

## Responses to reviewer 1

Comments to manuscript acpd-2017-1125: Cloud droplet size distribution broadening during diffusional growth: ripening amplified by deactivation and reactivation 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.

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. Reviewer comments are in blue, our response is in black and modifications of the manuscript are summarized in red text.

To sum up, we add several paragraphs in section 1 to give a “comprehensive presentation of earlier works on this topic”. To allow the “reproducibility of this study”, we add more details of our current model in section 2, including key mathematical equations. More sensitivity studies, including the effects of water accommodation coefficient and spectral discretization, and more discussion of the limitations of the presented approach are added in sections 3 and 4.

### Comments on the content

#### Abstract

The study builds upon the considerations presented by Korolev in 1995, what is dully acknowledged. However, the work of Celik 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 Celik 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).

Thank the reviewer for the helpful comment. We remove the reference in the abstract.

“Results show that the CDSO can be broadened during condensational growth as a result of Ostwald ripening amplified by droplet deactivation and reactivation, ~~which is consistent with earlier work, which is consistent with Korolev (1995).~~”

In the introduction, we add a paragraph to have an in-depth discussion of previous studies about the Ostwald ripening effect on cloud droplet size distribution broadening.

“Ostwald ripening for cloud droplets is the phenomenon when larger droplets grow and smaller droplets shrink due to curvature and/or solute effects and, thus, it can broaden the CDSO at both small and large sides of the distribution. Srivastava (1991) investigated the growth of cloud droplets in a rising air parcel. Results show that the variance of squared radius of the CDSO was constant during the condensational growth process if both curvature and solute effects were ignored, but it was increased if those effects were considered. This “condensational broadening” is more pronounced in clouds with high cloud droplet number concentration and low vertical velocity. In turbulent clouds, droplets will experience supersaturated/subsaturated conditions in updraft/downdraft regions. Korolev (1995) studied the evolution of the CDSO driven by supersaturation fluctuations in a vertically oscillating air parcel. Supersaturation fluctuations in his study mean that air is supersaturated in the updraft and subsaturated in the downdraft; however no spatial inhomogeneity of supersaturation is considered in the parcel. Results show that the growth and evaporation cycles during the CDSO evolution are irreversible if the solute and curvature effects are considered. This “CDSO irreversibility” (terminology used in his paper) will promote the growth of large cloud droplets, lead to evaporation or even deactivation of small cloud droplets, and thus broaden the CDSO. Korolev (1995) argued that stronger turbulent fluctuations of supersaturation would result in a broader CDSO. This is contrary to Celik and Marwitz (1999), who found that supersaturation fluctuations are not responsible for CDSO broadening and the formation of large droplets. The curvature and solute effects on Ostwald ripening, activation and deactivation have been the topics of study in recent years (e.g., Wood et al., 2002; Arabas and Shima, 2017, Chen et al., 2018; Sardina et al., 2018) but, to our knowledge, the relative roles of the curvature effect and solute effect on CDSO broadening have not been investigated.”

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

We changed “Lagrangian bin-microphysics” to “moving-bin” in the abstract and throughout the manuscript. “Lagrangian bin-microphysics” is included as a parenthetical when introduced in the methods.

## 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.

Thank you for the helpful comments. We split the paragraph into three: (1) the effects of the broadening of cloud droplet spectrum; (2) turbulence-induced CDSB broadening; (3) aerosol-induced CDSB broadening.

### **Paragraph about the effects of the CDSB broadening:**

“The broadening of the CDSB has a strong effect on precipitation and radiation. A broader CDSB implies larger differences in the terminal velocity of droplets. This is beneficial for collision coalescence and might cause the fast-rain process in the atmosphere (e.g., Göke et al., 2007). In addition, a broader CDSB increases the relative dispersion, which is the ratio of standard deviation to the mean CDSB. Previous studies show that an increase in relative dispersion is relevant to the albedo effect and increases albedo susceptibility (Feingold et al., 1997; Liu and Daum, 2002; Feingold and Siebert, 2009). An interesting question is why the CDSB is wider than predicted; in particular, why large droplet sizes are frequently observed in the clouds (e.g., Siebert and Shaw, 2017). Several mechanisms have been proposed that can be divided into two categories: turbulence-induced spectra broadening and aerosol-induced spectra broadening. A brief review is given next for each category.”

