

## Authors' Response to Reviews of

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

Jung-Sub Lim, Yign Noh, Hyunho Lee, Fabian Hoffmann  
Atmospheric Chemistry and Physics,

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RC: Reviewers' Comment, AR: Authors' Response, □ Manuscript Text

\*All line numbers in this response letter refer to the revised manuscript unless stated.

**RC:** *This paper presents a numerical investigation into the role of low-concentration, large-sized droplets, referred to as precipitation embryos (PEs), in the formation of rain in warm clouds. The study employs box simulations focused exclusively on the process of collision-coalescence. The authors utilize a "one-to-one" Lagrangian cloud model (LCM), which is an appropriate choice for capturing the detailed mechanisms of collision-coalescence. By modeling small cloud volumes (less than one cubic meter) and excluding other in-cloud processes, the study provides qualitative insights into the role of PEs in rain initiation. However, this narrow focus allows the authors to explore a broad range of droplet sizes and concentrations, as well as the effects of turbulence-induced collision enhancement (TICE). The results of the study corroborate previously observed phenomena, such as the competition between PEs and TICE, and the particularly significant role of PEs in clouds that otherwise would not precipitate. While the findings themselves may not be particularly groundbreaking, they contribute to a deeper understanding of warm-rain formation. Notably, the paper introduces a useful formalism for determining the minimum number and size of PEs required to influence rain formation. I believe this study is a valuable contribution to the field and would recommend its publication, provided the authors address the following points:*

**AR:** Dear Reviewer, we sincerely appreciate your time and effort in reviewing our manuscript and providing valuable feedback, which has helped clarify the discussion. We have revised the manuscript accordingly. Below you will find a point-by-point response from the authors.

### 1. Comment 1

**RC:** *Clarification of Non-PE Concentrations: It is unclear what non-PE concentrations were used. Based on line 81, these appear to be 238, 456, and 523 droplets per cubic centimeter, but the caption of Figure 1 provides conflicting information. It would also be beneficial to ensure that there is a case representing a clear marine concentration of approximately 100 droplets per cubic centimeter, given that sea-spray aerosols are a known source of PEs.*

**AR:** The text in line 81 contained a misprint and the number concentrations provided in the label of Figure 1 are correct. We changed the text accordingly. Note that the two cases with  $N_0 = 138$  and  $87 \text{ cm}^{-3}$  and  $\bar{r} = 12, 14 \text{ }\mu\text{m}$ , represent typical clean maritime conditions.

**L92:** In these cases,  $N_0 = 466, 138, \text{ and } 87 \text{ cm}^{-3}$  to achieve the same  $q_c = 1.0 \text{ g kg}^{-1}$  (Fig. 1).

## 2. Comment 2

- RC:** *Range of Dissipation Rates for TICE: The paper tests TICE for dissipation rates of 16, 80, and 100 cm<sup>2</sup> s<sup>-3</sup>. However, the values of 80 and 100 cm<sup>2</sup> s<sup>-3</sup> are quite similar, and all the tested rates fall within the typical range for cumulus clouds (10–100 cm<sup>2</sup> s<sup>-3</sup>). It would be more informative to include dissipation rates representative of other cloud types, such as stratocumulus, cumulus, and deep convection, to broaden the study’s applicability.*
- AR:** Now we change the considered  $\varepsilon$  values to 5, 10, 50, 100, 200 cm<sup>2</sup> s<sup>-3</sup> which accounts for typical stratocumulus, shallow cumulus, and deep convective clouds.

**L118:** To investigate the effect of TICE, five different kinetic energy dissipation rates  $\varepsilon = 5, 10, 50, 100,$  and 200 cm<sup>2</sup> s<sup>-3</sup> are considered for RM10.

## 3. Comment 3

- RC:** *Discussion of PE Characteristics in Real Clouds: The paper would benefit from a discussion of the expected number and size of different types of PEs in real clouds, along with an assessment of their likely significance in natural cloud systems.*
- AR:** We appreciate this comment. We now discuss PE concentrations compared to the atmospheric conditions:

**L45:** Lastly, there is a large uncertainty in the number concentration of PEs in clouds (Khain, 2009). For instance, PEs originating from 1–20  $\mu\text{m}$  sea salt aerosols exhibit a wide range of concentrations from  $10^{-4}$  to  $10^{-2}$  cm<sup>-3</sup> (Jung et al., 2015; Jensen & Nugent, 2017), with a strong environmental and spatial dependency (Woodcock, 1953; Jung et al., 2015). Based on the “one in a million” definition of “lucky droplet” acting as PEs (e.g., Kostinski & Shaw, 2005), typical cloud droplet concentrations over the ocean and continents ( $10^1$  to  $10^3$  cm<sup>-3</sup>) imply PE concentrations of  $10^{-5}$  to  $10^{-3}$  cm<sup>-3</sup>. On the other hand, for climate-engineering practices such as cloud seeding, the concentration of seeded particles can exceed natural values, ranging from  $10^{-1}$  to  $10^1$  cm<sup>-3</sup> (Kuba & Murakami, 2010). Due to this large variability, assessing the PE effect for a broad range of PE concentrations is important.

**L99:** We choose a minimum  $n_{\text{PE}} = 1$  to investigate whether a ‘one in a million’ PE can accelerate droplet collision, as highlighted in previous studies on lucky droplets (Kostinski & Shaw, 2005; Dziekan & Pawlowska, 2017). Within a given reference volume, the minimum and maximum  $n_{\text{PE}}$  values of 1 and 1000 correspond to concentrations of approximately  $2.97 \times 10^{-4}$  cm<sup>-3</sup> and  $2.97 \times 10^{-1}$  cm<sup>-3</sup>, respectively, reflecting the wide range of PE concentrations observed in nature (Khain, 2009; Jung et al., 2015).

**L147:** For RM10, when  $n_{\text{PE}} = 3$ , the PE number concentration is approximately  $10^{-3}$  cm<sup>-3</sup>. In this case, even PEs larger than 40  $\mu\text{m}$  are not effective in accelerating  $t_{10\%}$  (Fig. 2b). However, when the PE concentration increases to a relatively high value ( $n_{\text{PE}} = 30$ ), PEs larger than 22  $\mu\text{m}$  can substantially accelerate  $t_{10\%}$  (Fig. 2b). Such high PE concentrations are uncommon but have been observed in certain oceanic conditions (Jung et al., 2015). In contrast, for RM14, which represents typical maritime clouds in a pristine environment with  $N_0 = 87$  cm<sup>-3</sup>, the effect of PEs is reduced. PEs smaller than 33  $\mu\text{m}$  are unable to accelerate  $\mu_{100}$  regardless of  $n_{\text{PE}}$  (Fig. 2c). Moreover,  $t_{10\%}$  is accelerated only when both  $n_{\text{PE}}$  and  $r_{\text{PE}}$  are very large (Fig. 2d). However, such extreme conditions

are uncommon in typical maritime environments. This suggests that the impact of PEs depends on the initial DSD shape, requiring a collisionally stable cloud for a substantial effect.

**L315:** In this study, PEs larger than  $22 \mu\text{m}$  are found to effectively accelerate the precipitation ( $t_{10\%}$ ) for clouds in relatively polluted environments, when their concentration exceeds  $10^{-3} \text{ cm}^{-3}$ , consistent with Feingold et al. (1999). While this PE concentration falls within the range observed for giant sea-salt particles over the ocean (Jung et al., 2015), for clouds in a pristine environment, substantially higher numbers and sizes of PE are required to achieve effective precipitation acceleration. These observations are based on measurements of sea-salt aerosols ( $2\text{--}20 \mu\text{m}$ ), which have large solution masses and corresponding large equilibrium sizes. However, under atmospheric conditions, these particles might not have sufficient time to grow to their equilibrium size (Ivanova et al., 1977), potentially resulting in lower PE concentrations. On the other hand, it is possible that PE concentrations can increase through stochastic collisions (Kostinski & Shaw, 2005; Dziekan & Pawlowska, 2017) as the cloud evolves. Furthermore, because the critical threshold decreases in DSDs with higher *collisional stability*, the effect of PEs is expected to be especially strong in non-precipitating, or polluted clouds as suggested in previous studies (e.g., Johnson, 1993; Dziekan et al., 2021).

#### 4. Comment 4

**RC:** *Interpretation of Function  $\Phi$  in Equation (5): The function  $\Phi$ , defined in Equation (5), describes the impact of PEs by combining their number and size. However, it is a decreasing function of PE size and concentration, which makes the plots harder to interpret. It might be more intuitive if  $\Phi$  were an increasing function, and if  $\Phi = 0$  corresponded to the absence of PEs.*

**AR:** We appreciate this reviewer's comment. We have re-defined  $\Phi = n_{\text{PE}}^a r_{\text{PE}}^b$ . Now,  $\Phi = 0$  represents the absence of PEs. We believe that Equation (5) becomes more intuitive due to this change.

**L160:** Thus, we write

$$\mu_\alpha = c_\alpha - k_\alpha n_{\text{PE}}^{a_\alpha} r_{\text{PE}}^{b_\alpha} = c_\alpha - k_\alpha \Phi_\alpha(n_{\text{PE}}, r_{\text{PE}}) \quad (1)$$

for a  $\mu_\alpha$  exceeding the critical threshold.

#### 5. Comment 5

**RC:** *Definition of Rain in the 10 % Mass Conversion: It is unclear how rain is defined when calculating the time required to convert 10 % of the total cloud mass to rain. Is the initial presence of rainwater (e.g., from PEs already exceeding the rain threshold at  $t = 0$  s) included in the 10 % mass calculation?*

**AR:** It is defined as 10 % of initial  $q_c + q_r$ , where  $q_r > 0$  when  $r_{\text{PE}} > 40 \mu\text{m}$  and  $n_{\text{PE}} > 0$ . Now, we explain this in the manuscript more clearly.

**L131:** Adding PEs increases the initial  $q_c$ , or the rainwater mixing ratio  $q_r$  when  $r_{\text{PE}} > 40 \mu\text{m}$  and  $n_{\text{PE}} > 0$ , potentially limiting the comparability of simulated cases. To address this, we restricted the analysis of  $t_{10\%}$  and further conversion rates such as autoconversion rate (i.e., raindrop formation by

collisions between cloud droplets), and accretion rate (i.e., raindrop growth by raindrops collecting cloud droplets) to cases where the increase in the initial  $q_c + q_r$  due to the addition of PEs is below 2 %. In most cases, the increase in  $q_c$  and  $q_r$  is below 1 %. However, two exceptions,  $n_{PE} = 300$ , with  $r_{PE} = 40 \mu\text{m}$  and  $n_{PE} = 1000$ , with  $r_{PE} = 27 \mu\text{m}$ , show an increase of 1.9 %.

## 6. Comment 6

**RC:** *Interpretation of Results Regarding the 'Lucky Droplet' Theory: The statement on line 190—"This suggests that while PEs can accelerate the formation of the largest raindrops, these droplets may not substantially impact overall rain initiation after the initial period when they are few"—is described as contradicting the 'lucky droplet' theory. However, I disagree with this interpretation. The results show that when there are too few lucky droplets (PEs), they do not significantly impact the time to convert 10 % of the cloud mass into rain. Nonetheless, when more lucky droplets are present, they do affect this time ( $\pm 10$  %). Furthermore, even if  $t_{10\%}$  averaged over small cloud volumes is not affected, the presence of lucky droplets could still play an important role in rain formation in real clouds.*

**AR:** We agree that the original statement was misleading. We have revised the argument to state that while  $t_{100}$  does not necessarily ensure a shorter  $t_{10\%}$  when PEs are too few, the presence and continued generation of lucky droplets can indeed increase the PE concentration and influence rain initiation.

**L234:** This suggests that while PEs can accelerate the formation of the largest raindrop, these droplets may not directly impact the overall rain mass growth when the number of PEs is low.

**L321:** On the other hand, it is possible that PE concentrations can increase through stochastic collisions (Kostinski & Shaw, 2005; Dziekan & Pawlowska, 2017) as the cloud evolves.

## 7. Technical Comments

**RC:** *Line 135: There is an unclosed parenthesis.*

**AR:** Done.

## References

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\*All line numbers in this response letter refer to the revised manuscript unless stated.

**RC:** *This paper offers a fresh perspective on the role that rare large droplets, here named “precipitation embryos (PEs)”, can have on precipitation formation from a droplet distribution which would otherwise undergo slower collision-coalescence (akin to the “luck-drops” theory). The study is well-motivated and the use of the superdroplet model throughout is particularly appropriate. The conclusions drawn are well-grounded and are worthwhile for our understanding of the timing of rain formation. They certainly further our knowledge of how sufficiently numerous and large droplets can accelerate droplet collision-coalescence, and they also give particularly useful insight to how turbulence-induced collision enhancement (TICE) - another process known to accelerate rain formation - can either complement or inhibit the acceleration caused by large droplets. Indeed I believe the study to be within the scope of ACP and I recommend that the editor consider publishing this work. Nevertheless I have some specific comments I would like to see addressed first, particularly with regard to section 3 and the decision to truncate the droplet size distribution (DSD) for almost all the analysis.*

**AR:** Dear reviewer, we highly appreciate your effort and time to evaluate this manuscript and provide fruitful comments that greatly improved the quality of the paper. We have revised our manuscript accordingly. Below you will find a point-by-point response from the authors.

