Responses to Reviewer 2

Review of "Cloud droplet size distribution broadening during diffusional growth: ripening amplified by deactivation and reactivation" by Yang et al. submitted for Atmospheric Chemistry and Physics

Using idealized adiabatic parcel simulations with Lagrangian bin-microphysics, the authors investigate the broadening of a cloud droplet size distribution (CDSD). By including the effects of aerosol deactivation and reactivation, it is shown that process of Ostwald ripening, which has been assumed to be weak in warm clouds by other authors, can be significantly amplified, resulting in sufficiently large droplets that might be able to initiate collision and coalescence. The authors demonstrate convincingly that the deactivation of aerosols in a downdraft leads to a lower number of cloud droplets in a subsequent updraft, which enhances the growth of these droplets, resulting in superadiabatic droplets sizes.

Additionally, the reactivation of some aerosols leads to an additional broadening of the CDSD to smaller sizes. Although I feel that the presented results represent a rather extreme case of the amplification of Ostwald ripening due to aerosol deactivation/reactivation, which might not be the case in nature, it clearly demonstrates the effect and potential importance of a proper representation of deactivation/reactivation, which many cloud models lack. Accordingly, some minor additional simulations might be necessary to determine the limits of the presented microphysical processes and to fit it in the current literature. All in all, the manuscript is interesting, well written, and should be published after the following concerns are addressed.

We thank the reviewer for the constructive comments and for the improvements they have motivated in the revised paper. In this document we address all comments and detail the changes in response. Reviewer comments are in blue, our response is in black and modifications of the manuscript are summarized in red text.

General Comments

Model Description. Although plenty of references are given, the essential parts of the used microphysical model need to be stated. Only the abstract and the conclusions (Sec. 4) state, that the bin-microphysics is Lagrangian, i.e., it utilizes moving bins instead of fixed bins. This information is missing in Sec. 2 but essential for the model used in this study, which relies on a fixed relation of aerosol mass and droplet size (which is only possible in a Lagrangian (or moving bin) framework). Does the microphysical model include any other processes than diffusional droplet growth including activation/deactivation? Moreover, it would be nice (but not necessary) to present the used equation describing diffusional droplet growth including activation/deactivation. This would be also an opportunity to define quantities as Se and Ssat, which are used in other parts of the manuscript (e.g., Fig. 8).

We thank the reviewer for the helpful comments. Section 2 is rewritten with more details and mathematical equations for the employed model. Please see the revised manuscript for more details.

Idealized Setup. It is not disputable that the presented simulations represent an idealized setup. However, the probability that a parcel undergoes numerous oscillations of 150 m or more is rather unlikely. The results of Wood et al. (2002), who investigated CDSD ripening in a slightly more realistic setup including potential effects of aerosol deactivation and reactivation (last lines of their section 3), do not indicate a strong evidence of the proposed amplification of CDSD ripening by deactivation/reactivation. Therefore, I strongly suggest testing even thinner recirculation layers, i.e., fluctuations which are more likely to be observed in nature. I expect that if a certain depth of the recirculation layer is undercut, deactivation will be inhibited and the amplification of ripening due to deactivation/reactivation will stop. These additional investigations are not only necessary to understand the importance of the proposed amplification mechanism, but also connects the presented study to other work on spectral ripening (e.g., Wood et al. (2002), or Grabowski and Abade (2017) who extensively investigated the dependence of spectral broadening on the length scales of the involved turbulence in the absence of deactivation/reactivation).

We thank the reviewer for the helpful comments. To investigate whether deactivation or reactivation will be inhibited for thin recirculation layers, three more cases with ΔH=50m, 10m, and 1m are carried out. Results show that reactivation is inhibited, but deactivation always occurs. The evolution of CDSD is similar for those cases, which is similar to the evolution of CDSD due to Ostward ripening in still environment. We add a figure and more discussion in the manuscript.

