

We thank the two reviewers for their invaluable comments. We provided the point-by-point responses below. The comments are in black and the responses in blue. Changes made in the revised manuscript were also marked in blue.

Comment # 1

The revised study considers many of my previous concerns, and I feel much more comfortable with the results after the numerical issues have been resolved. However, there are still some issues that need to be addressed before publication. In particular, I feel that the aspect of “cloud seeding” requires some attention, as detailed below.

Major Comments

Cloud seeding and ultra-giant nuclei

First, I feel that the presumably overarching subject of “cloud seeding” is not well motivated. Cloud seeding is mentioned in the title and four times in the abstract, but it is only mentioned once at the end of the introduction (l. 79). Accordingly, the article lacks general background information on cloud seeding, e.g., how cloud seeding might be used to enhance precipitation in arid environments. Accordingly, I cannot find a continuous storyline that guides the reader through the manuscript, and the erratically occurring discussion of cloud seeding feels like a distraction. The authors owe their interesting results a more concise story. One leading idea for this study, which is discernable at some places already, might be the separation of the *artificial* influence of cloud seeding on the initiation of rain in comparison to *natural* cloud processes such as turbulence and the effect of aerosol hygroscopicity. In other words, one could simply ask: Does cloud seeding make sense?

We included motivation for using the idea of cloud seeding for studying the impact of aerosols on line 36-50:

“In this study, we implement the idea of in-cloud seeding, i.e., seeding hygroscopic particles near the cloud base to investigate the effects of aerosols in droplet growth and rain formation. Hygroscopic cloud seeding, owing to its potential effect of increasing rainfall, has been conducted in research and operational context globally to address the shortage of water resources in arid environments (e.g., Silverman and Sukarnjanaset, 2000; Terblanche et al., 2000). The general concept of hygroscopic cloud seeding in rain enhancement is that the introduction of artificial cloud condensation nuclei (seeding particles) into warm clouds can, on the one hand, suppress the activation of small natural aerosols, and on the other hand, generate large initial particles that accelerate or enhance the collision-coalescence process (Cooper et al., 1997). Regardless of its existence in operational weather modification for decades, the direct effect of seeding is still inconclusive, partly due to the chaotic nature of the convective cloud system making it impossible to conduct controllable seeding experiments and the limitation in detecting and assessing the seeding results with current instrumentations (Silverman, 2003; Flossmann et al., 2019). Nevertheless, the progress made in cloud seeding

does advance our understanding of cloud-aerosol-precipitation interactions. A leading idea of this study is to make use of the concept of cloud seeding experiments to separate the influence of aerosols on rain initiation from the effects of natural cloud processes such as turbulence and aerosol hygroscopicity, as well as to shed light on the long-existing question of whether cloud seeding could enhance precipitation.“

Second, the initialization of giant aerosols ($R_d > 1 \mu\text{m}$) with their equilibrium radius (ll. 131 – 132) needs to be commented on. In subsaturated environments, these aerosols need several hours to days to reach their equilibrium radius (e.g., Mordy 1959), which indicates that these particles might have wet radii significantly smaller than their equilibrium radius when entering a cloud. Accordingly, these aerosols will not trigger the precipitation process as suspected by the authors (ll. 5 – 6). I was initially not too concerned with this shortcoming since the authors find that the contribution of these particles is insignificant (ll. 6 – 7). However, in the case of cloud seeding, the *natural* giant aerosols compete with the *artificially* added particles. For these cases, it might matter how large the largest natural particles are at the beginning of the simulation. And I suspect that smaller-than-equilibrium particles might increase the importance of seeded particles in the precipitation process, which will certainly affect statements such as ll. 282 – 283.

Our original statement in the initialization of aerosol size was not correct. In our simulation, the parcel model had different treatments in computing the initial size below and above $R_d = 1 \mu\text{m}$. We only applied equilibrium radius to $R_d < 1 \mu\text{m}$, and for $R_d \geq 1 \mu\text{m}$ we assumed a doubled volume in wet size, i.e., $R = 2^{1/3} R_d$.