### 1. Comment 1

**RC:** *Most critically, I am unconvinced by the decision to truncate the DSD at 20 m and I believe the analysis and conclusions of the paper would be clearer if they were presented without this cut-off. The truncation is justified as creating a “stable initial DSD in which collisions are negligible” however, as for example the black lines in figure 6 show, the truncated DSDs do undergo frequent collisions and so the truncation appears unjustified. Contrary to the Author’s introduction, I believe such examples also mean the role of “precipitation embryos” (PEs) in inducing collisions in an otherwise stable distribution is not addressed in this paper. The paper is better introduced as showing how PEs can accelerate the timing of rain formation for a DSD in which droplets already can collide. I am of the opinion that the analysis would be clearer, and the conclusions more convincing, if the cut-off was removed. For example the red and green lines in figures 4(a) and 4(b) (i.e. the RM12 and RM14 setups) show large changes between figures 4(a) and 4(b), and 4(c) and 4(d), ie. when the un-truncated distribution is used instead. Likewise figure 5 shows large differences in the results without truncation. This means the behaviour of the critical threshold is heavily influenced by the cutoff at 20 m and the analysis of the role of PEs, particularly in section 3, would be more convincingly shown if that influence wasn’t present.*

**AR:** We also agree with the reviewer that DSD without cut-off represents a more natural DSD. The original

motivation for the cut-off was to isolate the effects of PEs by starting from an initially *stable* DSD, in which droplets are not large enough to initiate frequent collisions. However, as the reviewer correctly pointed out, collisions can still occur in the truncated DSDs, even without PEs. To address this, we have revised the manuscript by making the case without the DSD cut-off (*broad cases*) as the base case and renamed the cases with DSD cut-off as *narrow cases*.

AR: In particular, Sec. 3 now shows the results without the DSD cut-off. The *narrow cases* are only used to discuss how the PE critical value changes with DSD shape, particularly with respect to the largest initial droplet size. We note that in cases where  $\bar{r} = 10 \mu\text{m}$  (light blue lines in Fig.1), there are very few cloud droplets larger than  $20 \mu\text{m}$  even without the DSD cut-off. Thus, the conclusions in Sec. 3 are not substantially different from the original manuscript.

## 2. Comment 2

RC: **L3 (and repeated): please reconsider the use of the term “colloidally stable”. The terminology is misleading because it is conventionally used to discuss the condition of a colloid substance - i.e. the mixture of soluble and insoluble substances in a solute.**

AR: Although Squires (1958) originally used the term “colloidal stability” to describe the ability of a DSD to initiate precipitation, we agree with the reviewer that this term has a different conventional meaning in other areas of the scientific community. Therefore, we decided to use the term *collisional stability*, and defined it in the introduction.

**L16:** These collision-limited DSDs can be regarded as being in a *collisionally stable* state (Squires, 1958), where mechanisms that accelerate the collision-coalescence process to form raindrops and initiate precipitation are crucial for breaking this stability.

## 3. Comment 3

RC: **L22 (“so-called precipitation embryos (PEs)”): is the term “precipitation embryo” coined in this paper? If not, please provide a citation here. If so, please consider if there is any existing terminology you can use instead. Why for example is the term “lucky droplet” not appropriate?**

AR: The term precipitation embryos was originally used to describe large droplets that initiate warm rain in earlier studies (e.g., Pontikis et al., 1987; Johnson, 1993). In this study, we focus on the effects of large droplets that initiate warm rain, regardless of their origin—whether from giant aerosol particles, “lucky droplets” arising from stochastic collisions, or seeded particles. While the term “lucky droplet” implies a droplet form from a series of unlike stochastic collisions, and “giant aerosol” refers to initially large droplets, we find that the broader concept of precipitation embryos which encompasses large droplets formed through multiple potential mechanisms, most suitable for this study. We have cited Johnson (1993), as it provides a definition most closely aligned with the concept used in our research.

**L24:** ... and (iii) (iii) the role of so-called precipitation embryos (PEs) (e.g., Johnson, 1993), the primary focus of this study.

#### 4. Comment 4

RC: *L81 (“ $N_0 = 238 \text{ cm}^{-3}$ ,  $456 \text{ cm}^{-3}$  and  $523 \text{ cm}^{-3}$ ”): the values for  $N_0$  given here do not match those in the legend of figure 1 so please correct them. Also it would be clearer if their order was consistent with the ordering of in the sentence before (i.e. if  $\bar{r}$  is ascending,  $N_0$  is descending).*

AR: Thank you for pointing this out. We have corrected the values in the text to match those in the legend of Figure 1, which were correct. We also reordered the values to ensure that when  $\bar{r}$  is ascending,  $N_0$  is descending, as suggested.

**L92:** In these cases,  $N_0 = 466$ ,  $138$ , and  $87 \text{ cm}^{-3}$  to achieve the same  $q_c = 1.0 \text{ g kg}^{-1}$  (Fig. 1).

#### 5. Comment 5

RC: *Figure 1: These DSDs do not show very clearly the alteration you make by truncating these distributions, nor do they show the impact of adding PEs on them. It would be helpful to the reader to include a neighbouring figure (or to replace this one) with a plot that shows the truncated DSDs and some DSDs which example the distribution with  $n_{\text{PE}}, r_{\text{PE}}$  values.*

AR: Thank you for the recommendation. We have incorporated a second panel in Figure 1 to address your suggestion. This panel now illustrates the truncated DSDs and includes an example PE distribution with  $r_{\text{PE}} = 27 \mu\text{m}$  and different  $N_{\text{PE}}$  values to provide a comprehensive representation of the DSD cut-off and PE addition. Moreover, the DSD with added PE ( $r_{\text{PE}} = 27 \mu\text{m}$  and  $n_{\text{PE}} = 1000$ ) for RM10 is shown as the dashed line in panel (a).

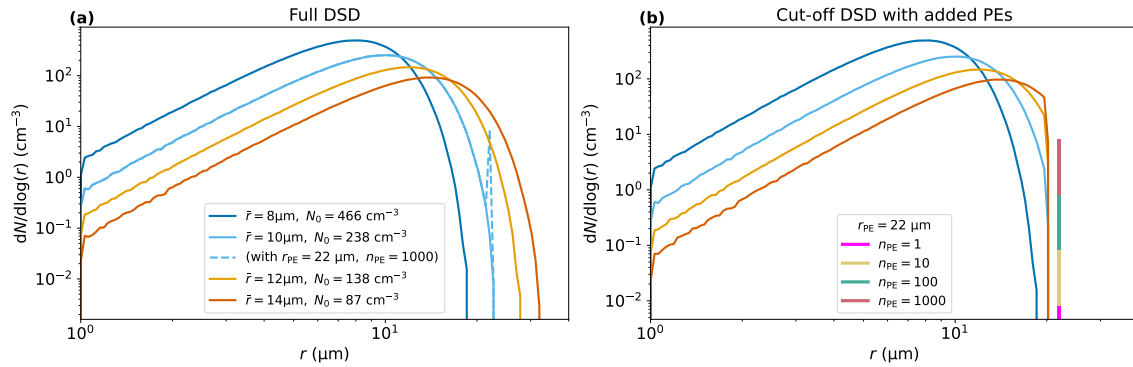


Figure 1: (a) Initial DSDs for various  $\bar{r}$  and their corresponding  $N_0$  values. The dashed line represents the DSD with  $r_{\text{PE}} = 27 \mu\text{m}$  and  $n_{\text{PE}} = 1000$ . b) Initial DSDs with a DSD cut-off for various  $\bar{r}$  and  $N_0$  values, along with a vertical bar plot showing various PE distributions for  $r_{\text{PE}} = 27 \mu\text{m}$  and various  $n_{\text{PE}}$  values. Excerpted from Fig. 1 of the manuscript.



## 6. Comment 6

- RC:** *L96 (“To explore the impact of PEs, we investigate 49 ensemble simulations...”): it’s quite important that this is justified with a comparison to the real world. How do these PE radii and number concentrations compare with observations? Especially relevant would be to put them in the context of shallow convective clouds, since that’s the range of TICE you consider later on, or cloud seeding. Context is particularly necessary because figure 2 shows both  $\mu_{100}$  and  $\mu_{10\%}$  have almost no dependency on a large portion of the  $n_{PE}, r_{PE}$  space spanned by the ensemble.*
- AR:** We agree with the reviewer that providing the observed range of  $n_{PE}$  and  $r_{PE}$  values is essential for context. In the revised manuscript, we have included references to observed PE number concentrations across different cloud types. Notably, the smallest  $n_{PE} = 1$  is lower than typically observed PE concentrations. However, this value was included to explore the “lucky droplet” effect, where a single “one-in-a-million” droplet is expected to accelerate the collision process. Now we relate the PE concentrations considered in this study to the observed values over the ocean to incorporate the reviewer’s comment.
- AR:** In addition, we have excluded cases with  $r_{PE} = 15 \mu\text{m}$  in the revised manuscript to improve clarity. While these cases were initially included to demonstrate the limited effect of small PEs ( $r_{PE} < 20 \mu\text{m}$ ), the effect is sufficiently represented by the cases with  $r_{PE} = 18 \mu\text{m}$  making the discussion clearer.

**L45:** Lastly, there is a large uncertainty in the number concentration of PEs in clouds (Khain, 2009). For instance, PEs originating from 1–20  $\mu\text{m}$  sea salt aerosols exhibit a wide range of concentrations from  $10^{-4}$  to  $10^{-2} \text{ cm}^{-3}$  (Jung et al., 2015; Jensen & Nugent, 2017), with a strong environmental and spatial dependency (Woodcock, 1953; Jung et al., 2015). Based on the “one in a million” definition of “lucky droplet” acting as PEs (e.g., Kostinski & Shaw, 2005), typical cloud droplet concentrations over the ocean and continents ( $10^1$  to  $10^3 \text{ cm}^{-3}$ ) imply PE concentrations of  $10^{-5}$  to  $10^{-3} \text{ cm}^{-3}$ . On the other hand, for climate-engineering practices such as cloud seeding, the concentration of seeded particles can exceed natural values, ranging from  $10^{-1}$  to  $10^1 \text{ cm}^{-3}$  (Kuba & Murakami, 2010). Due to this large variability, assessing the PE effect for a broad range of PE concentrations is important.

**L99:** We choose a minimum  $n_{PE} = 1$  to investigate whether a ‘one in a million’ PE can accelerate droplet collision, as highlighted in previous studies on lucky droplets (Kostinski & Shaw, 2005; Dziekan & Pawlowska, 2017). Within a given reference volume, the minimum and maximum  $n_{PE}$  values of 1 and 1000 correspond to concentrations of approximately  $2.97 \times 10^{-4} \text{ cm}^{-3}$  and  $2.97 \times 10^{-1} \text{ cm}^{-3}$ , respectively, reflecting the wide range of PE concentrations observed in nature (Khain, 2009; Jung et al., 2015).

**L147:** For RM10, when  $n_{PE} = 3$ , the PE number concentration is approximately  $10^{-3} \text{ cm}^{-3}$ . In this case, even PEs larger than 40  $\mu\text{m}$  are not effective in accelerating  $t_{10\%}$  (Fig. ??b). However, when the PE concentration increases to a relatively high value ( $n_{PE} = 30$ ), PEs larger than 22  $\mu\text{m}$  can substantially accelerate  $t_{10\%}$  (Fig. ??b). Such high PE concentrations are uncommon but have been observed in certain oceanic conditions (Jung et al., 2015). In contrast, for RM14, which represents typical maritime clouds in a pristine environment with  $N_0 = 87 \text{ cm}^{-3}$ , the effect of PEs is reduced. PEs smaller than 33  $\mu\text{m}$  are unable to accelerate  $\mu_{100}$  regardless of  $n_{PE}$  (Fig. ??c). Moreover,  $t_{10\%}$  is accelerated only when both  $n_{PE}$  and  $r_{PE}$  are very large (Fig. ??d). However, such extreme conditions are uncommon in typical maritime environments. This suggests that the impact of PEs depends on the initial DSD shape, requiring a collisionally stable cloud for a substantial effect.