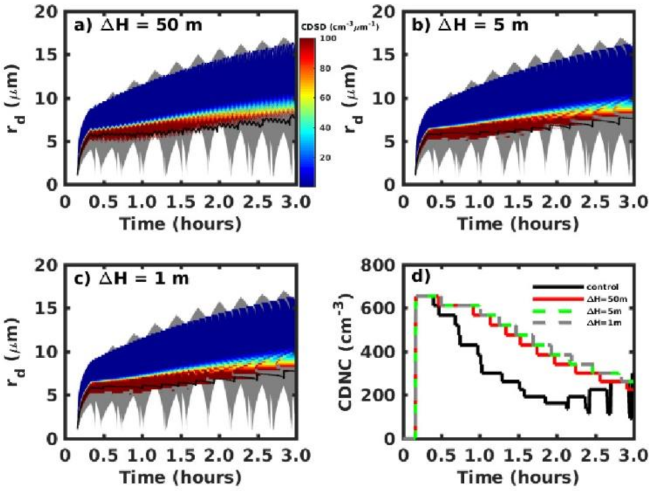

Figure 7: Cloud droplet size distribution (CDSD) changes with time for different thicknesses of recirculation layers: a) ΔH=50 m, b) ΔH=5 m, c) ΔH=1 m. d) Total cloud droplet number concentration changes with time for the different cases. The gray region in a-c represents the

range of the droplet size spectrum for the control case, and the black lines represent the mean cloud droplet radius change with time.

“We note that deactivation is suppressed for a thin recirculation layer ΔH=150 m as shown in Figure 6b, and therefore the CDSD broadening is not as efficient as the control case. However, the vertical oscillations of an air parcel due to turbulence might be much smaller than 150 m. Wood et al. (2002) did not observe the enhanced CDSD broadening by deactivation and reactivation with a shallower recirculation layer. One interesting question is whether deactivation or reactivation would be inhibited for a very thin recirculation layer. To answer this question, three more cases are carried out with recirculation layers of 50 m, 5 m and 1 m. All these cases have the same setup as the control case except for the thickness of recirculation layer. The CDSD and total cloud droplet number concentration for each case are shown in Figure 7. It can be seen that reactivation is inhibited for all cases, but deactivation always occurs. More interestingly, the CDSD for all these three cases are similar, and the decrease of total cloud droplet number concentration due to deactivation is also similar. The evolution of the CDSD for a thin recirculation layer is independent of air motion and degrades to a steady state where the CDSD broadening is due to Ostward ripening in a still environment.”

Minor Comments

P. 2, l. 2: Does the “linear growth rate” refer to the temporal change of the radius (dr/dt = …)?

Yes, “linear growth rate” means dr/dt. We delete “linear” in our manuscript as suggested by reviewer 1.

P. 3, ll. 30 – 31: Give a short explanation why the liquid water is slightly smaller in the ascending branch compared to the descending.

This is due to the kinetic effect or hysteretic effect that the liquid water content responds slower than the change in environment. In other words, liquid water content is smaller than it should be during condensation (ascent) and is larger than it should be during evaporation (descent). Therefore, the liquid water is slightly smaller in the ascending branch compared to the descending. This is consistent with Korolev et al. (2013). We modify the text as,

“Liquid water mixing ratio in the ascending branch is slightly smaller than that in the descending branch at the same height due to the kinetic effect (or hysteresis effect), which is consistent with Korolev et al. (2013).”

P. 4, ll. 7 – 8: Although it has been stated before, I would mention the development of a second mode in the CDSD by the reactivation of aerosols after about 2 hours.

We modify the text as:

"…Also notice that a second mode appears in the CDSD due to reactivation of aerosols after about 2 hours (see Figure 2d).…"

Fig. 2d: How do the aerosol masses (or dry radii) distribute across the CDSD? I expect that the largest droplets have been grown from the largest aerosols.

Yes, larger cloud droplets include more aerosol mass.

"…Droplet size in the moving bin monotonically increases with the dry aerosol mass associated with that moving bin.…"

P. 5, ll. 34 – 35: Give some more explanations on the setup of the ascending-only parcel simulation. What is its vertical velocity? The answer can be deduced from the following text but a clear statement would be helpful.

We add more details of the ascending-only parcel in the manuscript.

"…For the latter case, the cloud parcel ascends at a vertical velocity of 0.5 ms^{-1} for three hours with the same initial condition as the control case.…"

P. 6, l. 17 and p. 7, l. 12: "Recycling layer"? Based on the available literature, I would prefer the name "recirculation layer".

We replace "recycling layer" and "cycling layer" to "recirculation layer" throughout the manuscript.

