Comments to manuscript acpd-2017-1125:

Cloud droplet size distribution broadening during diffusional growth: ripening amplified by deactivation and reactivation

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The reviewed manuscript discusses the phenomenon of cloud droplet size spectrum broadening using an adiabatic parcel model. The authors highlight the role of the interplay between condensation/evaporation on small and large particles leading to an irreversible process analogous to Ostwald ripening. A methodology for discerning the contributions of deactivation and reactivation is developed and used to depict the amplifying role of deactivation and activation for the ripening-induced broadening. The topic is of prime relevance in the context of the ongoing developments of models comprehensively accounting for two-way aerosol-cloud interactions.

In general, the paper is concise and interesting, and I do recommend its publication pending revisions addressing concerns detailed below, and mainly related to:

- noncomprehensive presentation of earlier works on the topic,
- insufficient discussion of the limitations of the presented approach,
- limited reproducibility of the study.

Comments on the content

Abstract

The study builds upon the considerations presented by Korolev in 1995, what is dully acknowledged. However, the work of Çelik and Marwitz (1999), which is elsewhere (e.g., Wood et al. 2002) credited as the first to depict the Ostwald ripening in the context of cloud droplet growth, is not mentioned. Let me suggest not to include any references in the abstract, but rather revisit the introductory section to provide comprehensive references to earlier works on the topic including Srivastava (1991) and Çelik and Marwitz (1999).

The manuscript mentions turbulence only within the abstract and in the conclusions section (plus the somehow less relevant reference to turbulence-induced enhancement in collision efficiency on page 2). Some discussion is needed in the text to warrant statements that the study addresses turbulence-relevant vertical oscillations. In particular, the frequencies of oscillations studied are distant from those considered in recent studies on turbulence-induced effects in air-parcel activation models, e.g., (Ditas et al., 2012, Fig. 10 therein) or Hammer et al. (2015, Fig. 10 therein).

Let me also suggest using "moving-bin" instead of "Lagrangian bin-microphysics" in the abstract and throughout the text.

Section 1

A complete rewrite of the second paragraph (p. 2, lines 10–30) would be a good idea. The first sentence could likely be moved to the beginning of the third paragraph, perhaps made more precise by mentioning aerosol spectrum (or even moving-bin representation), and supported with some classic reference, e.g., the already referenced work of Mordy, but perhaps also the seminal work of Howell (1949). The second and third sentences could be merged in into the first paragraph where both narrow spectrum and cloud parcel are already mentioned. Then, I would suggest splitting the rest of the paragraph into two separate ones on: (i) the possible causes, and (ii) the possible effects of the broadening of cloud droplet spectrum.

Among the causes, the influence of aerosols highlighted in the already cited work of Chandrakar et al., the influence of in-cloud activation (e.g., Khain et al., 2000, sect. 3.5) as well as of turbulence (Devenish et al., 2012, e.g.,) could be mentioned additionally. The recent work of Grabowski and Abade, 2017 seems relevant to me as well.

Among the effects, along with the already mentioned enhancement of collision efficiency, the optical aspects should be listed given that they are highlighted even in the first sentence of the abstract. In fact, the last sentence of section 3.1 (p. 6, lines 12-13) seems to me to be more appropriate here.

It would be also beneficial to clarify the meaning of supersaturation fluctuations as the same term is used for studies assuming uniform supersaturation within an air parcel (as in Korolev, 1995) as well as studies resolving inhomogeneities of supersaturation in space (Devenish et al., 2012, and references therein).

The phrases "irreversibility of droplet size spectrum shape" (p. 2, line 26), "CDSD is irreversible" and "Irreversibility of the CDSD" (p. 4, line 13), while consistent with the original wording of Korolev (1995) sound somehow confusing to me as it is the process (i.e., the evolution in time) that is irreversible and not the spectrum shape – just a nomenclature issue. On a related note, the discussion on hysteretic effects in activation-deactivation cycles presented in Arabas and Shima (2017) might be of relevance (although limited to monodisperse spectra).

Section 2

ACP guidelines clearly state that "*paper should contain sufficient detail and references to public sources of information to permit the author's peers to replicate the work*"[1]. It is thus essential to either comprehensively define the mathematical formulation of the employed model or provide a straightforward way of obtaining the employed software in the very revision used for obtaining presented results.

