We greatly appreciate the invaluable comments from the reviewers. Below are the point-by-point responses. The comments are shown in black and responses are in blue.

Response letter to reviewer #1

Major Comments

Previous literature

The broadening of droplet size distributions has been studied intensively in the past, including the effects of aerosol hygroscopicity and turbulence (supersaturation fluctuations). Of course, the representation of these processes was highly idealized in the past, but the conclusions are not different from the submitted manuscript. For instance, Srivastava (1991) showed that the consideration of curvature in the diffusional growth equation is essential for the spectral broadening of a droplet size distribution in a lifted parcel. Furthermore, Korolev (1995) showed that curvature and solute terms lead to irreversible broadening if supersaturation fluctuations are applied. Further analysis has been carried out by Çelik and Marwitz (1999). Integrating these very simple, even analytical investigations into the highly complex framework of the submitted study will increase physical understanding and insight.

We integrated the above literature into our manuscript in Section 2.2 (Line 153-160): “Parcel model studies on droplet condensation in a lifted parcel show that the curvature term and the solute term can lead to condensational broadening on the droplet size spectrum. Srivastava (1991) demonstrated that the curvature effect is essential for DSD broadening in an ascending parcel. Korolev (1995) found that the curvature effect and the solute effect lead to irreversible broadening when supersaturation fluctuations are present. It is also found that aerosols of different sizes and different hygroscopicity can cause spectral broadening without supersaturation fluctuations (Çelik and Marwitz, 1999; Jensen and Nugent, 2017). Therefore, it is crucial to examine whether these effects are important in spectral broadening when they dynamically couple with droplet collisional growth in a turbulent environment. ”

Analysis

Many conclusions are based on Fig. 7. While the authors are candid about the fact the mean radius (panel a), the dispersion (panel b), the maximum radius (panel c), and the radar reflectivity (panel e) are calculated from droplet size distribution, I believe that this is also the case for the liquid water content (LWC) in panel d. Since the parcel is ascending with a constant velocity, the LWC is expected to increase linearly with time: $LWC = w\, t\, \Gamma$, with $\Gamma \approx 2.0 \times 10^{-6}$ kg m^-4. While the LWC)) follows this linear relationship for the first three minutes, it deviates significantly afterward. Of course, the LWC may deviate from this adiabatic behavior if the transfer of water vapor into the liquid phase is kinetically limited. However, this effect can be excluded since its magnitude decreases with droplet size and hence time. One reasonable explanation for this unphysical behavior is that the authors diagnosed the LWC from the droplet size distribution (as also suggested by the similarity to the mean radius in panel a) or made another error in the calculation of the LWC. Accordingly, any conclusions on LWC differences between the individual model runs are potentially false (e.g., ll. 9 – 10, 231 – 240, 265 – 268). Therefore, I strongly suggest repeating all simulations of this study, and to diagnose the LWC directly from the simulated droplets. I further suggest doing the same for the mean radius, dispersion, and maximum radius.

We appreciate the reviewer for pointing out this defect in our study. We identified that the unphysical behavior of the LWC and mean radius was caused by the round-off & truncation error of the single-precision droplet radius when solving the droplet growth equation. We have fixed the issue and re-run all 12 simulations in the revision.

The droplet growth equation in the present study includes the curvature effect and the solute effect. In previous version in Chen et al., (2018b, eq. (B1)), the equation we solved was $\frac{dR^2}{dt} = 2Kf_vS$, where K^{-1} is a temperature-dependent coefficient. In the present study, the droplet growth equation is modified to $\frac{dR^3}{dt} = 3Kf_v(S - f(solu, curv))$, where $f(solu, curv)$ includes the curvature term and the solute term. The $\frac{dR^3}{dt}$ format was used in the model for the convenience of calculating the condensation rate. It is more sensitive to the precision of calculation than $\frac{dR^2}{dt}$ because it's in a higher order. It should be pointed out that this truncation error issue only happened in the present study and did not affect the previous studies when the equation $\frac{dR^2}{dt} = 2Kf_vS$ was used. As shown in Fig. A, when using the $\frac{dR^2}{dt}$ scheme, the LWC evolution is not truncated by a lower precision.

In the $\frac{dR^3}{dt}$ scheme when supersaturation reaches below a certain critical value, a non-negligible round-off error occurred in calculating the new R^3: $R^3_{new} = R^3_{old} + dR^3$. dR^3 is rounded off to 0 when it is added to R^3_{old}, causing R^3 to stop growing (see dashed lines in Fig. B below). This caused the LWC and other spectra-derived statistics to plateau around T= 2min.

We switched all droplet-related variables to double-precision to minimize the round-off error and have rerun all simulations accordingly. In the new version, R^3 grows linearly with time (solid lines in Fig. B). All contents in the revised manuscript are based on the results of the new simulations.

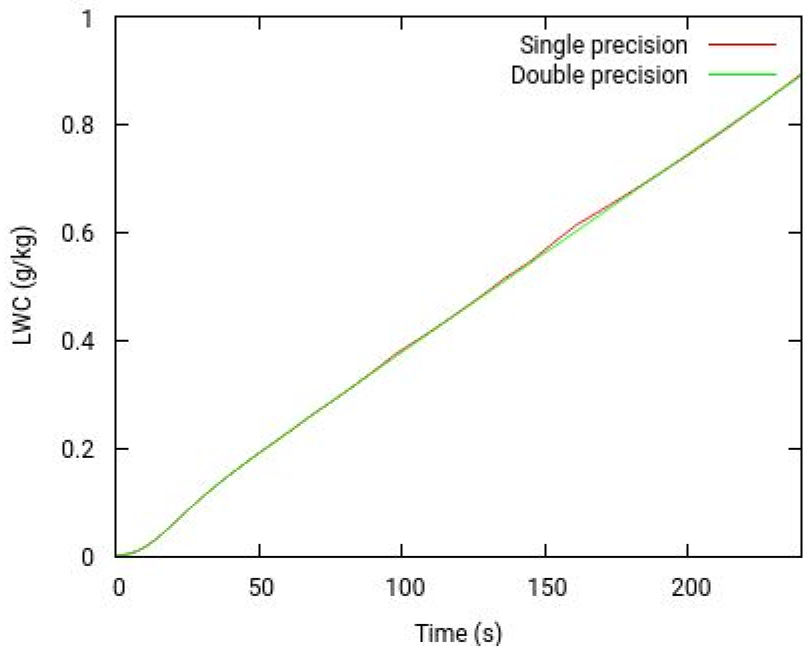

Fig. A: the time evolution of LWC when using the droplet growth equation $\frac{dR^2}{dt} = 2Kf_vS$ *(no solute effect or curvature effect). The green line is based on double-precision calculation, and the red line is based on single-precision calculation.*

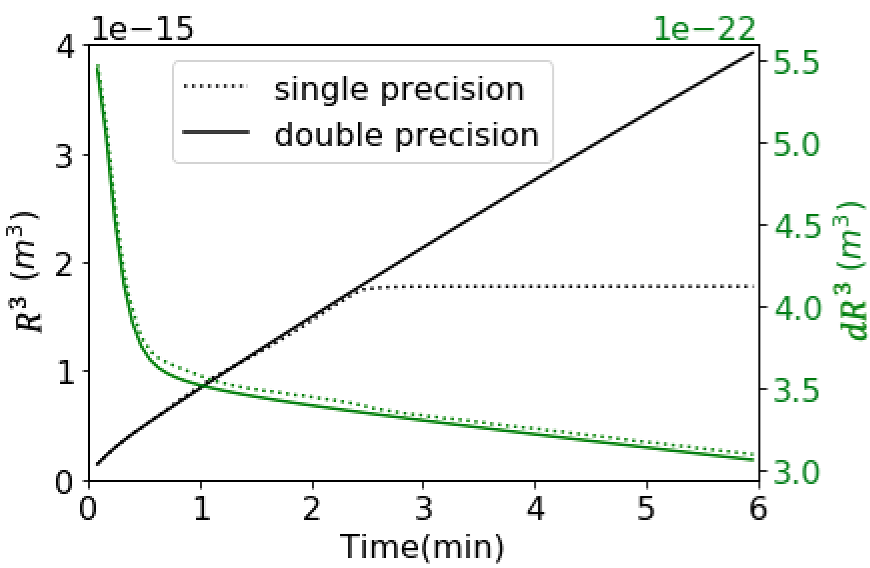

Fig. B: the time evolution of the mean R^3 *and* dR^3 *in Run A.* dR^3 *is the difference of* R^3 *from the previous time step. Dotted lines are from the single-precision run, and solid lines are from the double-precision (new version) run.*

Limitations of the Modeling framework

The authors state that the applied modeling framework promotes a more realistic assessment of cloud microphysical processes during the early development of a cloud. And I agree. However, there are certain limitations and restrictions inherent to this approach that need to be addressed. In fact, they might restrict the scope of the manuscript's conclusions significantly. **First,** the ascent of the air parcel is adiabatic. In cumulus clouds, entrainment dilutes the cloud constantly, reducing the liquid water content, while the activation of newly entrained aerosols might lead to a further broadening of the droplet size distribution (e.g., Lasher-Trapp et al. 2005).

We thank the reviewer's comments and have addressed the limitations and restrictions of this approach in the revised manuscript.

We agree that entrainment is a key process in cumulus clouds, in particular, close to cloud edges. Our study mainly looks at the adiabatic core region where entrainment mixing is minimum. It is found in Khain et al. (2013) that this region contains more large droplets due to higher liquid water content (LWC) than the rest of the cloud and is argued to favor the formation of raindrops.

In the revision, we added the description of the adiabatic region and explained the importance of this region to rain initiation in the introduction (lines 67-71):

"The adiabatic cores are regions free of entrainment of dry air. This region has a higher liquid water content (LWC) than the rest of the cloud and is argued to favor the formation of raindrops (Khain et al., 2013). To represent the DSD evolution at the core region, we prescribe here a dry aerosol size distribution in the sub-cloud region, and the aerosol activation process is explicitly simulated by a parcel model to provide a more physically-based initial DSD for the DNS."

In the result section, we discussed the supersaturation fluctuations in adiabatic core regions and the impact of the entrainment on condensational broadening at cloud edge on lines 219-224:

"Even though turbulence intensifies the collisional growth, the modulation on the droplet condensation is found insignificant. The DSDs in Run A and B in the condensation-only set are nearly identical (Fig. 4 (d-e)). This is because the supersaturation fluctuations are weak in an adiabatic core region. Vaillancourt et al. (2002) found that in a quasi-adiabatic environment both particle sedimentation and short-lived turbulent coherent structure reduce the supersaturation fluctuation and decrease the time that droplets are exposed to these fluctuations. We expect that the turbulent-induced condensational broadening is more significant in the cloud edge where entrainment mixing induces large variation in supersaturation fluctuations."

And lines 254-255:

"Thirdly, our idealized simulation focuses on the cloud adiabatic core which is devoid of entrainment. Inhomogeneous mixing by entrainment can possibly broaden the DSD."

In the conclusion section, we addressed the limitations inherent to this approach and the importance of including entrainment in future investigations on lines 339-350:

“Our idealized simulations focus on the cloud adiabatic core region and therefore exclude entrainment mixing which is highly active near the cloud edge. The activation of newly entrained aerosols might lead to a further broadening of the DSD (LasherTrapp, et al., 2005). In addition, the in-cloud mixing at a much larger scale than the DNS domain transports and mixes both the air and droplets from different parts of the cloud including the cloud edge, leading to a highly perturbed Lagrangian supersaturation experienced by droplets (Grabowski and Abade, 2017, eddy hopping effect). On the other hand, larger turbulent eddies can generate higher supersaturation fluctuations due to a higher variation in a vertical motion and thus may both affect the aerosol activation and broaden the DSD. Traditional DNS which is confined to a relatively small domain size (<1 m), and the impact of supersaturation fluctuations is significantly restricted. Methods such as an up-scaled DNS with superdroplets (e.g., Thomas et al., 2020) or representing the large-scale mixing with an external forcing on the thermodynamic fields (Paoli and Shariff, 2009) can be used for studying the impact of turbulent scales on the supersaturation fluctuations and thus on the condensational broadening of DSD. In conclusion, the relative importance of entrainment, eddy hopping effect, small-scale turbulence and aerosols requires further investigation.”

Second, the kinetic energy dissipation rate is assumed to be constant throughout the ascent, using a relatively high value only typical for the top of shallow cumulus clouds. This is a crude simplification since turbulence tends to increase with height (e.g., Seifert et al. 2010).

We used a non-turbulent parcel model to handle the ascending process below the cloud base, and use a statistically-stationary dissipation rate in DNS above the cloud base. The advantage of this idealized, simplified treatment is that the effect of turbulence can be easily distinguished and quantified from other effects/processes. We have addressed this limitation in the manuscript (lines 186-190): “It should be pointed out that the dissipation rate in cumulus clouds tends to increase with height. For simplicity, the eddy dissipation rate (ϵ) for all the turbulent cases is set statistically stationary ($\epsilon = 500\ cm^2\ s^{-3}$). The advantage of this idealized, simplified treatment is that the effect of turbulence can be easily separated from aerosol effects. A dissipation rate of $500\ cm^2\ s^{-3}$ represents a strongly turbulent environment in cumulus clouds to examine the upper-bound of turbulent effects on the DSD evolution.”

Fourth, the DNS domain is relatively small. Restricting vertical motions to 16.5 cm, the impact of supersaturation fluctuations is significantly restricted (e.g., Abade et al. 2018).

On the one hand, the turbulence in the adiabatic region is nearly homogeneous and isotropic (Vaillancourt et al. 2002), and the supersaturation fluctuations at local scales are mainly induced by droplet condensation and evaporation. We added a discussion on the impact of small-scale turbulence on supersaturation fluctuation (lines 220-224):

“... the supersaturation fluctuations are weak in an adiabatic core region. Vaillancourt et al. (2002) found that in a quasi-adiabatic environment both particle sedimentation and short-lived turbulent coherent structure reduce the supersaturation fluctuation and decrease the time that droplets are exposed to these fluctuations. We expect that the turbulent-induced condensational broadening is more significant in the cloud edge where entrainment mixing induces large variation in supersaturation fluctuations.”

On the other hand, the fluctuations in the vertical motions of scales larger than traditional DNS may be important to perturb the supersaturation. We discussed the limitation of DNS in the revision on lines 341-350:

“In addition, the in-cloud mixing at a much larger scale than the DNS domain transports and mixes both the air and droplets from different parts of the cloud including the cloud edge, leading to a highly perturbed Lagrangian supersaturation experienced by droplets (Grabowski and Abade, 2017, “eddy hopping effect”). On the other hand, larger turbulent eddies can generate higher supersaturation fluctuations due to a higher variation in a vertical motion and thus may both affect the aerosol activation and broaden the DSD. Traditional DNS which is confined to a relatively small domain size (<1m), and the impact of supersaturation fluctuations is significantly restricted. Methods such as an up-scaled DNS with superdroplets (e.g., Thomas et al., 2020) or representing the large-scale mixing with an external forcing on the thermodynamic fields (Paoli and Shariff, 2009) can be used for studying the impact of turbulent scales on the supersaturation fluctuations and thus on the condensational broadening of DSD. In conclusion, the relative importance of entrainment, eddy hopping effect, small-scale turbulence and aerosols requires further investigation.”