**L315:** In this study, PEs larger than  $22 \mu\text{m}$  are found to effectively accelerate the precipitation ( $t_{10\%}$ ) for clouds in relatively polluted environments, when their concentration exceeds  $10^{-3} \text{ cm}^{-3}$ , consistent with Feingold et al. (1999). While this PE concentration falls within the range observed for giant sea-salt particles over the ocean (Jung et al., 2015), for clouds in a pristine environment, substantially higher numbers and sizes of PE are required to achieve effective precipitation acceleration. These observations are based on measurements of sea-salt aerosols ( $2\text{--}20 \mu\text{m}$ ), which have large solution masses and corresponding large equilibrium sizes. However, under atmospheric conditions, these particles might not have sufficient time to grow to their equilibrium size (Ivanova et al., 1977), potentially resulting in lower PE concentrations. On the other hand, it is possible that PE concentrations can increase through stochastic collisions (Kostinski & Shaw, 2005; Dziekan & Pawlowska, 2017) as the cloud evolves. Furthermore, because the critical threshold decreases in DSDs with higher *collisional stability*, the effect of PEs is expected to be especially strong in non-precipitating, or polluted clouds as suggested in previous studies (e.g., Johnson, 1993; Dziekan et al., 2021).

## 7. Comment 7

**RC:** *L102 (“larger PEs can substantially increase the initial  $q_c$ ”): please state how much your simulations are perturbed by the presence of PEs. For example stating what the largest  $q_c$  of your ensemble is (when  $n_{\text{PE}}, r_{\text{PE}} = 1000, 18$ )*

**AR:** Thank you for this comment. In response, we have quantified the perturbations caused by the addition of PEs. The maximum increases in  $q_c + q_r$  are 12.5% and 6.4%, respectively, which were excluded from the original analysis. To address this, we have now introduced a criterion whereby only cases are included in the analysis in which  $q_c + q_r$  increase less than 2% due to the addition of PEs (see the values in Fig. 2 of this letter). For most cases, the increase in  $q_c + q_r$  caused by PEs is below 1%.

**L131:** Adding PEs increases the initial  $q_c$ , or the rainwater mixing ratio  $q_r$  when  $r_{\text{PE}} > 40 \mu\text{m}$  and  $n_{\text{PE}} > 0$ , potentially limiting the comparability of simulated cases. To address this, we restricted the analysis of  $t_{10\%}$  and further conversion rates such as autoconversion rate (i.e., raindrop formation by collisions between cloud droplets), and accretion rate (i.e., raindrop growth by raindrops collecting cloud droplets) to cases where the increase in the initial  $q_c + q_r$  due to the addition of PEs is below 2%. In most cases, the increase in  $q_c$  and  $q_r$  is below 1%. However, two exceptions,  $n_{\text{PE}} = 300$ , with  $r_{\text{PE}} = 40 \mu\text{m}$  and  $n_{\text{PE}} = 1000$ , with  $r_{\text{PE}} = 27 \mu\text{m}$ , show an increase of 1.9%.

## 8. Comment 8

**RC:** *L213 (“although  $t_{100}$  can be shorter than in the cases without PEs.”): please provide a suggested explanation or citation for this statement.*

**AR:** This sentence refers that  $t_{100}$  is shorter in the cases with small  $n_{\text{PE}}$  values compared to the  $t_{100}$  value in the case without PEs. We acknowledge the reviewer’s concern about the ambiguity and changed the sentence accordingly.

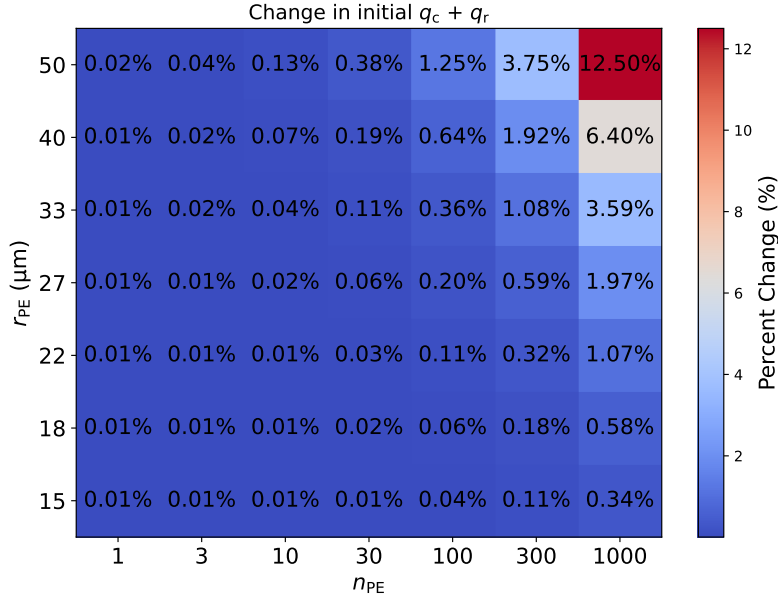


Figure 2: Heatmap of change in initial  $q_c + q_r$  by PEs for different  $r_{PE}$  and  $n_{PE}$  values.

**L253:** Thus, although larger PEs are more likely to collide, the overall collision frequency remains low when  $n_{PE}$  is small, resulting in slower PE autoconversion compared to non-PE autoconversion. While non-PE-autoconversion always decreases with increasing  $n_{PE}$ , PE-autoconversion increases substantially only for  $n_{PE} \geq 100$ . 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. Hence, shorter  $t_{100}$  does not necessarily lead to a shorter  $t_{10\%}$  when  $n_{PE}$  is small (Fig. 2).

## 9. Comment 9

**RC:** *Figure 6: With increasing PE concentration, the accretion and auto-conversion rates due to non-PE collisions maximise earlier. A acknowledgement and a possible reason for this behaviour would be beneficial because it is not self-evident why it should occur.*

**AR:** This behavior occurs because PE particles suppress non-PE autoconversion, as discussed in this paper. As shown in Fig. 6c, the non-PE autoconversion rate is nearly identical across all cases during the initial 1000–1200 s, regardless of PE concentration. However, when the PE effect is stronger, more non-PE droplets are collected by PEs, reducing the number of droplets available for autoconversion. As a result, in cases with higher PE concentrations, the autoconversion rate peaks earlier and then begins to decline. This suppression of non-PE autoconversion also decreases the number of non-PE raindrops, which reduces the non-PE accretion rate and causes it to peak earlier in these cases. These findings suggest that the primary role of PEs is to collect non-PE droplets, while indirectly suppressing both non-PE autoconversion and accretion. We have included this explanation in the revised manuscript.

**L272:** Interestingly, at high  $n_{PE}$ , the non-PE autoconversion and accretion rates reach their peak values earlier than in cases without PEs or with low  $n_{PE}$  (Fig. 6c, d, e, and f). During the initial 1000 s, the non-PE autoconversion rate is nearly identical across all cases, regardless of  $n_{PE}$ . However, when  $n_{PE}$  is high, more non-PE droplets are collected by PEs, reducing the number of droplets available for autoconversion. As a result, the non-PE autoconversion rate peaks and declines earlier in cases with higher PE concentrations. This suppression of non-PE autoconversion decreases the number of non-PE raindrops and the non-PE accretion rate. These findings highlight that the primary role of PEs is to collect non-PE droplets, which might suppress non-PE autoconversion and accretion.

## 10. Technical Comments

**RC:** *L11 (“A key question in warm rain initiation is to explain...”): There is no question posed here, consider rephrasing the second half of the sentence into a question, or rewording ”a key question”.*

**AR:** Done. Now, we address this as a key challenge.

**RC:** *L36 (“In particular, if we consider ... Thus, it is also important to account for stochastic fluctuations in the collision process.”): please rephrase this sentence because this logic is hard to follow. I was left asking myself: why is a 12.5  $\mu m$  droplet relevant to your study, and how does this citation justify the need for stochastic fluctuations? I assume you mean that by including stochasticity you don’t need as large lucky droplets to initiate collisions, but please make this more explicit if so.*

**AR:** We agree with the reviewer that the reference to a 12.5  $\mu m$  droplet is not directly relevant to our study. The original intention was to highlight that including stochasticity reduces the need for larger or more PEs to initiate rain. To address the reviewer’s concerns, we have rephrased the sentence to clarify this point and provide a better context for the importance of collision stochasticity.

**L39:** In the absence of PEs, DSDs with small-sized droplets barely initiate precipitation unless stochastic fluctuations in the collision process are considered. This phenomenon is known as the “lucky droplet” effect, which may produce PEs on its own (Telford, 1955; Kostinski & Shaw, 2005; Dziekan & Pawlowska, 2017). When this effect dominates, adding only a few PEs may not substantially accelerate rain initiation.

**RC:** *L56 (“frequently called superdroplets by introducing a weighting factor”): please add missing comma after “superdroplets”.*

**AR:** Done

**RC:** *L110 (“The timescale  $t_{10\%}$  represents the time when 10 % of the initial cloud droplet mass converts to rain...”): is rain here defined as above 40 or 100 microns?*

**AR:** A raindrop is defined as a droplet larger than 40  $\mu m$ , in radius in our study. We clarified this in the revised manuscript.

**RC:** *L145 (equation 6): Missing subscript on  $\alpha$  on  $\Phi$  the left-hand side of the equation.*

**AR:** Done.

**RC:** *Figure 4 and figure 5: Please reconsider the colours you use to plot these figures because they are unkind*

to colour-blindness (especially figure 4). Also in figure 4, the re-use of the same colours for subplots (e) and (f), although they are no longer denoting different  $\bar{r}$ , is undesirable.

AR: Thank you for your comment. We have now revised all figures to be accessible for color vision deficiency. Also, we assigned new colors to cases with the same  $\bar{r}$  but different  $\varepsilon$  in panels e and f of Figure 4, ensuring that these colors do not overlap with those used in panels a–d.

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# 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 find that 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 to accelerate rain initiation when collisions are already efficient without PEs, e.g., due to a broad droplet size distribution or strong turbulence effect. 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 climate intervention approaches, such as rain enhancement or marine cloud brightening.

## 1 Introduction

A key ~~question in~~ challenge in understanding warm rain initiation is ~~to explain~~ explaining the growth of cloud droplets in the radius range between 15 and 40  $\mu\text{m}$ , 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~~ In the droplet size distributions (DSDs) ~~in a colloidal stable state, where that are too narrow or consist of too small droplets,~~ collisions among droplets ~~are inefficient due to a narrow DSD or too small droplets, mechanisms accelerating and thus precipitation formation are inefficient.~~ These collision-limited DSDs can be regarded as being in a collisionally stable state (Squires, 1958), where mechanisms that accelerate the collision-coalescence process to form ~~a raindrop and initiate the precipitation are key to raindrops and initiate precipitation are crucial for~~ 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, 2024), (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;

25 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) (e.g., Johnson, 1993), the primary focus of this study.

The presence of PEs larger than 20  $\mu\text{m}$  can initiate the collision process as they are already larger than the size-gap range (e.g., Woodcock, 1953; Telford, 1955; Johnson, 1982; Extton et al., 1986; Johnson, 1993; Feingold et al., 1999; Teller and Levin, 2006; Alfonso et al., 2013; Hoffmann et al., 2017; Dziekan et al., 2021). The sources of these PEs can be giant aerosol  
30 particles, predominantly large sea-salt aerosols that form solution droplets having a ~~size-range-radius~~ between 1  $\mu\text{m}$  and 100  $\mu\text{m}$  (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”~~ “lucky droplets” 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 enhance precipitation (Bowen, 1952; Cotton, 1982). In this study, PEs  
35 are broadly defined as large droplets, irrespective of their origin.

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~~  $10^{-3} \text{ cm}^{-3}$  20  $\mu\text{m}$ -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-the~~ droplet number concentration and liquid water content (e.g., Yin et al., 2000). Other studies  
40 indicate that the 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  $\mu\text{m}$ -sized lucky droplet among 10  $\mu\text{m}$  droplets can initiate collisions. Thus, it is also important to account for the absence of PEs, DSDs with small-sized droplets barely initiate precipitation unless~~ stochastic fluctuations in the collision process ~~are considered. This phenomenon is known as the “lucky droplet” effect, which may produce PEs on its~~  
45 own (Telford, 1955; Kostinski and Shaw, 2005; Dziekan and Pawlowska, 2017). When this effect dominates, adding only a few PEs may not substantially accelerate rain initiation. In addition, 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 other in influencing collisional growth.

Lastly, there is a large uncertainty in the number concentration of PEs in clouds (Khain, 2009). For instance, PEs originating  
50 from 1–20  $\mu\text{m}$  sea salt aerosols exhibit a wide range of concentrations from  $10^{-4}$  to  $10^{-2} \text{ cm}^{-3}$  (Jung et al., 2015; Jensen and Nugent, 2017), with a strong environmental and spatial dependency (Woodcock, 1953; Jung et al., 2015). Based on the “one in a million” definition of “lucky droplet” acting as PEs (e.g., Kostinski and Shaw, 2005), typical cloud droplet concentrations over the ocean and continents ( $10^1$  to  $10^3 \text{ cm}^{-3}$ ) imply PE concentrations of  $10^{-5}$  to  $10^{-3} \text{ cm}^{-3}$ . On the other hand, for climate-engineering practices such as cloud seeding, the concentration of seeded particles can exceed natural values, ranging  
55 from  $10^{-1}$  to  $10^1 \text{ cm}^{-3}$  (Kuba and Murakami, 2010). Due to this large variability, assessing the PE effect for a broad range of PE concentrations is important.

