

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<sup>-4</sup>. 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

$\frac{dR^2}{dt} = 2K f_v S$ , where  $K^{-1}$  is a temperature-dependent coefficient. In the present study, the droplet growth equation is modified to  $\frac{dR^3}{dt} = 3K f_v (S - f(\text{solu}, \text{curv}))$ , where  $f(\text{solu}, \text{curv})$

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  $\frac{dR^2}{dt} = 2K f_v S$  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  $R^3$ :  $R_{new}^3 = R_{old}^3 + dR^3$ .  $dR^3$  is rounded off to 0 when it is added to  $R_{old}^3$ , 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.

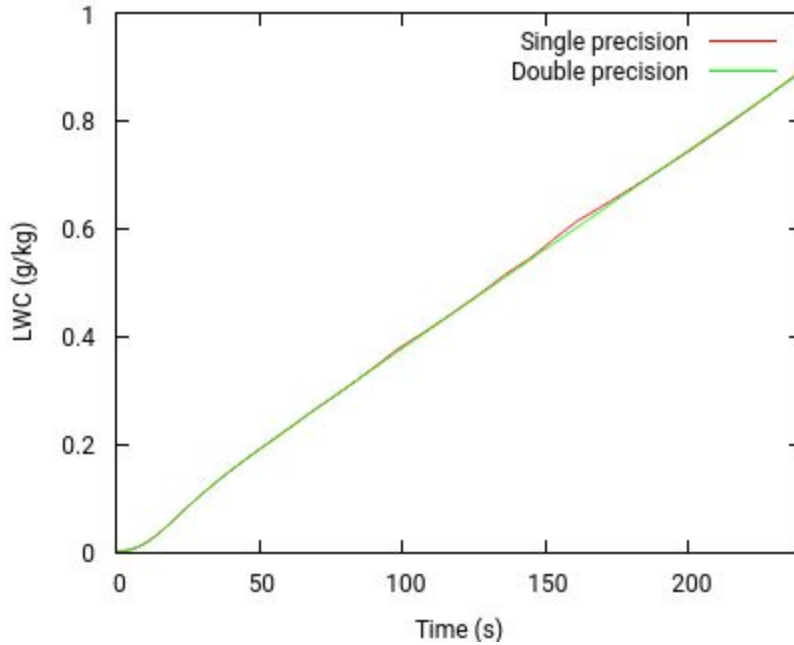


Fig. A: the time evolution of LWC when using the droplet growth equation  $\frac{dR^2}{dt} = 2K f_v S$  (no solute effect or curvature effect). The green line is based on double-precision calculation, and the red line is based on single-precision calculation.

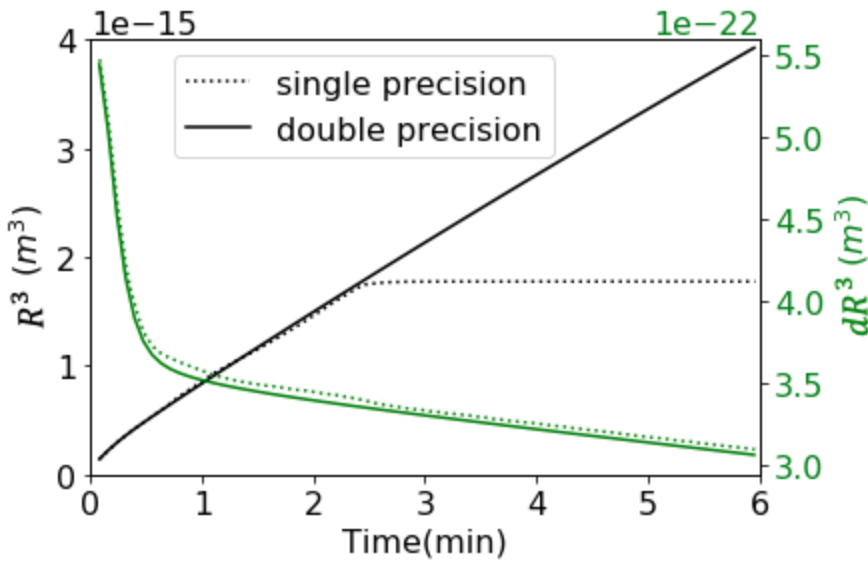


Fig. B: the time evolution of the mean  $R^3$  and  $dR^3$  in Run A.  $dR^3$  is the difference of  $R^3$  from the previous time step. Dotted lines are from the single-precision run, and solid lines are from the double-precision (new version) run.

### *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 ( $\epsilon$ ) for all the turbulent cases is set statistically stationary ( $\epsilon = 500 \text{ cm}^2 \text{ s}^{-3}$ ). The advantage of this idealized, simplified treatment is that the effect of turbulence can be easily separated from aerosol effects. A dissipation rate of  $500 \text{ cm}^2 \text{ s}^{-3}$  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 ( $N = 10, 20 \text{ cm}^{-3}$ , corresponding to 10 – 20% increase in the total number concentration). Conditions with more ultra-giant aerosols ( $R \gg 1 \mu m$ ), lower aerosol concentrations ( $N \ll 100 \text{ cm}^{-3}$ ), or highly polluted environment ( $N \gg 100 \text{ cm}^{-3}$ ) will be of interest to further assess the relative importance of aerosols and turbulence.”

