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 CDSD 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 curvature and/or solute effects and, thus, it can broaden the CDSD 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 CDSD 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 CDSD 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 CDSD evolution are irreversible if the solute and curvature effects are considered. This "CDSD 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 CDSD. Korolev (1995) argued that stronger turbulent fluctuations of supersaturation would result in a broader CDSD. This is contrary to Celik and Marwitz (1999), who found that supersaturation fluctuations are not responsible for CDSD 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 CDSD 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 CDSD broadening; (3) aerosol-induced CDSD broadening.

Paragraph about the effects of the CDSD broadening:

"The broadening of the CDSD has a strong effect on precipitation and radiation. A broader CDSD 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 CDSD increases the relative dispersion, which is the ratio of standard deviation to the mean CDSD. 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 CDSD 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 CDSD broadening:

"Turbulence is ubiquitous in the clouds and can cause CDSD 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 CDSD. 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 Muessle, 2016)."

Paragraph about the aerosol-induced CDSD broadening:

"Aerosols, which serve as condensation nuclei of cloud droplets, can also cause CDSD 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 µm) 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 CDSD 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 CDSD 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 CDSD 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 CDSD evolution are irreversible if the solute and curvature effects are considered. This "CDSD 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 CDSD. Korolev (1995) argued that stronger turbulent fluctuations of supersaturation would result in a broader CDSD. This is contrary to Celik and Marwitz (1999), who found that supersaturation fluctuations are not responsible for CDSD 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 CDSD 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 CDSD 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), "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).

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

"Results show that the growth and evaporation cycles during the CDSD evolution are irreversible if solute and curvature effects are considered. This "CDSD 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 CDSD."

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 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.

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 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 cm^{-3} and 10^4 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 cm^{-3}) and polluted clouds (10^4 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 CDSD is investigated next.~~ Two vertical velocities (0.1 ms^{-1} and 1.0 ms^{-1}) are used to test their effects on CDSD broadening. These values are chosen based on observations that updraft in stratocumulus clouds is on the order of 0.1 ms^{-1} and in cumulus clouds is on the order of 1.0 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 ~~cycling~~ recirculation layer depths are ~~also~~ tested, 150 m and 350 m, to investigate the effect of eddy size on CDSD 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 CDSD 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 CDSD 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 CDSD, two more sensitivity studies are conducted. One case is to set mass accommodation coefficient (α_m) to 0.06 based on Shaw and Lamb (1999). It is expected that a smaller value of α_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 rmin is around 5 µm 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 CDSD 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 CDSD observed in previous studies might have the problem of instrumental broadening due to low instrument resolution or long-distance averaging of the sampling volume (Brenguier et al., 2011; Devenish et al., 2012). A broad CDSD 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 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).

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 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 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 μm (Hocking, 1959)~~. On the other hand, collisional growth is efficient when the droplet diameter is larger than 38 μm (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 cm^{-3} and 10^4 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 P¨oschl, 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.

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.

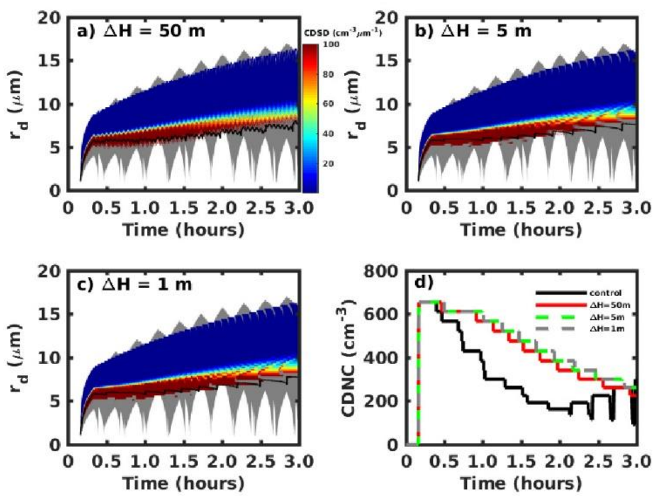

Figure 7: Cloud droplet size distribution (CDSD) changes with time for different thicknesses of recirculation layers: a) ΔH=50 m, b) ΔH=5 m, c) ΔH=1 m. d) Total cloud droplet number concentration changes with time for the different cases. The gray region in a-c represents the

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.