We have modified the original statement as following on line 154-156:

“For aerosols with dry radius $R_d \leq 1 \mu\text{m}$, the initial wet radius is set to the size when the droplet is in equilibrium at the given ambient humidity: $dR/dt=0$ (Jensen and Nugent, 2017). For giant aerosols with $R_d > 1 \mu\text{m}$, the initial wet size is assumed to be twice the dry volume, i.e., $R = 2^{1/3} R_d$. ”

It should be pointed out that the timescale of giant aerosols reaching an equilibrium size highly depends on the relative humidity. In a sub-saturated environment (initial relative humidity $RH_0 = 85.61\%$ in our case), the timescale is a few seconds. Fig. F below shows the parcel model results of the particle growth at $RH_0 = 85.61\%$ (our case) and $RH_0 = 100\%$, respectively. In both cases, the dry radius is set to $10 \mu\text{m}$ with a number concentration of 1 cm^{-3} , updraft velocity is set to 0 m/s (i.e., S is only modulated by droplet condensation). It is shown that in a sub-saturated environment, the time for the droplet to reach equilibrium is less than 20 seconds. In a saturated environment, it takes a few minutes to reach equilibrium. Overall, the higher the RH, the longer the aerosol takes to reach equilibrium size. But in both cases, they have a timescale shorter than an hour.

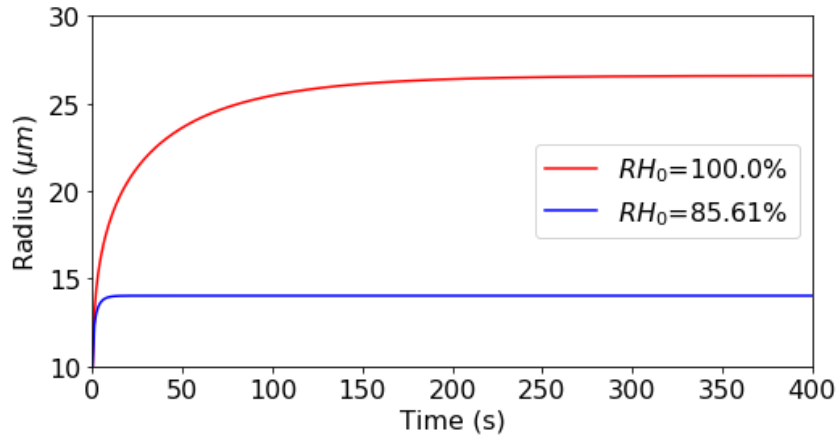


Fig. F. Radius varies with time with an initial $RH_0 = 100.0\%$ (red curve) and an initial $RH_0 = 85.61\%$ (blue curve).

Minor Comments

LI. 89 – 91: It feels redundant to mention twice that there is no more activation above the cloud base.

Removed “and aerosol activation is unimportant”

LI. 89 – 91: It might be the case that there is no activation in the cloud core or your parcel model. However, activation of laterally entrained aerosols might occur in “more complete” cumulus clouds (Slawinska et al. 2012; Hoffmann et al. 2015).

We have acknowledged this in the conclusion when addressing the shortcomings of the current study (line 357-360). We added the above-mentioned references in the revision.

Fig. 1: The supersaturation below cloud base increases approximately linear (see Fig. 2a). Please change this in your sketch.

Changed to linear increase

LI. 100 and 112 – 113: It feels redundant to mention twice that most droplets are smaller than 10 μm .

Removed “, and most droplets in the parcel model are below 10 μm ”

LI. 166 – 167: Is there a significant effect of the fallen-out droplets on the LWC? Figure 7 a does not show a (significant) change in the increase of LWC once droplets reach 50 μm in radius. Similarly, the mean radius seems not to be affected by the fallen-out droplets. Could you please

state the LWC of the fallen-out particles? This will give the reader a sense of how much water is lost due to precipitation.