Fifth, the range of investigated aerosol concentrations is relatively narrow. Especially the investigation of even lower aerosol concentrations might be of interest to assess the relative importance of aerosol hygroscopicity and turbulence even further.

We have included the discussion of the range of aerosol concentration in the revision:

“However, the seeding particles in this study only cover a limited range of dry radius ($R = 0.1,\ 1\ \mu m$) and number concentration ($N = 10,\ 20\ cm^{-3}$, corresponding to $10 - 20\%$ increase in the total number concentration). Conditions with more ultra-giant aerosols ($R \gg 1\ \mu m$), lower aerosol concentrations ($N \ll 100\ cm^{-3}$), or highly polluted environment ($N \gg 100\ cm^{-3}$) will be of interest to further assess the relative importance of aerosols and turbulence.”

Minor Comments

Ll. 3, 69: The term “aerosol loading” feels ambiguous here. I believe you want to investigate the effects resulting from changes in the number of CCN.

We have changed the aerosol loading to aerosol number concentration for clarification.

L. 8: Inhibiting is a very strong word here. As long as the smaller droplets are activated and the domain is supersaturated, their growth might be reduced but not inhibited.

We replaced “inhibiting” with “reducing”.

L. 9: “[...] seeding reduces the LWC [...]”. See major comment and revise accordingly.

We have addressed the unphysical behavior of LWC and rerun all the simulations. The new results show that LWC grows linearly with height. It is also found that the LWC is insensitive to aerosols and turbulence (Fig. 7 (a)). We have rewritten the discussion and conclusion based on the new results. For instance, lines 256-277 discussed the insensitivity of LWC to turbulence and aerosols and the implication on autoconversion parameterizations.

Ll. 9 ff.: The term *effective radius* is used interchangeably with *mean radius* within the entire manuscript. While the *mean volume radius* is usually close to the effective radius, this is not necessarily the case for the *arithmetic mean radius*. Please clarify.

We replaced all *effective radius* with *mean radius*. In the revision, we refer *mean radius* only to *the arithmetic mean radius* throughout the manuscript for consistency.

Ll. 35 – 36: The comment regarding the effects of cloud chamber walls is appreciated. However, I feel that this relatively specific effect might demand a reference, e.g., Thomas et al. (2019).

We expanded the argument to include more detail in wall effects on lines 33-37:

“On the other hand, laboratory facilities such as cloud chambers are difficult to create environments scalable to real clouds. Furthermore, the effects of chamber walls, such as the heat and moisture fluxes fed into the solid wall and the droplet loss due to their contact with the wall, are challenging to quantify with considerable uncertainties in the measurements. 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.”

Ll. 43 – 44, 89: Why is it necessary to divide the model into the two steps using a standard parcel model first and subsequently a DNS? I assume it is straightforward to consider the activation of aerosols within the DNS framework, similar to the parcel model. In fact, considering turbulent supersaturation fluctuations during activation might have important implications for the estimated number of cloud droplets (Abade et al. 2018). Overall, this could result in a more consistent modeling approach. And based on the data provided in the manuscript, one could easily estimate that the total computing increases by less than 30 % when the DNS is used for the entire ascent. I consider this increase as acceptable.

We agree that using a DNS model for the entire process will yield a more consistent and more accurate result. One of the purposes of this paper is to reduce the computational load, as not all aerosols are activated, and the unactivated aerosols have little impact on the supersaturations or the DSD. This approach is particularly useful and economical to deal with the situation of a large number of aerosols in the beginning and a small number of activated droplets subsequently.

In addition, it is discussed in the paper that the supersaturation fluctuations at DNS scales are small and therefore have a limited impact on the droplet activation and droplet condensational growth. The eddy-hopping effect discussed in Abade et al. (2018) mainly comes from large scale turbulence due to the fact that the variance of supersaturation increases with the turbulence scales. For the scales of DNS (less than 1m), the fluctuation/variance of supersaturation is too small to see the effect. This argument is also supported by Vaillancourt et al. (2002) which found that small-scale turbulence has a negligible effect on droplet condensational growth. The above discussion is included in the conclusion section (lines 341-350).

L. 84: Add the height of the maximum supersaturation to the text.

Included on lines 84-86:

“The first phase starts from the unsaturated sub-cloud region ($\approx 300\ m$ below cloud base) to the level where the supersaturation reaches a maximum ($\approx 43\ m$ above cloud base, see Fig. 2(a)).”

Caption to Fig. 2: Is it really the relative altitude? It is still the (absolute) height above cloud base. A *relative* altitude is usually divided by another quantity for normalization.

To avoid confusion, we switched to “the height from cloud base ($H - H_{CB}$)”

Ll. 92 – 93: The fact that droplets smaller than 10 μm in radius collide rarely is a well-known fact, and can be obtained from any text book on cloud physics.

This argument is true in the context of a still-air case but needs to be carefully interpreted for turbulent cases. We modified the sentence to “These droplets have very small collision rates even in strong turbulence” for clarification.

L. 94: I believe that the hygroscopicity parameter is represented by κ in the Eq. (1) and (2). Am I right? I suggest stating this explicitly.

Yes, we added κ on line 101, just before introducing equation (1).

Ll. 123 – 124: How do you distinguish between activated and unactivated aerosols? Large aerosols do not need to be activated (i.e., be larger than the critical radius) to behave like a droplet.

We first identified the smallest activated aerosol size by comparing it with its critical radius. All aerosols equal to or larger than this size are treated as “activated” and all aerosols below this size are “unactivated”. In this way, giant aerosols, even though they may not be activated are included in the DNS.

Ll. 126 – 129: Does this manipulation also change the liquid water content in the DNS simulation?

The change of liquid water content due to the fitted size distribution is negligible as the total number of droplets from each bin of the parcel model is much larger than the number of processors (=64) used in the DNS. The resulting difference in the LWC is 0.002g/kg at the initial time of DNS simulation (0.028 g/kg in the DNS and 0.030g/kg in the parcel model).

Ll. 136 – 139, Eq. (2): Why don’t you use the same diffusional growth equation for the parcel and the DNS model?

The equation (2) is almost the same as (1) except that the DNS version considers 1) the ventilation effect (reflected in fv coefficient) and 2) uses instantaneous S and T at droplet location instead of the parcel mean. In the revision, we removed equation (2) and discussed the differences between the parcel model and the DNS model when applying the equation in Section 2.1 (lines 110-115).

Table 1: I suggest using simpler names for the model runs, indicating directly the difference to the control run. E.g., “run NoTurb“ instead of “run B”.

We appended simple straightforward names (CTL, NoTurb, NoSolu, Seed_1N1R, Seed_1N2R, and Seed_2N1R) in Table 2 as well as in the context to explicitly indicate the difference between the six cases.

Ll. 165 – 166: This is misleading since the particles are still lifted with a mean updraft of 2 m s-1.

We have modified the description to “When turbulence is switched off, the background velocity fluctuation is set to $0\ m\ s^{-1}$. Therefore, particle motion is only affected by the mean updraft and gravitational settling… ”

L. 166: What is meant by “turbulent advection of the supersaturation fluctuation”? Is there no turbulent mixing in the DNS domain, only molecular diffusion, when ”turbulence is switched off”? This might overestimate the effects of supersaturation fluctuations caused by processes other than turbulence, e.g., the faster depletion of supersaturation in the vicinity of a large droplet.

Yes, when turbulence is off, there is no turbulent “mixing” of the supersaturation field, though advection will be a more precise term, as turbulence does not truly mix the field which enhances the subsequent mixing by diffusion. As such, the local supersaturation fluctuation is only affected by molecular diffusion and droplet condensation/evaporation.

To reduce the confusion, we changed the statement to “Therefore, particle motion is only affected by the mean updraft and gravitational settling, and the supersaturation fluctuation is only induced by droplet condensation and evaporation.” (lines 178-180)

L. 174: A kinetic energy dissipation rate of 500 cm2 s-3 at cloud base is too high. Typically, the dissipation rate increases with distance to the cloud base, and a value of 500 cm2 s-3 might only be representative for the top of a shallow cumulus cloud.

We addressed this question above under the second comment of the *Limitations of the Modeling framework.*

Sec. 3: I suggest introducing subsections to increase clarity of this section.

We divided the result section into two subsections: 3.1 natural cases and 3.2 seeded cases.

L. 177: You state that the parcel rises up to 1.2 km above cloud base. This ascent lasts 600 s = 10 min, assuming an updraft of 2 m s-1. Accordingly, the analysis after 6 min does not constitute the end of the simulation. Moreover, it is also not clear if this point in time is considered to be after the start of the DNS or after the start of the parcel model.

We thank the reviewer for pointing this out. The calculation was wrong and we corrected it in the revision (line 89)
In the revision, we have extended the simulated time to 500s (Table 1) which gives us a lifting height of 1 km.

L. 184: “Condensational growth after 1 min becomes extremely slow [...]”. “How can we distinguish between condensational growth and collision-coalescence in Fig. 5b (Fig. 3b in the revision)?”

We removed the statement, and the new discussion on distinguishing the condensational growth and collisional growth is based on the relative dispersion in Fig. 7 (c) (lines 243-245):

“Condensational growth narrows the DSD and decreases the relative dispersion in the condensation-only set (dotted lines in Fig. 7(c)). Droplet growth in the first two minutes is prevailed by condensation, as the relative dispersion in the condensation-collision set of experiments well overlaps with that in the condensation-only set.”

L. 187, Fig. 7b (Fig. 7(c) in the revision): The displayed dispersion values are much smaller than the values observed in real clouds. In fact, they are about one order in magnitude smaller. You should comment on this difference and name reasons.

In the previous version, the value of relative dispersion in some cases is << 0.1. The low values are mainly caused by the single-precision issue that truncated the growth of the large droplets (see Fig. A and the discussion under the comment of *"Analysis*"). This round-off error issue led to an overestimation of condensational narrowing and thus a lower dispersion. The issue also caused the LWC, mean radius, etc reached a plateau after a certain time point, which is unphysical. To solve this issue, we have switched all droplet-related variables to double precision.
In the new simulations, the dispersion in the condensation-collision set of experiments is around 0.1 at the end of 500s and is expected to grow.

In the revision, we listed a few reasons to explain the lower value in dispersion than observed in real clouds (lines 251-255):
"It is recognized that the relative dispersion of around 0.1 in this study is smaller than observed in most flight measurements. Firstly, the flight measurement is an average of a long-distance sampling ($\mathcal{O}(100m)$) which does not capture the local property of droplet size distribution and therefore is not comparable to our modeled results. Secondly, the simulations only last for 500s, and it is expected that the relative dispersion keeps growing. Thirdly, our idealized simulation is focused on the cloud adiabatic core which lacks entrainment. Inhomogeneous mixing by entrainment can possibly broaden the DSD. "

Ll.188 – 190: What is meant by "multi-modal feature"? For me, the droplet spectra seem to be almost monomodal. There might be a second mode developing between 20 and 25 µm, but it is not very distinct.

We agreed that the multi-modal feature is not significant. Therefore, we removed the description.

Fig. 3e : After 6 min, small (radius < 1 µm) droplets seem to appear in the droplet size distribution. Where do they come from?

We have rerun all simulations to correct the precision issue, and the new results did not show any small radius $< 1\mu m$.

Ll. 210 ff.: By CCN case you mean the case with solute effects (but no turbulence) or the control case?

It refers to the control case with both CCN and turbulence. To clarify, we have changed the description to “Run A (CTL)”, “Run B (NoTurb)”, and “Run C (NoSolu)” to refer to the control case, non-turbulent case, and no solute case, respectively and updated Table 1 accordingly.

Ll. 221 – 223: In what process are the hygroscopic CCN more effective in the first few minutes?

The original statement was not very clear. We removed the statement and modified the discussion on the effect of CCN hygroscopicity on lines 291-299:

“Finally, aerosol hygroscopicity is key to the onset time of autoconversion. All five aerosol-embedded cases see a similar onset time around T= 4 min. Removing the solute effect (hygroscopic material) in Run C delays the onset of autoconversion by about 1.5 min (green line Fig. 7 (f)). Nevertheless, after T = 6-7 min, the autoconversion rate in Run C exceeds all seeded cases. First, solute (CCN hygroscopicity) has a negligible effect on the growth of small aerosols, as the size distribution of small droplets in Run A and C remain almost identical. This is substantiated by the almost identical collision frequency of droplets below $20\ \mu m$ of the two cases (Fig. 6 (a-c)). Second, seeding reduces the mean radius of the droplets. This leads to a reduction in collisions for droplets over $20\ \mu m$ (Fig. 6(d)) and subsequently decelerates the autoconversion process. The above findings imply that increasing the aerosol size (ultra-giant aerosol) shortens the lifetime of the clouds through a fast onset of rain. And increasing the number of aerosols decelerates the rain process.”

L. 226: Define large droplets by their size range.

We removed the original discussion in the revision.

Ll. 234 – 238: It is impossible that a higher droplet concentration (as a result of the seeding) results in a lower liquid water content. In fact, in a rising parcel without (interactive) entrainment, one would assume a higher liquid water content due to the accelerated depletion of water vapor. See major comments above.

In the new simulations, a similar LWC is observed in both the unseeded and seeded cases. The value is only marginally higher in the seeded cases, but the difference is negligible.

L. 243: A “flatter and broader” droplet size distribution sounds tautological. Since you do not change the number of CCN significantly, a broader distribution needs to be flatter. Or does “flat” refer to another property of the distribution that I miss?

We have removed “flatter” and only used “broader”.

Ll. 231 – 247: All seeding experiments tend to address the effect of additional CCN. Those will always decrease the condensational growth of individual droplets since the water is distributed on a larger number of droplets. The more interesting case would be a reduction in the number of CCN.

A reduction in the number of CCN is arguably the opposite of adding additional CCN. In that sense, we can use the CTL case as a reduced-number case.

Fig. 7: The colors are barely distinguishable. Please change them. Why do you show the radar reflectivity (panel e), which is not discussed in the text?

We have changed the color scales and removed the radar reflectivity from the panel in Fig. 7 and added the autoconversion rate (f) and maximum radius (b).

Ll. 265 – 268: This conclusion is based on inadequate analysis and is not true as outlined in the major comments.

We have modified the conclusion based on the new results and updated discussion of LWC and autoconversion on lines 256-273.

Ll. 270 – 272: This claim is only true because of the limited range of CCN concentrations tested in this study. For the analyzed cloud, it will probably always rain if the CCN concentration is reduced to 10 cm-3 and it will probably never rain if it is increased to 1000 cm-3, irrespective of turbulence or aerosol hygroscopicity.