P. 6., ll. 27 – 30: Although I agree with the interpretation that deactivation/reactivation might amplify the ripening process, an additional explanation, originating directly from Korolev (1995, Section 2), needs to be considered: Broadening only occurs if the supersaturation is smaller than maximum of S + (r), a quantity which indicates the narrowing or broadening of the spectrum in the vicinity of a certain radius r. If the supersaturations are generally higher than S + , only narrowing of the CDSD occurs. Since in-cloud supersaturations generally decrease due to an increase in aerosol number concentration, it is to expect that only the more aerosol-laden simulations will be affected by Ostwald ripening while the cleaner simulation might be less affected (or not affected at all), which also agrees with the presented study.

We agree with the reviewer's comment. We add another explanation in the manuscript:

"Another explanation from Korolev (1995) is that the CDSD broadening occurs when air supersaturation (S_e) is smaller than the critical supersaturation for the smallest cloud droplets ($S_{sat}(r_{small})$). For this condition, the smallest cloud droplets evaporate and the largest cloud droplets might grow slightly if $S_e>S_{sat}(r_{large})$ or evaporate slightly if $S_e<S_{sat}(r_{large})$, thus leading to

broadening. If the water vapor mixing ratio in air on average is much larger than the saturated water vapor mixing ratio over droplet, only narrowing of the CDSD occurs. Because in-cloud supersaturation decreases with increased aerosol concentration, it is expected that the Ostwald ripening is more efficient in polluted cloud, which is also consistent with (Srivastava, 1991)."

Fig. 5b: Where do the high-frequent oscillations in the CDSD come from?

Here we keep the thickness of the recirculation layer constant. Therefore, larger vertical velocity suggests higher oscillation frequency. We add more description in the text:

"…Here we keep the thickness of the recirculation layer constant. Therefore, larger vertical velocity results in a higher oscillation frequency.…"

P. 8, l. 9: For clarity, state the underlying equation used for calculating S_{sat}.

We add the equation for S_{sat} in section 2.

P. 8, l. 24 – 34: This is an interesting result. Although I can imagine where the equation in l. 27 comes from, an extra step for its deviations might be illuminating for all readers. Moreover, I suggest discussing the underlying physics of the term sk in slightly more depth. A nice explanation is given for the case of a negative vertical velocity, in which the evaporation of a large number of small droplets maintains the supersaturation at a certain level. But how does sk act in an updraft?

Thank the reviewer for the helpful comments. We add extra step for the deviations in the text.

"This can be obtained from the analytical expression of supersaturation in an adiabatic cloud parcel: $dS_e/dt=Aw-Bnr(S_e-1)$, where A and B are parameters depending on thermodynamic properties (Korolev and Mazin, 2003). …because $dS_e/dt=Aw-Bnr(S_e-S_{sat})$ and thus…"

In the updraft region, all droplets grow and both solute and curvature effects are negligible. We add more discussion in the manuscript:

"In the updraft region, all droplets grow and the effect of s_k is negligible. In the downdraft region and for polydisperse cloud droplets, the environment conditions are buffered by the large number of small cloud droplets."

P. 9, ll. 24 – 25: What is meant by the right upper boundary of the CDSD?

We change "the right upper boundary of the CDSD" to "the large-size upper boundary of the CDSD"

P. 10, ll. 15 – 21: There are models with a similar treatment of microphysics, so-called Lagrangian cloud model. And a couple of publications investigation aerosol activation/deactivation in that framework (e.g., Andrejczuk et al. 2008; Hoffmann et al., 2015; Hoffmann 2017).

We add more discussion about aerosol activation and deactivation in manuscript.

"…The mechanism of CDSD broadening in this study requires the model to consider both solute and curvature effects all the time (i.e., before and after activation, deactivation and reactivation). Our results suggest the importance of solute and curvature effects to the deactivation and reactivation processes, which is consistent with previous studies (e.g., Andrejczuk et al., 2008; Hoffmann et al., 2015; Hoffmann, 2017; Chen et al., 2018).…"

Technical Comments

P. 1, l. 6: Usually, an abstract does not contain any citations.

We removed the citation in the abstract.

P. 1, l. 20: "of a warm cloud" or "of warm clouds"

We modify the text to be "of warm clouds"

P. 2, l. 23: "… GCCN not only **provide** an embryo … but also **enhance** droplet growth …"

We modify the sentence as:

"They found that GCCN ~~not only provides~~ provide an embryo for big droplets at the activation stage ~~but also enhances~~ and, more importantly, GCCN enhance droplet growth after activation due to the solute effect."

P. 3, l. 17: Since American English is used throughout the manuscript: "sulfate"

We change "sulphate" to "sulfate" throughout the text.