



The Critical Number and Size of Precipitation Embryos to Accelerate Warm Rain Initiation

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Abstract. Understanding warm rain initiation through droplet collision and coalescence is a fundamental yet complex challenge in cloud microphysics. Although it is well-known that sufficiently large droplets, so-called precipitation embryos (PEs), may accelerate droplet collisions, it is uncertain how many and how large these PEs should be to affect rain initiation substantially. We address this question using an ensemble of box simulations with Lagrangian cloud microphysics. We found that the warm rain initiation is substantially accelerated only if the PE size or number (or the product of those) exceeds a critical threshold necessary to compensate for the PE-induced suppression of collisions among non-PEs. The sensitivity of this threshold to the shape of the droplet size distribution and turbulence effects on the collision process is analyzed. It is shown that more and larger PEs are needed when collisions are already efficient without PEs. Beyond increasing our fundamental understanding of the precipitation process in warm clouds, our results may help to constrain the effect of PE-like particles intentionally or unintentionally added in geoengineering approaches, such as rain enhancement or marine cloud brightening.

1 Introduction

A key question in warm rain initiation is to explain the growth of cloud droplets in the radius range between 15 and 40 μm , the so-called size gap, in which neither condensational nor collisional growth is effective (e.g., Shaw, 2003; Devenish et al., 2012; Grabowski and Wang, 2013). Especially for droplet size distributions (DSDs) in a *colloidal stable* state (Squires, 1958), where collisions among droplets are inefficient due to a narrow DSD or too small droplets, mechanisms accelerating the collision-coalescence process to form a raindrop and initiate the precipitation are key to breaking this stability. Research over the past five decades has identified several key mechanisms: (i) DSD broadening by entrainment and mixing (Baker et al., 1980; Blyth, 1993; Krueger et al., 1997; Lasher-Trapp et al., 2005; Cooper et al., 2013; Hoffmann et al., 2019; Lim and Hoffmann, 2023), (ii) turbulence-induced collision enhancement (TICE), which increases the collision efficiency and reduces the size dependency of droplets to initiate collisions (e.g., Saffman and Turner, 1956; Kostinski and Shaw, 2005; Pinsky et al., 2008; Wang and Grabowski, 2009; Grabowski and Wang, 2013; Onishi et al., 2015; Hoffmann et al., 2017; Chen et al., 2020; Chandrakar et al., 2024), and (iii) the role of so-called precipitation embryos (PEs), the primary focus of this study.

The presence of PEs larger than 20 μm can initiate the collision process as they are already larger than the size-gap range (e.g., Woodcock, 1953; Telford, 1955; Johnson, 1982; Exton et al., 1986; Feingold et al., 1999; Teller and Levin, 2006; Alfonso



25 et al., 2013; Hoffmann et al., 2017; Dziekan et al., 2021). The sources of these PEs can be giant aerosol particles, predominantly
large sea-salt aerosols that form solution droplets having a size range between 1 μm and 100 μm (Johnson, 1982; Blyth, 1993;
O’Dowd et al., 1997; Feingold et al., 1999; Jensen and Nugent, 2017; Hudson and Noble, 2020; Hoffmann and Feingold, 2023),
rare (one in a million) ‘lucky’ that grow faster than the average droplet and initiate precipitation (Telford, 1955; Kostinski and
Shaw, 2005; Wilkinson, 2016; Alfonso and Raga, 2017; Alfonso et al., 2019), or particles from cloud seeding experiments to
30 enhance precipitation (Bowen, 1952; Cotton, 1982).

Although the aforementioned studies generally agree that PEs can accelerate warm rain initiation, it is uncertain how their
number and size affect the acceleration of droplet growth. Some studies suggest that a few 20 μm -sized droplets can effectively
accelerate the rain initiation (e.g., Feingold et al., 1999) and change the amount of precipitation and cloud properties such
as maximum droplet number concentration and liquid water content (e.g., Yin et al., 2000). Other studies indicate that the
35 effectiveness of PEs relies on the type of the cloud, with shallower clouds being more susceptible (e.g., Kuba and Murakami,
2010; Dziekan et al., 2021). In particular, if we consider the stochastic fluctuations of the collision process, only a 12.5 μm -
sized lucky droplet among 10 μm droplets can initiate collisions (Kostinski and Shaw, 2005). Thus, it is also important to
account for stochastic fluctuations in the collision process. Lastly, although a few previous studies have investigated these
mechanisms (Hoffmann et al., 2017; Chen et al., 2020), it remains unclear whether PE and TICE compete or complement each
40 other in influencing collisional growth.

A particle-based Lagrangian cloud model (LCM) is the natural choice for such investigation (e.g., Gillespie, 1972; Shima
et al., 2009; Hoffmann et al., 2017; Dziekan and Pawlowska, 2017; Unterstrasser et al., 2020; Li et al., 2022). Particularly,
it was shown that a “one-to-one” LCM, where each computational particle represents one single cloud drop is suitable to
consider stochastic fluctuations in collisional growth naturally (e.g., Dziekan and Pawlowska, 2017; Li et al., 2022). While
45 considering the numerous processes that also affect warm rain initiation (i.e., aerosol activation and condensation) is essential
for investigating rain initiation, a simple box model of the collision-coalescence process alone offers unique insights that cannot
be captured in a more complex model due to its tremendous computational costs when using one-to-one LCM. Therefore, this
study aims to investigate the early stages of collisional growth to determine the number and size of PEs needed to accelerate
collisional growth.

50 This paper is organized as follows. Section 2 introduces the LCM box model and the simulation setup. Section 3 presents
the results revealing the threshold on the minimum number and size of PEs to accelerate droplet collisions. Section 4 explores
the mechanism behind the existence of this threshold. We conclude our paper in Sec. 6.

2 Model and Simulation Setup

2.1 Lagrangian Cloud Box Model

55 In most applications, each computational particle of an LCM represents a large number of real droplets with identical prop-
erties, frequently called superdroplets by introducing a weighting factor (W_i) (e.g., Shima et al., 2009). Thus, the number



concentration of droplets is determined by

$$N = \sum_{i=1}^{n_{\text{ptcl}}} \frac{W_i}{\Delta V}, \quad (1)$$

where ΔV is a reference volume, and n_{ptcl} represents the number of computational particles in ΔV . In this study, we apply the “one-to-one” method, where each computational particle represents a single cloud droplet ($W_i = 1$). This approach fully captures the inherent stochasticity of the collision process (Shima et al., 2009; Dziekan and Pawlowska, 2017; Li et al., 2022).

The collision scheme follows the approach introduced by Shima et al. (2009) and Sölch and Kärcher (2010), in which a collision occurs with the probability

$$p_{m,n} = \frac{K}{\Delta V} \delta t, \quad (2)$$

primarily determined by the collection kernel

$$K = \pi(r_m + r_n)^2 E(r_m, r_n) |w(r_m) - w(r_n)|, \quad (3)$$

where r_m and r_n are the radii of the interacting droplets, E the collision efficiency of droplet pairs (Hall, 1980), w the droplet terminal velocity (Beard, 1976), and δt the model time step. Here, we assume the coalescence efficiency to be unity. In this study, a collected droplet is removed from the simulation after the collision-coalescence event, and the mass of the collecting droplet increases by the mass of the collected droplet.

The simulations do not consider other processes besides collisional growth, such as condensation or sedimentation, which are beyond the focus of our study. Therefore, our results should be regarded as representative for the early stages of collisional growth only. For a detailed explanation of the LCM collision scheme, readers are referred to Hoffmann et al. (2017), Noh et al. (2018), and Unterstrasser et al. (2020).