We agreed that the argument of this study holds under the condition of the limited range of CCN we tested. And we addressed this limitation on lines 315-319:

"However, the seeding particles in this study only cover a limited range of dry radius ($R = 0.1,\ 1\ \mu m$) and number concentration ($N = 10,\ 20\ cm^{-3}$, corresponding to $10 - 20\%$ increase in the total number concentration). Conditions with more ultra-giant aerosols ($R \gg 1\ \mu m$), lower aerosol concentrations ($N \ll 100\ cm^{-3}$), or highly polluted environment ($N \gg 100\ cm^{-3}$) will be of interest to further assess the relative importance of aerosols and turbulence."

Technical Comments

Language: While the language is understandable, several smaller mistakes slow down the reading process.

L. 3: Define DNS in the abstract.

Defined

L. 39: Remove “Lagrangian-tracking”.

Removed

L. 43: DNS has already been defined in l. 36.

Removed the definition

L. 56: It is odd citing Chen et al. (2018b) before citing Chen et al. (2018a).

We arrange the order according to the publication time in which 2018a was published before 2018b.

L. 61: I suggest adding “subsaturated” before downdrafts.

added

L. 81 ff.: Units are usually stated in upright characters and separated by a thin space (\, in LaTeX) from the numerical value.

corrected

L. 107, several other occasions: I suggest using exponents for units (m s-1 instead of m/s), consistent with the notation used throughout the manuscript.

We modified the units for consistency.

L. 200: Why do you introduce the abbreviation BR74, which is used only once in the manuscript?

Fig. 7d: It is g/kg, and not g/Kg.

We changed it to g/kg.

Response letter to reviewer #2

The problems addressed in the paper clearly match the scope of ACP. I concur with the first reviewer that Fig. 7 is a major riddle for the reader. Clearly, the piecewise- linear LWC profile needs to be explained and the “jumps in the statistics” need to be eliminated by deriving spectral properties from the droplet population and not from the binned spectrum.

We have identified the error causing the unphysical behavior of the LWC profile. See detailed discussion in the response to reviewer 1 (see *Analysis* under Major Comments). In the new simulations, the LWC follows a linear trend.

We have replotted Fig. 7 based on the calculation from the droplet population instead of from the binned spectrum.

I list below several other relatively major remarks that warrant requesting a major revision to the simulation protocol, result analysis and the manuscript itself.

1. First of all, I would argue that among all possible choices of the moment to switch on representation of turbulent inhomogeneities (i.e., the switch from parcel to DNS model), the level of peak supersaturation is the most unintuitive one. Numerically, it is likely one of the trickiest points for drop growth solver. Since the solute and curvature effects are resolved in the DNS, why not to benefit and resolve activation, especially as its sensitivity to supersaturation fluctuations is continuously being discussed in literature. It is all the more puzzling as the no-fluctuation activation is coupled with further growth in strongly turbulent environment.

We agree that performing a full DNS experiment and including the activation stage is most beneficial. The main focus of this paper is on the subsequent droplet growth after the activation stage. And the parcel-DNS framework provides an economical approach when the sub-cloud aerosol number concentration is much higher than the cloud droplet number concentration. By filtering out the unactivated aerosols, DNS can largely reduce the computation in tracing individual particles.

We justified this treatment on lines 92-97:

“Outputs from the parcel model at the height with maximum supersaturation are fed into DNS as initial conditions. Because unactivated aerosols have little influence on the subsequent droplet growth or on the water vapor field, only the activated aerosols from the parcel model are carried over to the DNS model as the initial background aerosol condition to decrease the computational load. The CCN size distribution and droplet size distribution are displayed in Fig. 2(c). This parcel-DNS hybrid model provides an economical approach and is the first step towards a fully DNS-resolved simulation of the entire ascending process.“

In the conclusion section (lines 351-355), we bring up the importance of implementing a full DNS from below the cloud base to include the effect of turbulence (and supersaturation fluctuations) on aerosol activation.

“This study proposes the first DNS model framework for scrutinizing the microphysical impact of cloud seeding and presents the first results of such a model. Full DNS modeling from below the cloud base will be the next step to include the effect of turbulence on aerosol activation. Additionally, more realistic scenarios resembling actual hygroscopic seeding conditions, such as utilizing multi-disperse size distributions, different hygroscopicity parameters, and seeding below the cloud base will be designed in the future development and deployment of this framework.”

2. The courageous assumption of 1.5 km adiabatic ascent with constant speed calls at least for more discussion on limitations of the study due to lack of representation of entrainment.

In the revision, we added more discussion about the limitation of a lack of entrainment in our model throughout the article, and in the meantime stressed that our study focuses on the adiabatic core region.

Please see detailed discussion in the response to reviewer 1 under the first point of "*Limitations of the Modeling framework*"

3. The numerical experiments presented in the paper lack any sensitivity analysis that would confirm the convergence of the results and quantify their sensitivity to spatial, spectral and temporal resolution as well as to the choice of setup parameters. For instance, the initial aerosol spectrum is discretised onto a grid of only 39 classes for the parcel simulations, which is a crude resolution. While the Lagrangian-in-radius treatment of particle size evolution is indeed free from numerical diffusion (not dispersion - p5/l117), it is highly sensitive to the spectral discretisation (see e.g. discussion of Fig. 8 in Kreidenweis et al. 2003, doi:10.1029/2002JD002697).

1) Sensitivity on the spatial and temporal resolution has been tested in Chen et al. (2016) for studies without hydrodynamic interactions and in Chen et al (2018) for studies with hydrodynamic interactions:

   The spatial resolution (dx) of the flow field is confined to be smaller than the Kolmogorov length scale (the smallest size of turbulent eddies), see detailed discussion in Chen et al. (2016, p625). In this way, all energy-containing eddies affecting the droplet motion are well-resolved.

   The temporal resolution (dt) is chosen to satisfy the CFL condition of the flow and at the same time well below the droplet inertial response timescale for an accurate representation of droplet trajectory, i.e., $dt = min(dt_{drop}, dt_{flow})$. dt_{flow} has to maintain courant number <0.25, and $dt_{drop} = 0.15\tau_{pmin}$, and τ_{pmin} is the droplet response time of the smallest inertial droplet in the domain. See Chen et al. (2018, Section 2e) for detailed validation.

2) Sensitivity on the bin resolution:

   The 39 classes applied in the parcel model follow the same bin resolution as Xue et al. (2010). The bins are discretized on a log scale with the bin width increased by doubling the mass., i.e., $r(n)^3 = r(1)^3 * 2^{n-1}$. In this way, the resolution is higher at small particle sizes and lower at large particle sizes. This configure is reasonable since the large size has a lower number concentration.

As increasing bin resolution reduces the number concentration for each bin, the bin width should be large enough that at the end we can assign a reasonably large integer number of particles of each size in the DNS domain. With our current resolution, for $r > 3\mu m$, each bin corresponds to less than one particle in each processor of the simulation (the number of droplets is evenly distributed across every processor initially); for $r_{dry} > 1\ \mu m$, each bin corresponds to less than 8 particles; for $r_{dry} > 0.4\ \mu m$, each bin has less than 100 particles.

At the small size end, we agree that the resolution will impact the activation rate of the aerosol, and thus the number of droplets present in the domain will vary. Fig. C below shows the number of concentration of unactivated aerosols varying with bin resolution. The number of bins spans from 32 to 253, corresponding to a multiplication factor of radius between two consecutive bins from 2.2 to 1.1. The red marker shows our current model configuration of 39 bins with a multiplication factor of 2. The total number of unactivated aerosols varies more in the range between 32 - 67 bins. However, 39-bin configuration produces a similar result with the higher resolution cases ($N = 19.66\ cm^{-3}$ in 39-bin vs $N = 20.25\ cm^{-3}$ in 253-bin).

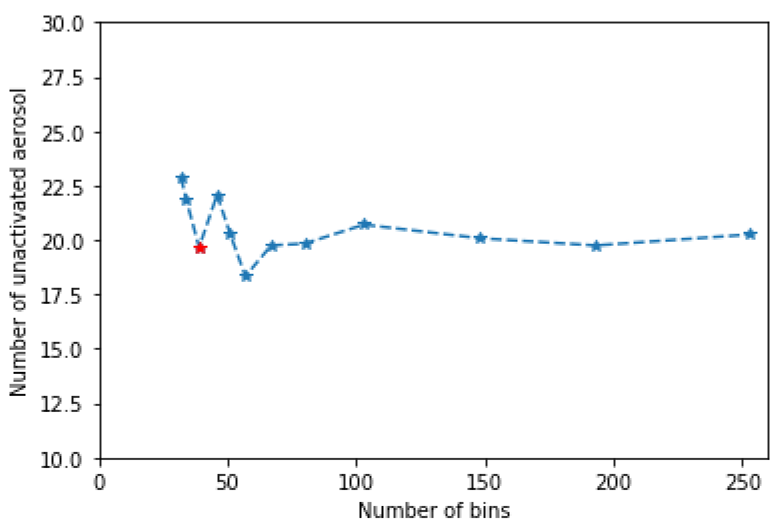

Fig. C: Sensitivity of the number concentration of unactivated aerosols to the number of bins using a multiplication factor from 2.2 to 1.1. The red marker shows our current model configuration of 39 bins with a multiplication factor of 2.

In the end, we stress that the bin resolution in our simulation only matters when comparing to the real observation of aerosol size distribution. First, the continuous lognormal distribution is only the approximated representation of the aerosol size distribution in one maritime condition. Adding the resolution does not refer to a more accurate representation of the observation.

Second, the moving-bin scheme does not produce numerical diffusion of the size distribution (we replaced the word “dispersion” with “diffusion” in the manuscript). It follows that the resolution of the size spectrum will also have no numerical impact on the evolution of the droplet spectrum.

4. Since the simulations feature collisional growth, perhaps it would be beneficial to analyse cloud and drizzle water separately (or is it already the case which could be related to the kink in the LWC profile in Fig. 7?), especially as the authors comment on autoconversion parameterisations. On a related note, the recent work by Noh et al. (2018, doi:10.1175/JAS-D-18-0080.1) is perhaps worth citing when discussing autoconversion rate dependence on spectral parameters (e.g., p8/l198,l204).

The kink was due to the single-precision round-off error, and in the new simulations, the LWC stays linear with height.

We have added the autoconversion rate in Fig. 7. And cited Noh et al. (2018) in the discussion (line 264-265).

5. It would be beneficial to switch from reporting particle concentrations per unit volume to concentrations per unit mass of air, so the variation stemming from diminishing density along the 1.5 km ascent would be excluded. This could also help to understand the difference between the total particle concentration in the log-normal distributions 133 + 66.6 + 3.06 = 202.66 cm^{-3} (in standard T,p conditions?) vs. total initial concentration of 112 cm^{-3} (page 5, lines 112-113).

The number concentration of [133, 66.6, 3.06] mentioned in the manuscript are parameters used for defining the three log-normal distributions. And the aerosol size distribution used in the simulation only took a limited size range. N=202.66 cm^{-3} is the total number concentration over the entire size range ($r = 0 - \infty \ \mu m$). The aerosol size range we chose was from $r = 10^{-3} - 49 \ \mu m$, resulting in $112 \ cm^{-3}$ in total concentration. The difference in concentration per unit volume due to diminishing density is small for an ascending of 1km (< 10%), therefore, we will still keep per unit volume as it is a preferably customary unit in both cloud measurement and modeling communities.

6. Mentioning seeding in the title of the paper would certainly better convey the focus of the study and, in my opinion, “an in-cloud seeding case study” could well replace the “parcel-DNS approach” subtitle.

We have changed the subtitle to “An in-cloud seeding case study using a parcel-DNS approach” to address both the physical process and the method.

7. A table summarising the simulations would be very helpful. Currently, model description is mixed with the set-up description, while some key parameters are hard to find in the text (e.g., domain size is just given in parenthesis in a sentence on particle

concentrations). Also, Table 1 (Table 2 in the revision) would be more helpful with added "collisions" column and with all 12 simulations listed. Same concerns all mentions of "six experiments" - there are 12 DNS runs.

We added Table 1 in the revision to summarize the model description.

To reduce the confusion, we defined the simulations without collisions as the "condensation-only" set and the ones with collisions as the "condensation-collision" set. And we updated the description of the two sets of experiments on lines 169-174:

"Two sets of experiments are performed. Each set consists of six cases, which gives 12 simulations in total. The first set of the experiment includes both condensational and collisional growth of droplets and will be referred to as the "condensation-collision" set. The second set excludes the droplet collision and will be referred to as the "condensation-only" set. The model setup for the two sets is the same other than the difference mentioned above. The configuration of the six cases is listed in Table 2. We focus on the condensation-collision set in the result section unless explicitly specified, and the condensation-only set is for the purpose of comparison to evaluate the influence by condensation and collision-coalescence. "

We modified the caption of Table 2 to:

"Model configuration of the six cases in each set of the experiment. Two sets of experiments are performed: set one includes both collision and condensation in the droplet growth and is referred to as the "condensation-collision" set; set two only considers droplet condensation and is referred to as the "condensation-only" set. This gives 12 cases in total. The natural DSD is taken from the parcel model output at $S = 1.59\%$. Monodisperse seeding is considered in "seeded" cases with CCN size (R_d) and initial droplet size (R) listed in the table."

8. Last but not least, please clarify if the study can be independently reproduced by providing information on the versions of the model code used and its availability.

We updated the description in the *Code and data availability* section.

Other remarks:

- p1/l17: "interaction" → interactions

Checked

- p1/l34: space before parenthesis missing

Checked

- p2/l23: framework → frameworks

Checked

- p2/l26: “certain microphysical processes” – please be more specific

We removed the sentences and condensed the first paragraph in the introduction section to a more concise form.

- p2/l38: “solve” → “solves”

Checked

- p2/l41: isn’t the motivation to reduce the computational cost, rather than to reduce uncertainty? (replacing DNS with a parcel model actually increases uncertainty...), I would suggest removing the whole paragraph actually (lines 41–51)

The purpose of this parcel-DNS hybrid framework is to reduce the computational cost without sacrificing the accuracy of the physical representation of droplet growth. The hybrid method increases the accuracy in terms of modeling the impact of individual aerosols after the activation stage and reduces the computational cost by excluding the unactivated aerosols.

To clarify the motivation and reduce confusion, we have revised the paragraph as below (lines 39-45):

“Only a few DNS studies to date investigated the evolution of the droplet size distribution (DSD) in an updraft environment (e.g., Chen et al. 2018; Gotoh et al. 2016; Saito and Gotoh 2018). However, the solute effect (aerosol hygroscopicity) and curvature effect were excluded in those works for simplicity. Other DNS studies focused on the steady-state conditions, i.e., zero updrafts with zero mean supersaturation (e.g., Li et al. 2020; Sardina et al. 2015). It is recognized that DNS is computationally expensive. To achieve an accurate representation of cloud microphysics while maintaining a feasible computational load, a hybrid modeling framework that combines a parcel model and a DNS model is proposed in this study.”