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

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Abstract. Cloud droplet size distributions (CDSDs), which are related to cloud albedo and lifetime, are usually broader in warm clouds than predicted from adiabatic parcel calculations. We investigate a mechanism for the CDSD broadening using a ~~Lagrangian bin-microphysics~~ **moving-bin** cloud parcel model that considers the condensational growth of cloud droplets formed on polydisperse, sub-micrometer aerosols in an adiabatic cloud parcel that undergoes vertical oscillations, such as those due to cloud circulations or turbulence. Results show that the CDSD can be broadened during condensational growth as a result of Ostwald ripening amplified by droplet deactivation and reactivation, **which is consistent with early work**.~~, which is consistent with Korolev (1995).~~ The relative roles of the solute effect, curvature effect, deactivation and reactivation on CDSD broadening are investigated. Deactivation of smaller cloud droplets, which is due to the combination of curvature and solute effects in the downdraft region, enhances the growth of larger cloud droplets and thus contributes particles to the larger size end of the CDSD. Droplet reactivation, which occurs in the updraft region, contributes particles to the smaller size end of the CDSD. In addition, we find that growth of the largest cloud droplets strongly depends on the residence time of cloud droplet in the cloud rather than the magnitude of local variability in the supersaturation fluctuation. This is because the environmental saturation ratio is strongly buffered by smaller cloud droplets. Two necessary conditions for this CDSD broadening, which generally occur in the atmosphere, are: (1) droplets form on polydisperse aerosols of varying hygroscopicity and (2) the cloud parcel experiences upwards and downwards motions. Therefore we expect that this mechanism for CDSD broadening is possible in real clouds. Our results also suggest it is important to consider both curvature and solute effects before and after cloud droplet activation in a cloud model. The importance of this mechanism compared with other mechanisms on cloud properties should be investigated through in-situ measurements and 3-D dynamic models.

1 Introduction

Warm clouds play a crucial role in water cycle and energy balance on Earth (Boucher et al., 2013) so understanding the whole life cycle of warm ~~cloud~~ **clouds**, including formation, development and precipitation, is important for better prediction of local weather and global climate. Cloud droplet growth is dominated by diffusion of water vapor at the early stage of cloud development, while collisional growth is ~~believed~~ **considered** to be the most important mechanism for drizzle formation and warm cloud precipitation (Pruppacher and Klett, 2010). The concept of a cloud parcel rising adiabatically in the atmosphere has been used to study cloud microphysical properties for decades. Imaging **Imagining** an initially sub-saturated air parcel rising adiabatically, cloud forms at the lifting condensation level and the growth of cloud droplets due to diffusional growth can be accurately predicted if we know the aerosol chemical composition. Because the linear growth rate of a cloud droplet is inversely proportional to droplet size, diffusional growth is inefficient when the droplet diameter is larger than 20 μm while collisional growth is efficient when the droplet diameter is larger than 38 μm (Hocking, 1959). **On the other hand, collisional growth is efficient when the droplet diameter is larger than** 38 μm **(Pruppacher and Klett, 2010).** In addition **Meanwhile**, the sizes of the smaller cloud droplets will approach those of the larger droplets and narrow the cloud droplet size distribution (CDSD), which is also unfavorable for collisional growth (Howell, 1949; Mordy, 1959). If only diffusional growth is considered, **the CDSD becomes narrower and** several tens of minutes even up to hours will be needed for a cloud droplet to reach efficient-collision size in an ascending cloud parcel. , which is much longer than the timescale typically observed. **However, the CDSD in a real cloud is usually wider than predicted by an adiabatic cloud parcel model and drizzle-size cloud droplets are frequently observed in warm clouds (e.g., Laird et al., 2000; Glienke et al., 2017; Siebert and Shaw, 2017).**

The broadening of the CDSD has a strong effect on precipitation and radiation. A broader CDSD 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 CDSD increases the relative dispersion, which is the ratio of standard deviation to the mean CDSD. 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 CDSD 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.

Turbulence is ubiquitous in the clouds and can cause CDSD 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 CDSD. 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 stochas-

tic 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 Muessle, 2016).

Aerosols, which serve as condensation nuclei of cloud droplets, can also cause CDSD 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 μm) 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.

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 CDSD 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 CDSD 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 CDSD 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 CDSD evolution are irreversible if the solute and curvature effects are considered. This “CDSD 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 CDSD. Korolev (1995) argued that stronger turbulent fluctuations of supersaturation would result in a broader CDSD. This is contrary to Çelik and Marwitz (1999), who found that supersaturation fluctuations are not responsible for CDSD 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 CDSD broadening have not been investigated.