The fallout of $R > 50 \mu\text{m}$ did not change the LWC in a discernible sense. Fig. G(a) shows the LWC including and excluding $R > 50 \mu\text{m}$. As the mass of $R > 50 \mu\text{m}$ is about two orders of magnitude smaller than the LWC (Fig. G(b)), the two curves of LWCs are almost identical. Our study emphasized on the processes at the rain initiation stage. Therefore we mainly simulated the period of time before a substantial amount of precipitation forms.

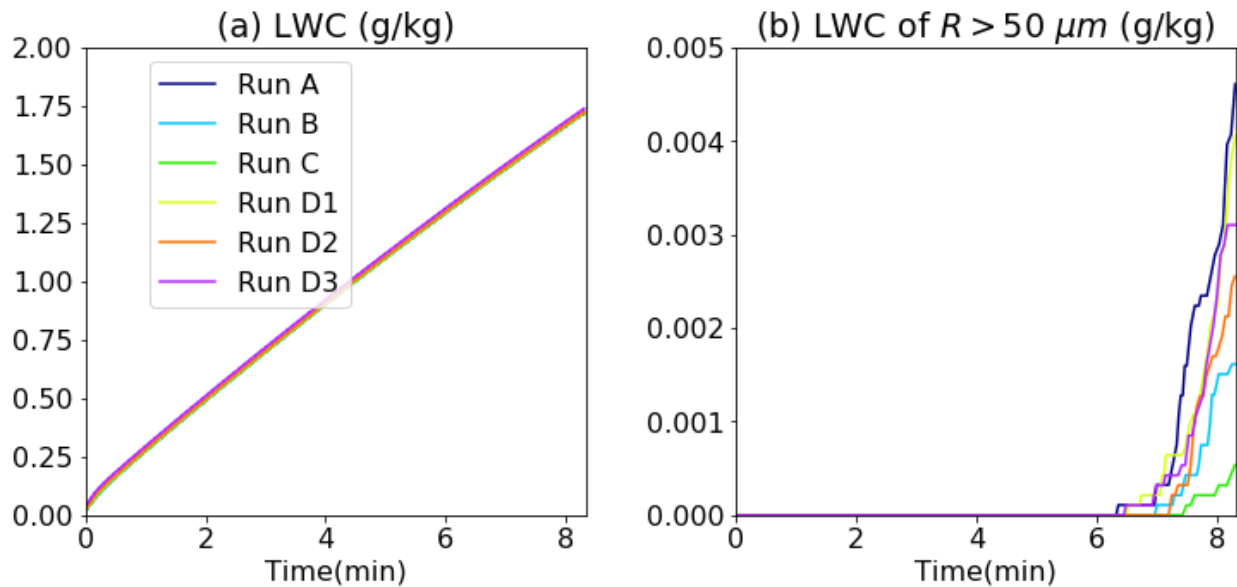


Fig. G: (a) The time series of LWC of the six simulations. Colors distinguish the cases. Solid lines denote the LWC including the mass of $R > 50 \mu\text{m}$ and dashed lines represent the LWC excluding $R > 50 \mu\text{m}$. (b) The time series of LWC of $R > 50 \mu\text{m}$.

Tab. 2, Figs. 3, 4, 5, 6, 7, and several places in the manuscript: When I previously suggested using more meaningful abbreviations for the individual model runs, I intended to use these abbreviations throughout the manuscript: Run NoTurb instead of Run B, Run NoSolu instead of Run C, ...

Switched to meaningful abbreviations in the figs and throughout the manuscript.

LI. 180 – 185: State explicitly that the seeded particles do not exhibit any variability in their initial radius, i.e., they are seeded using a delta distribution function.

We added “monodisperse” in the statement to stress the property of their size: “an extra number of **monodisperse** aerosols are introduced near the cloud base”.

L. 185: In addition to increasing the dry radius by a factor of 10, the wet radius in Run D3 is also doubled, according to Tab. 2.

We have modified the sentence to “In Run D3, the dry size of the seeding particles increases to tenfold of that in Run D1 with the wet size doubled”.

L. 236: For clarity, add “non-turbulent” before “gravitational collection process” if appropriate.

Modified to “non-turbulent gravitational collection process”.