2.2 Simulation Setups

The initial DSD is expressed as

$$N(m) = \frac{N_0}{\bar{m}} \exp\left(\frac{-m}{\bar{m}}\right), \quad (4)$$

where m is the mass of a droplet, $N_0 = 238 \text{ cm}^{-3}$ the initial droplet number concentration, and \bar{m} the mass of a droplet with $\bar{r} = 10 \text{ }\mu\text{m}$ (see orange line in Fig. 1). The DSD results in a cloud water mixing ratio (q_c) of approximately 1.0 g kg^{-1} . Additionally, cases with $\bar{r} = 8, 12, \text{ or } 14 \text{ }\mu\text{m}$ are considered to investigate the effect of PEs in different DSD shapes. In these cases, $N_0 = 238, 456 \text{ and } 523 \text{ cm}^{-3}$ to achieve the same $q_c = 1.0 \text{ g kg}^{-1}$ (Fig. 1). We name these cases ‘RM’, with the subsequent number denoting \bar{r} (e.g., RM10).

To establish a *colloidally stable* initial DSD in which collisions are negligible, droplets larger than $20 \text{ }\mu\text{m}$ are removed in selected simulations to prevent them from initiating collisions (Wang et al., 2006; Dziekan and Pawlowska, 2017); we will refer to such initialization as ‘cut-off DSD,’ (see dotted line in Fig. 1). In cases without this adjustment, where the initial DSD

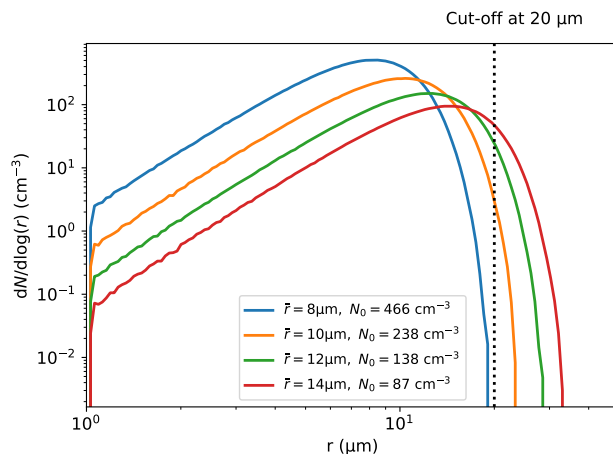


Figure 1. Initial DSDs for various \bar{r} and corresponding N_0 values. The dotted line indicates the cut-off radius of 20 μm , above which droplets are removed in some cases.

is *broader*, we refer to it as the case with a *broad DSD* and denote it by adding the letter 'B' to the naming convention (e.g., RM10B). In this study, we primarily discuss the simulation with $\bar{r} = 10 \mu\text{m}$ and cut-off DSD, i.e., the RM10 case, unless otherwise noted. Lastly, three different kinetic energy dissipation rates $\varepsilon = 16, 80, \text{ and } 100 \text{ cm}^2 \text{ s}^{-3}$ are considered for RM10 case to investigate the effect of TICE. TICE is incorporated in Eq. 3 using the parameterizations developed by Ayala et al. (2008) and Wang and Grabowski (2009), which are steered by ε . When TICE is considered, the case names are amended by a T followed by the value of ε (e.g., RM10-T100).

A total of 10^6 computational particles ($n_{\text{ptcl}} = 10^6$) are initialized to represent the initial DSD resulting in a reference volume $\Delta V = 3.36 \times 10^{-3} \text{ m}^3$. Every setup is simulated 100 times with different random numbers to ensure statistical convergence (cf. Fig. A1). Using a timestep $\delta t = 0.1 \text{ s}$, the model is integrated for 7200 s to account for the slowest realization to complete collisional growth, but the discussion is focused on the initial 2500 s, capturing the initiation of collisional growth.

To explore the impact of PEs, we investigate 49 ensemble simulations, each representing different combinations of PE radii ($r_{\text{PE}} = 15, 18, 22, 27, 33, 40, \text{ and } 50 \mu\text{m}$) and numbers ($n_{\text{PE}} = 1, 3, 10, 30, 100, 300, \text{ and } 1000$). Here, we define PEs as any droplets added to the original DSD, although the conventional definition of PEs requires $r_{\text{PE}} > 20 \mu\text{m}$. The largest PE size is chosen to correspond to the size of haze particles grown from 1–5 μm giant aerosols (Kuba and Murakami, 2010). We choose a minimum $n_{\text{PE}} = 1$ to investigate whether 'one in a million' lucky droplets could accelerate droplet collision, as highlighted in previous studies on lucky droplets (Kostinski and Shaw, 2005; Dziekan and Pawlowska, 2017). Maximum $n_{\text{PE}} = 1000$ is used for $r_{\text{PE}} = 15$ and 18 μm only, as larger PEs can substantially increase the initial q_c , limiting the comparability of the simulated cases. Within a given reference volume, the minimum and maximum n_{PE} can be interpreted as concentrations between $2.97 \times 10^{-4} \text{ cm}^{-3}$ and $2.97 \times 10^{-1} \text{ cm}^{-3}$, respectively.

In this study, the two specific timescales, t_{100} and $t_{10\%}$ are used to characterize precipitation efficiency. In previous studies, time for the first raindrop formation is used to quantify the efficiency of stochastic raindrop formation (Dziekan and Pawlowska,

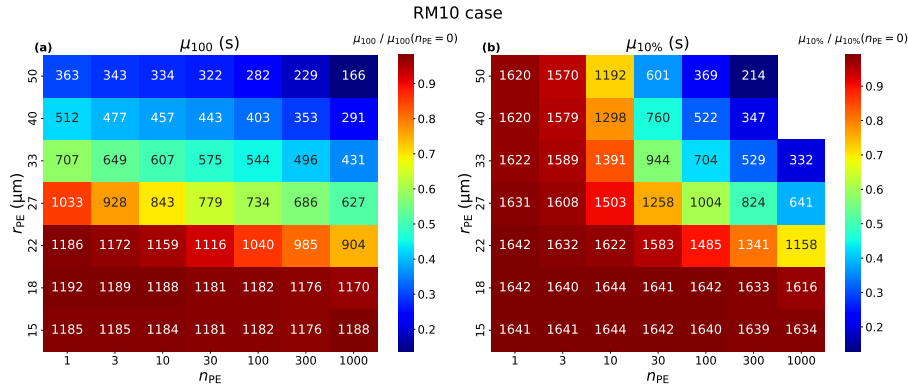


Figure 2. Ensemble-averaged values of (a) time for the first 100 μm raindrop formation, μ_{100} , (b) time for 10 % of cloud droplets to convert to raindrops, $\mu_{10\%}$, for RM10 case. The abscissa represents n_{PE} , the ordinate represents r_{PE} . The numbers in each box indicate the values of (a) μ_{100} , (b) $\mu_{10\%}$. Values for $r_{PE} \geq 40\mu\text{m}$ and $n_{PE} > 1000$ are not shown as the raindrop mass is already larger than 10 % due to large PEs. Colors in the plot represent the ratio of μ_{100} and $\mu_{10\%}$ to their values in the case without PEs ($n_{PE} = 0$). In the case without PE, $\mu_{100} = 1214$ s, and $\mu_{10\%} = 1660$ s respectively.

