et al., 2010); for example,  $b = 8.8 \times 10^{-3}$  cm<sup>-2</sup> s<sup>3</sup> (Noh et al., 2018) or  $1.5 \times 10^{-3}$  cm<sup>-2</sup> s<sup>3</sup> (Onishi et al., 2015).

Figure 9 shows that large values of  $\varepsilon$  are preferentially located in a region with larger  $\sigma_c$ 390 and  $R_c$  values and in a region with larger  $\sigma_c$  values when  $R_c$  is held constant. This may reflect 391 the fact that near the cloud top, where  $\varepsilon$  is larger, entrainment tends to increase  $\sigma_c$ . As a result, 392 the effect of turbulence in (3) should be expressed as  $(1+b_0\varepsilon)(1+b_1\varepsilon)$ , where  $1+b_0\varepsilon$  represents 393 the effect of TICE and  $1+b_1\varepsilon$  represents the effect of the preferential distribution of large  $\varepsilon$  values, 394 i.e.,  $\sigma_c$  tends to be larger at larger  $\varepsilon$  for a given  $R_c$ . As the simplest expression,  $(1+b_0\varepsilon)(1+b_1\varepsilon)$ 395 can be rewritten as  $1+b\varepsilon$  with  $b > b_0$ . In the box collision model, in which there are no 396 correlations between  $\varepsilon$ ,  $\sigma_c$ , and  $R_c$ , the increase of A induced by  $\varepsilon$  represents only the effect of 397 398 TICE.

The enhancement factor  $b_1$  may vary with  $R_c$ , as suggested in Figure 9. Consequently, the relation between  $\alpha$  and  $R_c - R_{c0}$  is no longer linear at large values of  $\varepsilon$ , as suggested in Figure 7. Nonetheless, the variation of *b* with  $R_c$  is neglected for simplicity in the present parameterization because the data are concentrated in a narrow  $R_c$  range at large  $\varepsilon$  (Figure 8) and  $\sigma_c$  varies widely for a given  $R_c$  (Figure 9).

![](_page_22_Figure_1.jpeg)

404

405 Figure 9 A scatter plot of  $\sigma_c$  vs.  $R_c$  at N60 (violet:  $0 \text{ cm}^2 \text{ s}^{-3} \le \varepsilon < 20 \text{ cm}^2 \text{ s}^{-3}$ , blue:  $20 \text{ cm}^2 \text{ s}^{-3}$ 406  $\le \varepsilon < 40 \text{ cm}^2 \text{ s}^{-3}$ , green:  $40 \text{ cm}^2 \text{ s}^{-3} \le \varepsilon < 60 \text{ cm}^2 \text{ s}^{-3}$ , orange:  $60 \text{ cm}^2 \text{ s}^{-3} \le \varepsilon < 80 \text{ cm}^2$ 407  $\text{s}^{-3}$ , red:  $80 \text{ cm}^2 \text{ s}^{-3} \le \varepsilon < 100 \text{ cm}^2 \text{ s}^{-3}$ ).

# 409 **3.6 Effects of DSD and turbulence on the dependence of** A **on** $q_c$ **and** $N_c$

In many parameterizations, *A* is expressed only in terms of  $q_c$  and  $N_c$  (e.g., Beheng, 1994; Khairoutdinov & Kogan, 2000; Kogan, 2013; Liu & Daum, 2004; Tripoli & Cotton, 1980) (Table 1). The present result shows that *m* and *n* in (1) include the effects of DSD and turbulence, which, in turn, are affected by  $q_c$  and  $N_c$ . In particular,  $\sigma_c$  and  $R_c$  tend to decrease with  $N_c$  (Figure 4), and  $\varepsilon$  tends to increase with  $q_c$  (Figure 10). Consequently, *m* and *n* cannot be constant, but they should vary depending on the characteristics of the given cloud if *A* is expressed in terms of only  $q_c$  and  $N_c$ .

![](_page_23_Figure_1.jpeg)

![](_page_23_Figure_2.jpeg)

Figure 10 Probability distribution of the data in the  $\varepsilon - q_c$  domain at N60 ( $\Delta \log \varepsilon = 3.74 \times 10^{-2}$ cm<sup>2</sup> s<sup>-3</sup>,  $\Delta \log q_c = 1.72 \times 10^{-2}$  kg kg<sup>-1</sup>)

Figure 11 shows the variations in *m* and *n* in the ranges 3 < m < 4 and 2 < n < 4, respectively, when *A* is parameterized only in terms of  $q_c$  and  $N_c$ , i.e.,  $A \propto q_c^m N_c^{-n}$ , using the least-squares method, as in Khairoutdinov and Kogan (2000). Here, we also include the results obtained regarding the surface heat and moisture fluxes enhanced by 1.5 times, in which the clouds develop to higher altitudes and result in larger  $\varepsilon$ ,  $q_c$ ,  $q_r$ , and  $R_c$  values. Figure 11 may explain the large variations in *m* and *n* (Table 1).