- p2/l45: “aerosol processing” in some contexts is used to refer to modification of ambient aerosol after evaporation of droplets (due to aqueous chemical reactions and collisions) – perhaps worth rephrasing

We have replaced the word “aerosol processing” with “aerosol activation”.

- p3/l60: “nuclei ... enhances” → “nuclei ... enhance” (or “representation of ...”)

Checked

- p3/l69: “Section 2.1-2.2” → “Sections 2.1-2.2”

Checked

- p3/l73: “droplet chemistry composition” → “hygroscopicity”

Checked

- p4/Fig2: suggest finding alternative wording for “stairs”, please rephrase the last sentence: “fitting the distributions to the DNS” seems awkward, typo in “processers”

We replaced “stairs” with “histogram” for consistency across the entire article.

We removed the sentence from the figure and explained the treatment of assigning initial droplets in DNS in Section 2.2 (lines 140-147):

“... only the activated aerosols from the parcel model are carried over to the DNS, reducing the particle number concentration to $N = 87\ cm^{-3}$. This treatment avoids the computation of tracking the inactivated particles. In the parcel model, the droplet size is calculated by using the moving-bin method. The dry radius of each bin remains constant, and the wet radius grows by condensation. To assign the initial droplet size and its dry radius in the DNS, we regrouped the activated droplet bins into 15 droplet size groups ($R = 2 - 16\ \mu m$) with an interval of $1\ \mu m$. Their CCN sizes remain the original value. Due to the parallelization setup in the model, the initial number of each droplet size group has to be an exact multiple of the number of processors in the simulation (64 processors are used in the present simulations). Therefore, a small difference in the resulting DSD between the two models is expected, as shown in Fig. 2(c).”

- p5/l109: are four significant digits really necessary when specifying initial RH?

We agreed that the four digits are not necessary regarding the impact on the result. We put it this way to list our model configuration as it was.

- p5/l118: “thermodynamic equilibrium” sounds puzzling, I suggest following Jensen and Nugent and explaining what is meant: “in equilibrium (dr/dt=0)”

Changed the statement

- p6/l136: “aerosol processing” – see comment p2/l45 above

Corrected

- p6/eq2: drop growth equation (2) implies that supersaturation is defined as S = e/es (as in Jensen and Nugent 2007), but in Chen et al. 2018b it is defined as S = qv/qvs – of course numerically almost the same, but perhaps worth clarifying

Added the definition when we introduced S below equation (1).

- p7/l167: why not replacing the inline fraction with just κ = 0?

Corrected

- p7/l171: k→ κ

Corrected

- p7/l166: “turbulent advection of the supersaturation fluctuation” suggests S′ is among the advected quantities

It is correct that S is an advected quantity since S is determined by two passive scalars: T and Qv, both are advected by turbulence

- p8/184: “extremely slow”: be more specific

We have modified the paragraph based on the new simulated results and thus removed this sentence.

- p8/187: “when” → “When”

Corrected

- p8/l192: o() → $\mathcal{O}$

Corrected

- p8/l197: space before parenthesis

Checked

- p8/l202: avoid word “claim”

Changed it to “demonstrates”

- p9/Fig2: mention in the caption that collisions were enabled

Added

- p11/Figs5-6 (Fig. 3 and Fig. 5 in the revision): mention in the caption that collisions were enabled
- p12/Fig7: mention in the caption that collisions were enabled

We explicitly mentioned in the captions that the three plots were based on results in the condensation-collision set of experiments.

- p14/l297-298: remove “which is a major facility”?

Removed

- p14/l300-301: rephrase “support from Cheyenne ... and from Graham and Cedar”

We rephrase the statement to “We would like to acknowledge high-performance computing (HPC) support from Cheyenne, Graham, and Cedar. HPC resources at Cheyenne (doi:10.5065/D6RX99HX) is provided by NCAR's Computational and Information Systems Laboratory and sponsored by the National Science Foundation. HPC resources at Graham and Cedar are provided by Compute Canada (www.computecanada.ca).”

- References: use journal abbreviations

Checked

- References: most entries have doi/url given twice
- References: if there is a doi assigned, do not list url (e.g.: Skamarock et al., Yang et al.)

Removed the URL

Impact of CCN hygroscopicity and turbulence on cloud droplet growth: An in-cloud seeding case study using a parcel-DNS approach

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Abstract.

This paper investigates the relative importance of turbulence and aerosol effects on the broadening of the droplet size distribution (DSD) during the early stage of cloud and raindrop formation. A parcel-direct numerical simulation (DNS) hybrid approach is developed to seamlessly simulate the evolution of cloud droplets in an ascending cloud parcel. The results show that turbulence and CCN hygroscopicity are key to the efficient formation of large droplets. The ultra giant aerosols can quickly form embryonic drizzle drops and thus determines the onset time of autoconversion. However, due to their scarcity in natural clouds, their contribution to the total mass of drizzle drops is insignificant. In the meantime, turbulence sustains the formation of large droplets by effectively accelerating the collisions of small droplets. The DSD broadening through turbulent collisions is significant and therefore yields a higher autoconversion rate compared to that in a non-turbulent case. It is argued that the level of autoconversion is heavily determined by turbulence intensity. This paper also presents an in-cloud seeding scenario designed to scrutinize the effect of aerosols in terms of number concentration and size. It is found that seeding more aerosols leads to higher competition for water vapor and reduces the mean droplet radius, and therefore slows down the autoconversion rate. on the other hand, increasing the seeding particle size can buffer such negative feedback. Despite that the autoconversion rate is prominently altered by turbulence and seeding, parcel-mean variables such as LWC stays nearly identical among all cases. Additionally, the lowest autoconversion rate is not co-located with the smallest mean droplet radius. The finding indicates that the traditional Kessler-type or Sundqvist-type autoconversion parameterizations which depend on the LWC or mean radius could not well-capture the drizzle formation process. Properties related to the width or the shape of the DSD are also needed, suggesting that the Berry-and-Reinhardt scheme is conceptually better. It is also suggested that a turbulence-dependent relative-dispersion parameter should be considered.

1 Introduction

Aerosol-cloud-precipitation interactions represent one of the major uncertainties in weather and climate prediction (Fan et al., 2016). Current atmospheric models can not resolve the microphysical processes and thus rely on parameterizations to represent those interactions. Studies show that model results of the location and intensity of precipitation are sensitive to microphysics schemes (Xue et al., 2017; White et al., 2017; Grabowski et al., 2019). For example, White et al. (2017) showed that the autoconversion scheme is the dominant factor to account for the difference in rain production, and the uncertainty due to the choice of microphysical parameterizations exceeds the effects of aerosols. Up to this date, no benchmark "truth" from either measurements or modeling exists to gauge the performance of various microphysics schemes. On the one hand, in-situ measurements cannot directly obtain the process rates, such as the rate of autoconversion and accretion, which prevents such microphysical processes from being accurately modeled. The community has to rely on laboratory experiments, indirect observations, or theoretical models to develop and validate microphysical schemes (e.g., Stoelinga et al., 2003; Wood et al., 2002; Wang et al., 2005). On the other hand, laboratory facilities such as cloud chambers are difficult to create environments scalable to real clouds. Furthermore, the effects of chamber walls, such as the heat and moisture fluxes fed into the solid wall and the droplet loss due to their contact with the wall, are challenging to quantify with considerable uncertainties in the measurements. 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.

Currently, direct numerical simulation (DNS) is believed to be the only numerical approach capable of simulating the growth of individual cloud particles in turbulent flows (Grabowski et al., 2019). Only a few DNS studies to date investigated the evolution of the droplet size distribution (DSD) in an updraft environment (e.g., Chen et al., 2018b; Gotoh et al., 2016; Saito and Gotoh, 2018). However, the solute effect (aerosol hygroscopicity) and curvature effect were excluded in those works for simplicity. Other DNS studies focused on the steady-state conditions, i.e., zero updrafts with zero mean supersaturation (e.g., Li et al., 2020; Sardina et al., 2015). It is recognized that DNS is computationally expensive. To achieve an accurate representation of cloud microphysics while maintaining a feasible computational load, a hybrid modeling framework that combines a parcel model and a DNS model is proposed in this study. The parcel model provides the mean state of the air parcel and can be used when the effect of turbulence is less prominent. The DNS model explicitly resolves all small-scale turbulent eddies which are key to cloud particle interactions. The Lagrangian particle method is employed in the DNS to track the evolution of individual cloud particles coupling with the turbulent flow. This hybrid parcel-DNS approach allows a close examination of the growth history of cloud particles from aerosol activation to drizzle formation. By comparing simulations with different aerosol and turbulent conditions, we are able to evaluate the contribution of each microphysical component to warm rain initiation. The ultimate goal is to provide a numerical benchmarking tool to better understand aerosol-cloud-precipitation interaction at fine scales and improve the sub-grid-scale representation of clouds and precipitation in numerical weather and climate prediction.

Chen et al. (2018b) found that the evolution of DSD in turbulence is different depending on whether droplets grow by condensation-only, collision-only, or condensation-collision (Fig. 1 in their paper). This reveals that droplet condensation and collisions when interacting with turbulence, cannot be treated as the linear addition of the two processes. Many past DNS studies focused on either the condensation-only process or the collision-only process which might yield biased results. It should be pointed out that autoconversion defined as the mass transfer from small droplets to embryonic drizzle drops via collision-coalescence should not exclude the impact of condensational growth, as the two processes dynamically interact with each other. This paper presents a sequel to the study of Chen et al. (2018b) by addressing several caveats mentioned in their paper. Firstly, Chen et al. (2018b) treated only pure water droplets as is commonly assumed in most DNS studies (e.g., Sardina et al., 2015; Vaillancourt et al., 2001, 2002; Paoli and Shariff, 2009). This simplification may underestimate the rate of droplet growth by condensation. Jensen and Nugent (2017) found that cloud condensation nuclei (CCN) strongly enhances the particle growth, and droplets with giant CCN can even grow in regions of sub-saturated downdrafts. In our new hybrid approach, we use an accurate droplet diffusional growth equation including both curvature effect and solute effect. Secondly, the initial DSD in Chen et al. (2018b) obtained from flight observations was a result of averages over a long-time period and along a long sampling path (including both core regions and cloud edges). The average might mask the local property of an adiabatic core that the DNS aims to simulate. The adiabatic cores are regions free of entrainment of dry air. This region has a higher liquid water content (LWC) than the rest of the cloud and is argued to favor the formation of raindrops (Khain et al., 2013). To represent the DSD evolution at the core region, we prescribe here a dry aerosol size distribution in the sub-cloud region, and the aerosol activation process is explicitly simulated by a parcel model to provide a more physically-based initial DSD for the DNS.

The main purpose of the present study is to investigate the relative importance of turbulence, CCN hygroscopicity and aerosols (size and number concentration) on the DSD broadening in cumulus clouds. The paper is organized as follows. Sections 2.1-2.2 introduce the hybrid model of a parcel-DNS framework, to seamlessly simulate early cloud development from aerosol activation to cloud droplet growth. In Section 2.3, the configuration of the 12 numerical simulations are described to compare the microphysical responses to turbulence (turbulent vs non-turbulent), hygroscopicity (pure-water droplets vs CCN-embedded droplets), aerosol size and number concentration (with or without extra aerosols injected), and droplet growth mechanisms (condensation-only vs condensation-collision). Results are presented in Section 3, showing that turbulence and CCN hygroscopicity are key to the formation of big droplets, and seeding slows down the broadening and lowers the autoconversion rate. Summary and outlook for future work are in Section 4.

2 Model setup

A hybrid model is used in this paper for simulating the droplet growth inside an ascending cloud parcel. The ascending process is divided into two phases based on the distinct dominant microphysical processes. A parcel model and a DNS model are combined to seamlessly simulate the two phases, as illustrated in the schematic diagram in Fig. 1. The first phase starts from the unsaturated sub-cloud region ($\approx 300\ m$ below cloud base) to the level where the supersaturation reaches a maximum ($\approx 43\ m$ above cloud base, see Fig. 2(a)). During this phase, supersaturation increases with height, and the microphysical process is dominated by aerosol activation. Cloud particles remain small and collisional growth is negligible. A non-turbulent parcel model is employed to calculate the droplet growth by condensation in this phase. The second phase starts from the level of maximum supersaturation (=1.59%) to 1 km above which takes 500 sec in simulated time (Table 1). At this stage, no new activation occurs as the supersaturation starts to decrease with height. This phase is dominated by cloud droplet growth, and aerosol activation is unimportant. DNS model is employed to calculate individual droplet growth by condensation and collision affected by its immediate local turbulent environment. Outputs from the parcel model at the height with maximum supersaturation are fed into DNS as initial conditions. Because unactivated aerosols have little influence on the subsequent droplet growth or on the water vapor field, only the activated aerosols from the parcel model are carried over to the DNS model as the initial background aerosol condition to decrease the computational load. The CCN size distribution and droplet size distribution are displayed in Fig. 2(c). This parcel-DNS hybrid model provides an economical approach and is the first step towards a fully DNS-resolved simulation of the entire ascending process.

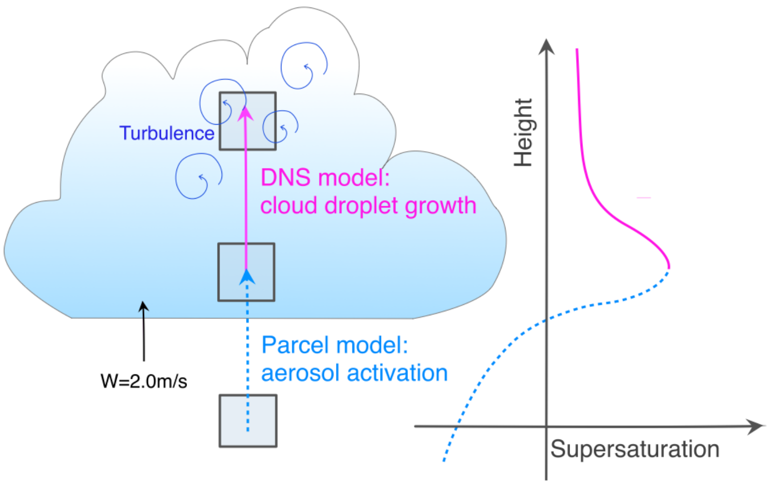

Figure 1. Schematic diagram of the parcel-DNS hybrid model along with the unscaled parcel-mean supersaturation with height. The parcel model simulates the ascending process below the height of maximum supersaturation (dashed blue line), and the DNS simulates the subsequent ascending process (solid violet line).