2017). As PEs considered in this study can be larger than the typical raindrop radius, i.e., over 40 μm , we define t_{100} as the time required for the formation of the first 100 μm droplet, i.e., a sufficiently large droplet that stimulates subsequent collisions Kostinski and Shaw (2005); Alfonso et al. (2019). Thus, t_{100} characterized the efficiency for *raindrop formation*. The timescale $t_{10\%}$ represents the time when 10 % of the initial cloud droplet mass converts to rain, measuring the efficiency of *rain initiation* from a mass perspective (Onishi et al., 2015; Dziekan and Pawlowska, 2017).

3 PE Effect on Precipitation Timescales

3.1 Critical Thresholds for Raindrop Formation and Rain Initiation

Figure 2 shows the ensemble-averaged t_{100} and $t_{10\%}$, named μ_{100} and $\mu_{10\%}$, for RM10. In general, increasing r_{PE} and n_{PE} both shorten μ_{100} and $\mu_{10\%}$, indicating accelerated rain initiation. However, when $r_{PE} \leq 18 \mu\text{m}$, i.e., smaller than the cut-off radius, μ_{100} and $\mu_{10\%}$ are not substantially accelerated compared to those cases without PEs regardless of n_{PE} . Note that, in the case without PEs, μ_{100} and $\mu_{10\%}$ are 1213 s and 1660 s, respectively, for RM10. This indicates that the addition of PEs smaller than the maximum droplet radius of the DSD, even in large numbers (e.g., $n_{PE} = 1000$), has a negligible effect on raindrop formation. Interestingly, for $\mu_{10\%}$, n_{PE} plays a more crucial rule than for the μ_{100} . For $n_{PE} \leq 3$, $\mu_{10\%}$ is not accelerated (Fig. 2b) even for large PEs, whereas μ_{100} is accelerated (Fig. 2a). Thus, a faster μ_{100} does not always ensure a shorter $\mu_{10\%}$.

Overall, Fig. 2 shows μ_{100} and $\mu_{10\%}$ can be shortened with increasing n_{PE} and r_{PE} , but only if a critical threshold is exceeded. Below this critical threshold, the effect of PEs on rain initiation is negligible. This raises the following question: What

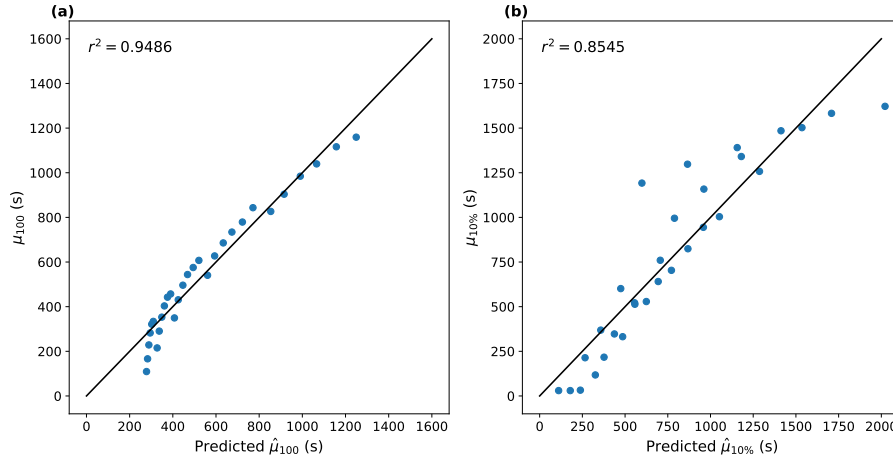


Figure 3. Scatter plots of simulated (a) μ_{100} and (b) $\mu_{10\%}$ (ordinate) and predicted values (abscissa) using Eq. (5) for RM10 case. Black solid lines indicate the one-to-one line.

are the specific size and number of PEs required to accelerate rain initiation substantially? To identify the critical threshold, we first express μ_{100} and $\mu_{10\%}$ as functions of n_{PE} and r_{PE} . As shown in Fig. 2, μ_{100} and $\mu_{10\%}$ are inversely related to the product of n_{PE} and r_{PE} once the critical threshold is exceeded. Thus, we write

$$\mu_{\alpha} = k_{\alpha} \Phi_{\alpha}(n_{PE}, r_{PE}) + c_{\alpha} = k_{\alpha} n_{PE}^{-a_{\alpha}} r_{PE}^{-b_{\alpha}} + c_{\alpha} \quad (5)$$

for a μ_{α} exceed the critical threshold. Here k_{α} is a rate-of-change coefficient, c_{α} a constant, $\Phi_{\alpha}(n_{PE}, r_{PE})$ represents the composite relationship of n_{PE} and r_{PE} with scaling exponents a_{α} and b_{α} , and the subscript α is 100 or 10% for μ_{100} and $\mu_{10\%}$, respectively.

To determine the parameters of Eq. (5), we fit a_{α} , b_{α} , c_{α} , and k_{α} , using μ_{100} and $\mu_{10\%}$ from cases with $r_{PE} \geq 22$ and $n_{PE} \geq 10$. In these cases, both μ_{100} and $\mu_{10\%}$ are directly affected by changes in r_{PE} and n_{PE} (Fig. 2), i.e., the PE critical threshold is exceeded. The fitted parameters are $a_{100} = 0.086$, $b_{100} = 3.086$, $c_{100} = 244$ s, and $k_{100} = 17308248$ for μ_{100} , and $a_{10\%} = 0.13$, $b_{10\%} = 1.13$, $c_{10\%} = -310$ s, and $k_{10\%} = 104016$ for $\mu_{10\%}$ with r_{PE} in μm . Note that, while units of each parameter are detailed in Appendix B, our focus will be on $\Phi_{\alpha}(n_{PE}, r_{PE})$, i.e., a_{α} and b_{α} first. The parameters c_{α} and k_{α} will be discussed in more detail after we expand Eq. (5) with more physically meaningful terms. The values of a_{α} and b_{α} indicate that both μ_{100} and $\mu_{10\%}$ are more sensitive to r_{PE} than n_{PE} . When comparing a_{100} and b_{100} to $a_{10\%}$ and $b_{10\%}$, $\mu_{10\%}$ seems to depend more on n_{PE} and less on r_{PE} , than μ_{100} , which is consistent with the results shown in Fig. 2. Figure 3 juxtaposes the simulated and predicted μ_{100} and $\mu_{10\%}$ values using Eq. (5). This result indicates that μ_{100} and $\mu_{10\%}$ can be expressed with Φ_{α} relatively well. However, Eq. (5) tends to overestimate $\mu_{10\%}$ when it is below 750 s, and generally fails to predict $\mu_{10\%}$ when it is over 1000 s (Fig. 3b). This is due to the cases with $n_{PE} < 10$ and $r_{PE} < 22$ μm , which show almost no dependency on r_{PE} .