### **Paragraph about the turbulence-induced CDSB broadening:**

“Turbulence is ubiquitous in the clouds and can cause CDSB broadening in both condensation and collision processes (e.g., Shaw, 2003; Devenish et al., 2012). Turbulence induces vertical oscillations of air parcels and causes fluctuations in temperature, water vapor concentration, and supersaturation (e.g., Ditas et al., 2012; Hammer et al., 2015). The effects of supersaturation fluctuations on droplet condensational growth in turbulent environments have been studied for several decades (e.g., Cooper, 1989; Khvorostyanov and Curry, 1999). A qualitative description of this mechanism is that some “lucky” cloud droplets experience relatively larger supersaturation or stay a relatively longer time in the cloud compared with the other cloud droplets; therefore they can grow larger in size and broaden the CDSB. Recent theoretical and experimental studies support this mechanism and provide ways to quantify the resulting width of the droplet size distribution (e.g., McGraw and Liu, 2006; Sardina et al., 2015; Chandrakar et al., 2016; Grabowski and Abade, 2017; Siewert et al., 2017). Turbulence can also modulate the condensational growth of cloud droplets through mixing and entrainment (e.g., Lasher-Trapp et

al., 2005; Cooper et al., 2013; Korolev et al., 2013; Yang et al., 2016). In addition, turbulence can enhance the collision efficiency between droplets and produce “lucky” cloud droplets through stochastic collisions, which has been confirmed by direct numerical simulations and Lagrangian drop models (e.g., Paluch, 1970; Kostinski and Shaw, 2005; Falkovich and Pumir, 2007; Grabowski and Wang, 2013; Naumann and Seifert, 2015; de Lozar and Muesle, 2016).”

**Paragraph about the aerosol-induced CDSB broadening:**

“Aerosols, which serve as condensation nuclei of cloud droplets, can also cause CDSB broadening in turbulent environments through several mechanisms. First, turbulence-induced mixing and entrainment can trigger in-cloud activation of haze particles, which can broaden the left branch of size distribution (e.g., Khain et al., 2000; Devenish et al., 2012; Yang et al., 2016; Grabowski et al., 2018). Secondly, giant cloud condensational nuclei (GCCN, usually defined as aerosols with dry diameter larger than a few  $\mu\text{m}$ ) provides an embryo for large droplets, which can broaden the right branch of size distribution (e.g., Feingold et al., 1999; Yin et al., 2000; Jensen and Lee, 2008; Cheng et al. 2009). Recently, Jensen and Nugent (2017) investigated the effect of GCCN on droplet growth and rain formation using a cloud parcel model. They found that GCCN provides an embryo for big droplets at the activation stage and, more importantly, GCCN enhances droplet growth after activation due to the solute effect. For example, droplets formed on GCCN can still grow through the condensation of water vapor in the downdraft region even though the environment is subsaturated with respect to pure water (Jensen and Nugent, 2017). This, in fact, is an extreme case of Ostwald ripening.”

**Paragraph about Ostwald ripening:**

“Ostwald ripening for cloud droplets is the phenomenon when larger droplets grow and smaller droplets shrink due curvature and/or solute effects and, thus, it can broaden the CDSB at both small and large sides of the distribution. Srivastava (1991) investigated the growth of cloud droplets in a rising air parcel. Results show that the variance of squared radius of the CDSB was constant during the condensational growth process if both curvature and solute effects were ignored, but it was increased if those effects were considered. This “condensational broadening” is more pronounced in clouds with high cloud droplet number concentration and low vertical velocity. In turbulent clouds, droplets will experience supersaturated/subsaturated conditions in updraft/downdraft regions. Korolev (1995) studied the evolution of the CDSB driven by supersaturation fluctuations in a vertically oscillating air parcel. Supersaturation fluctuations in his study mean that air is supersaturated in the updraft and subsaturated in the downdraft; however no spatial inhomogeneity of supersaturation is considered in the parcel. Results show that the growth and evaporation cycles during the CDSB evolution are irreversible if the solute and curvature effects are considered. This “CDSB irreversibility” (terminology used in his paper) will promote the growth of large cloud droplets, lead to evaporation or even deactivation of small cloud droplets, and thus broaden the CDSB. Korolev (1995) argued that stronger turbulent fluctuations of supersaturation would result in a broader CDSB. This is contrary to Celik and Marwitz (1999), who found that supersaturation fluctuations are not responsible for CDSB broadening and the formation of large droplets. The curvature and solute effects on Ostwald ripening, activation and deactivation have been the topics of study in recent years (e.g., Wood et

al., 2002; Arabas and Shima, 2017, Chen et al., 2018; Sardina et al., 2018) but, to our knowledge, the relative roles of the curvature effect and solute effect on CDSB broadening have not been investigated.”

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.

We add more discussion about the influence of turbulence and in-cloud activation.

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.

We add a paragraph about the effects of the broadening of cloud droplet spectrum (second paragraph in the manuscript).

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).

We clarify the meaning of supersaturation fluctuations in the text:

“Korolev (1995) studied the evolution of the CDSB driven by supersaturation fluctuations in a vertically oscillating air parcel. Supersaturation fluctuations in his study mean that air is supersaturated in the updraft and subsaturated in the downdraft; however no spatial inhomogeneity of supersaturation is considered in the parcel. ”

The phrases “irreversibility of droplet size spectrum shape” (p. 2, line 26), “CDSB is irreversible” and “Irreversibility of the CDSB” (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).