The parameterization given by (6) suggests that *m* and *n* in (1) should be larger than m =7/3 and n = 1/3, respectively, when *A* is parameterized only in terms of  $q_c$  and  $N_c$  (e.g., Beheng (1994), Kogan (2013) in Table 1). The decrease in  $\sigma_c$  with increasing  $N_c$  (Figure 4) and the proportionality of  $N_c$  to  $N_0$  (Figure 3e) make *n* larger than 1/3. The increase in  $\varepsilon$  with increasing  $q_c$  (Figure 10) makes *m* larger than 7/3. Figure 11 also suggests that *n* tends to be larger at higher aerosol concentrations and smaller surface fluxes because a smaller  $R_c - R_{c0}$  in (6) is more sensitive to the variation in  $N_c$ .

![](_page_24_Figure_1.jpeg)

![](_page_24_Figure_2.jpeg)

Figure 11 Variations in m and n with  $N_0$  (blue: original surface heat and moisture fluxes; red: enhanced (1.5 times) surface heat and moisture fluxes).

#### 437 **4** Evaluation of the New Parameterization and the Effects of DSD and Turbulence

## 438 4.1 Comparison of the parameterizations of A with LCM data

Figure 12 compares the new parameterizations (NP) of A (6) with LCM data. To clarify 439 the effects of DSD and turbulence on the parameterization of A, we also examine the cases in 440 441 which their contributions are excluded in the parameterization; that is, constant values are used instead of  $N_c^{-X}(R_c - R_{c0})$  and  $(1 + b\varepsilon)$  in (6), referred to as XDSD and XTURB, respectively. 442 Here, the constant values are determined based on the averages of their corresponding in-cloud 443 values. Furthermore, we include the parameterization obtained from the box collision model, in 444 which there exist no correlations between the parameters  $\varepsilon$ ,  $\sigma_c$ , and  $N_c$  (Noh et al., 2018), which 445 is referred to as XCOR. 446

Since collisions occur stochastically with a probability that is proportional to the collection kernel in the LCM, only subgroups  $\{q_c, N_c, \varepsilon\}$  with sufficiently large numbers of grid data have statistical significance. The criterion regarding the minimum number of grid data is set to 500 in Figure 12 and Table 3. All data obtained from the simulations with and without the activation process are analyzed together since the difference is not significant compared to the differences among individual simulations. Note that the effect of the activation process is mainly limited to the determination of  $N_c$ .

NP shows a good correlation, which confirms that the formula and optimized constant 454 values given by (6) indeed represent the LCM data properly. XDSD and XTURB result in more 455 scattering and weaker correlation compared to NP (Table 3, Figure 12). In particular, they cannot 456 produce large values of A (>  $10^{-6}$  kg kg<sup>-1</sup>s<sup>-1</sup>), which represent intermittent bursts of raindrop 457 formation near the cloud top under the influence of large  $R_c$ ,  $\sigma_c$ , and  $\varepsilon$  (Figure 2). This illustrates 458 the importance of the inclusion of the effects of DSD and turbulence on the parameterization of A. 459 XCOR cannot reproduce large values of A either; this is partially attributed to the underestimation 460 of b, which does not consider the correlation between turbulence and DSD. 461 462

![](_page_25_Figure_3.jpeg)

Figure 12 Probability distribution of A from the LCM data vs. bulk parameterizations ( $r^* = 25$ 465 µm): (a) NP, (b) XTURB, (c) XDSD, (d) XCOR.

	NP	XTURB	XDSD	XCOR
RMSE	4 52	7 88	7 28	6.00
$(10^{-8} \text{ kg kg}^{-1} \text{ s}^{-1})$	4.52	/.00	1.20	0.90
correlation	0.77	0.59	0.58	0.53
coefficient				

466 **Table 3** The root mean square errors (RMSEs) and correlation coefficients obtained from NP,
467 XTURB, XDSD, and XCOR.