To better capture the behavior of μ_{100} and $\mu_{10\%}$, especially near the critical threshold where the dependency on r_{PE} and n_{PE} vanishes, we expand Eq. (5) by a Heaviside step function \mathcal{H} , such that

$$145 \quad \mu_{\alpha}(\Phi) = \mu_{\alpha,c} - k_{\alpha}(\Phi_{\alpha,c} - \Phi_{\alpha}) \cdot \mathcal{H}(\Phi_{\alpha,c} - \Phi_{\alpha}), \quad (6)$$

where $\mu_{\alpha,c}$ is the baseline value of μ_{α} in the absence of PEs incorporating parameter c_{α} from above. When fitting Eq. (6) all results, including cases where $r_{PE} < 22$ and $n_{PE} < 10$, are used, with parameters a_{α} and b_{α} fixed to the values obtained previously. The specific parameters for Eq. (6) and their r-squared values are detailed in Appendix B. In general, r-squared values exceed 0.95 for μ_{100} , and range from 0.75 to 0.9 for $\mu_{10\%}$. The results of $\mu_{100}(\Phi)$ and $\mu_{10\%}(\Phi)$ for the RM10 case are shown in Fig. 4a and b as blue solid lines. Until exceeding the critical thresholds ($\Phi_{100,c} = 5.04 \times 10^{-5}$ and $\Phi_{10\%,c} = 1.51 \times 10^{-2}$; see Tabs. B1 and B2), μ_{100} and $\mu_{10\%}$ remain constant at $\mu_{100,c} = 1190$ s and $\mu_{10\%,c} = 1608$ s. These values agree well with μ_{100} and $\mu_{10\%}$ without PEs, 1214 s and 1660 s, respectively. However, once Φ_{α} becomes smaller than $\Phi_{\alpha,c}$, i.e., exceeds the critical threshold, μ_{100} and $\mu_{10\%}$ decrease as expected from Eq. (5).

3.2 Factors Controlling the Critical Threshold

155 Using Eq. (6), we are now able to investigate how the critical threshold varies for different initial DSD shapes (characterized by \bar{r} and the consideration of a cut-off radius) and the presence of TICE. To achieve this, we fit the results to Eq. (6) for RM8, RM10, RM12, and RM14 with or without cut-off DSD (Fig. 4a, b, c, and d). Additionally, we consider TICE for RM10 (Fig. 4e and f). Although a_{α} and b_{α} parameters for Φ_{100} and $\Phi_{10\%}$ may vary for different cases, we fix them to the values obtained earlier (see Fig. 3) to directly compare $\mu_{\alpha,c}$, $\Phi_{\alpha,c}$ and k_{α} across different initial conditions. The fitted parameters for these initial conditions are detailed in Appendix B. Figure 4 shows that all cases exhibit the same fundamental feature: the presence of a critical threshold $\Phi_{\alpha,c}$.

We first discuss the results for μ_{100} . As \bar{r} increases, $\mu_{100,c}$ decreases (Figs. 4a and 5b), indicating that it takes less time to produce a large raindrop even without PEs. This is due to the increased number of large droplets when \bar{r} increases, although the largest droplet size remains unchanged due to the cut-off DSD. In this case, $\Phi_{100,c}$ also decreases with increasing \bar{r} , implying more and larger PEs are needed to exceed the critical threshold (Fig. 5a).

Results from the cases with a broad DSD with different \bar{r} are shown in Fig. 4c and d. In these cases, also the maximum radius of the droplet and hence the DSD width increases, making droplet collisional growth more efficient (cf. Fig. 4a and b). For the same \bar{r} , we see that $\Phi_{100,c}$ is smaller in broad DSD cases (Fig. 5a). This is because of the presence of larger droplets in the initial DSD, which are equally efficient as PEs in the collision process, reducing the importance of the PE effect. Moreover, both $\mu_{100,c}$ and $\Phi_{100,c}$ decrease with increasing \bar{r} (Fig. 4c and d and Fig. 5a). This is because, without cut-off DSD, both the size and the number of large droplets increase with increasing \bar{r} , (cf. Fig. 1). The results for $\mu_{10\%,c}$ and $\Phi_{10\%,c}$ show a similar pattern to those of $\mu_{100,c}$ and $\Phi_{100,c}$ (Fig. 4c and Fig. 5c and d), where both $\mu_{10\%,c}$ and $\Phi_{10\%,c}$ decrease with increasing \bar{r} in the absence of the cut-off DSD.

175 Additionally, the TICE effect is considered for RM10 (Fig. 4e and f). TICE is considered with three different $\varepsilon = 16, 80$ and $100 \text{ cm}^2 \text{ s}^{-3}$ which are typically found within different cloud types: $1 - 10 \text{ cm}^2 \text{ s}^{-3}$ in stratocumulus clouds, $10 - 100 \text{ cm}^2 \text{ s}^{-3}$ in shallow

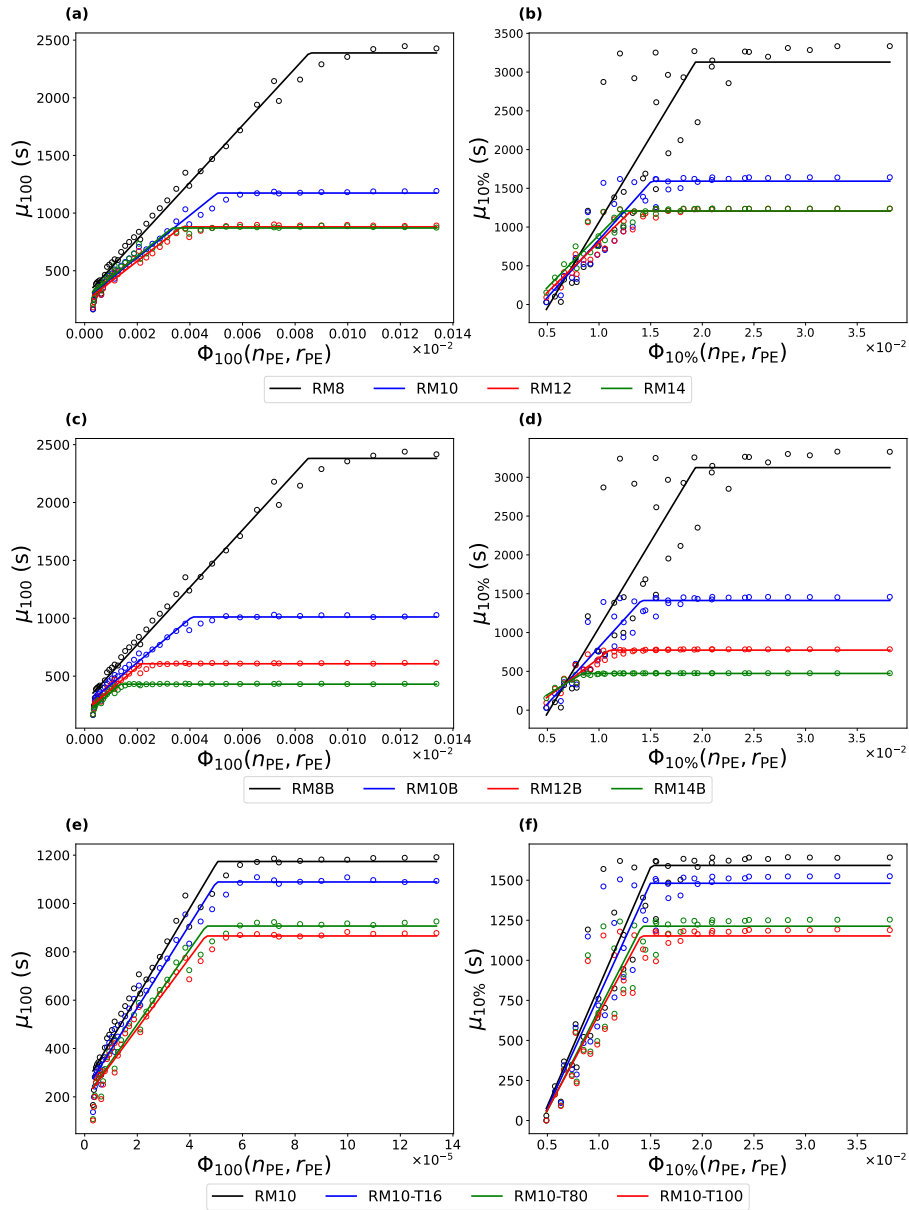


Figure 4. μ_{100} (left column) and $\mu_{10\%}$ (right column) as a function of Φ_{α} for different initial conditions are shown. Each point represents the simulation results, while solid lines indicate the fitted Eq. (6). The first row (a and b) represents cases with cut-off DSD (RM8, RM10, RM12, and RM14), and the second row (c and d) represents cases without cut-off DSD (RM8B, RM10B, RM12B, and RM14B). The third row (e and f) represents RM10 with different ε values (RM10-T16, RM10-T80, and RM10-T100).