Fig. 4: Panel “(b) Run C (cond+coll)” needs to be labeled “(c) Run C (cond+coll)”.

Corrected.

Fig. 5: Change units on ordinate to “ $\text{cm}^{-3} \text{s}^{-1}$ ”

Changed.

L. 259: Consider moving the definition of the autoconversion rate from the caption of Fig. 7 to the main text.

We have added the definition on line 276-277 while keeping the definition in the caption for clarification.

L. 289: What is meant by “the inhibition effect”? I know that you are referring to the last sentence, but it is not obvious that the previous sentence describes the “inhibition effect”.

We have modified the sentence to “Increasing the size of seeding particles in Run D3 (Seed-1N2R) buffers the above-mentioned inhibition effect caused by increasing aerosol number concentration.” And we also replaced “prohibited” by “inhibited” in the previous sentence.

LI. 321 – 327: Since and the amount of water lost due to precipitation is presumably negligible (see also comment for II. 166 – 167 above), and the range of tested aerosol concentrations and droplet sizes is rather small, it should not surprise that parcel-mean values “are not sensitive to turbulence level and aerosol conditions”. It is expected that these quantities are approximately adiabatic. It might be interesting to add some lines to Fig. 7 showing the adiabatic LWC or the adiabatic mean radius, which will allow one to quantify the degree of non-adiabaticity in the conducted simulations.

Fig. F illustrated that the mean value is very close to the adiabatic value. Adding adiabatic LWC would complicate the figure. On line 255-256 we added: “This is because the fall out mass of drizzle drops of $R > 50 \mu\text{m}$ before $T=500 \text{ s}$ is negligible, and the bulk LWC of the six cases is approximately adiabatic.” to explain the negligible effect of turbulence & aerosols on modulating the bulk LWC.

Technical Comments

L. 13: “On the other hand” not “on the other hand”

Corrected.

L. 40 ff.: It is awkward to cite “Chen et al. 2018b” before “Chen et al. 2018a”.

The order of reference was automatically arranged by the ACP latex typesetting, which determines the index of “a” and “b” in the manuscript. I am not sure how to change that manually.

L. 85 ff.: Units should be displayed with upright characters, not italics.

It is also the default font in ACP latex format when using “\$” for numbers, units, and math symbols.

L. 89: The SI symbol for seconds is “s” not “sec”.

Replaced all “sec” with “s” in the manuscript.

L. 111: There is a space missing between “flow.” and “Studies”.

Corrected.

L. 232 and other places: I suggest to abbreviate “minute” here because it is part of an equation ($T = 6 \text{ min}$).

Has abbreviated “minute” to “min” in all equation forms.

References

Hoffmann, F., Raasch, S., & Noh, Y. (2015). Entrainment of aerosols and their activation in a shallow cumulus cloud studied with a coupled LCM–LES approach. *Atmospheric Research*, 156, 43-57.

Mordy, W. (1959). Computations of the growth by condensation of a population of cloud droplets. *Tellus*, 11(1), 16-44.

Slawinska, J., Grabowski, W. W., Pawlowska, H., & Morrison, H. (2012). Droplet activation and mixing in large-eddy simulation of a shallow cumulus field. *Journal of the atmospheric sciences*, 69(2), 444- 462.

Comment # 2

Reproducibility

According to ACP Data Policy¹ “Data do not comprise the only information which is important in the context of reproducibility. Therefore, Copernicus Publications encourages authors to also deposit software, algorithms, model code, ... on suitable FAIR-aligned repositories/archives whenever possible.”

While the mentioned policy merely encourages to do so, let me urge the authors to do so, to do better than “parcel model and DNS model used to produce the dataset are available upon request” by making the code publicly available on a persistent repository (and/or as an electronic supplement to the paper).

We acknowledge that it is beneficial to share the model code with the readers if the code is in a user-friendly format. However, our model is a complex piece of code developed over two decades. Different parts of the code are written by different authors and a number of them are not co-authors of this paper. We simply do not have permission to share their part of the code with the general public. Additionally, the current version of the code is not well-documented, and it would be very difficult for general interested users to understand and to use it without a proper user’s manual. Considerable effort is required to document the meaning of each variable, functionality of each module, etc. For these reasons, we do not feel proper to share the code with the general public at this moment.