### **Minor Comments**

LI. 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.

LI. 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.

LI. 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.”

LI. 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 ( $\approx 300\text{ m}$  below cloud base) to the level where the supersaturation reaches a maximum ( $\approx 43\text{ m}$  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 ( $H - H_{CB}$ )”

LI. 92 – 93: The fact that droplets smaller than  $10\text{ }\mu\text{m}$  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  $\kappa$  in the Eq. (1) and (2). Am I right? I suggest stating this explicitly.

Yes, we added  $\kappa$  on line 101, just before introducing equation (1).



LI. 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.

LI. 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).

LI. 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  $f_v$  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.

LI. 165 – 166: This is misleading since the particles are still lifted with a mean updraft of  $2 \text{ m s}^{-1}$ .

We have modified the description to “When turbulence is switched off, the background velocity fluctuation is set to  $0 \text{ m s}^{-1}$ . 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  $500 \text{ cm}^2 \text{ s}^{-3}$  at cloud base is too high. Typically, the dissipation rate increases with distance to the cloud base, and a value of  $500 \text{ cm}^2 \text{ s}^{-3}$  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  $600 \text{ s} = 10 \text{ min}$ , assuming an updraft of  $2 \text{ m s}^{-1}$ . 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  $\ll 0.1$ . 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. ”

LI.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  $\mu m$ , 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  $< 1 \mu m$ ) 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  $< 1 \mu m$ .

LI. 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.

LI. 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.

LI. 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”.

LI. 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).

LI. 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.

LI. 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  $10 \text{ cm}^{-3}$  and it will probably never rain if it is increased to  $1000 \text{ cm}^{-3}$ , 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 ( $R = 0.1, 1 \mu\text{m}$ ) and number concentration ( $N = 10, 20 \text{ cm}^{-3}$ , corresponding to 10 – 20% increase in the total number concentration). Conditions with more ultra-giant aerosols ( $R \gg 1 \mu\text{m}$ ), lower aerosol concentrations ( $N \ll 100 \text{ cm}^{-3}$ ), or highly polluted environment ( $N \gg 100 \text{ cm}^{-3}$ ) 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 ( $\text{m s}^{-1}$  instead of  $\text{m/s}$ ), 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  $\text{g/kg}$ , and not  $\text{g/Kg}$ .

We changed it to  $\text{g/kg}$ .

## 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 - p5/1117), 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  $\tau_{pmin}$  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:

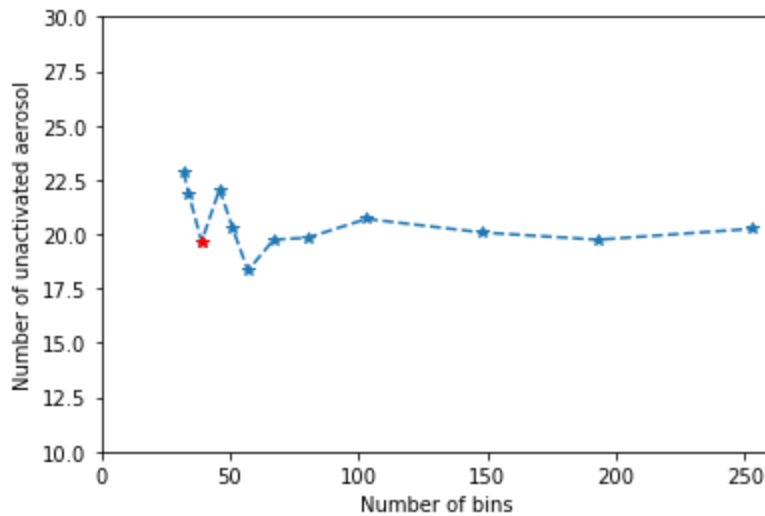
The 39 classes applied in the parcel model follow the same bin resolution as Xue et al. (2010). The bins are discretized on a log scale with the bin width increased by doubling the mass., i.e.,  $r(n)^3 = r(1)^3 * 2^{n-1}$ . In this way, the resolution is higher at small



particle sizes and lower at large particle sizes. This configuration 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 ( $N = 19.66\text{ cm}^{-3}$  in 39-bin vs  $N = 20.25\text{ cm}^{-3}$  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., p8/1198,1204).

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  $133 + 66.6 + 3.06 = 202.66 \text{ cm}^{-3}$  (in standard T,p conditions?) vs. total initial concentration of  $112 \text{ cm}^{-3}$  (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.  $N=202.66 \text{ cm}^{-3}$  is the total number concentration over the entire size range ( $r = 0 - \infty \mu\text{m}$ ). The aerosol size range we chose was from  $r = 10^{-3} - 49 \mu\text{m}$ , resulting in  $112 \text{ cm}^{-3}$  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 have added the GitHub links to the parcel model and DNS model used in this study in the *Code and data availability* section.

Other remarks:

- p1/117: “interaction” → interactions

Checked

- p1/134: 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 “processors”

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  $N = 87 \text{ cm}^{-3}$ . 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 ( $R = 2 - 16 \mu\text{m}$ ) with an interval of  $1 \mu\text{m}$ . 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  $S = e/e_s$  (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  $\kappa = 0$ ?

Corrected

- p7/l171:  $k \rightarrow \kappa$

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  $Q_v$ , 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”  $\rightarrow$  “When”

Corrected

- p8/l192:  $o() \rightarrow \mathcal{O}$

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](http://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