Review of "Impact of hygroscopic CCN and turbulence on cloud droplet growth: A parcel-DNS approach" by Chen et al. (acp-2019-886)

The submitted study investigates the effects of turbulence and aerosols (number and hygroscopicity) on cloud droplet growth by condensation and collection. By applying a unique combination of direct numerical simulation (DNS) with a parcel model, the authors show that turbulence increases droplet collection, while aerosol hygroscopicity increases spectral broadening, in agreement with several previous studies. Furthermore, the authors show that additional aerosols lead to a reduced mean droplet radius as expected. However, the simultaneously reduced liquid water content — in the presented magnitude and within the applied framework — remains a riddle to the reviewer.

The general topic of this study is of interest and the applied modeling framework is expected to lead to new insights. However, (potentially false) conclusions are based on an inadequate analysis of the modeling results. Additionally, the manuscript lacks appropriate consideration of previously published literature and requires some clarification in writing. While I believe that these weaknesses can be remedied, there are too many and too severe concerns at this stage. Therefore, I must suggest rejecting the manuscript from publication in Atmospheric Chemistry and Physics in the current form. I will support my suggestion in more detail below.

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.

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: $LWC \ = \ w \ t \ \Gamma_W$, with $\Gamma_W \approx 2.0 \times 10^{-6}\ \mathrm{kg\ m^{-4}}$. 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.

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). 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). 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). 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.

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.

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.

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

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.

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).

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.

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

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.

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.

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.

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.

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

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

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".

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

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.

L. 174: A kinetic energy dissipation rate of 500 $cm^2 s^{-3}$ at cloud base is too high. Typically, the dissipation rate increases with distance to cloud base, and a value of 500 $cm^2 s^{-3}$ might only be representative for the top of a shallow cumulus cloud.

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

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.

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

L. 187, Fig. 7b: 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.

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.

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

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

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

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

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.

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 a another property of the distribution that I miss?

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.

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?

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

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.

Technical Comments

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

L. 3: Define DNS in the abstract.

L. 39: Remove “Lagrangian-tracking”.

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

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

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

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

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.

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.

References

Abade, G. C., Grabowski, W. W., and Pawlowska, H. (2018). Broadening of cloud droplet spectra through eddy hopping: Turbulent entraining parcel simulations. *Journal of the Atmospheric Sciences*, *75*(10), 3365-3379.

Çelik, F., and Marwitz, J. D. (1999). Droplet spectra broadening by ripening process. Part I: Roles of curvature and salinity of cloud droplets. *Journal of the atmospheric sciences*, *56*(17), 3091-3105.

Korolev, A. V. (1995). The influence of supersaturation fluctuations on droplet size spectra formation. *Journal of the atmospheric sciences*, *52*(20), 3620-3634.

Lasher-trapp, S. G., Cooper, W. A., and Blyth, A. M. (2005). Broadening of droplet size distributions from entrainment and mixing in a cumulus cloud. *Quarterly Journal of the Royal Meteorological Society: A journal of the atmospheric sciences, applied meteorology and physical oceanography*, *131*(605), 195-220.

Seifert, A., Nuijens, L., and Stevens, B. (2010). Turbulence effects on warm-rain autoconversion in precipitating shallow convection. *Quarterly Journal of the Royal Meteorological Society*, *136*(652), 1753-1762.

Srivastava, R. C. (1991). Growth of cloud drops by condensation: Effect of surface tension on the dispersion of drop sizes. *Journal of the Atmospheric Sciences*, *48*(13), 1596-1599.

Thomas, S., Ovchinnikov, M., Yang, F., van der Voort, D., Cantrell, W., Krueger, S. K., and Shaw, R. A. (2019). Scaling of an atmospheric model to simulate turbulence and cloud microphysics in the Pi Chamber. *Journal of Advances in Modeling Earth Systems*, *11*(7), 1981-1994.