convective clouds, and $100 - 1000 \text{ cm}^2 \text{ s}^{-3}$ in deep convective clouds (Siebert et al., 2006; Seifert et al., 2010; Pruppacher and Klett, 2012). With strong turbulence ($\varepsilon = 80 - 100 \text{ cm}^2 \text{ s}^{-3}$), $\Phi_{100,c}$ is lower (Figs. 4a and 5a) than in the case with no or

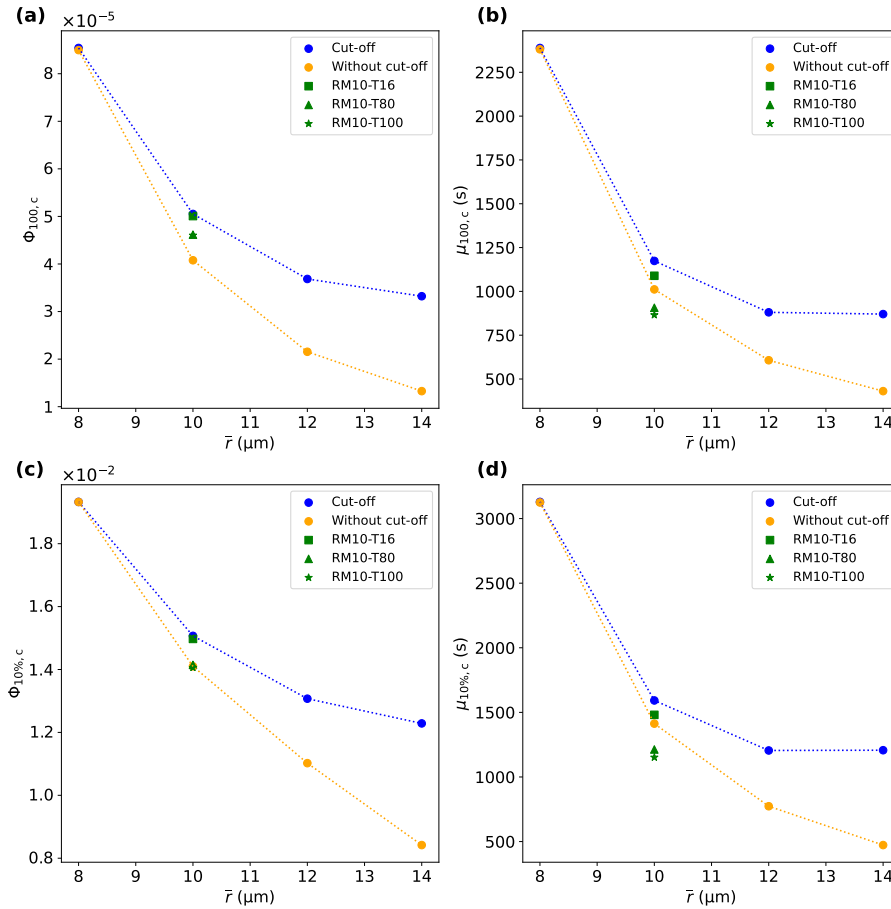


Figure 5. Results of (a) $\Phi_{100,c}$, (b) $\mu_{100,c}$, (c) $\Phi_{10\%,c}$ and (d) $\mu_{10\%,c}$ for different \bar{r} . Blue circles depict the cases with cut-off DSD while orange circles depict cases without cut-off DSD. Green square, triangle, and star shape represent the results with $\varepsilon = 16, 80, 100 \text{ cm}^2 \text{ s}^{-3}$, respectively, for the RM10 case.

weaker TICE ($\varepsilon = 16 \text{ cm}^2 \text{ s}^{-3}$). Thus, more and larger PEs are required to substantially accelerate μ_{100} with TICE. In other words, the PE effect becomes weaker when TICE is strong. Both $\Phi_{100,c}$ and $\Phi_{10\%,c}$ decrease but only slightly with TICE but they never fall below the values observed in the broad DSD case (Fig. 5a and c). In contrast, $\mu_{100,c}$ and $\mu_{10\%,c}$ decrease more substantially with increasing ε , becoming even shorter than in the broad DSD case (Fig. 5b and d). This suggests that TICE enhances the efficiency of every collision event, leading to a faster $\mu_{10\%,c}$, due to collisions among all cloud droplets in the entire system. However, the impact of TICE on the critical threshold is less pronounced than that of the DSD shape (\bar{r} and cut-off DSD).

In summary, when droplet collisions are efficient without PEs, whether due to a large \bar{r} , the presence of large droplets (i.e., broad DSD), or TICE, the PE size and number needs to be larger to accelerate rain initiation substantially. Although we have identified the existence of the critical threshold, there remains uncertainty regarding why $t_{10\%}$ is not always shorter than



the case without PEs when n_{PE} is small, even though t_{100} is decreased (e.g., $n_{PE} < 10$ cases in Fig. 2). This discrepancy may arise because $t_{10\%}$ involves interactions among multiple droplets and PEs, whereas t_{100} depends on the behavior of an individual droplet or PEs. This suggests that while PEs can accelerate the formation of the largest raindrop, these droplets may not substantially impact the overall rain initiation after the initial period when they are few. This contradicts the ‘lucky droplet’ theory that a few lucky droplets trigger the subsequent runaway growth and rain initiation (Kostinski and Shaw, 2005; Alfonso and Raga, 2017). In the following section, we will explore how PEs affect $t_{10\%}$ to explain why a shorter t_{100} does not ensure a shorter $t_{10\%}$.

195 4 PE Effects on Rain Initiation

In order to understand the effects of PE size and number on rain initiation more clearly, we consider the time series of raindrop mixing ratio q_r , autoconversion rate (i.e., raindrop formation by collisions between cloud droplets), and accretion rate (i.e., raindrop growth by raindrops collecting cloud droplets) using $r_{PE} = 22 \mu\text{m}$ and $27 \mu\text{m}$ with different n_{PE} ranging from 0 to 300 for RM10 (Fig. 6). Overall, q_r evolves faster for larger r_{PE} and n_{PE} (Fig. 6a and b). However, with PEs below the critical threshold (i.e., for $n_{PE} \leq 30$ at $r_{PE} = 22 \mu\text{m}$ and $n_{PE} \leq 3$ at $r_{PE} = 27 \mu\text{m}$), the difference from the case with and without PE is insignificant, implying that PEs do not substantially enhance rain initiation, although raindrop formation ($q_r > 0$) starts earlier (Fig. 6a and b). This result is consistent with Fig. 2, in which μ_{100} is smaller than $\mu_{100,c}$, but $\mu_{10\%}$ is comparable to $\mu_{10\%,c}$.