Here we consider an adiabatic cloud parcel that experiences vertical oscillations, with cloud droplets that are formed on polydisperse, sub-micrometer aerosols. Results show that the CDSD is broadened during diffusional growth due to Ostwald ripening and associated droplet deactivation and reactivation, which is consistent with previous studies (e.g., Korolev, 1995; Çelik and Marwitz, 1999). In this study, we investigate (1) what are the relative roles of the solute and curvature effects on CDSD broadening, and (2) what other factors can affect this broadening? This paper is organized as follows. Section 2 introduces the basic setup for cloud parcel model, which is similar to Jensen and Nugent (2017) except that there are no GCCN. Results related to CDSD broadening and the associated ~~sensitivities~~ **sensitivity** studies are detailed in Section 3. Conclusions are summarized in Section 4, including a discussion of implications in cloud observations and modeling.

2 Methods

Historically there are two types of bin microphysics: fixed-bin scheme and moving-bin scheme (see section 4.2.1 in Khain et al. (2015) and references therein). The advantage of the moving-bin method (i.e., Lagrangian bin-microphysics) is that it can avoid artificial CDSD broadening. In this study, we use a cloud parcel model with moving-bin microphysics. The original version of the model was designed to study cirrus clouds by Heymsfield and Sabin (1989), and then the warm clouds (Feingold and Heymsfield, 1992; Feingold et al., 1998). In recent years, this model has been modified and applied to investigate various of microphysical problems (e.g., Feingold and Kreidenweis, 2000; Xue and Feingold, 2004; Ervens and Feingold, 2012; Yang et al., 2012; Li et al., 2013; Yang et al., 2016). In the current version of parcel model, air pressure (p), parcel height (h), air temperature (T), water vapor mixing ratio (q_v), and radii of haze and cloud droplets (r_i) are prognostic variables, which are calculated using the variable-coefficient ordinary differential equation solver (VODE) (Brown et al., 1989). Specifically, p is calculated from hydrostatic equation and h depends on the vertical velocity (w). Similar to Eq. 11 in Heymsfield and Sabin (1989), T is calculated from,

$$\mathbf{\frac{dT}{dt} = -\frac{g}{c_{p,air}} w + \frac{l_v}{c_{p,air}} \frac{dq_w}{dt},} \tag{1}$$

where g is the gravitational acceleration, $c_{p,air}$ is the heat capacity of air, l_v is the latent heat of water vaporization, and q_w is the liquid water mixing ratio. The first term in Eq. 1 is the cooling due to dry adiabatic ascent, and the second term is the microphysical contribution due to the release of latent heat of condensation. Because the total water mixing ratio is conserved in the parcel, a decrease in water vapor mixing ratio ($-dq_v$) equals an increase in liquid water mixing ratio (dq_w). Air supersaturation (S_e), which controls the growth of haze and cloud droplets, is calculated from T, p and q_v. A brief introduction of the model setup and the main mathematical formulations used for cloud microphysical processes are described below.

In this study, the parcel starts rising at about 300 m below cloud base and starts descending at about 300 m above cloud base, which is similar to Jensen and Nugent (2017), except that our cloud parcel then experiences upward and downward oscillations between 50 m above cloud base and 300 m above cloud base (see Figure 1a). The ascending and descending velocities are set to be 0.5 $m\ s^{-1}$ and $-0.5\ m\ s^{-1}$ for the control case. At the parcel's initial altitude of 600 m, the initial air temperature is 284.3 K, pressure is 938.5 hPa, and saturation ratio is 0.856, which are as same as Jensen and Nugent (2017).

The initial dry aerosols are ammonium ~~sulphate~~ **sulfate** with a log-normal size distribution range of 10 nm to 500 nm in radius (no GCCN). The sub-micrometer aerosols are parsed into 100 bins, **where the**~~. The~~ median radius is 50 nm ~~with a~~**and the** geometric standard deviation ~~of~~**is** 1.4. The total number mixing ratio is 1000 mg^{-1} for the control case, which is about 1000 cm^{-3} (see Figure 1b). The model first calculates the equilibrium size of haze droplets for each bin at 85.6% relative humidity, as does Jensen and Nugent (2017). **The equilibrium size of haze particles for the ith bin (r_i) at initial relative humidity is obtained by solving the equation $S_{sat}(r_i) = RH(t=0)$ iteratively, where S_{sat} is the saturation ratio for a solution droplet, calculated from Köhler equation (Pruppacher and Klett, 2010, p. 172),**

$$\mathbf{S_{sat}} \equiv \frac{\mathbf{e}}{\mathbf{e_s(T)}} = \mathbf{a_s(r_{d,i}, r_i)} \exp\left(\frac{\mathbf{2\sigma_s}}{\rho_\mathbf{w}\mathbf{R_v T r_i}}\right), \tag{2}$$

where e is the water vapor pressure in air, e_s is the saturated water vapor pressure over a solution droplet at T, ρ_w is the density of water, and R_v is the gas constant for water vapor. σ_s is the water activity of the haze droplets, which is a function of temperature and solute (Pruppacher and Klett, 2010, p. 133). a_s is the water activity of haze droplets, which depends on the composition of aerosol, size of dry aerosol (r_d), and size of haze droplets (r). In this study, a_s for cloud droplets is calculated from laboratory-based parameterizations (Eq. 2 in Tang and Munkelwitz (1994)).