LWC profile and single/double precision issue

The origin of the problem is somewhat surprising. Let me just note that in the reply to the first round of reviews, the authors made an apparent error in definition of the growth rate of particles expressed in the third power of radius. From the introduced equation for the growth rate dR^2/dt it is evident that $KfvS$ has the unit of m^2/s , which is inconsistent with the later definition of dR^3/dt . It should read $dR^3/dt = 3RKfv(S - f(solu, curv))$.

We apologize for the typo in the first round of review, and the model code used the right format of the equation. The correct format of the equation in R^3 form should be

$$\frac{dR^3}{dt} = 3RKfv_v(S - f(solu, curv)), \text{ as addressed by the reviewer.}$$

Spectral discretisation

The figure C provided in the reply to the first round of reviews confirms significant sensitivity to the spectral discretisation with low number of “moving bins”. Consequently, it is hard to agree with the statement that “the resolution of the size spectrum will also have no numerical impact

on the evolution of the droplet spectrum” (reply to the reviews). Please acknowledge the sensitivity in the text. I suggest including Fig. C in the manuscript.

Based on the sensitivity test, the relative error caused by changing bin resolution is below 2.3% of the total aerosol number concentration (2.6cm^{-3} higher than the 253-bin case). In particular, the 39-bin case is only 0.6 cm^{-3} lower than the 253-bin case. Therefore, we think the sensitivity is not significant.

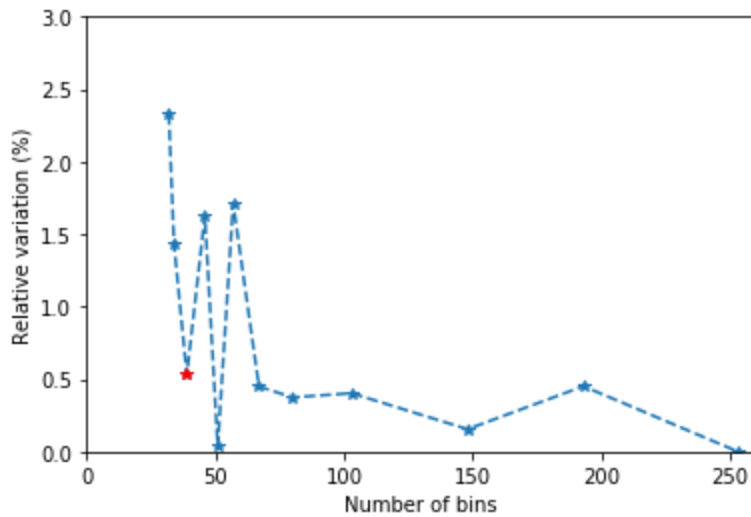


Fig. D. Relative variation = $(N_{act} - N_{act0})/N_{tot}$, where N_{act} is the number of activated aerosols at each bin resolution, N_{act0} is the number of activated aerosols at 253-bin, and N_{tot} is the total number of aerosols.

We have included the sensitivity analysis in the text.

”The result shows that the variation caused by changing bin resolution has a decreasing trend with increasing resolution, with a maximum variation of 2.3% of the total aerosol number concentration in the 32-bin case. In particular, the 39-bin case has only 0.6 cm^{-3} more aerosols activated than in the 253-bin case.”

Lognormal spectrum initialisation

The geometric standard deviation seems to be mismatched with the spread parameter (see caption of Table. 1 in Xue et al. 2010) as the geometric standard deviation must have values above unity. On a related note, doesn’t the vertical axis of Fig. 2c denote $dN/d(\ln(r))$ as in Fig. 4? Same concerns colour scale in Fig. 3.