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



60 consider stochastic fluctuations in collisional growth naturally (e.g., Dziekan and Pawlowska, 2017; Li et al., 2022). While 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 the 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.

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 ~~See 6.~~ Section 5.

## 2 Model and Simulation Setup

### 70 2.1 Lagrangian Cloud Box Model

In most applications, each computational particle of an LCM represents a large number of real droplets with identical properties, 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

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

75 where  $\Delta V$  is a reference volume, and  $n_{\text{ptcl}}$  represents the number of computational particles in  $\Delta V$ . 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

$$80 \quad p_{m,n} = \frac{K}{\Delta V} \frac{K_{m,n}}{\Delta V} \delta t, \quad (2)$$

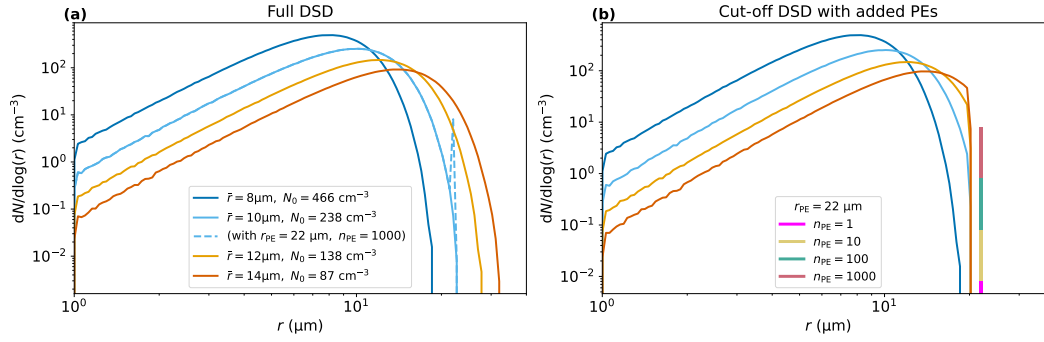
primarily determined by the ~~collection kernel~~ gravitational collection kernel

$$K_{\underline{m},\underline{n}} = \pi(r_m + r_n)^2 E(r_m, r_n) |w(r_m) - w(r_n)|, \quad (3)$$

where  $r_m$  and  $r_n$  are the radii of the interacting droplets,  $E$  the collision efficiency of droplet pairs (Hall, 1980), and  $w$  the droplet terminal velocity (Beard, 1976), ~~and~~  $\delta t$  is 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





**Figure 1.** (a) Initial DSDs for various  $\bar{r}$  and their corresponding  $N_0$  values. The dotted-dashed line indicates represents the DSD with  $r_{PE} = 27 \mu\text{m}$  and  $n_{PE} = 1000$ . (b) Initial DSDs with a DSD cut-off radius of  $20 \mu\text{m}$  for various  $\bar{r}$  and  $N_0$  values, above which droplets are removed in some cases along with a vertical bar plot showing various PE distributions for  $r_{PE} = 27 \mu\text{m}$  and various  $n_{PE}$  values.

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

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

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  $\bar{r} = 10 \mu\text{m}$  (see orange light blue line in Fig. 1). The DSD results in a cloud water mixing ratio ( $q_c$ ) of approximately  $1.0 \text{ g kg}^{-1}$ . Additionally, cases with  $\bar{r} = 8, 12$ , or  $14 \mu\text{m}$  are considered to investigate the effect of PEs in different DSD shapes. In these cases,  $N_0 = 238, 456$  and  $523 \text{ cm}^{-3}$   $N_0 = 466, 138$ , and  $87 \text{ cm}^{-3}$  to achieve the same  $q_c = 1.0 \text{ g kg}^{-1}$  (Fig. 1). We name these cases ‘RM’, ‘RM’, where RM stands for the mean radius 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  $20 \mu\text{m}$  are removed in selected simulations to prevent them from initiating collisions; 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 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 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  $\epsilon = 16, 80$ , 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 and , which are steered by  $\epsilon$ . When TICE is considered, the case names are amended by a T followed by the value of  $\epsilon$  (e.g., RM10-T100).

A total of  $10^6$  computational particles ( $n_{\text{PEI}} = 10^6$ ) 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  $\delta t = 0.1$  s, the model is integrated for 7200 to account for the slowest realization to complete

110 collisional growth, but the discussion is focused on the initial 2500, capturing the initiation of collisional growth.

To explore the impact of PEs, we investigate 49–42 ensemble simulations, each representing different combinations of PE radii ( $r_{\text{PE}} = 15, 18, 22, 27, 33, 40$ , and  $50$   $\mu\text{m}$ ) and numbers ( $n_{\text{PE}} = 1, 3, 10, 30, 100, 300$ , 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  $1 - 5$   $\mu\text{m}$  giant-sea-salt aerosols (Kuba and Murakami, 2010). We choose a minimum  $n_{\text{PE}} = 1$  to investigate whether a ‘one in a million’ lucky droplets could PE can 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  $r_{\text{PE}} = 15$   $\mu\text{m}$ . Within a given reference volume, the minimum and maximum  $n_{\text{PE}}$  values of 1 and 18  $\mu\text{m}$  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 1000 correspond to concentrations of approximately  $2.97 \times 10^{-4} \text{ cm}^{-3}$  and  $2.97 \times 10^{-1} \text{ cm}^{-3}$ , respectively, reflecting the wide range of PE concentrations observed in nature (Khain, 2009; Jung et al., 2015).

Every setup is simulated 100 times with different random numbers to ensure statistical convergence (cf. Fig. A1). Using a timestep  $\delta t = 0.1$  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. A total of  $10^6$  computational particles ( $n_{\text{ptcl}} = 10^6$ ) are initialized to represent the initial DSD of RM10, resulting in a reference volume  $\Delta V = 3.36 \times 10^{-3} \text{ m}^3$ . For cases with different  $N_0$ ,  $n_{\text{ptcl}}$  scales with  $N_0$  from  $10^6$  at RM10 ( $N_0 = 238 \text{ cm}^{-3}$ ) to  $n_{\text{ptcl}} = 1,953,125$  for RM8 ( $N_0 = 466 \text{ cm}^{-3}$ ), and  $n_{\text{ptcl}} = 364,431$  for RM14 ( $N_0 = 87 \text{ cm}^{-3}$ ). This adjustment applies only to non-PE particles, with  $n_{\text{PE}}$  can be interpreted as concentrations between  $2.97 \times 10^{-4} \text{ cm}^{-3}$  and  $2.97 \times 10^{-1} \text{ cm}^{-3}$ , respectively being varied from 1 to 1000 for all  $N_0$ .

130 Changing  $\bar{m}$  also alters the number and mean radius of droplets larger than  $20$   $\mu\text{m}$ , which are critical for initiating droplet collisions. For instance, the radii of the largest initialized droplets are  $24$  and  $34$   $\mu\text{m}$  for RM10 and RM14, respectively. To isolate the dependency of the PE effect on the DSD shape for smaller droplets, we remove droplets larger than  $20$   $\mu\text{m}$  in specific simulations (Wang et al., 2006; Dziekan and Pawlowska, 2017). This initialization is referred to as a “cut-off DSD” (see the dotted line in Fig. 1). We denote these cases by adding the letter “N” to the naming convention (e.g., RM10N), referring to the resulting narrower DSD.

135 To investigate the effect of TICE, five different kinetic energy dissipation rates  $\varepsilon = 5, 10, 50, 100$ , and  $200 \text{ cm}^2 \text{ s}^{-3}$  are considered for RM10. These  $\varepsilon$  values are chosen to explore the TICE effect across different cloud types, where typical values range from  $1 - 10 \text{ cm}^2 \text{ s}^{-3}$  in stratocumulus clouds,  $10 - 100 \text{ cm}^2 \text{ s}^{-3}$  in shallow 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). TICE is incorporated in Eq. 3 using the parameterizations developed by Ayala et al. (2008) and Wang and Grabowski (2009), which are steered by  $\varepsilon$ . When TICE is considered, the case names are amended by a T followed by the numerical value of  $\varepsilon$  in  $\text{cm}^2 \text{ s}^{-3}$  (e.g., RM10-T100).

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

(Dziekan and Pawlowska, 2017). In this study, a raindrop is defined as a droplet larger than  $40\text{ }\mu\text{m}$ . As PEs considered in this study can be ~~larger than the typical raindrop radius, i.e., over  $40\text{ }\mu\text{m}$ ,~~ raindrops already, we define  $t_{100}$  as the time required for the formation of the first  $100\text{ }\mu\text{m}$  droplet, i.e., a sufficiently large droplet that stimulates subsequent collisions (Kostinski and Shaw, 2005; Alfonso et al., 2019). Thus,  $t_{100}$  ~~characterized~~ characterizes 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).

Adding PEs increases the initial  $q_c$ , or the rainwater mixing ratio  $q_r$  when  $r_{\text{PE}} > 40\text{ }\mu\text{m}$  and  $n_{\text{PE}} > 0$ , potentially limiting the comparability of simulated cases. To address this, we restricted the analysis of  $t_{10\%}$  and further conversion rates such as autoconversion rate (i.e., raindrop formation by collisions between cloud droplets), and accretion rate (i.e., raindrop growth by raindrops collecting cloud droplets) to cases where the increase in the initial  $q_c + q_r$  due to the addition of PEs is below 2 %. In most cases, the increase in  $q_c$  and  $q_r$  is below 1 %. However, two exceptions,  $n_{\text{PE}} = 300$ , with  $r_{\text{PE}} = 40\text{ }\mu\text{m}$  and  $n_{\text{PE}} = 1000$ , with  $r_{\text{PE}} = 27\text{ }\mu\text{m}$ , show an increase of 1.9 %.

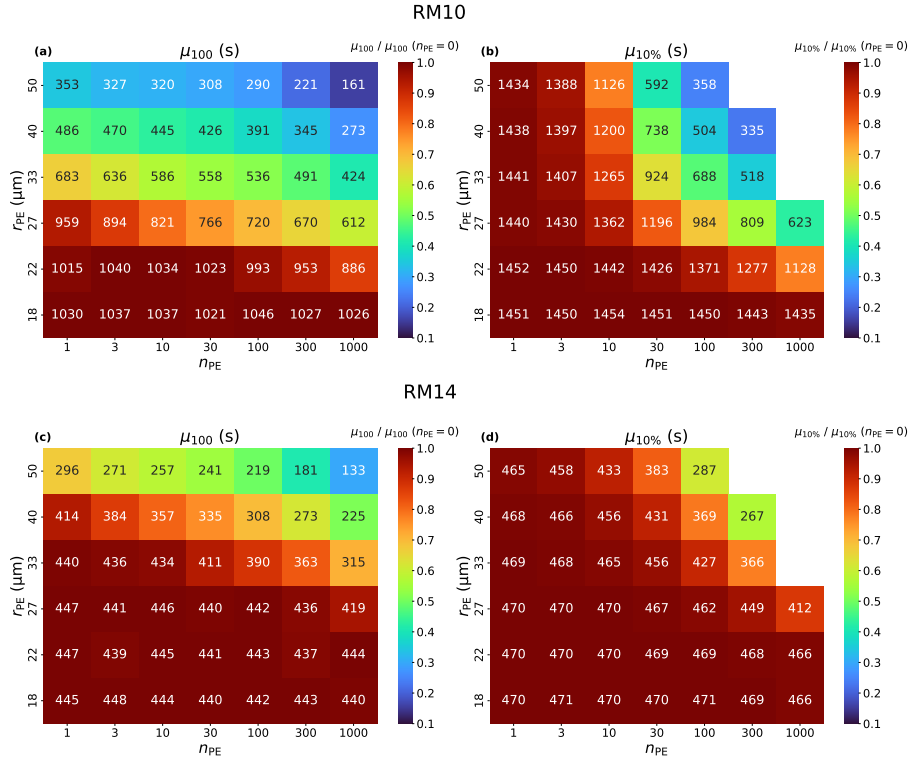
### 3 PE Effect on Precipitation Timescales

#### 3.1 Critical Thresholds for Raindrop Formation and Rain Initiation

Figure 2 shows the ensemble-averaged  $t_{100}$  and  $t_{10\%}$ , named  $\mu_{100}$  and  $\mu_{10\%}$ , for RM10 and RM14. In general, increasing  $r_{\text{PE}}$  and  $n_{\text{PE}}$  both shorten  $\mu_{100}$  and  $\mu_{10\%}$ , indicating accelerated rain initiation. However, when  ~~$r_{\text{PE}} \leq 18\text{ }\mu\text{m}$~~   $r_{\text{PE}} = 18\text{ }\mu\text{m}$ , i.e., smaller than the ~~cut-off radius~~ maximum droplet radius of the initial DSD (Fig. 1a),  $\mu_{100}$  and  $\mu_{10\%}$  are not substantially accelerated compared to those cases without PEs regardless of  $n_{\text{PE}}$ . Note that, in the case without PEs,  $\mu_{100}$  and  $\mu_{10\%}$  are ~~1213~~ 1027 s and ~~1660~~ 1452 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.,  $n_{\text{PE}} = 1000$ ), has a negligible effect on raindrop formation. Interestingly, ~~for  $\mu_{10\%}$ ,~~  $n_{\text{PE}}$  plays a more crucial ~~rule than for the role for  $\mu_{10\%}$  than for  $\mu_{100}$ .~~ For  $n_{\text{PE}} \leq 3$ ,  $\mu_{10\%}$  is not accelerated (Fig. 2b) even for large PEs, whereas  $\mu_{100}$  is accelerated (Fig. 2a). Thus, a faster  $\mu_{100}$  does not always ensure a shorter  $\mu_{10\%}$ .