We rephrase our discussion about “irreversibility” and make the statement clearer.

“Results show that the growth and evaporation cycles during the CDSB evolution are irreversible if solute and curvature effects are considered. This “CDSB irreversibility” (terminology used in his paper) will promote the growth of large cloud droplets, lead to evaporation or even deactivation of small cloud droplets, and thus broaden the CDSB.”

## 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”. 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 referred 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.

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

## 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.

We change “the sizes of cloud droplets” to the more appropriate description “**droplet size for a moving bin**” throughout the manuscript.

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.

We change “totally evaporated” to “**deactivates and becomes a haze particle**” throughout the manuscript.

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

We modify this sentence as:

“...For the latter case, the cloud parcel ascends at a vertical velocity of  $0.5 \text{ ms}^{-1}$  for three hours with the same initial condition as the control case. At the end of the simulation, the cloud parcel reaches about 6000 m and cloud droplets are supercooled (around 248 K), but we ignore ice nucleation in this study.”

### 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.

We thank the reviewer for the helpful comments. We add more discussion and references about the choice of the parameters as follows.

#### **For aerosol number concentration:**

“We test two other aerosol number concentrations ~~by increasing and decreasing the number concentration of the control by an order of magnitude~~,  $10^2 \text{ cm}^{-3}$  and  $10^4 \text{ cm}^{-3}$ , and keep the median radius and geometric standard deviation the same as the control case (see Figures 4 a and c). These values are chosen to represent the conditions for clean clouds ( $10^2 \text{ cm}^{-3}$ ) and polluted clouds ( $10^4 \text{ cm}^{-3}$ ), which are consistent with previous studies (Xue and Feingold, 2004; Chen et al., 2018).”

#### **For updraft velocity:**

“~~The effect of vertical velocity on the CDS is investigated next.~~ Two vertical velocities ( $0.1 \text{ ms}^{-1}$  and  $1.0 \text{ ms}^{-1}$ ) are used to test their effects on CDS broadening. These values are chosen based on observations that updraft in stratocumulus clouds is on the order of  $0.1 \text{ ms}^{-1}$  and in cumulus clouds is on the order of  $1.0 \text{ ms}^{-1}$  (Ditas et al., 2012; Katzwinkel et al., 2014).”

#### **For the recirculation layer:**

“Turbulence driven by cloud-top radiative cooling can result in various eddy sizes in the stratocumulus-topped boundary layer (Wood, 2012). Two different ~~eyeling~~ recirculation layer depths are ~~also~~ tested, 150 m and 350 m, to investigate the effect of eddy size on CDS broadening.”

### 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.

We thank the reviewer for the helpful comments. To test the effects of mass accommodation coefficient and spectrum discretization, two more sensitivity studies are added. One case sets the mass accommodation coefficient to 0.06 based on Shaw and Lamb (1999). The other case changes the number of bins from 100 to 200. Both cases show similar results compared with the control case. We also change the three-significant-digit precision to two-significant-digit precision in Table 1 and throughout the paper. We modify the text as:

“We have studied the effects of total aerosol number concentration, updraft velocity, and thickness of the recirculation layer on CDSB broadening. However we note that there are other parameters used in this study that can lead to the uncertainties in the results. For example, Takeda and Kuba (1982) found that using an insufficient number of model bins will lead to the narrow CDSB reported by Mordy (1959). Kreidenweis et al. (2003) found that both the spectral discretisation and the uncertainty in the value of mass accommodation coefficient can lead to uncertainty in the results. To test the effects of mass accommodation coefficient and spectrum discretization on the CDSB, two more sensitivity studies are conducted. One case is to set mass accommodation coefficient ( $\alpha_m$ ) to 0.06 based on Shaw and Lamb (1999). It is expected that a smaller value of  $\alpha_m$  might suppress the growth of cloud droplets. The other case is to change the number of bins from 100 to 200, while keeping other parameters the same as in the control case.”

~~“...In addition if reactivation also occurs, the relative dispersions are closer to observations, which are usually larger than 0.1 (Liu and Daum, 2002; Chandrakar et al., 2016). If reactivation also occurs, the smallest cloud droplet radius associated with a moving bin  $r_{min}$  is around 5  $\mu m$  and the relative dispersion is larger than 0.1. It is interesting to note that low mass accommodation has a negligible effect on  $r_{max}$ , but it has a stronger impact on  $r_{min}$ . This will result in a broader CDSB compared with the control case. In addition, results for 200 bins are similar to that for the control case, which means that the 100 bins used in this study are enough to limit the uncertainty due to spectrum discretization...”~~

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).



We extend the discussion of the observed cloud droplet size distribution and relative dispersion in the manuscript.