The geometric standard deviation σ of the maritime particle size distribution in Xue et al. 2010 was originally taken from Jaenicke (1988) with σ value taken as the logarithm with base 10: $\sigma' = \log_{10}(\sigma) = [0.657\ 0.210\ 0.396]$, and it was then converted to the natural logarithm value (see

footnote b of Table. 1): $\sigma' = \log_{10}(\sigma) = [1.5128 \ 0.4835 \ 0.9118]$. To avoid confusion, we changed to its original value $\sigma = [4.5394 \ 1.6218 \ 2.4889]$.

The vertical axis in Fig. 2c denotes the number concentration of each assigned bin in the model, for easier comparison between the parcel model and DNS. Therefore, it will be N . We added “The vertical axis denotes the number concentration of the assigned particle size in the model.” in the caption of Fig. 2 for clarification.

We corrected the unit in Fig. 3 to “ $dN/d(\ln(r)) \ (cm^{-3}\mu m^{-1})$ ”

Supersaturation definition inconsistency

Note that in order to arrive at eq. (1), one needs to define supersaturation as $S = e/e_s$ (passage from 7.15-7.16 to 7.17 in the Rogers & Yau book) – worth clarifying as the manuscript involves the alternative definition of S using mixing ratios just below eq. (1).

On the one hand, it is more convenient to use mixing ratio (q_v) instead of vapor pressure (e) for calculating supersaturation, because q_v is a predicted variable of the model. On the other hand, S of the atmosphere using mixing-ratio is a very good approximation to the original form.

q_v is defined as $q_v = \varepsilon \frac{e}{P-e}$ (eq. (2.18) in Rogers & Yau 1989), where $\varepsilon \approx 0.615$. One can derive $\frac{q_v}{q_{vs}} = \frac{e/(P-e)}{e_s/(P-e_s)} \approx \frac{e}{e_s}$, given $e \ll P$ and $e_s \ll P$ in the atmosphere. In our simulation, the relative error is below 0.02% (Fig. E below), which is negligible.

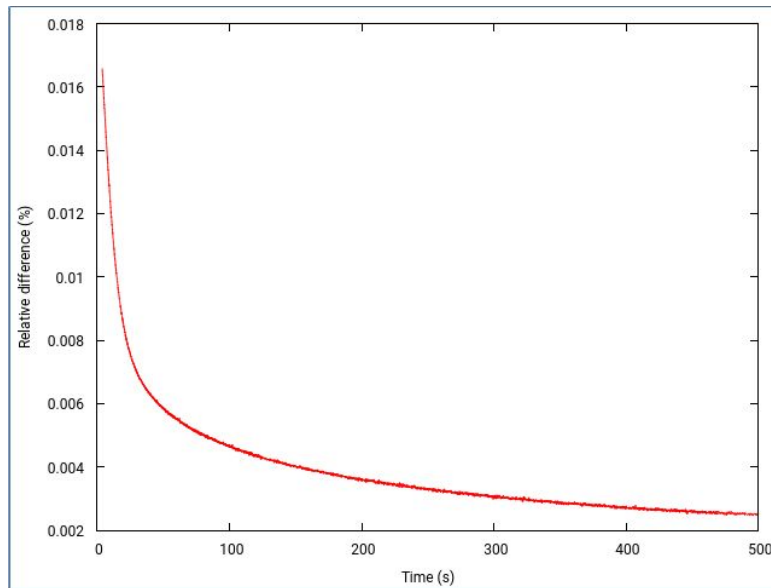


Fig. E. The relative error in calculating supersaturation due to using mixing ratio.

Seeding nomenclature

Starting from the abstract, the manuscript mentions “seeding more aerosols”, “seeding aerosol”, “aerosols injected”, etc; while in fact the seeded particles, given their size would classify as

droplets being four or eight micron in radius. In the case of D1 and D2 runs, these particles are well above their critical sizes, hence would also classify as droplets because of being activated (the D3 seeding particles are below their critical sizes). To sum up, I suggest elaborating on the somewhat arbitrary choice of dry/wet sizes of the seeded particles, and switching from calling them “seeding aerosol” to “seeding particles”.

Used the nomenclature “seeding particles” throughout the manuscript.