For RM10, when  $n_{\text{PE}} = 3$ , the PE number concentration is approximately  $10^{-3}\text{ cm}^{-3}$ . In this case, even PEs larger than  $40\text{ }\mu\text{m}$  are not effective in accelerating  $t_{10\%}$  (Fig. 2b). However, when the PE concentration increases to a relatively high value ( $n_{\text{PE}} = 30$ ), PEs larger than  $22\text{ }\mu\text{m}$  can substantially accelerate  $t_{10\%}$  (Fig. 2b). Such high PE concentrations are uncommon but have been observed in certain oceanic conditions (Jung et al., 2015). In contrast, for RM14, which represents typical maritime clouds in a pristine environment with  $N_0 = 87\text{ cm}^{-3}$ , the effect of PEs is reduced. PEs smaller than  $33\text{ }\mu\text{m}$  are unable to accelerate  $\mu_{100}$  regardless of  $n_{\text{PE}}$  (Fig. 2c). Moreover,  $t_{10\%}$  is accelerated only when both  $n_{\text{PE}}$  and  $r_{\text{PE}}$  are very large (Fig. 2d). However, such extreme conditions are uncommon in typical maritime environments. This suggests that the impact of PEs depends on the initial DSD shape, requiring a collisionally stable cloud for a substantial effect.

Overall, Fig. 2 shows  $\mu_{100}$  and  $\mu_{10\%}$  can be shortened with increasing  $n_{\text{PE}}$  and  $r_{\text{PE}}$ , 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 are the specific size and number of PEs required to accelerate rain initiation substantially? To identify the critical threshold,



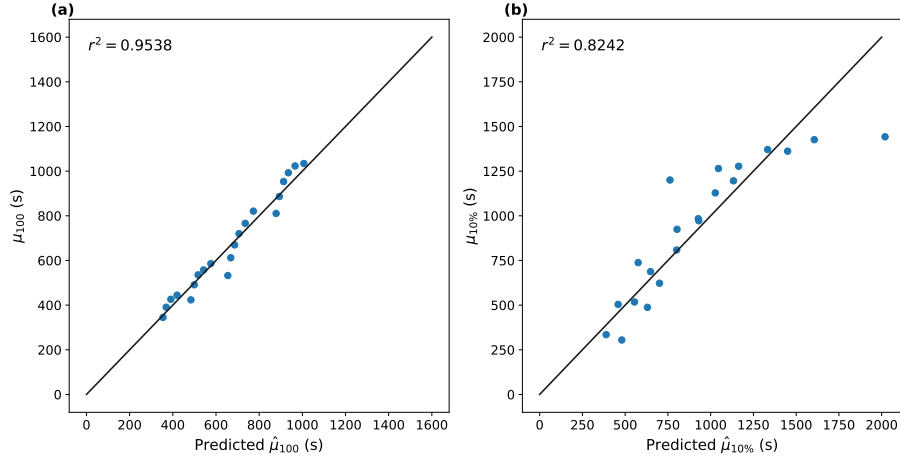
**Figure 2.** Ensemble-averaged values of (a and c) time for the first 100  $\mu\text{m}$  raindrop formation,  $\mu_{100}$ , and (b and d) time for 10 % of cloud droplets to convert to raindrops,  $\mu_{10\%}$ , for RM10 case (first row) and RM14 (second row). The abscissa represents  $n_{\text{PE}}$ , the ordinate represents  $r_{\text{PE}}$ . The numbers in each box indicate the values of (a)  $\mu_{100}$ , (b)  $\mu_{10\%}$ .  $\mu_{10\%}$  values for  $r_{\text{PE}} \geq 40 \mu\text{m}$  and  $n_{\text{PE}} > 1000$  are not shown as the raindrop mass is already larger than 10 % cases where the initial  $q_c + q_i$  increase due to large PEs by more than 2 % are not shown. Colors in the plot represent the ratio of  $\mu_{100}$  and  $\mu_{10\%}$  to their values in the case without PEs ( $n_{\text{PE}} = 0$ ). In the case without PEs,  $\mu_{100} = 1214 \text{ s}$ ,  $\mu_{100} = 1027 \text{ s}$  and  $\mu_{10\%} = 1452 \text{ s}$  for RM10, and  $\mu_{100} = 1660 \text{ s}$  respectively  $\mu_{100} = 442 \text{ s}$  and  $\mu_{10\%} = 470 \text{ s}$  for RM14.

we first express  $\mu_{100}$  and  $\mu_{10\%}$  as functions of  $n_{\text{PE}}$  and  $r_{\text{PE}}$ . As shown in Fig. 2,  $\mu_{100}$  and  $\mu_{10\%}$  are inversely related to the product of decrease as both  $n_{\text{PE}}$  and  $r_{\text{PE}}$  increase once the critical threshold is exceeded. Thus, we write

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

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

To determine the parameters of Eq. (5), we fit  $a_{\alpha}$ ,  $b_{\alpha}$ ,  $c_{\alpha}$ , and  $k_{\alpha}$ , using  $\mu_{100}$  and  $\mu_{10\%}$  from cases with  $r_{\text{PE}} \geq 22$  and  $n_{\text{PE}} \geq 10$ . In these cases, both  $\mu_{100}$  and  $\mu_{10\%}$  are directly affected by changes in  $r_{\text{PE}}$  and  $n_{\text{PE}}$  (Fig. 2), i.e., the PE critical threshold is exceeded. The fitted parameters are  $a_{100} = 0.086$ ,  $b_{100} = 3.086$ ,  $c_{100} = 244 \text{ s}$ , and  $k_{100} = 17308248$ ,  $c_{100} = 3363 \text{ s}$ , and



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

$k_{100} = 1.035$  for  $\mu_{100}$ , and  $a_{10\%} = 0.13$ ,  $b_{10\%} = 1.13$ ,  ~~$c_{10\%} = -310$  s, and  $k_{10\%} = 104016$~~   $c_{10\%} = 165592$  s, and  $k_{10\%} = 3.018$  for  $\mu_{10\%}$  with  $r_{PE}$  in  $\mu\text{m}$ . ~~Note that, while units~~ Units of each parameter are detailed in Appendix B; ~~our~~ our.

Our focus will be on  $\Phi_{\alpha}(n_{PE}, r_{PE}, \text{i.e.,})$  with  $a_{\alpha}$  and  $b_{\alpha}$  first. The parameters  $c_{\alpha}$  and  $k_{\alpha}$  will be discussed in more detail after we expand Eq. (5) with more physically meaningful terms. The values of  $a_{\alpha}$  and  $b_{\alpha}$  indicate that both  $\mu_{100}$  and  $\mu_{10\%}$  are more sensitive to  $r_{PE}$  than  $n_{PE}$  ~~as expected from Fig. 2.~~ When comparing  $a_{100}$  and  $b_{100}$  to  $a_{10\%}$  and  $b_{10\%}$ ,  ~~$\mu_{10\%}$  seems to depend more the dependency~~ on  $n_{PE}$  ~~and less on  $r_{PE}$ , than is stronger in  $\mu_{10\%}$  than in  $\mu_{100}$ ,~~ which is also consistent with the results shown in Fig. 2. Figure 3 juxtaposes the simulated and predicted  $\mu_{100}$  and  $\mu_{10\%}$  ~~values using Eq. (5).~~ This result indicates that  $\mu_{100}$  and  $\mu_{10\%}$  can be expressed with  $\Phi_{\alpha}$  relatively well. However, Eq. (5) ~~tends to overestimate overestimates~~  $\mu_{10\%}$  when it is ~~below 750 over 1400 s, and generally fails to predict  $\mu_{10\%}$  when it is over 1000~~ (Fig. 3b). This is due to the cases with  $n_{PE} < 10$  ~~and  $r_{PE} < 22 \mu\text{m}$ ,~~ which show almost no dependency on  $r_{PE}$ . The reasons behind this behavior will be further discussed in detail in Sec. 4.

To better capture the behavior of  $\mu_{100}$  and  $\mu_{10\%}$ , especially near the critical threshold where the dependency on  $r_{PE}$  and  $n_{PE}$  vanishes, ~~e.g., cases where  $r_{PE} < 22 \mu\text{m}$  and  $n_{PE} < 10$  for RM10,~~ we expand Eq. (5) by a Heaviside step function  $\mathcal{H}$ , such that

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

where  $\mu_{\alpha,c}$  is the baseline value of  $\mu_{\alpha}$  in the absence of PEs incorporating parameter  $c_{\alpha}$  from above. When fitting Eq. (6) to all results, ~~including cases where  $r_{PE} < 22$  and  $n_{PE} < 10$ , are used, with the~~ parameters  $a_{\alpha}$  and  $b_{\alpha}$  were fixed to the values ~~obtained previously.~~ previously obtained from RM10 to enable a more direct comparison between different cases, focusing solely on the parameters in Eq. (6). 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  $\mu_{100}$ , and range from ~~0.75 to 0.9~~ 0.67 to 0.84 for  $\mu_{10\%}$ . The results of  ~~$\mu_{100}(\Phi)$  and~~

$\mu_{10\%}(\Phi)$  for the  $\mu_{100}(\Phi_{100})$  and  $\mu_{10\%}(\Phi_{10\%})$  for RM10 case are shown in Fig. 4a and b as blue solid lines. Until exceeding the their critical thresholds ( $\Phi_{100,c} = 5.04 \times 10^{-5}$  and  $\Phi_{10\%,c} = 1.51 \times 10^{-2}$ ,  $\Phi_{100,c} = 1.91 \times 10^4$  and  $\Phi_{10\%,c} = 7.23 \times 10^1$  for RM10; see Tabs. B1 and B2),  $\mu_{100}$  and  $\mu_{10\%}$  remain constant at  $\mu_{100,c} = 1190$  s and  $\mu_{10\%,c} = 1608$  s,  $\mu_{100,c} = 1025$  s and  $\mu_{10\%,c} = 1405$  s. These values agree well with  $\mu_{100}$  and  $\mu_{10\%}$  without PEs, 1214/1027 s and 1660/1452 s, respectively. However, once  $\Phi_\alpha$  becomes smaller/larger than  $\Phi_{\alpha,c}$ , i.e., exceeds the critical threshold,  $\mu_{100}$  and  $\mu_{10\%}$  decrease as expected from Eq. (5).

### 3.2 Factors Controlling the Critical Threshold

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 the values of parameters  $a_\alpha$  and  $b_\alpha$  parameters for  $\Phi_{100}$  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  $k_\alpha$  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  $\mu_{100}$ . As  $\bar{r}$  increases, both  $\mu_{100,c}$  decreases and  $\mu_{10\%,c}$  decrease (Figs. 4a and 5b), indicating that it takes less time to produce a large raindrop even without PEs; and 5b and d). This is due to the increased number of large droplets making collisions more likely when  $\bar{r}$  increases. Results from the cases with a cut-off DSD with different  $\bar{r}$  are shown in Fig. 4c and d. As before,  $\mu_{100,c}$  and  $\mu_{10\%,c}$  also decrease with increasing  $\bar{r}$  although the largest droplet size remains unchanged due to the cut-off DSD. In this case, (Fig. 5b). Here, this is due to the increased number of droplets in the 15 – 20  $\mu\text{m}$  size range among non-PE droplets (Fig. 1b), which can initiate collisions through stochastic processes. However,  $\mu_{100,c}$  and  $\mu_{10\%,c}$  remain nearly unchanged for the  $\bar{r} = 12 \mu\text{m}$  and  $\bar{r} = 14 \mu\text{m}$  cases because the difference in the number concentration of 15 – 20  $\mu\text{m}$  droplets between these cases is minimal (cf. Fig. 1b), even though the number concentration of smaller droplets is substantially lower for  $\bar{r} = 14 \mu\text{m}$ . This suggests that  $\mu_{100,c}$  is more sensitive to the number concentration of larger droplets (e.g., those with radii of 15–20  $\mu\text{m}$ ) than to smaller droplets, particularly when considering the cut-off DSD.

The critical threshold  $\Phi_{100,c}$  also decreases/increases with increasing  $\bar{r}$ , implying indicating that more and larger PEs are needed/required to exceed the critical threshold (Fig. 5a).