The time series of autoconversion and accretion evolution provides more details on how PEs affect rain initiation. In Fig. 6c to f, solid lines represent droplet growth without PEs (i.e., between non-PE droplets exclusively), while dotted lines represent droplet growth involving PEs (i.e., collisions between PEs and non-PE droplets or among PEs). We found that non-PE autoconversion decreases with increasing n_{PE} (Fig. 6c and d). This is because large PEs have an advantage in the autoconversion process, growing faster and collecting non-PE droplets, which in turn suppresses the autoconversion of non-PE droplets.

For $r_{PE} = 22 \mu\text{m}$, both autoconversion and accretion initiate earlier with PEs than in the case without PEs, but only when $n_{PE} \geq 100$ (Fig. 6c). When $n_{PE} < 30$, autoconversion and consequently accretion by PEs are even slower than those of non-PE droplets. This is because autoconversion depends heavily on stochastic events. Thus although larger PEs are more likely to collide, a small n_{PE} reduces the likelihood of these collisions, making PE autoconversion slower than non-PE autoconversion. Thus, when n_{PE} is small, PEs may not decrease $t_{10\%}$, although t_{100} can be shorter than in the cases without PEs.

For $r_{PE} = 27 \mu\text{m}$, while non-PE-autoconversion always decreases with increasing n_{PE} , PE-autoconversion increases substantially only when $n_{PE} \geq 100$. Therefore, before exceeding the critical threshold, PEs suppress non-PE autoconversion more than they enhance autoconversion which can even lead to a decrease in the total (PE and non-PE) autoconversion. Interestingly, in this case, the time to initiate PE autoconversion remains unchanged with n_{PE} affecting only its magnitude (Fig. 6c). The initiation time for PE autoconversion is influenced by r_{PE} since this process is closely related to the number of collisions or time required for droplets to grow larger than $40 \mu\text{m}$, which occurs more quickly for larger PEs (cf. Fig. 6c and d). Thus, r_{PE}

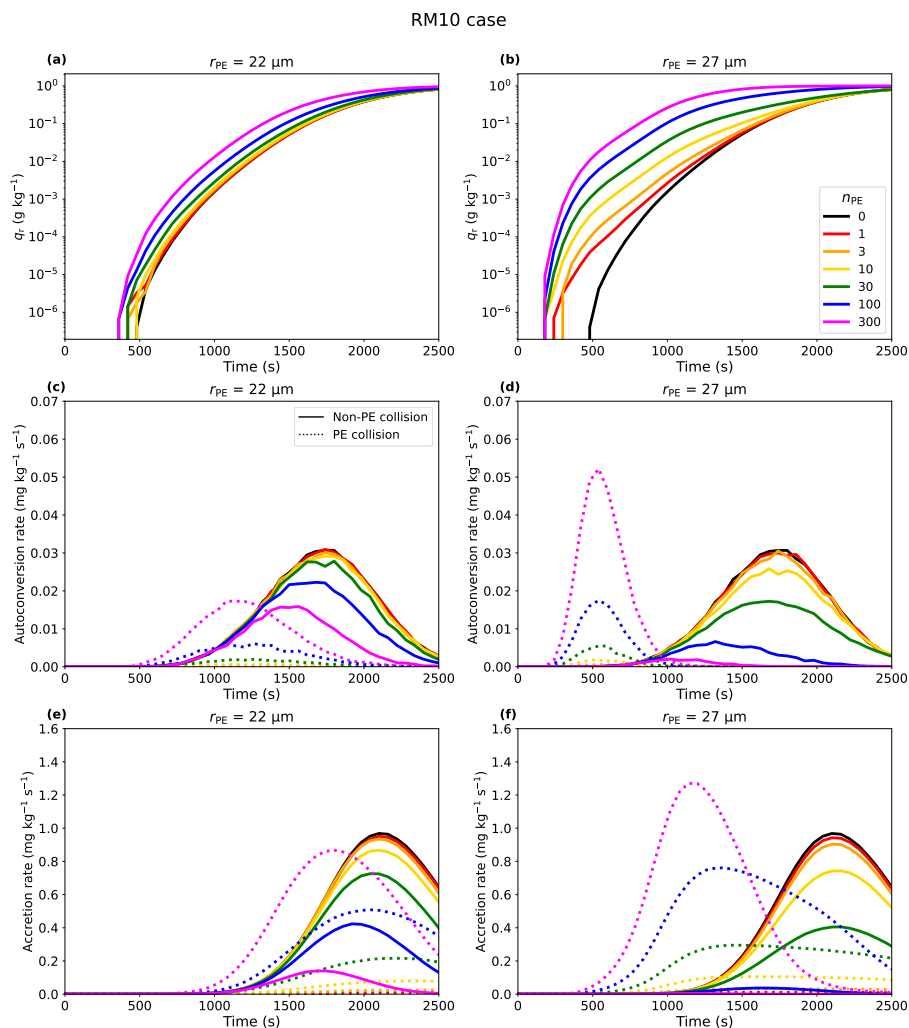


Figure 6. Time series of (a/b) raindrop mixing ratio, (c/d) autoconversion rate, and (e/f) accretion rate, for the RM10 case, shown for two different values of r_{PE} : $22 \mu\text{m}$ (first column) and $27 \mu\text{m}$ (second column). The colors of the lines represent different n_{PE} values, with the black solid line representing the result from the simulation without PEs ($n_{PE} = 0$). In (c) to (f), the solid lines denote autoconversion and accretion without PEs (between non-PE droplets exclusively), while the dotted line depicts autoconversion and accretion by PEs.

220 determines the initiation time for autoconversion, especially when $r_{PE} \geq 27 \mu\text{m}$, while n_{PE} determines how much non-PE droplet autoconversion and accretion are suppressed.

Accretion starts earlier when $r_{PE} = 22 \mu\text{m}$ and $n_{PE} > 100$ and any n_{PE} for $r_{PE} = 27 \mu\text{m}$ (Fig. 6e and f), which is triggered by the earlier raindrop formation by autoconversion (Fig. 6c and d). However, even for $r_{PE} = 27 \mu\text{m}$, accretion by PEs increases only slightly when $n_{PE} \leq 30$, i.e., below the critical threshold. Once the critical threshold is exceeded, particularly for n_{PE} ,
 225 accretion is substantially increased and accelerated compared to the case for $n_{PE} = 0$ (Fig. 6e and f). In this case, accretion is