2.1 Parcel model

The parcel model is adopted from Jensen and Nugent (2017) with two main modifications: (1) The droplet collision-coalescence is excluded for simplicity because most particles in this phase are smaller than 10 μm. These droplets have very small collision rates even in strong turbulence (Chen et al., 2016, 2018a), and the growth is dominated by condensation. (2) The hygroscopicity

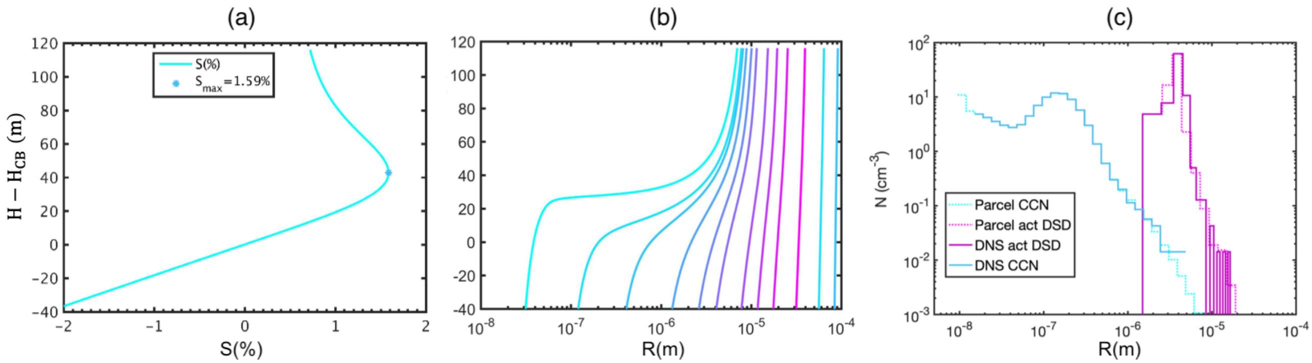

Figure 2. (a) Supersaturation and (b) radius of droplets with different initial wet sizes varying with the height from cloud base ($H - H_{CB}$). Only bins of activated particles are illustrated in (b). (c) The background natural CCN (dry particle) size distribution in the parcel model (light dotted blue histogram) and in the DNS model (darker solid blue histogram), and the droplet size distribution at maximum supersaturation ($S_{max} = 1.59\%$) in the parcel model (light dotted violet histogram) and in the DNS model (darker solid violet histogram).

parameter, κ, proposed by Petters and Kreidenweis (2007, their equation (6)) is employed in the droplet diffusional growth equation:

$$R\frac{dR}{dt} = \frac{S - \frac{R^3 - R_d^3}{R^3 - R_d^3(1-\kappa)} exp(\frac{2\sigma_w}{R_v \rho_w T R})}{\frac{\rho_w R_v T}{e_s D'} + \frac{\rho_w L_v}{K' T}(\frac{l_v}{R_v T} - 1)} f_v, \quad (1)$$

here R is droplet radius, R_d is the radius of CCN, $\sigma_w = 7.2 \times 10^{-2}\ Jm^{-2}$ is surface tension of water against air, $R_v = 467\ Jkg^{-1}K$ is individual gas constant for water vapor, ρ_w and ρ_a are the density of water and air, respectively, T is air temperature, and e_s is the saturated water vapor pressure. D' and K' are respectively the water vapor diffusivity and thermal conductivity that include kinetic effects (see equation (11a)-(11b) in Grabowski et al., 2011), and $L_v = 2.477 \times 10^6\ Jkg^{-1}$ is the latent heat of vaporization. S is supersaturation ratio defined as $\frac{q_v}{q_{vs}} - 1$ where q_v and q_{vs} are water vapor mixing ratios at the current condition and at saturated condition, respectively. f_v is ventilation coefficient which takes into account the distortion in water vapor field around the droplet surface when the droplet moves relative to the flow.Studies show that the effect is negligible when droplets are smaller than 10 μm in radius (Rogers and Yau, 1989, p116), and most droplets in the parcel model are below 10 μm. Therefore, the ventilation effect is excluded in this phase, i.e., $f_v = 1$. In DNS, we apply the empirical formulas of f_v from Beard and Pruppacher (1971) which depends on the droplet Reynolds number and Schmidt number (see also equation (B2)-(B3) in Chen et al., 2018b).

There are two advantages of using the hygroscopicity parameter: 1) The chemical information of the aerosol (i.e., molecular weight, van Hoff factor, density, etc.) is simplified into a single parameter in the solute term; 2) the hygroscopicity parameter of mixed solute due to collision-coalescence can be simply calculated by a weighted average of the volume fractions of each component in the mixture (Petters and Kreidenweis, 2007).

The initial environmental conditions are taken from the cumulus cloud case of Jensen and Nugent (2017, Table 2). The parcel ascends from $H = 600\ m$ ($\approx 284\ m$ below cloud base) with a constant updraft velocity of $2.0\ ms^{-1}$, resembling a fair-weather cumulus cloud condition. The detailed information is listed in Table 1. The CCN (dry aerosol) size distribution fits a lognormal distribution, taken from the pristine case of Xue et al. (2010) (light blue histogram in Fig. 2(c)). The distribution consists of three log-normal modes in which the geometric mean dry radii in the three modes are $R = [0.0039, 0.133, 0.29]\ \mu m$, the geometric standard deviations are $\sigma = [1.5128, 0.4835, 0.9118]$, and the total number concentrations of the whole size range are $N = [133, 66.6, 3.06]\ cm^{-3}$. The initial size is discretized into 39 bins between $0.006\ \mu m$ and $49\ \mu m$, which gives a total number concentration $N = 112\ cm^{-3}$. It is worth noting that the number concentration of CCN larger than $10\ \mu m$ is below $10^{-4}\ cm^{-3}$, corresponding to less than one particle in the DNS domain ($L = 16.5\ cm$). The hygroscopicity parameter of all aerosols is assumed $\kappa = 0.47$. The moving-bin method or moving-size-grid method (see discussion in Yang et al., 2018) is applied to calculate the evolution of the DSD. In this way, the numerical diffusion caused by the Eulerian bins can be avoided (Morrison et al., 2018; Grabowski et al., 2019). at the given ambient humidity (Jensen and Nugent, 2017). As illustrated in Fig. 2(b), the droplets with initial radius below $1\ \mu m$ grow quickly by condensation between $20-40\ m$ above the cloud base before the maximum supersaturation is reached, and droplets larger than $1\ \mu m$ grow slower, creating a narrow DSD near the cloud base.

Table 1. Model description and initial conditions of the parcel model and the DNS model.

	Parcel	DNS
Model description		
Domain size	0D air parcel	$0.165 \times 0.165 \times 0.165\ m^3$
Δx	-	$1.289 \times 10^{-3}\ m$
Δt	$10^{-4}\ s$	$3.15 \times 10^{-5}\ s$
Microphysics treatment	moving-bin method	Lagrangian particle method
Initial conditions		
Initial temperature	284.3 K	281.2 K
Initial pressure	938.5 hPa	902.2 hPa
Initial number concentration of natural background aerosols	$112\ cm^{-3}$	$85\ cm^{-3}$
Initial saturation ratio	85.61%	101.59%
Updraft velocity	$2.0\ m\ s^{-1}$	$2.0\ m\ s^{-1}$
Simulated time	300 sec	500 sec

2.2 DNS model

All DNS simulations are initialized with an identical mean state listed in Table 1. A constant mean updraft speed of $2\ m\ s^{-1}$ is prescribed to lift the air parcel. The initial mean-state variables for DNS are obtained from the parcel model output at maximum supersaturation ($S = 1.59\%$). Above this altitude, no further activation is expected in the parcel due to the decreasing supersaturation. The inactivated aerosols, corresponding to the first two bins of the light blue histogram in Fig. 2(c), do not influence the subsequent evolution of the DSD. Therefore, only the activated aerosols from the parcel model are carried over to the DNS, reducing the particle number concentration to $N = 85\ cm^{-3}$. This treatment avoids the computation of tracking the inactivated particles. In the parcel model, the droplet size is calculated by using the moving-bin method. The dry radius of each bin remains constant, and the wet radius grows by condensation. To assign the initial droplet size and its dry radius in the DNS, we regrouped the activated droplet bins into 15 droplet size groups ($R = 2 - 16\ \mu m$) with an interval of $1\ \mu m$. Their CCN sizes remain the original value. Due to the parallelization setup in the model, the initial number of each droplet size group has to be an exact multiple of the number of processors in the simulation (64 processors are used in the present simulations). Therefore, a small difference in the resulting DSD between the two models is expected, as shown in Fig. 2(c).

The DNS model in the present study is initially developed by Vaillancourt et al. (2001) and has undergone a few modifications since then (Franklin et al., 2005; Chen et al., 2016, 2018a, b). The model employs two sets of equations: 1) the macroscopic equations to calculate the base-state (parcel-mean) variables, and 2) the microscopic equations to calculate the fluctuation of the variables affected by the small-scale turbulence and the local droplet condensation. A detailed description of the DNS model can be found in Chen et al. (2018b, Section 2 and Appendix B).

Two modifications are made in the present study. First, we use equation (1) to replace the simplified version of the droplet growth equation in Chen et al. (2018b, equation (B1)) where the curvature term and the solute term are excluded. Parcel model studies on droplet condensation in a lifted parcel show that the curvature term and the solute term can lead to condensational broadening on the droplet size spectrum. Srivastava (1991) demonstrated that the curvature effect is essential for DSD broadening in an ascending parcel. Korolev (1995) found that the curvature effect and the solute effect lead to irreversible broadening when supersaturation fluctuations are present. It is also found that aerosols of different sizes and different hygroscopicity can cause spectral broadening without supersaturation fluctuations (Çelik and Marwitz, 1999; Jensen and Nugent, 2017). Therefore, it is crucial to examine whether these effects are important in spectral broadening when they dynamically couple with droplet collisional growth in a turbulent environment. Second, droplets with $R < 5\ \mu m$ are treated as non-inertial particles due to their small Stokes number, i.e., their velocity is equal to the flow velocity. The length of a timestep is constrained by the inertial response time of the smallest inertial particle (see discussion in Chen et al., 2018a, on the length of the timestep). The treatment above avoids using too small a timestep when small aerosols are introduced. For droplets between $5 - 40\ \mu m$, their motion is determined by both the Stokes drag force and gravity, and for droplets over $40\ \mu m$ nonlinear drag force is considered (see full description below the equation (B10) in Chen et al., 2018b). Droplets over $50\ \mu m$ are treated as fall-out and are removed from the simulation.

2.3 DNS experimental design

Two sets of experiments are performed. Each set consists of six cases, which gives 12 simulations in total. The first set of the experiments includes both condensational and collisional growth of droplets and will be referred to as the "condensation-collision" set. The second set excludes the droplet collision and will be referred to as the "condensation-only" set. The model setup for the two sets is the same other than the difference mentioned above. The configuration of the six cases is listed in Table

2. We focus on the condensation-collision set in the result section unless explicitly specified, and the condensation-only set is for the purpose of comparison to evaluate the influence by condensation and collision-coalescence.

Table 2. Model configuration of the six cases in each set of the experiment. Two sets of experiments are performed: set one includes both collision and condensation in the droplet growth and is referred to as the "condensation-collision" set; set two only considers droplet condensation and is referred to as the "condensation-only" set. This gives 12 cases in total. The natural DSD is taken from the parcel model output at $S = 1.59\%$. Monodisperse seeding is considered in "seeded" cases with CCN size (R_d) and initial droplet size (R) listed in the table.

Experiments		Turbulence	Solute effect	Initial DSD
Natural cases	Run A (CTL)	on	on	Natural DSD
	Run B (NoTurb)	off	on	Natural DSD
	Run C (NoSolu)	on	off	Natural DSD
"Seeded" cases	Run D1 (Seed-1N1R)	on	on	Natural DSD+ “seeding” aerosol ($R_d = 0.1\ \mu m$, $R = 4\ \mu m$, $N = 10\ cm^{-3}$)
	Run D2 (Seed-2N1R)	on	on	Natural DSD + “seeding” aerosol ($R_d = 0.1\ \mu m$, $R = 4\ \mu m$, $N = 20\ cm^{-3}$)
	Run D3 (Seed-1N2R)	on	on	Natural DSD + “seeding” aerosol ($R_d = 1\ \mu m$, $R = 8\ \mu m$, $N = 10\ cm^{-3}$)

Run A (CTL) is the control run. Only one condition is changed in each of the other five cases. Runs A-C use the same initial DSD from the parcel model and are referred to as the “natural” cases. Turbulence and solute effect are switched off in Run B (NoTurb) and Run C (NoSolu), respectively, to gauge the effects of turbulence and CCN hygroscopicity on the DSD. When turbulence is switched off, the background velocity fluctuation is set to $0\ m\ s^{-1}$. Therefore, particle motion is only affected by the mean updraft and gravitational settling, and the supersaturation fluctuation is only induced by droplet condensation and evaporation. When the solute term is switched off, i.e., $\kappa = 0$, droplets consist of only pure water. Runs D1-D3 are referred to as "seeded" cases because an extra number of aerosols are introduced near the cloud base (at the beginning of DNS). Two seeding sizes and two number concentrations are considered, as described in Table 2. Different than the traditional cloud seeding, the same hygroscopicity of $\kappa = 0.47$ is assumed for both the natural aerosols and the seeding aerosols. In Run D1, we introduce seeding aerosol of dry radius $R_d = 0.1\ \mu m$, wet radius $R = 4\ \mu m$, and number concentration $N = 10\ cm^{-3}$. We double the seeding aerosol number concentration in Run D2 and increase tenfold the dry size in Run D3 (see Table 2). It should be pointed out that the dissipation rate in cumulus clouds tends to increase with height (Seifert et al., 2010). For simplicity, the eddy dissipation rate (ϵ) for all the turbulent cases is set statistically stationary ($\epsilon = 500\ cm^2\ s^{-3}$). The advantage of this idealized, simplified treatment is that the effect of turbulence can be easily separated from aerosol effects. A dissipation rate of $500\ cm^2\ s^{-3}$ represents a strongly turbulent environment in cumulus clouds to examine the upper-bound of turbulent effects on the DSD evolution.

3 Results

3.1 Natural cases

We first compare the results of the natural cases (Run A-C) to examine the effect of turbulence and hygroscopicity (solute) on the droplet evolution. Fig. 3 shows that aerosol and turbulence effectively broaden the DSD at different times. Without hygroscopic aerosols in Run C (NoSolu), the DSD broadening is suppressed within the first six minutes. However, the tail evolution quickly catches up and converges to that in Run A (CTL) afterwards. Meanwhile, switching off turbulence in Run B (NoTurb) suppresses the DSD broadening at a later time (Fig. 3). The tail of the spectrum in Run A and Run B stays similar in the first two minutes and starts to differ by a large amount afterwards.