Results from the cases with a broad DSD with different  $\bar{r}$  are shown in Fig. 4e and f. Interestingly, for  $\bar{r} = 8 \mu\text{m}$ , almost all sizes and numbers of PEs are effective in shortening  $\mu_{100}$  (Fig. 4e 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 2a). In contrast, for  $\bar{r} = 14 \mu\text{m}$ ,  $\Phi_{100,c}$  becomes very high, making most PEs ineffective in shortening  $\mu_{100}$ . This suggests that the PE effect is more pronounced for DSDs where collisions among droplets are less efficient, i.e., cases with smaller  $\bar{r}$  and slower  $\mu_{100}$ .

In cases with cut-off DSD,  $\Phi_{100,c}$  is smaller in broad DSD cases lower compared to those without, for the same  $\bar{r}$  (Fig. 5a). This is because of the presence due to the absence of larger droplets in the initial DSD, which are equally efficient as as effective as PEs in the collision process, reducing the importance of the PE effect. Moreover, both  $\mu_{100,c}$  and  $\Phi_{100,c}$  decrease with

240 increasing  $\bar{r}$  (Fig. 4e and d and Fig. 5a). This is because, without ~~Thus, employing a 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. 4e and Fig. 5e and d), where both  $\mu_{10\%,c}$  and  $\Phi_{10\%,c}$  increase as  $\bar{r}$  becomes larger. Now, we examine the relationship between the radius of the largest initialized non-PE droplet and the radius of a single PE which can accelerate  $t_{100}$ . We employ  $\Phi_{100,c}$  for RM10 and  $\Phi_{10\%,c}$  decrease with increasing RM10N, from Table B1. When  $n_{PE} = 1$ , the critical  $r_{PE} = 24.4$ , and  $22.9 \mu\text{m}$  for RM10 and RM10N, respectively. In each case, the radius of the largest initialized non-PE droplet is  $24 \mu\text{m}$  (RM10) and  $20 \mu\text{m}$  (RM10N). Therefore, for RM10, a single PE only slightly larger than the largest initialized non-PE droplet radius is sufficient to exceed the critical threshold. In contrast, RM10N requires a PE much larger than the largest non-PE droplet. This difference implies that the critical PE size is influenced not only by the radius of the largest initialized non-PE droplet but also by the collision efficiency among non-PE droplets, which depends on  $\bar{r}$  in the absence of the cut-off DSD and whether a DSD cut-off is present.~~

Additionally, the TICE effect is considered for RM10 (Fig. 4e and f). TICE is considered with three different  $\epsilon = 16, 80$  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 convective clouds, and  $100 - 1000 \text{ cm}^2 \text{ s}^{-3}$  in deep convective clouds. With strong turbulence ( $\epsilon = 80 - 100 \text{ cm}^2 \text{ s}^{-3}$ ),  $\Phi_{100,c}$  is lower (Fig. 4a and 5a) than in the case with no or weaker TICE ( $\epsilon = 16 \text{ cm}^2 \text{ s}^{-3}$ ). Specifically,  $\mu_{\alpha,c}$  decreases as  $\epsilon$  increases (Fig. 4a and 5a) than in the case with no or weaker TICE ( $\epsilon = 16 \text{ cm}^2 \text{ s}^{-3}$ ). Thus, more and larger PEs are required to substantially accelerate  $\mu_{100}$  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 e, f and Fig. 5b, d), whereas  $\Phi_{\alpha,c}$  exhibits only a slight increase (Fig. 5a and c). In contrast,  $\mu_{100,c}$  and  $\mu_{10\%,c}$  decrease more substantially with increasing  $\epsilon$ , becoming even shorter than in the broad DSD case (4e, f and Fig. 5b and d, a, c). This suggests indicates 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).

265 , making  $\mu_{\alpha,c}$  shorter. Therefore, more PEs are required in the presence of TICE compared to cases without. Notably, the critical PE threshold increases substantially only when  $\epsilon \geq 200 \text{ cm}^2 \text{ s}^{-3}$  (Fig. 5a and c; Table B1). This indicates that the influence of TICE on limiting the PE effect is primarily important in deep convective clouds or in regions within shallow clouds where  $\epsilon$  is locally high (e.g., Pruppacher and Klett, 2012). In summary, when droplet collisions are already efficient without PEs, whether due to a large  $\bar{r}$ , either due to the presence of large droplets (i.e., broad DSD), or TICE, the a large  $\bar{r}$  or the absence of a DSD cut-off) or under the influence of TICE—a larger PE size and number needs to be larger to are necessary to substantially accelerate rain initiation substantially.

270 Although we have identified the existence of the critical threshold for the PE effect, there remains uncertainty a question regarding why  $t_{10\%}$  is not always shorter than the case without PEs when  $n_{PE}$  is small affected by the presence of PEs, 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 PE. This suggests that while PEs can accelerate the formation of the largest raindrop, these droplets may not substantially directly impact the overall rain



275 ~~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~~ mass growth when the number of PEs is low. In the following section, we will explore how PEs affect  $t_{10\%}$  to explain why a shorter  $t_{100}$  does not ensure a shorter  $t_{10\%}$ .

#### 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  
280 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  
285 earlier (Fig. 6a and b). This result is consistent with Fig. 2, in which  $\mu_{100}$  is smaller than  $\mu_{100,c}$ , but  $\mu_{10\%}$  is comparable to  $\mu_{10\%,c}$ .

The time series of autoconversion and accretion ~~evolution provides~~ provide 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~~ find that  
290 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~~ for  $n_{PE} \geq 100$  (Fig. 6c). When  $n_{PE} < 30$ , autoconversion and consequently accretion by PEs are even slower than those of  
295 non-PE droplets. This ~~is because autoconversion depends heavily on stochastic events. Thus implies that the collisional growth of PEs is not necessarily faster than the collisional growth between non-PE droplets. Thus,~~ although larger PEs are more likely to collide, ~~a small~~ the overall collision frequency remains low when  $n_{PE}$  ~~reduces the likelihood of these collisions, making PE autoconversion slower than~~ is small, resulting in slower PE autoconversion compared to non-PE autoconversion. Thus, autoconversion. While non-PE autoconversion always decreases with increasing  $n_{PE}$ , PE autoconversion increases  
300 substantially only for  $n_{PE} > 100$ . 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. Hence, shorter  $t_{100}$  does not necessarily lead to a shorter  $t_{10\%}$  when  $n_{PE}$  is small, PEs may not decrease  $t_{10\%}$ , although  $t_{100}$  can be shorter than in the cases without PEs. (Fig. 2).

For  $r_{PE} = 27 \mu\text{m}$ , while non-PE autoconversion always decreases with increasing  $n_{PE}$ , PE autoconversion increases sub-  
305 stantially 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, increasing  $n_{PE}$  does not affect~~ the time to initiate PE autoconversion remains unchanged with  $n_{PE}$  affecting, but



affects only its magnitude (Fig. 6c). The initiation time for PE autoconversion is influenced by  $r_{\text{PE}}$  since this process is closely related to the number of collisions or time required for droplets to grow larger than  $40\text{ }\mu\text{m}$ , which occurs more quickly for larger PEs (cf. Fig. 6c and d). Thus,  $r_{\text{PE}}$  determines the initiation time for autoconversion, especially when  $r_{\text{PE}} \geq 27\text{ }\mu\text{m}$ , while  $n_{\text{PE}}$  determines how much non-PE droplet autoconversion and accretion are suppressed.

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

Interestingly, at high  $n_{\text{PE}}$ , the non-PE autoconversion and accretion rates reach their peak values earlier than in cases without PEs or with low  $n_{\text{PE}}$  (Fig. 6c, d, e, and f). During the initial 1000 s, the non-PE autoconversion rate is nearly identical across all cases, regardless of  $n_{\text{PE}}$ . However, when  $n_{\text{PE}}$  is high, more non-PE droplets are collected by PEs, reducing the number of droplets available for autoconversion. As a result, the non-PE autoconversion rate peaks and declines earlier in cases with higher PE concentrations. This suppression of non-PE autoconversion decreases the number of non-PE raindrops and the non-PE accretion rate. These findings highlight that the primary role of PEs is to collect non-PE droplets, which might suppress non-PE autoconversion and accretion.

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  $n_{\text{PE}} = 100$  (blue and purple solid lines in  $n_{\text{PE}} \geq 100$  (Fig. 7bd), 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  $n_{\text{PE}}$  substantially exceeds the critical threshold ( $r_{\text{PE}} = 27\text{ }\mu\text{m}$  and  $n_{\text{PE}} = 300$ ), 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 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 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 time required for the first 100  $\mu\text{m}$  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 ~~non-PE~~ droplet radius of ~~non-PE droplets~~the initial DSD. This is because  $t_{100}$  is more related to the growth of a single droplet where larger droplets, such as PEs, are expected 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~~collisional 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~~increases the collisional stability of the DSD and makes the collisional process less susceptible to PE perturbations because non-PE droplet collisions are already sufficient for 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. Although TICE does not directly alter the PE critical threshold, it reduces the difference in rain initiation acceleration between cases with and without PEs. Consequently, more and larger PEs are required to achieve the same acceleration in droplet growth as in cases without TICE.

While PEs In this study, PEs larger than 22  $\mu\text{m}$  are found to effectively accelerate the precipitation ( $t_{10\%}$ ) for clouds in relatively polluted environments, when their concentration exceeds  $10^{-3} \text{ cm}^{-3}$ , consistent with Feingold et al. (1999). While this PE concentration falls within the range observed for giant sea-salt particles over the ocean (Jung et al., 2015), for clouds in a pristine environment, substantially higher numbers and sizes of PE are required to achieve effective precipitation acceleration. These observations are based on measurements of sea-salt aerosols (2–20  $\mu\text{m}$ ), which have large solution masses and corresponding large equilibrium sizes. However, under atmospheric conditions, these particles might not have sufficient time to grow to their equilibrium size (Ivanova et al., 1977), potentially resulting in lower PE concentrations. On the other hand, it is possible that PE concentrations can increase through stochastic collisions (Kostinski and Shaw, 2005; Dziekan and Pawlowska, 2017) as the cloud evolves. Furthermore, because the critical threshold decreases in DSDs with higher collisional stability, the effect of PEs is expected to be especially strong in non-precipitating, or polluted clouds as suggested in previous studies (e.g., Johnson, 1993; Dziekan et al., 2021).

While PEs can accelerate the rain initiation by collecting other droplets, they may reduce the number of raindrops by suppressing 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

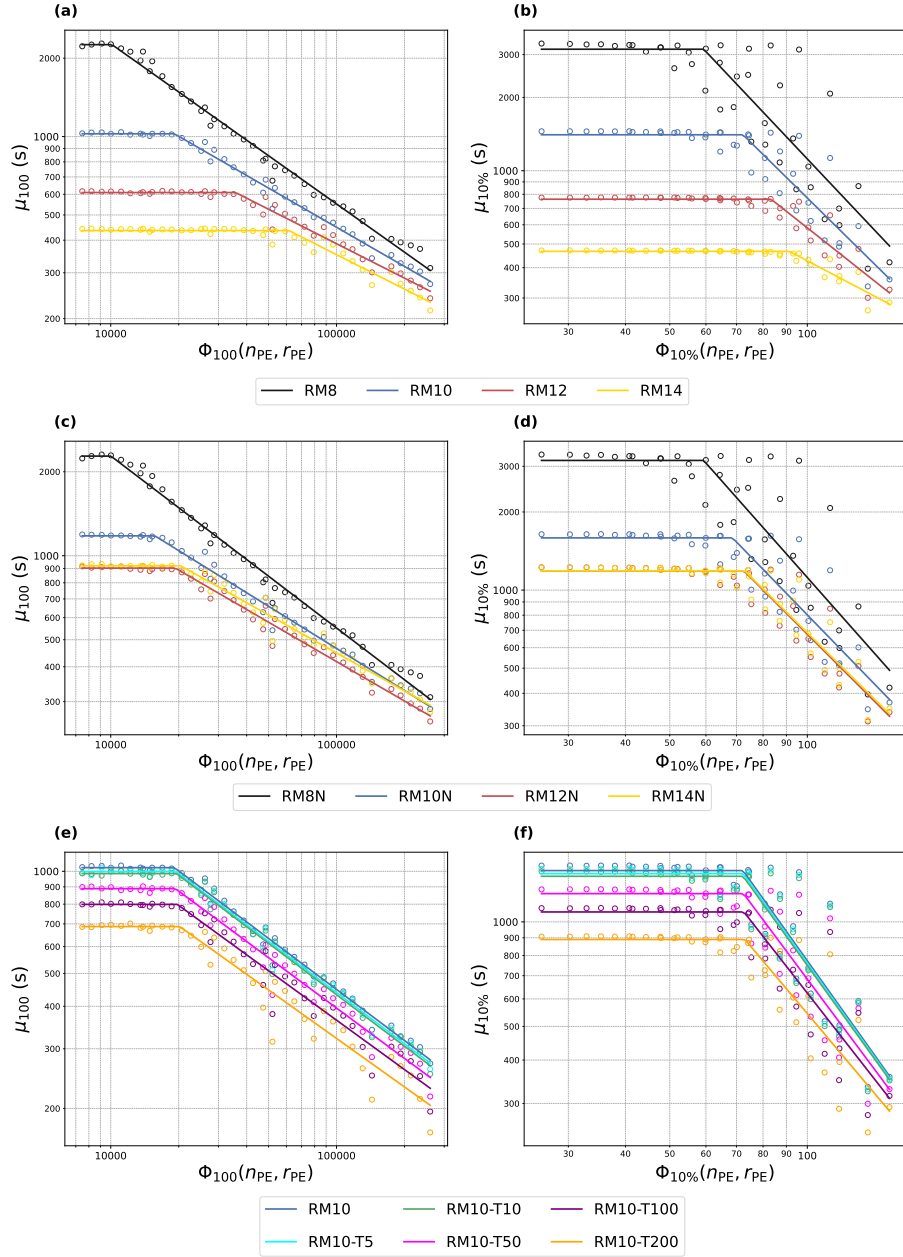
differently. Thus, ~~validating-confirming~~ this study's findings in more complex scenarios is ~~mandatory, for the future. These~~ necessary. Modeling efforts should incorporate additional processes such as aerosol activation, condensation, ~~entrainment,~~ and ~~especially, and entrainment. Especially,~~ collisional droplet breakup (Low and List, 1982) ~~which increases-, is expected to~~ increase the small number of PEs, causing more PE accretion afterward ~~or droplet sedimentation which decreases-, and droplet~~ sedimentation is expected to decrease the effect of PEs by making large raindrops precipitate and ~~prevents-prevent~~ 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 underscores the need for caution in ~~geoengineering-climate-engineering~~ 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 and minimal rain production to maximize efficiency.