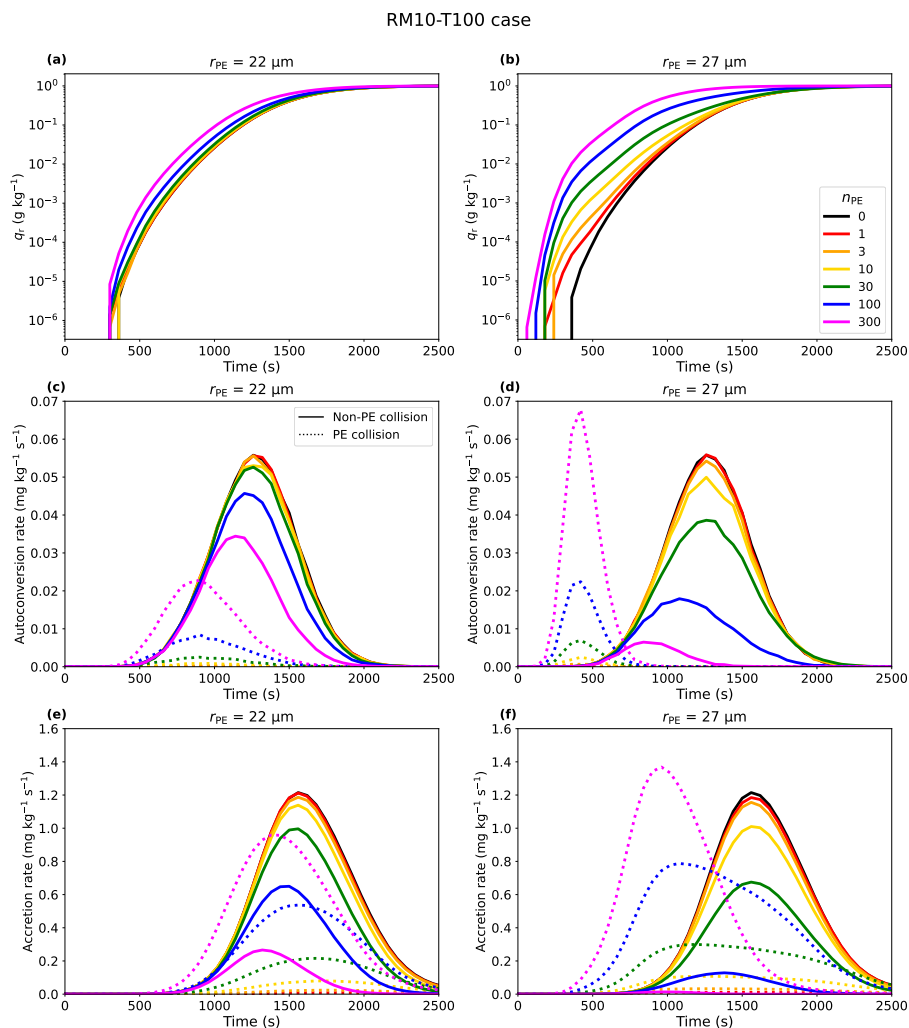


Figure 7. Same as for Fig. 6 but for cases with $\varepsilon = 100 \text{ cm}^2 \text{ s}^{-3}$.

dominated by PEs, outweighing the decrease in non-PE autoconversion (Fig. 6e and f), and initially larger q_r persists (Fig. 6a and b).

Results with TICE ($\varepsilon = 100 \text{ cm}^2 \text{ s}^{-3}$, Fig. 7) also highlight the importance of PEs in suppressing non-PE autoconversion. With TICE, collisions between small and similar-sized droplets are more efficient (Pinsky et al., 2008). Thus, with TICE, non-PE autoconversion is still substantial when $n_{PE} = 100$ (blue and purple solid lines in Fig. 7b), while it is almost totally suppressed without TICE (blue and purple solid lines in Fig. 6d). Thus, more and larger PEs are needed to outweigh non-PE accretion, making droplet growth less sensitive to PEs when TICE is considered. However, even with TICE, if n_{PE} substantially exceeds the critical threshold ($r_{PE} = 27 \mu\text{m}$ and $n_{PE} = 300$), droplet collisional growth is entirely dominated by PEs (purple



solid line in Fig. 7f). Thus, while both PEs and TICE accelerate droplet collisional growth, each effect becomes weaker when
235 the other effect dominates rain initiation (e.g., Chandrakar et al., 2024).

5 Summary and Conclusion

Understanding whether precipitation embryos (PEs), particles larger than the so-called size gap range, can accelerate the
droplet collision process remains a key question in warm rain initiation. Despite decades of research on the effect of PEs on
rain initiation (e.g., Telford, 1955; Johnson, 1982; Feingold et al., 1999; Teller and Levin, 2006; Alfonso et al., 2013), this
240 challenge persists and is still highlighted in recent studies (e.g., Chen et al., 2020; Dziekan et al., 2021; Chandrakar et al.,
2024), underscoring the need for further investigation.

In this study, we systematically investigated how PEs affect droplet collisional growth using ensembles of Lagrangian cloud
model (LCM) collision simulations. Our primary focus was to identify the minimal PE size and number necessary to accelerate
the droplet collision-coalescence process substantially. We evaluated the droplet collision efficiency using two timescales: the
245 time required for the first 100 μm droplet to form (t_{100}) and the time to convert 10% of the total initial cloud mass to rain mass
($t_{10\%}$).

We found that the droplet collision process does not substantially accelerate when the number or size of PEs is below
a critical threshold. t_{100} is accelerated only when the radii of PEs are larger than the maximum droplet radius of non-PE
droplets. This is because t_{100} is more related to the growth of a single droplet where larger droplets, such as PEs, are expected
250 to grow faster than smaller droplets. In contrast, $t_{10\%}$ depends more on the number of PEs. Even with substantially large PEs, a
faster formation of the first large raindrop does not always ensure faster rain initiation when the number of PEs is small. This is
because PEs increase autoconversion and accretion only when their number is sufficient while simultaneously suppressing the
autoconversion of non-PE droplets to become raindrops. Thus, when autoconversion of non-PE droplets is already efficient,
more or larger PEs are required to accelerate $t_{10\%}$.

To determine the critical threshold for rain initiation by PEs, we derived a simple equation that relates the number and size
of PEs to t_{100} and $t_{10\%}$. The equation revealed that the critical threshold depends on the colloidal stability of the droplet size
distribution (DSD) characterized by the DSD shape or turbulence-induced collision enhancement (TICE). We showed that
increasing the droplet mean radius and hence the size of pre-existing large droplets decreases the colloidal stability of DSD and
makes the collisional process less susceptible to PE perturbations because non-PE droplet collisions are already sufficient for
260 initiating rain. Equivalently, more and larger PEs are needed to substantially accelerate the droplet growth with TICE, which
increases the collision frequency among smaller non-PE droplets making the collision process less reliant on PEs.

While PEs can accelerate the rain initiation by collecting other droplets, they may reduce the number of raindrops by sup-
pressing non-PE droplets to grow as raindrops. As a result, clouds without PEs may have more and larger raindrops, as PEs
do not collect those before reaching the cloud top. This might lead to longer-lasting clouds and affect the precipitation differ-
265 ently. Thus, validating this study's findings in more complex scenarios is mandatory, for the future. These should incorporate
additional processes such as aerosol activation, condensation, entrainment, and especially, collisional droplet breakup (Low



and List, 1982) which increases the small number of PEs, causing more PE accretion afterward or droplet sedimentation which decreases the effect of PEs by making large raindrops precipitate and prevents PEs from further collisions

In conclusion, we confirm that a DSD barely producing raindrops is more sensitive to PEs (e.g., Dziekan et al., 2021). This
270 underscores the need for caution in geoengineering approaches like marine cloud brightening (Latham et al., 2012), aiming to
create highly reflective clouds by artificially adding aerosol particles, where the unintended initiation of rain by adding large
particles could be counterproductive (Hoffmann and Feingold, 2021). Indeed, this study found that PEs surpassing a critical
threshold can initiate rain, while numerous PEs with a sufficiently small size are harmless. In addition, approaches to enhance
precipitation, such as cloud seeding (Bowen, 1952; Cotton, 1982), should prioritize identifying target clouds with high stability
275 and minimal rain production to maximize efficiency.

Code and data availability. A Python version of the LCM code is available on the link (https://github.com/jslim93/PyLCM_edu). The simulations were conducted using the FORTRAN version of the code, which employs the same collision routine as the Python version but provides faster computation. Simulation data will be made available upon request to the authors.