### 469 **4.2 Application of the new parameterization to a BCM**

It is important to understand how the parameterization of *A* affects the BCM results when it is applied to it. Figure 13 compares the profiles of various horizontal mean variables obtained from the LCM and the BCM with the new parameterization of *A*, similar to Figure 3. Additionally, the cases without the effects of DSD and turbulence (XDSD and XTURB, respectively) are included. Here, both the LCM and BCM simulations include the activation process that starts with an aerosol concentration  $N_a = 120$  cm<sup>-3</sup>.

Figure 13 shows that the differences between the LCM and BCM results appear not only 476 in the profiles of A but also in various other profiles. Here, A,  $q_c$ , and cloud fraction are calculated 477 478 by horizontal averaging, and  $\sigma_c$ ,  $\varepsilon$ ,  $N_c/N_0$ , and  $R_c$  are averaged only within clouds, similar to Figure 3. In the BCM,  $N_c$  does not decrease with z, and  $q_c$  does not increase near the cloud top; 479 these findings are in contrast with the profiles obtained from the LCM. The contrast in the profiles 480 of  $q_c$  and  $N_c$  agrees with the previous comparison of the LCM and BCM results conducted by 481 Sato et al. (2018) for shallow cumulus cloud field simulations. This difference is attributed to the 482 excessive turbulent diffusion of  $q_c$  and  $N_c$  across the cloud edge in the BCM, as suggested by 483 Sato et al. (2018), causing a substantial loss of cloud droplets (or  $q_c$  and  $N_c$ ) from the cloud in 484 the BCM. This excessive loss of cloud droplets also eliminates the peak of  $q_c$  near the cloud top. 485 However, the loss of  $N_c$  is immediately restored by the activation scheme in the BCM, in which 486 the increase in  $N_c$  is calculated by the difference between the predicted and existing  $N_c$  values at 487

every time step. This results in the overestimation of activation in the BCM, as pointed out by Hoffmann (2016). The LCM results show that the in-cloud activation is negligible in the present simulation (Hoffmann et al., 2015). Slawinska et al. (2012) also showed that in a BCM,  $N_c$ decreases as the height increases without activation above the cloud base, but it remains uniform with activation in shallow cumulus clouds.

As a result, the BCM makes  $N_c$  larger and  $q_c$  smaller as the height increases, and this 493 leads to a slower increase in  $R_c$  with height than that provided by the LCM. The underestimation 494 of  $q_c$  and  $R_c$  and the overestimation of  $N_c$  in the BCM cause the underestimation of A. Sato et al. 495 (2018) showed that raindrops are formed from the LCM but not from the BCM. This suggests that 496 the parameterization of the activation and condensation processes may require modification to 497 match the LCM results, but this task is beyond the scope of the present paper. This fact also makes 498 it difficult to evaluate the performance of the parameterizations of A. In this paper, we increase the 499 proportional constant a in (6) by a factor of 2 as a way to cope with this difference, and the main 500 focus is given to how the effects of DSD and turbulence in a parameterization affect the output 501 result. 502

Figure 13 also shows that raindrop formation starts at heights that are too low in the BCM results. Too early onset of precipitation is also observed in the BCM compared to the SBM (Cotton & Anthes, 1989; Khain et al., 2015; Shipway & Hill, 2012; Straka & Rasmussen, 1997). This may also reflect the limitation of the BCM, in which *A* is calculated based on the mean values of parameters.

![](_page_28_Figure_2.jpeg)

Figure 13 Vertical profiles of the horizontal mean variables averaged over the last 3 hours of simulations (black: LCM, red: BCM-NP, blue: BCM-XTURB, green: BCM-XDSD): (a) A, (b)  $q_c$ , (c)  $N_c$ , (d)  $\varepsilon$ , (e) cloud fraction, (f)  $R_c$  (Here  $N_c$ ,  $\varepsilon$ , and  $R_c$  are averaged only within clouds.)

## 515 4.3 Effects of DSD and turbulence in a BCM

Figure 13 shows that A becomes much weaker when the effects of DSD and turbulence 516 (XDSD and XTURB, respectively) are not included in the parameterization of A, although  $q_c$  and 517  $R_c$  are almost the same (or even slightly larger in the case of  $q_c$ ). The effects of DSD and 518 turbulence can be more clearly identified in the time series of  $q_c$  and A (Figure 14). It is difficult 519 to reproduce intermittent bursts of raindrop formation with large values of A, as shown in Figure 520 2, without the effects of DSD or turbulence in the parameterization of A (XDSD, XTURB) (Table 521 4). This result is consistent with that shown in Figure 12, in which large values of  $A \ge 10^{-6} \text{ kg kg}^{-1}$ 522 <sup>1</sup>s<sup>-1</sup>) cannot be reproduced in XDSD and XTURB. Figure 14 illustrates an important role of DSD 523 and turbulence in the raindrop formation in shallow cumulus clouds, as shown by Hoffmann et al. 524 (2017). 525

![](_page_29_Figure_3.jpeg)

526

Figure 14 Time series of the vertically integrated (a)  $q_c$ , (b) A (black: LCM, red: BCM-NP, blue: BCM-XTURB, green: BCM-XDSD).