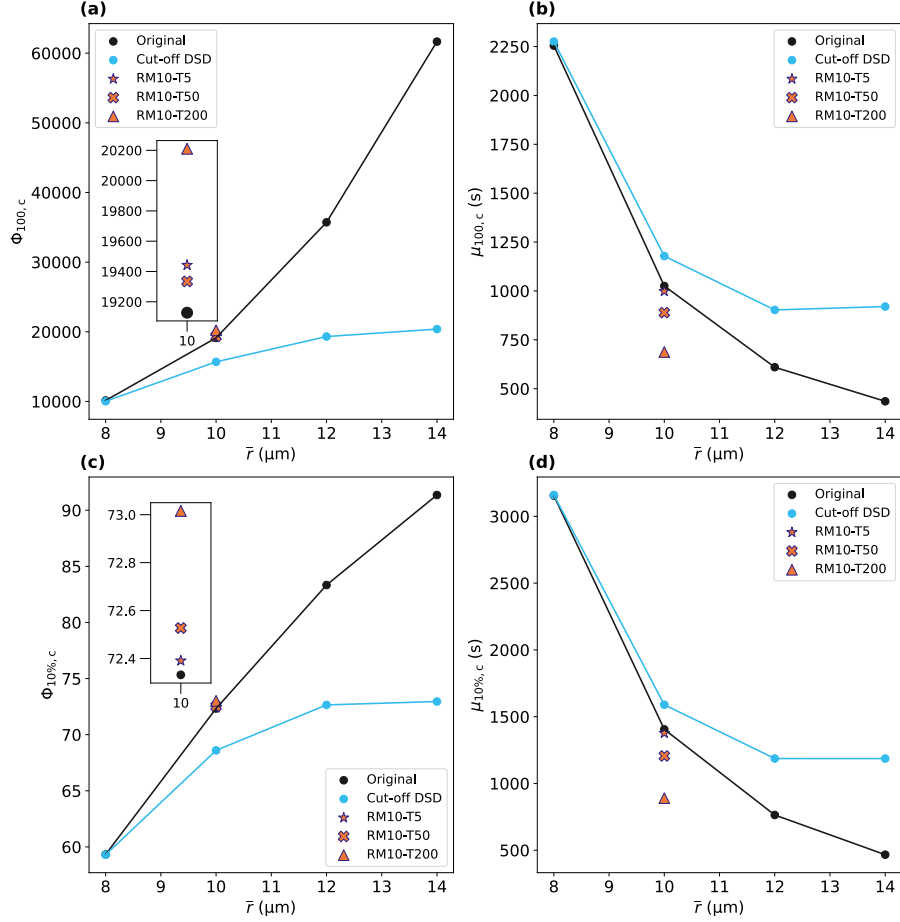
*Code and data availability.* A Python version of the LCM code is available on the link ([https://github.com/jslim93/PyLCM\\_edu](https://github.com/jslim93/PyLCM_edu)). The simulations were conducted using the FORTRAN version of the code, which employs the same collision routine as the Python version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

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

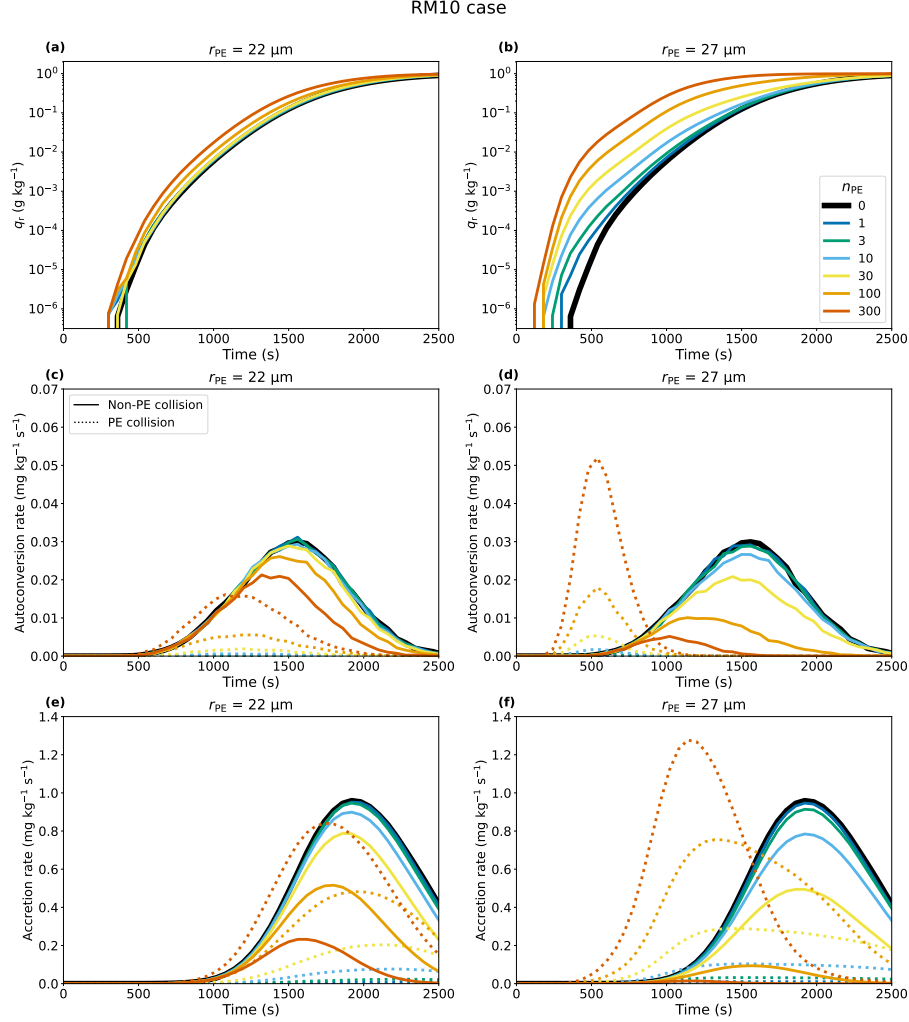
Figure A1 illustrates how the mean and ~~relative~~ standard deviation of  $t_{100}$  ( $\mu_{100}$  and  ~~$\sigma_{100}\sigma_{100}/\mu_{100}$~~ , respectively) and  $t_{10\%}$  ( $\mu_{10\%}$  and  ~~$\sigma_{10\%}\sigma_{10\%}/\mu_{10\%}$~~ , respectively) evolve as the ensemble size increases from 1 to ~~200 for 200~~. Different colors of the dots represent RM8 (black), RM10 (blue), RM12 (red), RM14 (yellow) without PEs ( $n_{PE} = 0$ ). The mean values and ~~standard~~ deviations begin to the relative standard deviations converge when the ensemble size exceeds 100. Therefore, we consider an ensemble size of 100 adequate for obtaining reliable results.



**Figure 4.**  $\mu_{100}$  (left column) and  $\mu_{10\%}$  (right column) as a function of  $\Phi_{\alpha}$  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 without cut-off DSD (RM8, RM10, RM12, and RM14), and the second row (c and d) represents cases without with cut-off DSD (RM8N, RM10N, RM12N, and RM14N). The third row (e and f) represents RM10 with different  $\varepsilon$  values (RM10-T10, RM10-T5, RM10-T50, RM10-T100, and RM10-T200).

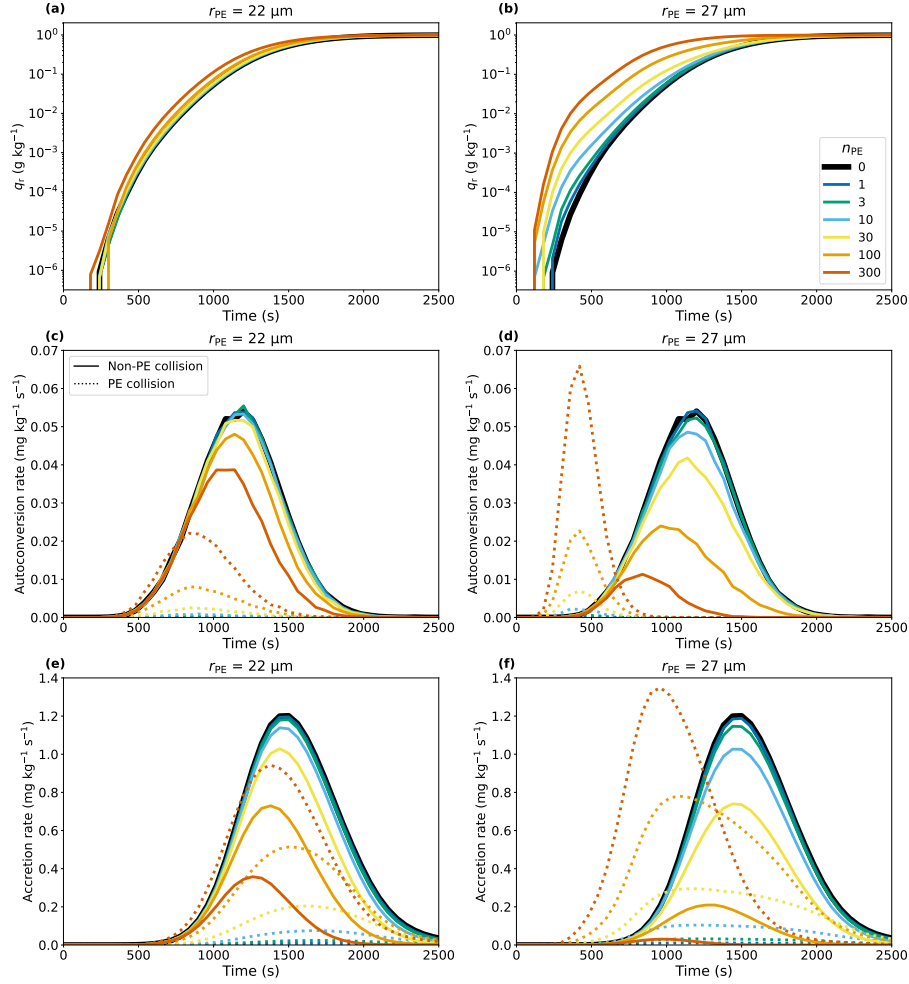


**Figure 5.** Results of (a)  $\Phi_{100,c}$ , (b)  $\mu_{100,c}$ , (c)  $\Phi_{10\%,c}$  and (d)  $\mu_{10\%,c}$  for different  $\bar{r}$ . Blue-Black circles depict the cases with/without cut-off DSD (original), while orange-light blue circles depict the cases without/with cut-off DSD. Green-square, Orange star, triangle, cross, and star triangle shape represent the results with  $\epsilon = 16, 80, 100 \text{ cm}^2 \text{ s}^{-3}$ ,  $\epsilon = 5, 50, 200 \text{ cm}^2 \text{ s}^{-3}$ , respectively, for the RM10 case. The insets show a zoomed-in view of the cases with different  $\epsilon$  at  $\bar{r} = 10 \mu\text{m}$ .