Appendix A: Ensemble Size Sensitivity of t_{100} and $t_{10\%}$

280 Figure A1 illustrates how the mean and standard deviation of t_{100} (μ_{100} and σ_{100} , respectively) and $t_{10\%}$ ($\mu_{10\%}$ and $\sigma_{10\%}$, respectively) evolve as the ensemble size increases from 1 to 200 for RM10 without PEs ($n_{PE} = 0$). The mean values and standard deviations begin to converge when the ensemble size exceeds 100. Therefore, we consider an ensemble size of 100 adequate for obtaining reliable results.

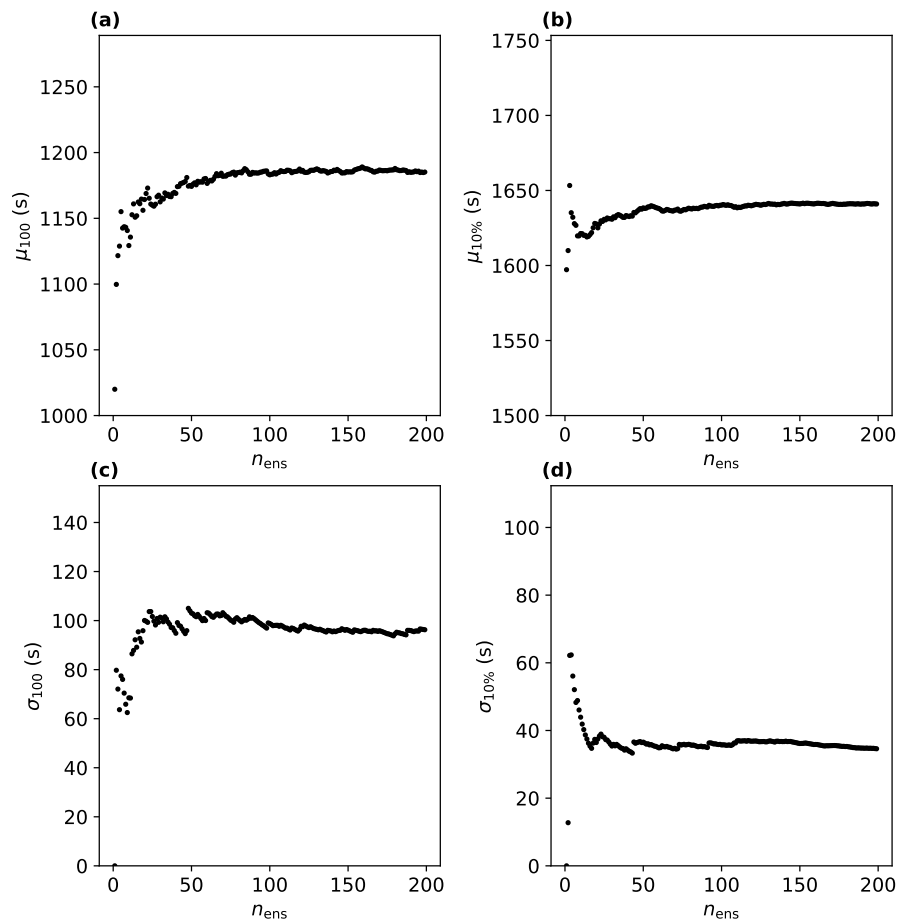


Figure A1. Variation of (a) μ_{100} , (b) $\mu_{10\%}$, (c) σ_{100} and (d) $\sigma_{10\%}$ with ensemble size (n_{ens}) for RM10 case and without PEs ($n_{PE} = 0$).



Appendix B: Parameters for the Fitting Function

285 Table B1 depicts the parameters and r^2 values derived from curve fitting Eq. (6) to μ_{100} for each result shown in Fig. 4a. Similarly, Table B2 shows the parameters and the r^2 values obtained from fitting Eq. (6) to $\mu_{10\%}$ for the results shown in Fig. 4b. The naming conventions for each case are as follows: Numbers following ‘RM’ denote \bar{r} (e.g., ‘RM8’ corresponds to cases with $\bar{r} = 8 \mu\text{m}$). ‘B’ denotes cases without cut-off DSD. Numbers following ‘T’ indicate ε (e.g., T16 corresponds to cases with $\varepsilon = 16 \text{ cm}^2 \text{ s}^{-3}$). The units of $\mu_{100,c}$ and $\mu_{10\%,c}$ are in s, Φ_{100} in $\mu\text{m}^{-1.852}$ and $\Phi_{10\%}$ in $\mu\text{m}^{-6.78}$. Units of Φ_{100} and $\Phi_{10\%}$ are determined by Eq. 5 with respective a_α , b_α parameters, where the unit of n_{PE} is μm and n_{PE} is unit-less. The subscript α is 100 or 10% for μ_{100} and $\mu_{10\%}$, respectively. Thus, the units of k_α for μ_{100} and $\mu_{10\%}$ are $\mu\text{m}^{1.852} \text{ s}$ and $\mu\text{m}^{6.78} \text{ s}$, respectively. In this study, these parameters are mainly used to compare how critical threshold varies in different cases than to obtain actual values.

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Table B1. Parameters for fitting function of μ_{100}

	RM8	RM10	RM12	RM14	RM8B	
$\Phi_{100,c}$	8.54×10^{-5}	5.05×10^{-5}	3.69×10^{-5}	3.32×10^{-5}	8.50×10^{-5}	
$\mu_{100,c}$	2388.73	1173.81	880.46	870.40	2381.12	
k_{100}	2.47×10^7	1.83×10^7	1.77×10^7	1.85×10^7	2.48×10^7	
r^2	0.99	0.98	0.98	0.97	0.99	
	RM10B	RM12B	RM14B	RM10-T16	RM10-T80	RM10-T100
$\Phi_{100,c}$	4.08×10^{-5}	2.16×10^{-5}	1.33×10^{-5}	5.01×10^{-5}	4.62×10^{-5}	4.60×10^{-5}
$\mu_{100,c}$	1011.30	607.01	430.81	1089.05	906.69	865.67
k_{100}	1.89×10^7	1.92×10^7	2.05×10^7	1.73×10^7	1.55×10^7	1.47×10^7
r^2	0.98	0.97	0.95	0.98	0.97	0.97



Table B2. Parameters for fitting function of $\mu_{10\%}$

	RM8	RM10	RM12	RM14	RM8B	
$\Phi_{10\%,c}$	1.93×10^{-2}	1.51×10^{-2}	1.31×10^{-2}	1.23×10^{-2}	1.93×10^{-2}	
$\mu_{10\%,c}$	3128.99	1592.18	1205.14	1207.07	3124.57	
$k_{10\%}$	2.21×10^5	1.49×10^5	1.30×10^5	1.35×10^5	2.21×10^5	
r^2	0.78	0.85	0.89	0.90	0.78	
	RM10B	RM12B	RM14B	RM10-T16	RM10-T80	RM10-T100
$\Phi_{10\%,c}$	1.41×10^{-2}	1.10×10^{-2}	8.42×10^{-3}	1.50×10^{-2}	1.41×10^{-2}	1.41×10^{-2}
$\mu_{10\%,c}$	1412.81	773.39	472.91	1480.90	1212.93	1151.98
$k_{10\%}$	1.47×10^5	1.00×10^5	8.00×10^4	1.41×10^5	1.26×10^5	1.20×10^5
r^2	0.87	0.92	0.91	0.85	0.85	0.85

Author contributions. JSL, YN, HL, and FH conceived the original conceptualization and interpretation of results and contributed to discussions. FH provided the base model and JSL modified the base model and formally analyzed the results. JSL wrote the original draft and JSL, YN, FH, and HL contributed to the review and editing. YN and FH provided the funding acquisition for the study and project administration.

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