On a related note, “CCN-embedded droplets” and “aerosol-embedded cases” seem misleading, if not incorrect, to me. Please rephrase.

We rephrased it to “solute-containing droplets” instead of “CCN-embedded” or “aerosol-embedded”.

Parcel-mean term

For easier reading, I suggest refraining from using the “parcel-mean” term when referring to DNS simulations, to avoid confusion with the parcel model used for initialisation. Bulk or macroscopic might be better terms.

Replaced “parcel-mean” with “bulk”.

Miscellaneous notes

p2/l28-29: the sentence “no benchmark “truth” from either measurements or modelling exists to gauge the performance of various microphysics schemes” arguably does not require the “Up to this date” opening – do the authors envisage that such “truth” will ever exist?;

Removed from the sentence

p2/l34-37: The mention of wall effects in cloud chambers in the very first paragraph of the introduction seem misplaced – suggest not to deviate too far from the scope of the paper in the introduction;

We have removed the sentence of “For example, Thomas et al. (2019) used a flux-balance model to estimate the wall effect on the mean temperature and mean water vapor mixing ratio and found that the results highly depend on the geometry of the chamber.”

p2/l74-75: suggest removing second part of the sentence (“to seamlessly ...”);

Removed

p3/l82: “ascending process” Y “ascent”;

Replaced with “ascent”

p4/l91: suggest removing “and aerosol activation is unimportant” (previous sentence mentions that no new activation occurs);

Removed

p4/l92: “Outputs from the parcel model” Y “Parcel model state”

Changed as suggested.

p6/l130-131: suggest removing the sentence “In this way, the numerical diffusion...” (out of scope);

Removed

p6/l131: “at the given ...” – missing beginning of the sentence

Changed to “The initial wet size is set to the size when droplet is in equilibrium at the given ambient humidity: $dr/dt = 0$ (Jensen and Nugent, 2017).”

p6/Table 1: moving-bin method is contrasted with Lagrangian particle method – both are Lagrangian- in-particle-size; is there, in practice, any difference in the context of this work? Please elaborate or avoid contrasting them;

We changed the method name to “Lagrangian particle-by-particle method” to distinguish the two methods.

p7/l156-161: background/motivation mixed with model description;

We moved the motivation to line 55-61 in the introduction section.

p7/l164: “when small aerosols are introduced” – no small aerosols are introduced, right?

We changed to “when small droplets are present” for clarification.

p7/l172: correct “same other then”;

We believe “other than” is the right spelling.

p9/l194: “aerosol ... broaden the DSD” – rephrase so that particular property of the aerosol population is mentioned

We changed it to “including solute and turbulence effectively broaden the DSD at different times” to reflect the comparison between the control case (Run A), no turbulence case (Run B), and no solute case (Run C).

p13/l279: ditto (“modulation by aerosols” – concentration, size, hygroscopicity?)

We changed it to “the modulation of droplet mean radius by seeding particles is larger than the modulation by collision-coalescence” to reflect that the modulation was made by introducing additional aerosols of different sizes and of different number concentration (seeding particles).

p11/l251: please add a reference

We have removed the paragraph as the information is inaccurate. The theoretical study by Liu et al. (2006, Fig. 1) and observations by Hudson and Yum (1997) demonstrated that for particle number concentration around 100 cm^{-3} and mean radius between 5-15 μm , the relative dispersion ranges from 0.01-0.1. The value of our cases at the end of the simulation is also from 0.01-0.1, consistent with those studies. We, therefore, added “The values among the six cases at the end of the simulation range from 0.01-0.1, which is highly consistent with the theoretical study by Liu et al. (2006, Fig. 1) for an aerosol number concentration close to 100 cm^{-3} .” on lines 261-262.

p14/fig 7: x axis label missing in subplots (a) and (b)

x labels added.

p16/l343: Lagrangian supersaturation: please elaborate

Modified to “a highly perturbed Lagrangian **history of** supersaturation experienced by droplets.”

Raster graphics

Please replot all figures ensuring vector graphics format (not raster as it is currently provided). Hope that helps.

Changed to vector graphics