

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 (CDS). 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 CDS 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.

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 S_e and S_{sat} , which are used in other parts of the manuscript (e.g., Fig. 8).

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 CDS 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 CDS 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).

Minor Comments

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

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

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

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

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.

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

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 CDS 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.

Fig. 5b: Where do the high-frequency oscillations in the CDS come from?

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

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 s_k 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 s_k act in an updraft?

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

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

Technical Comments

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

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

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

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

References

Andrejczuk, M., Reisner, J.M., Henson, B., Dubey, M.K. and Jeffery, C.A., 2008. The potential impacts of pollution on a nondrizzling stratus deck: Does aerosol number matter more than type?. *Journal of Geophysical Research: Atmospheres*, 113(D19).

Grabowski, W.W. and Abade, G.C., 2017. Broadening of Cloud Droplet Spectra through Eddy Hopping: Turbulent Adiabatic Parcel Simulations. *Journal of the Atmospheric Sciences*, 74(5), pp.1485-1493.

- Hoffmann, F., Raasch, S. and Noh, Y., 2015. Entrainment of aerosols and their activation in a shallow cumulus cloud studied with a coupled LCM–LES approach. *Atmospheric Research*, 156, pp.43-57.
- Hoffmann, F., 2017. On the limits of Köhler activation theory: how do collision and coalescence affect the activation of aerosols? *Atmospheric Chemistry and Physics*, 17(13), pp.8343-8356.
- Korolev, A.V., 1995. The influence of supersaturation fluctuations on droplet size spectra formation. *Journal of the Atmospheric Sciences*, 52(20), pp.3620-3634.
- Wood, R., Irons, S. and Jonas, P.R., 2002. How important is the spectral ripening effect in stratiform boundary layer clouds? Studies using simple trajectory analysis. *Journal of the Atmospheric Sciences*, 59(18), pp.2681-2693.