	$q_c~(10^8{ m kg})$	A (10 <sup>4</sup> kg)	$A (> 20 \text{ kg s}^{-1}) (10^4 \text{ kg})$
LCM	1.49	2.16	1.82
NP	1.29	1.57	1.14
XTURB	1.15	0.56	0.01
XDSD	1.22	0.77	0.34

530 **Table 4** Accumulated values of  $q_c$ , A, and  $A(> 20 \text{ kg s}^{-1})$ .

531

## 532 5 Conclusions

In the present work, we investigated the roles of DSD and turbulence in *A* by analyzing the LCM data of shallow cumulus clouds. This goal was accomplished through the following steps while producing many results that have important implications for cloud microphysics.

First, the correlations of DSD and turbulence with cloud parameters  $q_c$  and  $N_c$  (or  $R_c$ ) are investigated, and the results are applied to parameterize their effects in A. The physical meaning of a new parameterization of A is discussed.

• A new parameterization of A is developed as  $A = \alpha q_c^{7/3} N_c^{-1/3} H(R_c - R_{c0})$  with 539  $\alpha = aN_c^{-X}(R_c - R_{c0})(1 + b\varepsilon)$ , where  $A \propto q_c^{7/3}N_c^{-1/3}$  represents the collisional process based on 540 gravitational collision for a given DSD and turbulence, and  $\alpha$  represents the effects of DSD 541 and turbulence  $(N_c^{-X}(R_c - R_{c0}))$  and  $(1 + b\varepsilon)$ , respectively). It reveals that the dependence of A 542 on  $q_c$  and  $N_c$  is generated via these two different mechanisms. Analysis of a box collision 543 544 model may not include the latter mechanism in general. The correlation of  $\varepsilon$  with  $q_c$  and  $N_c$ (or  $R_c$ ) is not parameterized here, because the information of  $\varepsilon$  can be obtained from the 545 atmospheric models. 546

• Both  $R_c$  and  $\sigma_c$  are found to affect  $\alpha$ . The effect of DSD can be represented in terms of  $R_c$ and  $N_c$  since  $\sigma_c / R_c$  varies with  $N_c$ .  $\alpha$  increases linearly with  $R_c - R_{c0}$  when turbulence is weak, while the slope decreases with  $N_0$  because of smaller  $\sigma_c / R_c$ . Based on this, the amplification effect of DSD on A is parameterized as  $N_c^{-X}(R_c - R_{c0})$  with X = 1.0 and  $R_{c0} = 9$ um.

- Large values of  $\varepsilon$  are preferentially located in a region affected by entrainment, in which  $\sigma_c$
- tends to be large for a given  $R_c$ . This makes b in the amplification effect of turbulence on A,
- 554  $(1+b\varepsilon)$ , much larger than that estimated from TICE.
- The effects of DSD and turbulence and their correlations with  $q_c$  and  $N_c$  explain a wide range of exponent values of  $q_c$  and  $N_c$  in many existing parameterizations of A.

557 Second, the new parameterization is compared with the LCM data, and applied to a bulk 558 cloud model (BDM) while clarifying the difference between the cloud droplet mixing processes 559 of the LCM and BCM. The roles of DSD and turbulence in the parameterization of *A* are 560 investigated by comparing the results from *A* with and without these effects.

- The new parameterization of *A* is shown to exhibit a good correlation with LCM data. The parameterizations without the effects of DSD and turbulence show a lower correlation, and cannot produce large values of *A*.
- It is found from the comparison of the LCM and BCM results that the overestimation of the turbulent diffusion of q<sub>c</sub> and N<sub>c</sub> across the cloud edge in the BCM causes the overestimation of N<sub>c</sub> due to excessive activation and the underestimation of q<sub>c</sub> and R<sub>c</sub>. This makes autoconversion more difficult in the BCM than in the LCM.

An intermittent burst of raindrop formation with a large value of A can be reproduced by the
 new parameterization of A, but it cannot be reproduced without the inclusion of the effects of
 DSD and turbulence. It illustrates an important role of DSD and turbulence in the raindrop
 formation in shallow cumulus clouds.