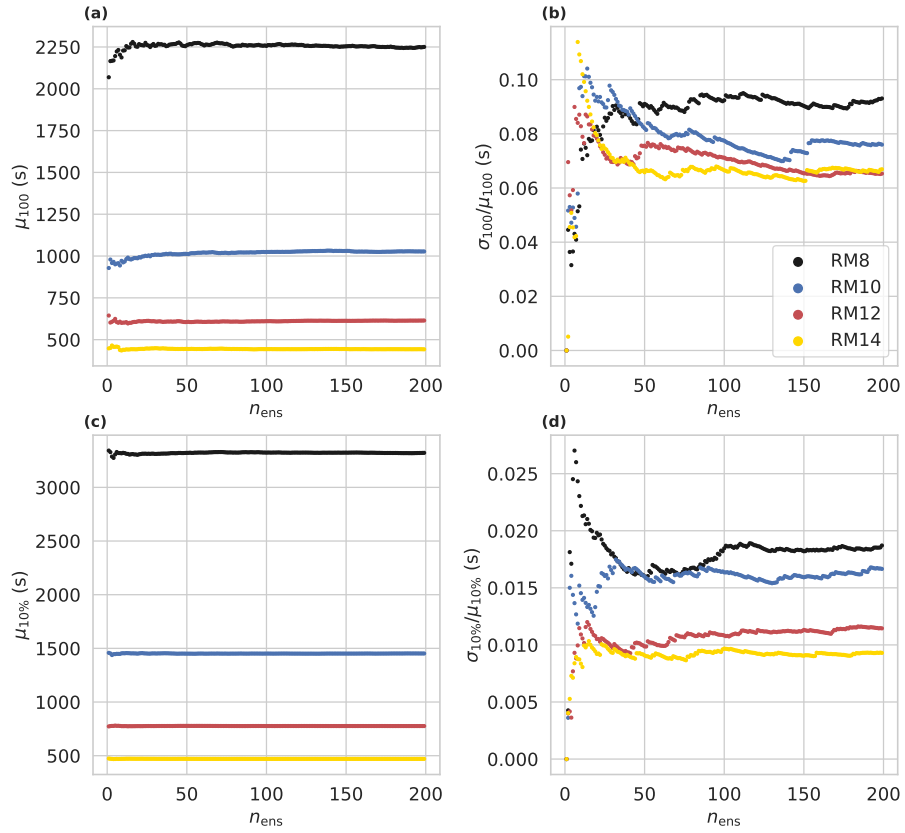


**Figure 6.** Time series of (a/b) raindrop mixing ratio, (c/d) autoconversion rate, and (e/f) accretion rate, for the RM10 case, shown for two different values of  $r_{PE}$ :  $22 \mu\text{m}$  (first column) and  $27 \mu\text{m}$  (second column). The colors of the lines represent different  $n_{PE}$  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.

RM10-T100 case



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



**Figure A1.** Variation of (a)  $\mu_{100}$ , (b)  $\mu_{10\%}$ , (c)  $\sigma_{100}/\mu_{100}$  and (d)  $\sigma_{10\%}/\mu_{10\%}$  with ensemble size ( $n_{\text{ens}}$ ) for RM8, RM10, RM12, and RM14 without PEs ( $n_{\text{PE}} = 0$ ).



## Appendix B: Parameters for the Fitting Function

Table B1 depicts the parameters and  $r^2$  values derived from curve fitting Eq. (6) to  $\mu_{100}$  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  $\bar{r} = 8 \mu\text{m}$ ). ‘BN’ denotes cases ~~without~~ with cut-off DSD. Numbers following ‘T’ indicate  $\varepsilon$  (e.g., ~~T16~~ T50 corresponds to cases with  ~~$\varepsilon = 16 \text{ cm}^2 \text{ s}^{-3}$~~   $\varepsilon = 50 \text{ cm}^2 \text{ s}^{-3}$ ). The units of  $\mu_{100,c}$  and  $\mu_{10\%,c}$  are in s,  $\Phi_{100}$  in  ~~$\mu\text{m}^{-1.852}$~~   $\mu\text{m}^{3.086}$  and  $\Phi_{10\%}$  in  ~~$\mu\text{m}^{-6.78}$~~   $\mu\text{m}^{1.13}$ . Units of  $\Phi_{100}$  and  $\Phi_{10\%}$  are determined by Eq. 5 with respective  $a_\alpha$ ,  $b_\alpha$  parameters, where the unit of  $n_{\text{PE}}$  is  $\mu\text{m}$  and  $n_{\text{PE}}$  is unit-less. The subscript  $\alpha$  is 100 or 10% for  $\mu_{100}$  and  $\mu_{10\%}$ , respectively. Thus, the units of  $k_\alpha$  for  $\mu_{100}$  and  $\mu_{10\%}$  are  ~~$\mu\text{m}^{1.852} \text{ s}$~~   $\mu\text{m}^{-3.086} \text{ s}$  and  ~~$\mu\text{m}^{6.78} \text{ s}$~~   $\mu\text{m}^{-1.13} \text{ s}$ , respectively. In this study, these parameters are mainly used to compare how critical threshold varies in different cases than to obtain actual values.

**Table B1.** Parameters for fitting function of  $\mu_{100}$

	RM8	RM10	RM12	RM14 <del>RM8B-</del>
$\Phi_{100,c}$	<del><math>8.54 \times 10^{-5}</math></del> <u><math>1.01 \times 10^4</math></u>	<del><math>5.05 \times 10^{-5}</math></del> <u><math>1.91 \times 10^4</math></u>	<del><math>3.69 \times 10^{-5}</math></del> <u><math>3.57 \times 10^4</math></u>	<del><math>3.32 \times 10^{-5}</math></del> <del><math>8.50 \times 10^{-5}</math></del> <u><math>6.17 \times 10^4</math></u>
$\mu_{100,c}$	<del>2388.73</del> <u>2254.69</u>	<del>1173.81</del> <u>1025.24</u>	<del>880.46</del> <u>609.73</u>	<del>870.40</del> <del>2381.12</del> <u>435.49</u>
$k_{100}$	<del><math>2.47 \times 10^7</math></del> <u>7.30</u>	<del><math>1.83 \times 10^7</math></del> <u>6.34</u>	<del><math>1.77 \times 10^7</math></del> <u>4.42</u>	<del><math>1.85 \times 10^7</math></del> <del><math>2.48 \times 10^7</math></del> <u>2.68</u>
$r^2$	0.99	<del>0.98</del> <u>0.99</u>	0.98	<del>0.97</del> <del>0.99</del> <u>0.95</u>

	<del>RM10B</del> <del>RM8N</del>	<del>RM12B</del> <del>RM10N</del>	<del>RM14B</del> <del>RM12N</del>	<del>RM10-T16</del> <del>RM14N</del>
<u><math>\Phi_{100,c}</math></u>	<del>RM10-T80</del> <u><math>1.00 \times 10^4</math></u>	<u><math>1.57 \times 10^4</math></u>	<u><math>1.93 \times 10^4</math></u>	<u><math>2.04 \times 10^4</math></u>
<u><math>\mu_{100,c}</math></u>	<u>2275.34</u>	<u>1178.18</u>	<u>902.79</u>	<u>920.13</u>
<u><math>k_{100}</math></u>	<u>7.30</u>	<u>6.13</u>	<u>5.81</u>	<u>5.70</u>
<u><math>r^2</math></u>	<u>0.99</u>	<u>0.99</u>	<u>0.98</u>	<u>0.98</u>

	<u>RM10-T5</u>	<u>RM10-T10</u>	<u>RM10-T50</u>	RM10-T100	<u>RM10-T200</u>
$\Phi_{100,c}$	<del><math>4.08 \times 10^{-5}</math></del> <u><math>1.94 \times 10^4</math></u>	<del><math>2.16 \times 10^{-5}</math></del> <u><math>1.96 \times 10^4</math></u>	<del><math>1.33 \times 10^{-5}</math></del> <u><math>1.93 \times 10^4</math></u>	<del><math>5.01 \times 10^{-5}</math></del> <u><math>1.97 \times 10^4</math></u>	<del><math>4.62 \times 10^{-5}</math></del> <del><math>4.60 \times 10^{-5}</math></del> <u><math>2.00 \times 10^4</math></u>
$\mu_{100,c}$	<del>1011.30</del> <u>998.16</u>	<del>607.01</del> <u>984.67</u>	<del>430.81</del> <u>888.95</u>	<del>1089.05</del> <u>798.79</u>	<del>906.69</del> <del>865.67</del> <u>687.12</u>
$k_{100}$	<del><math>1.89 \times 10^7</math></del> <u>6.29</u>	<del><math>1.92 \times 10^7</math></del> <u>6.23</u>	<del><math>2.05 \times 10^7</math></del> <u>5.85</u>	<del><math>1.73 \times 10^7</math></del> <u>5.39</u>	<del><math>1.55 \times 10^7</math></del> <del><math>1.47 \times 10^7</math></del> <u>5.00</u>
$r^2$	<del>0.98</del> <u>0.99</u>	<del>0.97</del> <u>0.99</u>	<del>0.95</del> 0.98	<del>0.97</del> <u>0.96</u>	<del>0.97</del> <u>0.94</u>

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

	RM8	RM10	RM12	RM14	<del>RM8B</del>
$\Phi_{10\%,c}$	<del><math>1.93 \times 10^{-2}</math></del> <u><math>5.93 \times 10^1</math></u>	<del><math>1.51 \times 10^{-2}</math></del> <u><math>7.23 \times 10^1</math></u>	<del><math>1.31 \times 10^{-2}</math></del> <u><math>8.33 \times 10^1</math></u>	<del><math>1.23 \times 10^{-2}</math></del> <u><math>1.93 \times 10^{-2}</math></u>	<del><math>9.14 \times 10^1</math></del>
$\mu_{10\%,c}$	<del>3128.99</del> <u>3155.62</u>	<del>1592.18</del> <u>1405.09</u>	<del>1205.14</del> <u>763.82</u>	<del>1207.07</del> <u>3124.57</u>	<del>466.49</del>
$k_{10\%}$	<del><math>2.21 \times 10^5</math></del> <u>7.30</u>	<del><math>1.49 \times 10^5</math></del> <u>6.34</u>	<del><math>1.30 \times 10^5</math></del> <u>4.42</u>	<del><math>1.35 \times 10^5</math></del> <u><math>2.21 \times 10^5</math></u>	<del>2.68</del>
$r^2$	<del>0.78</del> <u>0.75</u>	<del>0.85</del> <u>0.83</u>	<del>0.89</del> <u>0.88</u>	<del>0.90</del> <u>0.78</u>	<del>0.88</del>

	<del>RM10B</del> <u>RM8N</u>	<del>RM12B</del> <u>RM10N</u>	<del>RM14B</del> <u>RM12N</u>	<del>RM10-T16</del> <u>RM14N</u>
$\Phi_{10\%,c}$	<del>RM10-T80</del> <u><math>5.93 \times 10^1</math></u>	<u><math>6.86 \times 10^1</math></u>	<u><math>7.27 \times 10^1</math></u>	<u><math>7.30 \times 10^1</math></u>
$\mu_{10\%,c}$	<u>3160.14</u>	<u>1589.32</u>	<u>1186.52</u>	<u>1186.31</u>
$k_{10\%}$	<u>7.30</u>	<u>6.13</u>	<u>5.81</u>	<u>5.70</u>
$r^2$	<u>0.75</u>	<u>0.82</u>	<u>0.86</u>	<u>0.89</u>

	<u>RM10-T5</u>	<u>RM10-T10</u>	<u>RM10-T50</u>	RM10-T100	<u>RM10-T200</u>
$\Phi_{10\%,c}$	<del><math>1.41 \times 10^{-2}</math></del> <u><math>7.24 \times 10^1</math></u>	<del><math>1.10 \times 10^{-2}</math></del> <u><math>7.23 \times 10^1</math></u>	<del><math>8.42 \times 10^{-3}</math></del> <u><math>7.25 \times 10^1</math></u>	<del><math>1.50 \times 10^{-2}</math></del> <u><math>7.27 \times 10^1</math></u>	<del><math>1.41 \times 10^{-2}</math></del> <u><math>1.41 \times 10^{-2}</math></u>
$\mu_{10\%,c}$	<del>1412.81</del> <u>1376.65</u>	<del>773.39</del> <u>1352.81</u>	<del>472.91</del> <u>1205.85</u>	<del>1480.90</del> <u>1067.95</u>	<del>1212.93</del> <u>1151.98</u>
$k_{10\%}$	<del><math>1.47 \times 10^5</math></del> <u>6.29</u>	<del><math>1.00 \times 10^5</math></del> <u>6.23</u>	<del><math>8.00 \times 10^4</math></del> <u>5.85</u>	<del><math>1.41 \times 10^5</math></del> <u>5.39</u>	<del><math>1.26 \times 10^5</math></del> <u><math>1.20 \times 10^5</math></u>
$r^2$	<del>0.87</del> <u>0.82</u>	<del>0.92</del> <u>0.82</u>	<del>0.91</del> <u>0.81</u>	<del>0.85</del> <u>0.80</u>	<del>0.85</del> <u>0.85</u>

*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.

*Competing interests.* There are no competing interests.

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