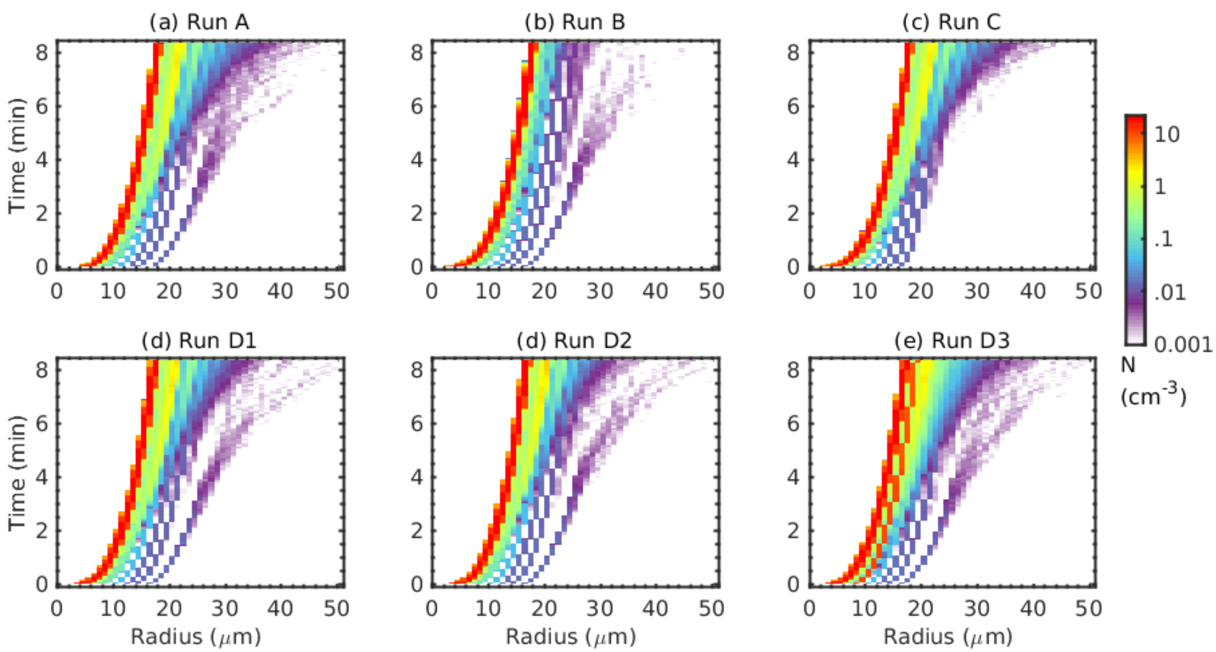

Figure 3. Time evolution of the droplet size distribution in the condensation-collision set of experiments. The droplet number concentration (cm^{-3}) is indicated by colors with its value shown in the color bar. The configuration of each experiment is listed in Table 2.

Turbulence effects on the DSD broadening is minor before T=6 min (Fig. 4(a-b). Both Run A and Run B produce a similar number of droplets over 25 μm at T=6 min. The majority of this size group is grown from the ultra giant aerosol with an initial dry and wet size of $R_d = 4.9\ \mu m$ and $R = 16\ \mu m$. They grow rapidly to 25 μm by condensation within the first two minutes in both Run A and B. However, droplets can hardly reach beyond 30 μm solely by condensation (Fig. 4 (d-e)). The tail over 30 μm is mainly formed by the subsequent collision-coalescence process. Once droplets are over 25 μm, the gravitational collection becomes effective, leading to a similar DSD tail with or without turbulence. However, gravitational collection of droplets below 25 μm in Run B is ineffective to sustain the formation of large droplets. After T=6 min, the tail of DSD in

Run B becomes quasi-stationary for droplets over 20 μm (red and blue histograms in Fig. 4(b)) due to negligible gravitational collisions. This can be illustrated by a negligible collision frequency in Run B in Fig. 6(e). In contrast, a substantial number of droplets $> 20\ \mu m$ are constantly formed in Run A after T=2 min through rapid turbulent collisions. Comparing to collision frequency in Run B (Fig. 5 (b)), turbulence substantially enhances the collisional growth of droplets of $R < 20\ \mu m$. The total collisions in turbulent cases increase by a factor of 20. It is also found that the turbulent enhancement of collisions is strongest among droplet pairs of similar sizes, i.e., with a radius ratio of $r/R > 0.8$. Similar-sized collisions increase by nearly a factor of 50 in turbulent cases, contributing to over 80% of the total collisions as opposed to 34% in Run B. This is because a non-turbulent environment does not favor similar-sized collisions due to a similar droplet settling speed. Turbulence, on the one hand, increases the relative motion between droplets and on the other hand, induces a stronger clustering of similar-sized droplets. The two effects jointly strengthen the similar-sized collisions. The turbulent enhancement on similar-sized collisions is then amplified by the condensational process. Chen et al. (2018b) also demonstrated that as the condensation process reduces the DSD width and generates more similar-sized droplets, turbulence enhances the similar-sized collision and thus broadens the DSD.

Even though turbulence intensifies the collisional growth, the modulation on the droplet condensation is found insignificant. The DSDs in Run A and B in the condensation-only set are nearly identical (Fig. 4 (d-e)). This is because the supersaturation fluctuations are weak in an adiabatic core region. Vaillancourt et al. (2002) found that in a quasi-adiabatic environment both particle sedimentation and short-lived turbulent coherent structure reduce the supersaturation fluctuation and decrease the time that droplets are exposed to these fluctuations. We expect that the turbulent-induced condensational broadening is more significant in the cloud edge where entrainment mixing induces large variation in supersaturation fluctuations.

When solute effect is removed in Run C (NoSolu), droplets can hardly reach beyond 30 μm before T=6 minute (Fig. 4 (c)) because of a lack of ultra-giant aerosols ($R_d > 4\ \mu m$). Embryonic drizzle drops at the early-stage (T<6 min) are formed from the fast growth of the ultra-giant aerosols as seen in both Run A and Run B. No significant change is found in the mean droplet radius and the relative dispersion between Run A and Run C (Fig. 7(d)). Only a slightly lower collision frequency in the droplet size group of $R > 20\ \mu m$ results from a lack of ultra-giant aerosols (see the green histograms in Fig. 5). This implies that the solute effect on droplet condensation in DSD broadening is small for aerosols below $R_d < 4\ \mu m$. The ultra giant aerosols ($R_d = 4.9\ \mu m$ in this study), due to their scarcity, have a negligible contribution in shifting the mean radius and relative dispersion (Fig. 7). As shown in Fig. 3(c), an efficient broadening is triggered at T= 6 minute, resulting in a similar DSD as in Run A at the end of the simulation. It is shown that droplets between $20-30\ \mu m$ are produced through turbulent collisions by the end of T = 6 minutes (Fig. 4(c)), causing a boost in collisions of droplets over 20 μm (Fig. 6(d)).

The time evolution of collision frequency in Fig. 6 shows that all five turbulent cases show a similar trend in total collisional frequency, even though the trend at the four size groups varies. The gravitational collection process is very weak with the collision frequency lowered by at least one order of magnitude in Run B. Still, a slightly higher droplet number concentration at $R > 40\ \mu m$ is observed in both Run A and B than in Run C, because of the presence of ultra giant aerosols. At the same time, the collision frequency of the four size groups in Run A and Run C are almost identical. Even though the ultra giant

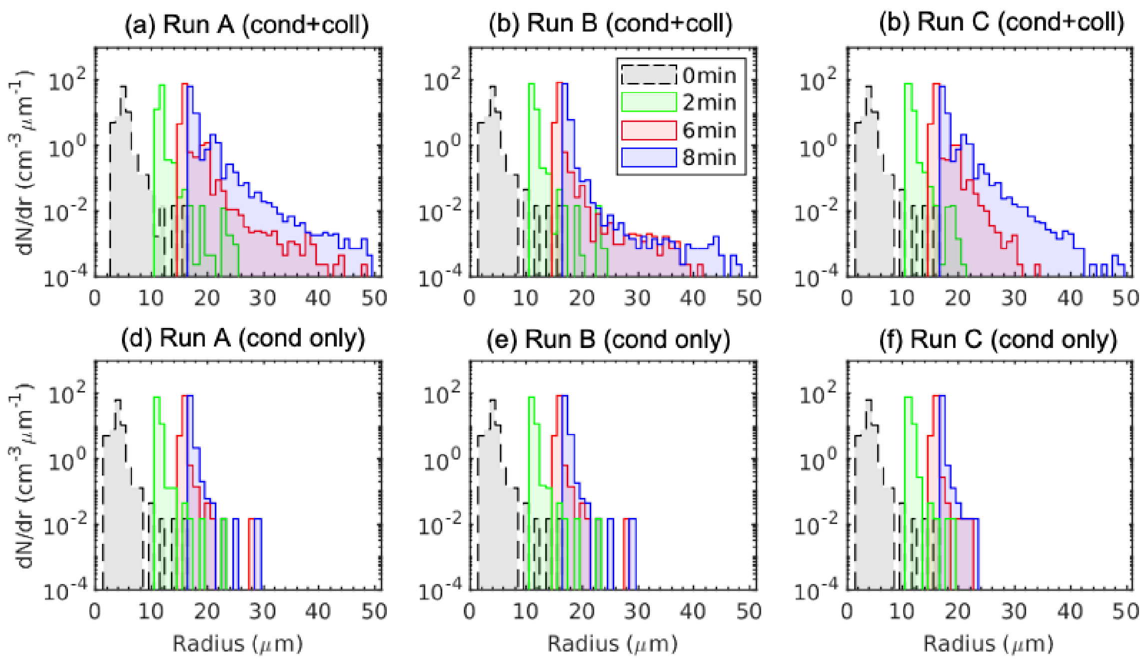

Figure 4. Droplet size distributions (DSDs) at $T = 0\ min$ (grey), $T = 2$ min (green), $T = 6\ min$ (red), and $T = 8\ min$ (blue) of the natural cases (Run A-C). The upper panel (a-c) is the DSD in the condensation-collision set of experiments, and the lower panel (d-e) is the DSD in the condensation-only set of experiments.

aerosols are important in forming early drizzle embryos, due to a low number concentration, they do not sustain an efficient collectional process.

The relative dispersion, defined as the ratio between the standard deviation of the DSD and the mean droplet radius, is an indicator of the width of the DSD. Condensational growth narrows the DSD and decreases the relative dispersion in the condensation-only set (dotted lines in Fig. 7 (c)). Droplet growth in the first two minutes is prevailed by condensation, as the relative dispersion in the condensation-collision set of experiments well overlaps with that in the condensation-only set. After T = 2 min the relative dispersion in the condensation-collision set and the condensation-only set starts to deviate from one another. This is mainly due to two factors: 1) the condensation narrowing slows down as droplets get larger and supersaturation gets lower; 2) the collision rate increases with the increasing droplet mean radius and thus leads to a higher collision rate to strengthen the DSD broadening. In Run B, the collision rate stays the lowest of all cases throughout the simulation (Fig. 6 (e)), leading to the smallest relative dispersion of all the six cases.

It is recognized that the relative dispersion of around 0.1 in this study is smaller than observed in most flight measurements. Firstly, the flight measurement is an average of a long-distance sampling ($\mathcal{O}(100m)$) which does not capture the local property of droplet size distribution and therefore is not comparable to our modeled results. Secondly, the simulations only last for 500s,

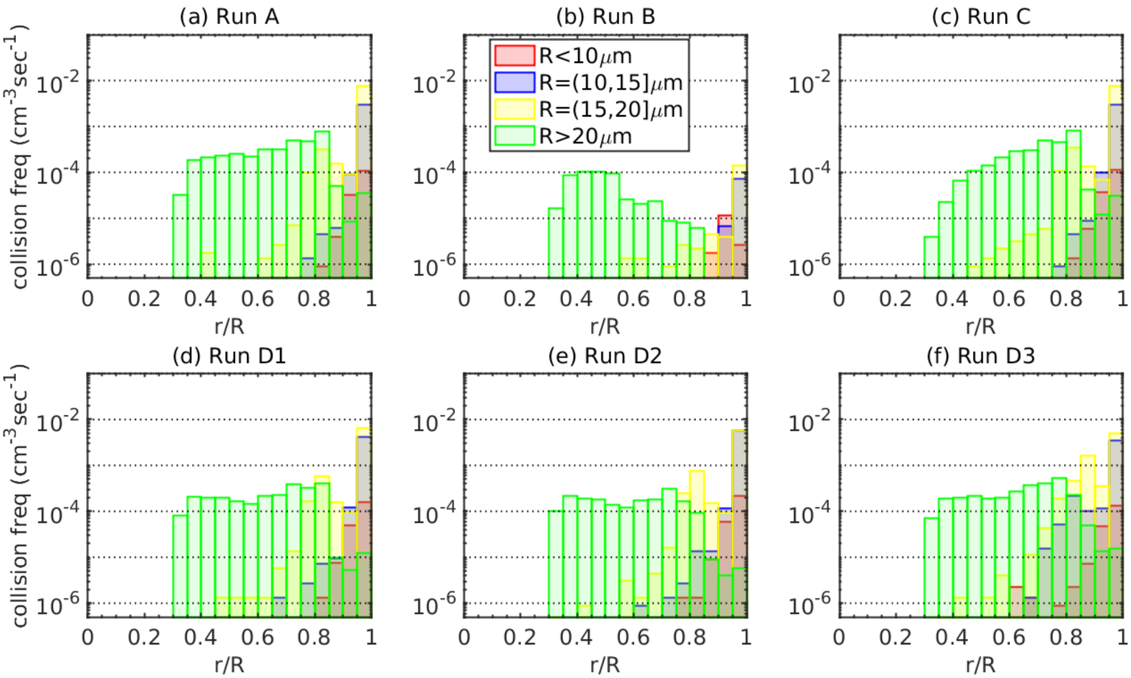

Figure 5. Collision frequency varying with r/R in the condensation-collision set of experiments. r/R is the radius ratio between the small droplet and the large droplet of collided droplet pairs. The droplet pairs are divided into four size groups by the big droplet radius, R, shown in the legend.

and it is expected that the relative dispersion keeps growing. Thirdly, our idealized simulation focuses on the cloud adiabatic core which is devoid of entrainment. Inhomogeneous mixing by entrainment can possibly broaden the DSD.