To highlight the problem, let me point out that the Feingold, Walko, et al. (1998) paper referenced as describing "*the original version of the model*" actually covers simulations with a 2D LES-type model "*that uses lognormal basis functions to represent cloud and drizzle drop spectra*". The Feingold, Kreidenweis, and Zhang (1998) reference was likely meant, although therein the reader is refereed to Feingold and Heymsfield (1992) for "*further details of the microphysical model*". There, in turn, the reader will learn that "*the model used ... is discussed in detail by Heymsfield and Sabin (1989)*". While a parcel model might be considered a very simple tool, the numerical nuances (e.g., spectral discretisation, implicit vs. explicit supersaturation calculation, choice of values for parameters such as mass accommodation coefficient) do cause significant differences among results from different implementations as depicted for instance in the intercomparison study of Kreidenweis et al. (2003) which actually included the model used in the refereed manuscript. While properly attributing the authors of model formulation and implementation, and giving the readers the ability to reproduce the results is crucial, elaborating on the model details shall make the manuscript easier to comprehend as well.

Section 3.1

The references to sizes of single cloud droplets (p. 4, lines 2, 3, 24, 26, 30, 31; p. 8, l. 3) contrast the more appropriate description of "droplet size for a bin" (p. 3, lines 20, 21, 25; p. 4 line 4). There is also a statement on "reactivation of that bin" (caption of Fig. 2). I suggest unifying the way the size associated with a moving bin is referred to.

The notion of "totally evaporated" droplet (p. 4, line 30) seems misleading to me. The model describes a population of solution droplets, likely under the assumption of the salt mass being negligible in comparison with water mass. Conditions imposed to disable reactivation should be clarified.

I suggest rephrasing the passage on supercooled parcel at 6000 m to underline the technical (not physical) nature of this element of the analysis.

[1] http://www.atmospheric-chemistry-and-physics.net/for_authors/obligations_for_authors.html

Section 3.2

This section lacks any references to other studies which would be very appropriate here and which should help to give support to the choice of parameters used.

As will be pointed out below, the analysis of sensitivity to spectral discretisation would also be very beneficial.

Section 3.3

As a general comment, let me point out that neither the sensitivity analysis nor the discussion of the results touches upon the numerical limitations of the employed parcel model. As pointed out in Kreidenweis et al. (2003, e.g., discussion of Fig. 8 therein) both the spectral discretisation and the uncertainty in the value of mass accommodation coefficient translate into significant uncertainty in the results (obtained with the very same parcel model as used in this study). In Takeda and Kuba (1982, sect. 2.5 therein) it was pointed out that the narrowness of size distributions reported by Mordy (1959) was actually likely influenced by the spectrum discretisation. As a more technical remark, the analysis presented in Arabas and Pawlowska (2011, Fig. 4 therein) shall discourage the authors from using three-significant-digit precision in Table 1 and throughout the paper.

The last sentence of the first paragraph (p. 7, lines 24-26) shall likely be extended into a separate paragraph to allow for referencing the discussion that followed from the work of Liu and Daum – see e.g. Lu and Seinfeld (2006, sect. 6 therein) and Brenguier, Burnet, and Geoffroy (2011, sect. 2 therein). Also, the issue of instrumental broadening shall be mentioned (sect. 3.2 in Devenish et al., 2012, and references therein).

The discussion of residence time in the third paragraph (p. 8) could benefit from referencing other studies discussing in-cloud residence time in context of aerosol recycling (see e.g. section 4.2 in Andrejczuk, Reisner, et al., 2008, and references therein).

Section 4

The discussion on the limitations of the presented analysis given in second and third paragraph of the section (p. 9 lines 31-33, p.10 lines 1-13) is somehow imbalanced, in my opinion. On the one hand, the lack of entertainment and mixing is commented just with a short statement. On the other hand, a separate paragraph is presented in support of the assumption of polydisperse aerosol and the presence of both upward and downward motions (if to be kept, this paragraph calls for references and more quantitative discussion, e.g. by discussing the relevant dynamical and microphysical timescales as in Korolev 1995, sect. 6). I suggest placing much more attention on the adiabaticity assumption, especially given the three-hour-long simulation time. The discussion of the importance of mixing based on LES and TEM simulations presented in Ovchinnikov and Easter (2010) shall come in handy, especially that the TEM used therein is based on the same parcel-model formulation from Feingold, Kreidenweis, and Zhang (1998).

The discussion presented in the last paragraph (p. 10, lines 15-21 also referenced in the last sentence of the abstract) calls for a mention of particle-based microphysics techniques, some of which do fulfil the mentioned requirement of considering "*both solute and curvature effects before and after activation*", and in particular – also deactivation and reactivation. Several references to works published throughout the last decade are given, e.g., in Hoffmann, Raasch, and Noh (2015), where discussion on the role of reactivation can also be found (sect. 3.1 therein).

While it might likely be considered out of scope of the present paper, let me point out that the presented discussion is a very counterargument to the simplification of the particle-based condensation schemes recently suggested in Grabowski, Dziekan, and Pawlowska (2017), and based on the assumption that detailed modelling of reactivation is only relevant if aerosol processing by collisions or chemical reactions is addressed. The earlier discussions of the consequences of neglecting pre-activation droplet growth in models of clouds (e.g., Srivastava, 1991; Chuang, Charlson, and Seinfeld, 1997) seem relevant as well.