“Results from sensitivity studies show that relative dispersion is larger than 1.5 when both deactivation and reactivation occur (see Table 1), which is consistent with the values from observations and simulations (e.g., Miles et al., 2000; Liu and Daum, 2002; Lu and Seinfeld, 2006; Chandrakar et al., 2016). It should be mentioned that the CDSO observed in previous studies might have the problem of instrumental broadening due to low instrument resolution or long-distance averaging of the sampling volume (Brennguier et al., 2011; Devenish et al., 2012). A broad CDSO is also observed by recent holographic measurements, which limit the effect of instrument broadening and have much higher temporal and spatial resolution than other instruments, such as particle-counting probes (Beals et al., 2015; Glienke et al., 2017; Desai et al., 2018).”

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).

We add more discussion about residence time of cloud droplets and its impact on droplet size.

“...Previous studies show that although the mean lifetime of cloud droplets is usually less than half an hour, the residence time for some lucky cloud droplets can be longer than one hour (e.g., Feingold et al., 1996; Kogan, 2006; Andrejczuk et al., 2008). Those long-lifetime cloud droplets might contribute to large droplets in the cloud, similar to long-lifetime ice particles in mixed-phase clouds (Yang et al., 2015).”

#### 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 entrainment 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).

We add more discussion about the effect of entrainment and mixing and adiabatic assumption.

“It should be mentioned that one limitation of this study arises from the use of the adiabatic assumption for three-hour simulations. Turbulence can result in not only upward and downward

oscillations but also in entrainment and mixing (Shaw, 2003; Devenish et al., 2012). The latter can cause cloud droplet evaporation, deactivation and reactivation (Korolev et al., 2013; Yang et al., 2016). In addition, the lifetime of the cloud parcel is usually less than one hour (Andrejczuk et al., 2008). Therefore, one should be aware that results in this study are based on a very idealized state. More realistic studies should consider mixing processes where for example a trajectory ensemble model would be a suitable tool (Ovchinnikov and Easter, 2010; Feingold et al., 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.

We thank the reviewer for pointing out that our results contradict several previous studies. We add more discussion in the manuscript:

“...The mechanism of CDSB 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 are consistent with previous studies (e.g., Andrejczuk et al., 2008; Hoffmann et al., 2015; Hoffmann, 2017; Chen et al., 2018). However the results are counter to some other studies where details of activation and deactivation are argued to be unimportant in the cloud simulation (e.g., Srivastava, 1991; Chuang et al., 1997; Grabowski et al., 2018)....”

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).

We add more discussion about 2d-bin aerosol-cloud spectra scheme in the manuscript,

“...Tracking the solute distribution for each bin of cloud droplet is possible using a joint 2-D bin aerosol-cloud microphysical scheme, but it is very computationally expensive (e.g., Bott, 2001; Andrejczuk et al., 2010; Ovchinnikov and Easter, 2010; Lebo and Seinfeld, 2011)...”

Comments on the composition and technical remarks

p. 1, l. 23 please avoid the word “believed”

We change “believed” to “considered”.

p. 1, l. 24 Imagining rather than imaging?

We change “Imaging” to “Imagining”.

p. 2, l. 2 please explain or remove the word “linear”

We remove “linear” throughout the manuscript.

p. 2, l. 3 please rephrase the sentence so that collisional growth efficiency is not logically coupled with inverse proportionality of condensational growth rate

We rephrase the sentence:

~~“...while collisional growth is efficient when the droplet diameter is larger than 38  $\mu\text{m}$  (Hocking, 1959). On the other hand, collisional growth is efficient when the droplet diameter is larger than 38  $\mu\text{m}$  (Pruppacher and Klett, 2010).”~~

p. 2, l. 3-4 I suggest using approximate sizes and perhaps referencing a more recent textbook instead of the work of Hocking

We change “Hocking” to “Pruppacher and Klett”.

p. 2, l. 15 please indicate causation instead of just saying “be related to”

We change “be related to” to “cause”

p. 2, l. 23 GCCN provide (and not provides)?

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. 3 sensitivity studies (not sensitivities)

We change “sensitivities” to “sensitivity”.

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)?

We change “kinematic” to “kinetic”.

p. 6, l. 20 please rephrase “number concentrations of the control”

We rephrase that sentence:

“We test two other aerosol number concentrations ~~by increasing and decreasing the number concentration of the control by an order of magnitude~~,  $10^2 \text{ cm}^{-3}$  and  $10^4 \text{ cm}^{-3}$ , and keep the median radius and geometric standard deviation the same ~~as the control case~~”

p. 7, l. 15 “larger than **in** the control case”

We add “in” there.

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.

We thank the reviewer for the detailed comments on reference. We corrected our references in the manuscript. DOI number labels for all references are also added.

## Figures

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

All figures are high-resolution images with \*.eps format now.