Despite that DSDs differ among the six cases, the modulation of the bulk condensation by both turbulence and aerosol is negligible, as supported by an almost identical LWC of the six cases (Fig. 7(a)). Turbulence and aerosols redistribute water mass among different droplet sizes by modifying the condensational and collisional growth of individual droplets, thus shifting the droplet statistics such as the mean radius and relative dispersion, and eventually alters the autoconversion rate (Fig. 7(f)). It is also found that even though Run B produces the second largest mean radius, the autoconversion rate stays the lowest, accompanied by the smallest relative-dispersion. Therefore, properties such as the shape of the DSD and relative dispersion are more relevant to autoconversion than the LWC. The traditional autoconversion parameterizations such as the Kessler-type parameterization (Kessler, 1969; Liu and Daum, 2004) and the Sundqvist-type parameterizations (Sundqvist, 1978; Liu et al., 2006) customarily use a threshold function based on the mean radius and/or the LWC. It is suggested that autoconversion rate is also influenced by various other parameters (see Noh et al., 2018, and references therein). The present study demonstrates that both parameters, in particular, the LWC cannot properly capture the trend of the autoconversion. The autoconversion rate by Berry and Reinhardt (1974), and its modified versions which include both the mean droplet size and dispersion parameter,

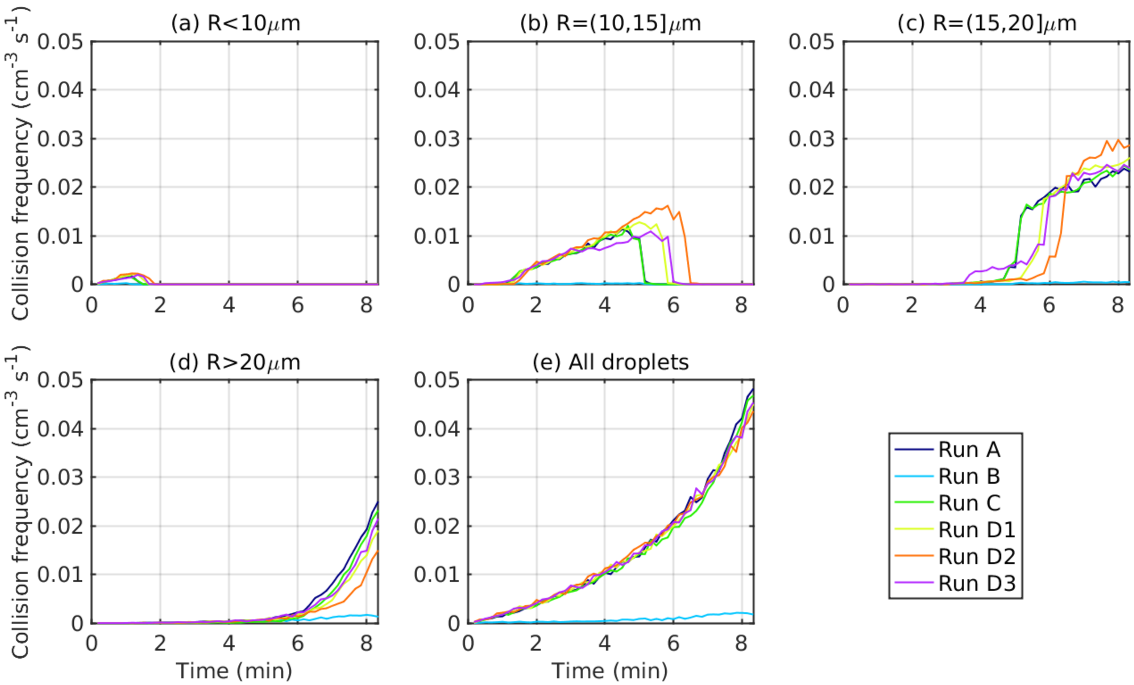

Figure 6. (a-d) Time evolution of collision frequency for droplet pairs of four different size groups mentioned in Fig. 5. (e) Time evolution of collision frequency for all droplet pairs.

is conceptually better than the Kessler-type schemes. Our results thus agree with Gilmore and Straka (2008) which found that the scheme of Berry and Reinhardt (1974) is more sophisticated and requires less tuning to match the observed onset of rain and proportions of cloud and rain. They also found that the growth rate of rain mass and number concentration are highly sensitive to the shape and dispersion parameters. Additionally, it is worth noting that turbulence modifies the collision rate and thus shifts the DSD shape and relative dispersion. Therefore, a turbulence-dependent relative-dispersion parameter is needed in developing the autoconversion scheme.

3.2 Seeded cases

Seeding reduces the mean droplet radius due to higher competition for water vapor among individual droplets (Fig. 7 (d)). Therefore seeding slows down the autoconversion process. Nevertheless, the LWC is not affected by seeding (Fig. 7)a)), which again indicates that the LWC is not a well-related quantity to autoconversion in this case.

When investigating the relative importance of aerosol and turbulence to droplet growth, it is found that the modulation of droplet mean radius by aerosols is larger than the modulation by collision-coalescence. In Fig. 7 (d), the difference between seeded and unseeded cases exceeds the difference between the condensation-only set (dotted lines) and condensation-collision set (solid lines) of each case. Regardless, turbulent collision-coalescence yields large droplets over 30 μm and increases the

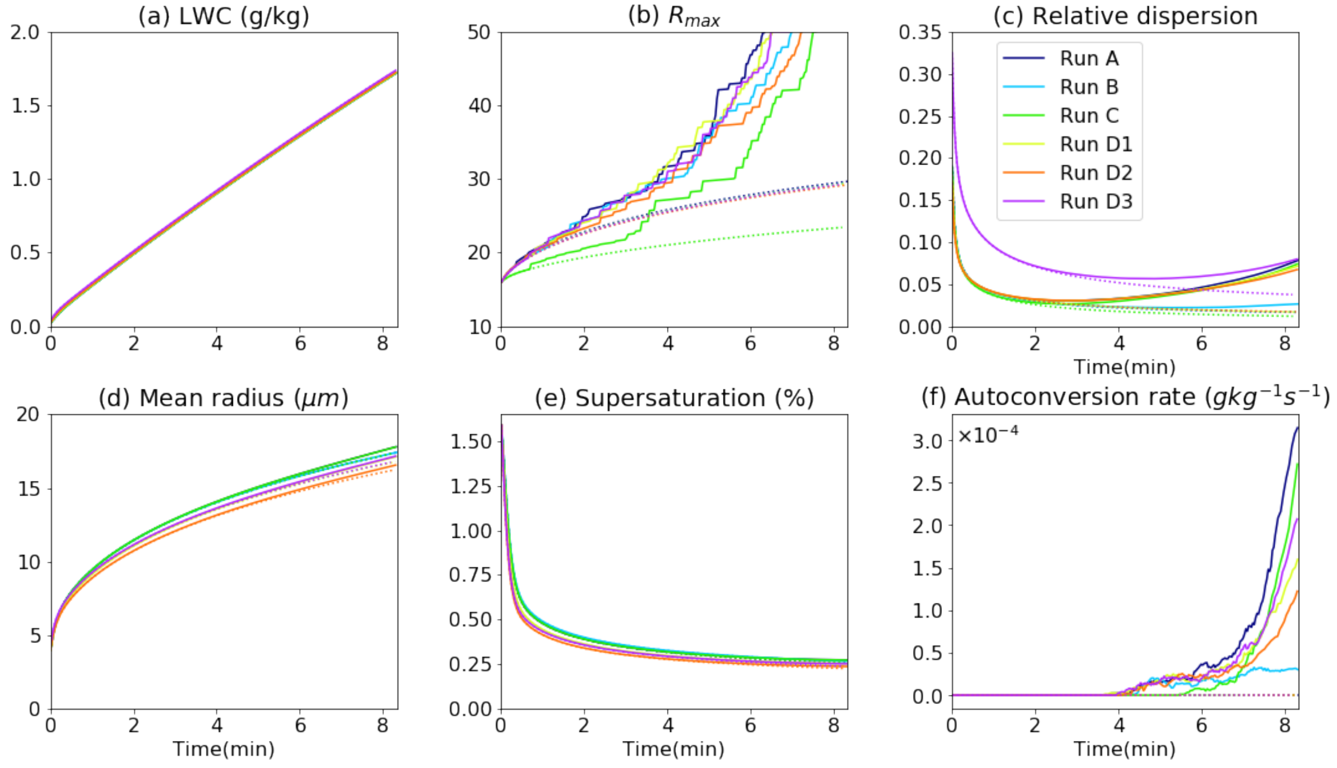

Figure 7. The temporal variation of parcel-mean (a) liquid water content (LWC), (b) maximum droplet radius (R_{max}) , (c) relative dispersion, (d) droplet mean radius, (e) supersaturation ratio, and (f) autoconversion rate in the condensation-collision set of experiments (solid lines) and in the condensation-only set of experiments (dotted lines). The relative dispersion is defined as the standard deviation of the droplet radius divided by the mean radius. The autoconversion rate here is defined as the mass transfer rate from droplet smaller than $R = 30$ μm to droplet larger than 30 μm. The droplets over 50 μm are treated as fall-outs and removed from the domain. Thus (b) only shows a maximum droplet size at 50 μm.

width of the DSD. The total collision rate is heavily determined by the turbulence level and mildly affected by seeding or CCN hygroscopicity (Fig. 6(e)). Besides, the change in R_{max} and relative dispersion due to collisions exceeds that from changing the aerosol condition. As condensational growth can hardly produce droplets over 30 μm, turbulent enhancement of collision is determinant in the mass conversion from small droplets to drizzle embryos. Meanwhile, seeding increases the competition for water vapor among droplets and reduces the mean droplet size, leading to more collisions of small droplets and fewer collisions of large droplets (Fig. 6(a-d)). Specifically, by doubling the seeding aerosol number in Run D2 (Seed-2N1R), the condensational growth of small droplets is further prohibited due to a higher competition of water vapor, resulting in more small droplets. Increasing the size of seeding aerosols in Run D3 (Seed-1N2R) buffers the inhibition effect. The resulting autoconversion rate is Run A > Run D3 > Run D1> Run D2.

Finally, aerosol hygroscopicity is key to the onset time of autoconversion. All five aerosol-embedded cases see a similar onset time around T= 4 min. Removing the solute effect (hygroscopic material) in Run C delays the onset of autoconversion by about 1.5 min (green line Fig. 7 (f)). Nevertheless, after T = 6-7 min, the autoconversion rate in Run C exceeds all seeded cases. First, solute (CCN hygroscopicity) has a negligible effect on the growth of small aerosols, as the size distribution of small droplets in Run A and C remain almost identical. This is substantiated by the almost identical collision frequency of droplets below 20 μm of the two cases (Fig. 6 (a-c)). Second, seeding reduces the mean radius of the droplets. This leads to a reduction in collisions for droplets over 20 μm (Fig. 6(d)) and subsequently decelerates the autoconversion process. The above findings imply that increasing the aerosol size (ultra-giant aerosol) shortens the lifetime of the clouds through a fast onset of rain. And increasing the number of aerosols decelerates the rain process.

4 Summary and discussion

This paper investigates the effects of turbulence and aerosol properties (hygroscopicity, number concentration, and size) on the microphysics during early cloud and rain development. A parcel-DNS hybrid modeling framework is developed. The parcel model is used to generate the initial size distribution of activated aerosols, and the DNS model calculates the subsequent growth of those activated aerosols affected by both the microscopic (turbulent fluctuation) and the macroscopic (parcel mean) environment. By using this economical modeling framework, continuous particle growth from sub-cloud aerosols to cloud droplets is accurately represented.

Overall, ultra-giant aerosols in the natural cases quickly form the drizzle embryo and thus determine the onset time of autoconversion. However, they only form a few big raindrops due to their scarcity, which has little impact on the level of autoconversion. Turbulence enhances the collision frequency by more than one order of magnitude and determines the level of autoconversion. Specifically, turbulence enhances the collisions among similar-sized droplets that are less likely to happen in a non-turbulent environment, effectively broadening the DSD. Therefore, the autoconversion in a turbulent environment is significantly greater than in a non-turbulent environment. It is also found that seeding (increasing aerosol number and size) modifies the level of autoconversion. On the one hand, increasing the aerosol number reduces the mean radius due to stronger competition for water vapor, and therefore slows down the autoconversion. On the other hand, increasing the seeding size can buffer such negative feedback. However, the seeding particles in this study only cover a limited range of dry radius ($R = 0.1,\ 1\ \mu m$) and number concentration ($N = 10,\ 20\ cm^{-3}$, corresponding to $10 - 20\%$ increase in the total number concentration). Conditions with more ultra-giant aerosols ($R \gg 1\ \mu m$), lower aerosol concentrations ($N \ll 100\ cm^{-3}$), or highly polluted environment ($N \gg 100\ cm^{-3}$) will be of interest to further assess the relative importance of aerosols and turbulence. It is argued that predicting the rain onset time requires accurate information and representation of ultra-giant aerosols. And an accurate autoconversion scheme requires a well-quantified turbulent collisions kernel.

Even though the autoconversion rate differs among the six cases, it is found that the parcel-mean variables such as LWC, mean radius, and supersaturation are not sensitive to turbulence level and aerosol conditions. In this case the LWC and mean droplet radius, which are key parameters in Kessler-type or Sundqvist-type autoconversion parameterizations, are not well-related quantities to autoconversion rate, and information of turbulence intensity and aerosols are essential to determine the autoconversion rate. It is argued that these parcel-mean variables are mainly affected by the updraft speed which is held the same among the six cases. Sensitivity studies are needed in the future to investigate the effect of the LWC on the autoconversion rate due to a change in the updraft.

Cloud models are sensitive to microphysics schemes, and the autoconversion parameterization is one of the main sources of uncertainty in the representation of warm clouds and rain with few observations to verify against. The large uncertainty may be ascribed to the decoupling of microphysics from subgrid-scale turbulence and a lack of aerosol information in the parameterization. Therefore, the aerosol effect evaluated by the models should be cautiously interpreted. The hybrid parcel-DNS model can be used for verifying the autoconversion rate affected by turbulence and aerosols at the sub-grid scale of large-eddy simulation (LES).

Despite a good number of improvements made, the current modeling framework still presents the following shortcomings: for simplicity, the same hygroscopic parameter ($\kappa = 0.47$) is assumed among the natural aerosols and the seeding aerosols. Besides, seeding is initialized $40\ m$ above the cloud base while traditional hygroscopic seeding introduces particles around $100 - 300\ m$ below the cloud base. This treatment might affect the model results as seeding below the cloud base influences the activation and growth of the background aerosols and thus modifies the DSD at the cloud base (Cooper et al., 1997).

Our idealized simulations focus on the cloud adiabatic core region and therefore exclude entrainment mixing which is highly active near the cloud edge. The activation of newly entrained aerosols might lead to a further broadening of the DSD (e.g., Lasher-Trapp et al., 2005). In addition, the in-cloud mixing at a much larger scale than the DNS domain transports and mixes both the air and droplets from different parts of the cloud including the cloud edge, leading to a highly perturbed Lagrangian supersaturation experienced by droplets (Grabowski and Abade, 2017, "eddy hopping effect"). On the other hand, larger turbulent eddies can generate higher supersaturation fluctuations due to a higher variation in a vertical motion and thus may both affect the aerosol activation and broaden the DSD. Traditional DNS which is confined to a relatively small domain size (<1 m), and the impact of supersaturation fluctuations is significantly restricted. Methods such as an up-scaled DNS with superdroplets (e.g., Thomas et al., 2020) or representing the large-scale mixing with an external forcing on the thermodynamic fields (Paoli and Shariff, 2009) can be used for studying the impact of turbulent scales on the supersaturation fluctuations and thus on the condensational broadening of DSD. In conclusion, the relative importance of entrainment, eddy hopping effect, small-scale turbulence and aerosols requires further investigation.