Finally, the authors shall consider citing Ovchinnikov and Easter (2010) along the work of Lebo and Seinfeld (p. 10, line 18), while the reference to the work of Bott, focused on the coalescence numerics, seems less relevant. References to earlier works employing joint "2d-bin" aerosol-cloud spectra can be found e.g. in paragraph 3 of Andrejczuk, Grabowski, et al. (2010) and in paragraph 10 of Ovchinnikov and Easter (2010).

Comments on the composition and technical remarks

p. 1, l. 23 please avoid the word "believed"
p. 1, l. 24 Imagining rather than imaging?
p. 2, l. 2 please explain or remove the word "linear"
p. 2, l. 3 please rephrase the sentence so that collisional growth efficiency is not logically coupled with inverse proportionality of condensational growth rate
p. 2, l. 3-4 I suggest using approximate sizes and perhaps referencing a more recent textbook instead of the work of Hocking
p. 2, l. 15 please indicate causation instead of just saying "be related to"
p. 2, l. 23 GCCN provide (and not provides)?
p. 3, l. 3 sensitivity studies (not sensitivities)
p. 5, l. 24 "kinetic" (i.e., relate to the pace of the process as in chemical kinetics) rather than "kinematic" (i.e., related to motion)?
p. 6, l. 20 please rephrase "number concentrations of the control"
p. 7, l. 15 "larger than **in** the control case"

Within references, please correct capitalisation in journal names and use abbreviated versions following the ACP guidelines[2]. I strongly suggest adding a doi label for each reference (this will not be added by Copernicus editors). Here are corrections to several entries in the bibliography:

- Bott reference volume should be 59–60.
- Cheng et al. reference is missing page identifier: D08201.
- Falkovich and Pumir reference has wrong year (2015, should be 2007), wrong volume (should be 64) and is missing page numbers: 4497–4505.
- Feingold and Siebert reference is missing book title, editor and publisher information.
- Heintzenberg et al. reference has a truncated title and missing booktitle information, it should likely be replaced with Pöschl, Rose, and Andreae (2009).
- Laird et al. reference requires correction in capitalisation of "Iii".
- Li et al. reference is missing page range: 11213–11227.
- Lozar and Muessle reference should be cited as "de Lozar and Muessle" (at least according to ACP website).
- Pruppacher and Klett book reference mistakenly includes an additional author and is missing publisher name.
- Xue and Feingold reference is missing page identifier: D18204.

Figures

It is essential to replace the raster low-resolution image files used in figures 1–6 with vector graphics (PostScript/SVG/PDF formats).

Hope that helps!
Sylwester Arabas

[2] http://www.atmospheric-chemistry-and-physics.net/Copernicus_Publications_Reference_Types.pdf

References (entries present in the paper omitted)

Andrejczuk, M., W.W. Grabowski, et al. (2010). Cloud-aerosol interactions for boundary layer stratocumulus in the Lagrangian Cloud Model. *J. Geophys. Res.* 115, D22214. DOI: 10.1029/2010JD014248.

Andrejczuk, M., J.M. Reisner, et al. (2008). The potential impacts of pollution on a nondrizzling stratus deck: Does aerosol number matter more than type? *J. Geophys. Res.* 113, D19204. DOI: 10.1029/2007JD009445.

Arabas, S. and H. Pawlowska (2011). Adaptive method of lines for multi-component aerosol condensational growth and CCN activation. *Geosci. Model Dev.* 4, 15–31. DOI: 10.5194/gmd-4-15-2011.

Arabas, S. and S. Shima (2017). On the CCN (de)activation nonlinearities. *Nonlin. Proc. Geophys.* 24, 535–542. DOI: 10.5194/npg-24-535-2017.

Brenguier, J.-L., F. Burnet, and O. Geoffroy (2011). Cloud optical thickness and liquid water path - does the k coefficient vary with droplet concentration? *Atmos. Chem. Phys.* 11, 9771–9786. DOI: 10.5194/acp-11-9771-2011.

Çelik, F. and J.D. Marwitz (1999). Droplet Spectra Broadening by Ripening Process. Part I: Roles of Curvature and Salinity of Cloud Droplets. *J. Atmos. Sci.* 56, 3091–3105. DOI: 10.1175/1520-0469(1999)056<3091:DSBBRP>2.0.CO;2.

Chuang, P.Y., R.J. Charlson, and J.H. Seinfeld (1997). Kinetic limitations on droplet formation in clouds. *Nature* 390, 594–596. DOI: 10.1038/37576.

Devenish, B. J. et al. (2012). Droplet growth in warm turbulent clouds. *Q.J.R. Meteorol. Soc.* 138, 1401–1429. DOI: 10.1002/qj.1897.