Finally, it should be mentioned that the present work is based on the LCM results of 572 shallows cumulus clouds. The correlation between parameters may vary depending on the given 573 cloud conditions, and therefore, constant values in the parameterization  $(X, a, b, \text{ and } R_{c0})$ , which 574 are obtained for shallow cumulus clouds in the present study, can be modified for other clouds 575 such as stratocumulus clouds and deep convective clouds. The comparison between the LCM and 576 BCM results suggests that the optimization of the parameterization of A must be carried out in 577 combination with the optimizations of other parameterizations, such as activation and 578 condensation, because of the fundamental differences between cloud droplet mixing processes. 579

It also suggests the necessity for further comparison and evaluation of cloud microphysicsreproduced by the LCM and SBM.

582

## 583 Appendix

To examine the validity of the LCM data used in the present study, it is necessary to 584 investigate the sensitivities of the domain size and grid resolution in the LCM simulations. Figure 585 A1 compares the vertical profiles of variables from the LCM simulations, corresponding to Figure 586 3, with a double horizontal domain size (DDS) of 4.8 km × 4.8 km × 2.88 km and a double grid 587 resolution (DGR) of 10 m for the case of N60. The figure shows that the vertical profiles are 588 hardly modified at DDS, but raindrop formation increases slightly with DGR; this is mainly 589 because larger  $\varepsilon$  values are associated with better resolved convective eddies (Sato et al., 2018). 590 The temporal variation in profiles becomes weaker at DDS as the number of generated clouds 591 592 increases (not shown), but DDS does not affect the mean vertical profiles. The scatter plots of  $\alpha$ vs.  $R_c$ , corresponding to Figure 7, also do not show significant differences (Figure A2). This 593 confirms that the correlations between the parameters used for the parameterization of A (6) 594 remain valid. At DGR, a larger  $\varepsilon$  can produce a larger A from the same parameterization (6). 595

![](_page_33_Figure_1.jpeg)

597

Figure A1 Vertical profiles of the variables averaged over the last 3 hours of simulations (black: original, red: DDS, blue: DGR): (a) A, (b)  $q_c$ , (c)  $N_c / N_0$ , (d)  $\varepsilon$ , (e)  $\sigma_c$ , (f)  $R_c$  (Here  $N_c / N_0$ ,  $\varepsilon$ ,  $\sigma_c$ , and  $R_c$  are averaged only within clouds.)

![](_page_34_Figure_1.jpeg)

Figure A2 Scatter plots of  $\alpha$  vs.  $R_c$  (violet:  $0 \text{ cm}^2 \text{ s}^{-3} \le \varepsilon < 20 \text{ cm}^2 \text{ s}^{-3}$ , blue:  $20 \text{ cm}^2 \text{ s}^{-3} \le \varepsilon < 60 \text{ cm}^2 \text{ s}^{-3}$ ,  $20 \text{ cm}^2 \text{ s}^{-3} \le \varepsilon < 60 \text{ cm}^2 \text{ s}^{-3}$ ,  $20 \text{ cm}^2 \text{ s}^{-3} \le \varepsilon < 80 \text{ cm}^2 \text{ s}^{-3}$ , red:  $40 \text{ cm}^2 \text{ s}^{-3}$ , green:  $40 \text{ cm}^2 \text{ s}^{-3} \le \varepsilon < 60 \text{ cm}^2 \text{ s}^{-3}$ , orange:  $60 \text{ cm}^2 \text{ s}^{-3} \le \varepsilon < 80 \text{ cm}^2 \text{ s}^{-3}$ , red:  $80 \text{ cm}^2 \text{ s}^{-3} \le \varepsilon < 100 \text{ cm}^2 \text{ s}^{-3}$ ). The means (circles) and standard deviations (crosses) of  $\alpha$  vs.  $R_c$  are included. A dashed blue line represents the variation in the mean  $\alpha$  with  $R_c$  for  $0 \text{ cm}^2 \text{ s}^{-3} \le \varepsilon < 20 \text{ cm}^2 \text{ s}^{-3}$ : (a) DDS, (b) DGR.

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# 615 Data Availability Statement

This LES/LCM model used in this study is publicly available (https://palm.muk.uni-616 hannover.de/trac/browser?rev=4464). For analysis purposes, the model was extended, and 617 additional analysis tools were developed. The code is available from the authors upon request. The 618 data used for the figures in this available 619 paper are at https://doi.org/10.6084/m9.figshare.14843004.v3 620

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