This study proposes the first DNS model framework for scrutinizing the microphysical impact of cloud seeding and presents the first results of such a model. Full DNS modeling from below the cloud base will be the next step to include the effect of turbulence on aerosol activation. Additionally, more realistic scenarios resembling actual hygroscopic seeding conditions, such as utilizing multi-disperse size distributions, different hygroscopicity parameters, and seeding below the cloud base will be designed in the future development and deployment of this framework.

Code and data availability. The data produced by the Direct Numerical Simulation (DNS) model and parcel model can be accessed in the Harvard Dataverse repository (Chen et al., 2019, doi:10.7910/DVN/HBIKKV). The parcel model and DNS model used to produce the dataset are available upon request.

Author contributions. This study was co-designed by Sisi Chen, Lulin Xue, and M.K. Yau. Sisi Chen conducted the model simulation, did the data analysis, and wrote the manuscript. Lulin Xue and M.K. Yau provided advice and discussions on the model results and revised the manuscript.

Competing interests. The authors declare that they have no conflict of interest.

Acknowledgements. We thank the two anonymous reviewers for their invaluable comments. This work is supported by the National Center for Atmospheric Research, sponsored by the National Science Foundation under Cooperative Agreement No. 1852977. Part of this work is supported by the National Center of Meteorology, Abu Dhabi, UAE under the UAE Research Program for Rain Enhancement Science. We would like to acknowledge high-performance computing (HPC) support from Cheyenne, Graham, and Cedar. HPC resources at Cheyenne (doi:10.5065/D6RX99HX) is provided by NCAR's Computational and Information Systems Laboratory and sponsored by the National Science Foundation. HPC resources at Graham and Cedar are provided by Compute Canada (www.computecanada.ca).

References

Beard, K. V. and Pruppacher, H. R.: A Wind Tunnel Investigation of the Rate of Evaporation of Small Water Drops Falling at Terminal Velocity in Air, J. Atmos. Sci., 28, 1455–1464, https://doi.org/10.1175/1520-0469(1971)028<1455:awtiot>2.0.co;2, 1971.

Berry, E. X. and Reinhardt, R. L.: An Analysis of Cloud Drop Growth by Collection Part II. Single Initial Distributions, J. Atmos. Sci., 31, 1825–1831, https://doi.org/10.1175/1520-0469(1974)031<1825:aaocdg>2.0.co;2, 1974.

Çelik, F. and Marwitz, J. D.: Droplet Spectra Broadening by Ripening Process. Part I: Roles of Curvature and Salinity of Cloud Droplets, Journal of the Atmospheric Sciences, 56, 3091–3105, https://doi.org/10.1175/1520-0469(1999)056<3091:dsbbrp>2.0.co;2, 1999.

Chen, S., Bartello, P., Yau, M. K., Vaillancourt, P. A., and Zwijsen, K.: Cloud Droplet Collisions in Turbulent Environment: Collision Statistics and Parameterization, J. Atmos. Sci., 73, 621–636, https://doi.org/10.1175/JAS-D-15-0203.1, 2016.

Chen, S., Yau, M. K., and Bartello, P.: Turbulence Effects of Collision Efficiency and Broadening of Droplet Size Distribution in Cumulus Clouds, J. Atmos. Sci., 75, 203–217, https://doi.org/10.1175/JAS-D-17-0123.1, 2018a.

Chen, S., Yau, M.-K., Bartello, P., and Xue, L.: Bridging the condensation–collision size gap: a direct numerical simulation of continuous droplet growth in turbulent clouds, Atmos. Chem. Phys., 18, 7251–7262, https://doi.org/10.5194/acp-18-7251-2018, 2018b.

Chen, S., Xue, L., and Yau, M.: Data support for "Impact of CCN hygroscopicity and turbulence on cloud droplet growth: An in-cloud seeding case study using parcel-DNS approach", https://doi.org/10.7910/DVN/HBIKKV, https://doi.org/10.7910/DVN/HBIKKV, 2019.

Cooper, W. A., Bruintjes, R. T., and Mather, G. K.: Calculations Pertaining to Hygroscopic Seeding with Flares, J. Appl Meteorol., 36, 1449–1469, https://doi.org/10.1175/1520-0450(1997)036<1449:cpthsw>2.0.co;2, 1997.

Fan, J., Wang, Y., Rosenfeld, D., and Liu, X.: Review of Aerosol–Cloud Interactions: Mechanisms, Significance, and Challenges, J. Atmos. Sci., 73, 4221–4252, https://doi.org/10.1175/JAS-D-16-0037.1, 2016.

Franklin, C. N., Vaillancourt, P. A., Yau, M. K., and Bartello, P.: Collision Rates of Cloud Droplets in Turbulent Flow, J. Atmos. Sci., 62, 2451–2466, https://doi.org/10.1175/jas3493.1, 2005.

Gilmore, M. S. and Straka, J. M.: The Berry and Reinhardt Autoconversion Parameterization: A Digest, J. Appl. Meteorol. Clim., 47, 375–396, https://doi.org/10.1175/2007jamc1573.1, 2008.

Gotoh, T., Suehiro, T., and Saito, I.: Continuous growth of cloud droplets in cumulus cloud, New Journal of Physics, 18, 043 042, https://doi.org/10.1088/1367-2630/18/4/043042, 2016.

Grabowski, W. W. and Abade, G. C.: Broadening of Cloud Droplet Spectra through Eddy Hopping: Turbulent Adiabatic Parcel Simulations, Journal of the Atmospheric Sciences, 74, 1485–1493, https://doi.org/10.1175/jas-d-17-0043.1, 2017.

Grabowski, W. W., Andrejczuk, M., and Wang, L.-P.: Droplet growth in a bin warm-rain scheme with Twomey CCN activation, Atmos. Res., 99, 290–301, https://doi.org/10.1016/j.atmosres.2010.10.020, 2011.

Grabowski, W. W., Morrison, H., Shima, S.-I., Abade, G. C., Dziekan, P., and Pawlowska, H.: Modeling of Cloud Microphysics: Can We Do Better?, B. Am. Meteorol Soc., 100, 655–672, https://doi.org/10.1175/BAMS-D-18-0005.1, 2019.

Jensen, J. B. and Nugent, A. D.: Condensational Growth of Drops Formed on Giant Sea-Salt Aerosol Particles, J. Atmos. Sci., 74, 679–697, https://doi.org/10.1175/jas-d-15-0370.1, 2017.

Kessler, E.: On the Distribution and Continuity of Water Substance in Atmospheric Circulations, in: On the Distribution and Continuity of Water Substance in Atmospheric Circulations, pp. 1–84, American Meteorological Society, https://doi.org/10.1007/978-1-935704-36-2_1, 1969.

Khain, A., Prabha, T. V., Benmoshe, N., Pandithurai, G., and Ovchinnikov, M.: The mechanism of first raindrops formation in deep convective clouds, Journal of Geophysical Research: Atmospheres, 118, 9123–9140, https://doi.org/10.1002/jgrd.50641, 2013.

Korolev, A. V.: The Influence of Supersaturation Fluctuations on Droplet Size Spectra Formation, Journal of the Atmospheric Sciences, 52, 3620–3634, https://doi.org/10.1175/1520-0469(1995)052<3620:tiosfo>2.0.co;2, 1995.

Lasher-Trapp, S. G., Cooper, W. A., and Blyth, A. M.: Broadening of droplet size distributions from entrainment and mixing in a cumulus cloud, Quarterly Journal of the Royal Meteorological Society, 131, 195–220, https://doi.org/10.1256/qj.03.199, 2005.

Li, X.-Y., Brandenburg, A., Svensson, G., Haugen, N. E. L., Mehlig, B., and Rogachevskii, I.: Condensational and Collisional Growth of Cloud Droplets in a Turbulent Environment, Journal of the Atmospheric Sciences, 77, 337–353, https://doi.org/10.1175/jas-d-19-0107.1, 2020.

Liu, Y. and Daum, P. H.: Parameterization of the Autoconversion Process.Part I: Analytical Formulation of the Kessler-Type Parameterizations, J. Atmos. Sci., 61, 1539–1548, https://doi.org/10.1175/1520-0469(2004)061<1539:potapi>2.0.co;2, 2004.

Liu, Y., Daum, P. H., McGraw, R., and Wood, R.: Parameterization of the Autoconversion Process. Part II: Generalization of Sundqvist-Type Parameterizations, J. Atmos. Sci., 63, 1103–1109, https://doi.org/10.1175/jas3675.1, 2006.

Morrison, H., Witte, M., Bryan, G. H., Harrington, J. Y., and Lebo, Z. J.: Broadening of Modeled Cloud Droplet Spectra Using Bin Microphysics in an Eulerian Spatial Domain, J. Atmos. Sci., 75, 4005–4030, https://doi.org/10.1175/jas-d-18-0055.1, 2018.

Noh, Y., Oh, D., Hoffmann, F., and Raasch, S.: A Cloud Microphysics Parameterization for Shallow Cumulus Clouds Based on Lagrangian Cloud Model Simulations, Journal of the Atmospheric Sciences, 75, 4031–4047, https://doi.org/10.1175/jas-d-18-0080.1, 2018.

Paoli, R. and Shariff, K.: Turbulent Condensation of Droplets: Direct Simulation and a Stochastic Model, J. Atmos. Sci., 66, 723–740, https://doi.org/10.1175/2008JAS2734.1, 2009.

Petters, M. D. and Kreidenweis, S. M.: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, Atmos. Chem. Phys., 7, 1961–1971, https://doi.org/10.5194/acp-7-1961-2007, 2007.

Rogers, R. R. and Yau, M. K.: A Short Course in Cloud Physics, Butterworth-Heinemann, 3rd edn., 1989.

Saito, I. and Gotoh, T.: Turbulence and cloud droplets in cumulus clouds, New Journal of Physics, 20, 023 001, https://doi.org/10.1088/1367-2630/aaa229, 2018.

Sardina, G., Picano, F., Brandt, L., and Caballero, R.: Continuous Growth of Droplet Size Variance due to Condensation in Turbulent Clouds, Phys. Rev. Lett., 115, 184 501, https://doi.org/10.1103/PhysRevLett.115.184501, 2015.

Seifert, A., Nuijens, L., and Stevens, B.: Turbulence effects on warm-rain autoconversion in precipitating shallow convection, Quarterly Journal of the Royal Meteorological Society, 136, 1753–1762, https://doi.org/10.1002/qj.684, 2010.

Srivastava, R. C.: Growth of Cloud Drops by Condensation: Effect of Surface Tension on the Dispersion of Drop Sizes, Journal of the Atmospheric Sciences, 48, 1596–1599, https://doi.org/10.1175/1520-0469(1991)048<1596:gocdbc>2.0.co;2, 1991.

Stoelinga, M. T., Hobbs, P. V., Mass, C. F., Locatelli, J. D., Colle, B. A., Houze, R. A., Rangno, A. L., Bond, N. A., Smull, B. F., Rasmussen, R. M., Thompson, G., and Colman, B. R.: Improvement of Microphysical Parameterization through Observational Verification Experiment, B. Am. Meteorol Soc., 84, 1807–1826, https://doi.org/10.1175/BAMS-84-12-1807, 2003.

Sundqvist, H.: A parameterization scheme for non-convective condensation including prediction of cloud water content, Q. J. Roy. Meteor. Soc., 104, 677–690, https://doi.org/10.1002/qj.49710444110, 1978.

Thomas, L., Grabowski, W. W., and Kumar, B.: Diffusional growth of cloud droplets in homogeneous isotropic turbulence: DNS, scaled-up DNS, and stochastic model, Atmospheric Chemistry and Physics Discussions, 2020, 1–21, https://doi.org/10.5194/acp-2020-159, https://www.atmos-chem-phys-discuss.net/acp-2020-159/, 2020.

Thomas, S., Ovchinnikov, M., Yang, F., Voort, D., Cantrell, W., Krueger, S. K., and Shaw, R. A.: Scaling of an Atmospheric Model to Simulate Turbulence and Cloud Microphysics in the Pi Chamber, J. Adv. Model Earth Sy., 11, 1981–1994, https://doi.org/10.1029/2019ms001670, 2019.

Vaillancourt, P. A., Yau, M. K., and Grabowski, W. W.: Microscopic Approach to Cloud Droplet Growth by Condensation. Part I: Model Description and Results without Turbulence, J. Atmos. Sci., 58, 1945–1964, https://doi.org/10.1175/1520-0469(2001)058<1945:MATCDG>2.0.CO;2, 2001.

Vaillancourt, P. A., Yau, M. K., Bartello, P., and Grabowski, W. W.: Microscopic Approach to Cloud Droplet Growth by Condensation. Part II: Turbulence, Clustering, and Condensational Growth, J. Atmos. Sci., 59, 3421–3435, https://doi.org/10.1175/1520-0469(2002)059<3421:MATCDG>2.0.CO;2, 2002.

Wang, L.-P., Ayala, O., Kasprzak, S. E., and Grabowski, W. W.: Theoretical Formulation of Collision Rate and Collision Efficiency of Hydrodynamically Interacting Cloud Droplets in Turbulent Atmosphere, J. Atmos. Sci., 62, 2433–2450, https://doi.org/10.1175/JAS3492.1, 2005.

White, B., Gryspeerdt, E., Stier, P., Morrison, H., and Thompson, G.: Uncertainty from the choice of microphysics scheme in convection-permitting models significantly exceeds aerosol effects, Atmos. Chem. Phys. Discuss., 17, https://doi.org/10.5194/acp-2016-760, 2017.

Wood, R., Field, P., and Cotton, W.: Autoconversion rate bias in stratiform boundary layer cloud parameterizations, Atmos. Res., 65, 109–128, https://doi.org/10.1016/s0169-8095(02)00071-6, 2002.

Xue, L., Teller, A., Rasmussen, R., Geresdi, I., and Pan, Z.: Effects of Aerosol Solubility and Regeneration on Warm-Phase Orographic Clouds and Precipitation Simulated by a Detailed Bin Microphysical Scheme, J. Atmos. Sci., 67, 3336–3354, https://doi.org/10.1175/2010jas3511.1, 2010.

Xue, L., Fan, J., Lebo, Z. J., Wu, W., Morrison, H., Grabowski, W. W., Chu, X., Geresdi, I., North, K., Stenz, R., Gao, Y., Lou, X., Bansemer, A., Heymsfield, A. J., McFarquhar, G. M., and Rasmussen, R. M.: Idealized Simulations of a Squall Line from the MC3E Field Campaign Applying Three Bin Microphysics Schemes: Dynamic and Thermodynamic Structure, Mon. Weather Rev., 145, 4789–4812, https://doi.org/10.1175/MWR-D-16-0385.1, 2017.

Yang, F., Kollias, P., Shaw, R. A., and Vogelmann, A. M.: Cloud droplet size distribution broadening during diffusional growth: ripening amplified by deactivation and reactivation, Atmos. Chem. Phys., 18, 7313–7328, https://doi.org/10.5194/acp-18-7313-2018, 2018.