Ditas, F. et al. (2012). Aerosols-cloud microphysics-thermodynamics-turbulence: evaluating supersaturation in a marine stratocumulus cloud. *Atmos. Chem. Phys.* 12, 2459–2468. DOI: 10.5194/acp-12-2459-2012.

Feingold, G. and A.J. Heymsfield (1992). Parameterizations of Condensational Growth of Droplets for Use in General Circulation Models. *J. Atmos. Sci.* 49, 2325–2342. DOI: 10.1175/1520-0469(1992)049<2325:POCGOD>2.0.CO;2.

Feingold, G., S.M. Kreidenweis, and Y. Zhang (1998). Stratocumulus processing of gases and cloud condensation nuclei, part I, Trajectory ensemble model. *J. Geophys. Res.* 103, 19527–19542. DOI: 10.1029/98JD01750.

Feingold, G., R.L. Walko, et al. (1998). Simulations of marine stratocumulus using a new microphysical parameterization scheme. *Atmos. Res.* 47–48, 505–528. DOI: 10.1.1.453.2319.

Grabowski, W.W. and G.C. Abade (2017). Broadening of Cloud Droplet Spectra through Eddy Hopping: Turbulent Adiabatic Parcel Simulations. *J. Atmos. Sci.* DOI: 10.1175/JAS-D-17-0043.1.

Grabowski, W.W., P. Dziekan, and H. Pawlowska (2017). Lagrangian condensation microphysics with Twomey CCN activation. *Geosci. Model. Dev.* accepted. DOI: 10.5194/gmd-2017-214.

Hammer, E. et al. (2015). Sensitivity estimations for cloud droplet formation in the vicinity of the high-alpine research station Jungfraujoch (3580 m a.s.l.) *Atmos. Chem. Phys.* 15, 10309–10323. DOI: 10.5194/acp-15-10309-2015.

Heymsfield, A.J. and R.M. Sabin (1989). Cirrus Crystal Nucleation by Homogeneous Freezing of Solution Droplets. *J. Atmos. Sci.* 46, 2252–2264. DOI: 10.1175/1520-0469(1989)046<2252:CCNBHF>2.0.CO;2.

Hoffmann, F., S. Raasch, and Y. Noh (2015). Entrainment of aerosols and their activation in a shallow cumulus cloud studied with a coupled LCM–LES approach. *Atmos. Res.* 156, 43–57. DOI: 10.1016/j.atmosres.2014.12.008.

Howell, W. (1949). The growth of cloud drops in uniformly cooled air. *J. Meteorol.* 6.2, 134–149. DOI: 10.1175/1520-0469(1949)006<0134:TGOCDI>2.0.CO;2.

Khain, A. et al. (2000). Notes on the state-of-the-art numerical modeling of cloud microphysics. *Atmos. Res.* 55, 159–224. DOI: 10.1016/S0169-8095(00)00064-8.

Kreidenweis, S.M. et al. (2003). Modification of aerosol mass and size distribution due to aqueous phase SO_2 oxidation in clouds: comparisons of several models. *J. Geophys. Res.* 108.D7, 4213. DOI: 10.1029/2002JD002697.

Lu, M.-L. and J.H. Seinfeld (2006). Effect of aerosol number concentration on cloud droplet dispersion: A large-eddy simulation study and implications for aerosol indirect forcing. *J. Geophys. Res.* 111, D02207. DOI: 10.1029/2005JD006419.

Ovchinnikov, M. and R.C. Easter (2010). Modeling aerosol growth by aqueous chemistry in a nonprecipitating stratiform cloud. *J. Geophys. Res.* 115, D14210. DOI: 10.1029/2009JD012816.

Pöschl, U., D. Rose, and M.O. Andreae (2009). Climatologies of Cloud-related Aerosols. Part 2: Particle Hygroscopicity and Cloud Condensation Nucleus Activity. In: *Clouds in the perturbed climate system: their relationship to energy balance, atmospheric dynamics, and precipitation.* Ed. by J. Heintzenberg and R.J. Charlson. MIT Press. DOI: 10.7551/mitpress/9780262012874.001.0001.

Srivastava, R.S. (1991). Growth of cloud drops by condensation: Effect of surface tension on the dispersion of drop sizes. *J. Atmos. Sci.* 48, 1596–1605. DOI: 10.1175/1520-0469(1991)048<1596:GOCDBC>2.0.CO;2.

Takeda, T. and N. Kuba (1982). Numerical study of the effect of CCN on the size distribution of cloud droplets. Part I. Cloud droplets in the stage of condensational growth. *J. Meteorol. Soc. Jpn* 60, 978–993. DOI: 10.2151/jmsj1965.61.3_375.