Only diffusional growths of haze and cloud droplets are considered in our model. Collision coalescence, sedimentation, mixing, and entrainment are ignored. The growth of haze or cloud droplet for the ith bin is calculated from,

$$\frac{\mathbf{dr_i}}{\mathbf{dt}} = \frac{\mathbf{1}}{\mathbf{r_i}} \frac{\mathbf{S_e - S_{sat}}}{\mathbf{G}}, \tag{3}$$

where G is the growth parameter given by,

$$\mathbf{G} = \left[\frac{\rho_\mathbf{w}\mathbf{R_v T}}{\mathbf{D'_v e_s(T)}} + \frac{\rho_\mathbf{w}\mathbf{l_v}}{\mathbf{k'_T T}}\left(\frac{\mathbf{l_v}}{\mathbf{R_v T}} - \mathbf{1}\right)\right]. \tag{4}$$

D'_v and k'_T are, respectively, the modified diffusion coefficient and the modified thermal diffusion coefficient (Lamb and Verlinde, 2011, p. 337-338),

$$\mathbf{D'_v} = \frac{\mathbf{D_v}}{\frac{\mathbf{r_i}}{\mathbf{r_i}+\lambda} + \frac{\mathbf{4D_v}}{\alpha_\mathbf{m}\mathbf{\bar{c}_{air} r_i}}}, \tag{5}$$

and

$$\mathbf{k'_T} = \frac{\mathbf{k_T}}{\frac{\mathbf{r_i}}{\mathbf{r_i}+\lambda} + \frac{\mathbf{4k_T}}{\alpha_\mathbf{T}\mathbf{\bar{c}_{air} n_{air} c_{p,air} r_i}}}. \tag{6}$$

Here D_v is the physical diffusion coefficient, k_T is the thermal diffusion coefficient, λ is the mean free path of air, $\overline{c}_{air}$ is the mean molecular speed of air, and n_{air} is the number concentration of air. α_m is the mass accommodation coefficient and α_T is the thermal accommodation coefficient. In this study, we choose $\alpha_m = 1.0$ and $\alpha_T = 1.0$.

S_{sat} in the growth equation (Eq. 3) is calculated from the Köhler equation (Eq. 2). Therefore, the curvature effect (exponential part in Eq. 2) and the solute effect (a_s in Eq. 2) are considered during the growth process for each bin. It should be noted that there are several methods to calculate the solute effect with the relative deviations for activation ranging up to 20%, but the differences are small for droplet growth (Pöschl et al., 2009). **In addition, different choices of parameters–such as σ_s, α_m and α_T–can also cause differences in droplet growth (Kreidenweis et al., 2003). How the choices of different parameters would affect our results is worth studying in the future.** The total simulation time is 3 hours, and variables are recorded every 1 s that include temperature, pressure, height, water vapor mixing ratio, as well as droplet size and number concentration for each bin.

3 Results and discussions

3.1 Cloud droplet size distribution broadening

For the control case, the liquid water mixing ratio increases linearly with height in the ascending branches and decreases in the descending branches as shown in Figure 2a. 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)**. The saturation ratio has an increasing trend in the ascending branch after each cycle, but has a decreasing trend in the descending branch (indicated by red and blue arrows in Figure 2b). ~~Droplet size as a function of height in two bins~~ **Droplet size for two moving bins** are shown in Figure 2c. **Droplet size in the bins monotonically increase with the dry aerosol mass associated with the bin.** The solid line is for the cloud droplet that formed on a dry aerosol of 503 nm and represents the largest droplet in our simulation. It grows in the ascending branch but it evaporates in the descending branch. Also, the droplet size for this bin increases after each cycle. The dashed line in Figure 2c is for the cloud droplet that formed on a dry aerosol of 51 nm. For this cloud droplet, the changes in radius with height are similar for the initial few cycles, after which the cloud droplet ~~totally evaporates~~ **deactivates and becomes a haze particle**. Ultimately, the aerosol is reactivated again as a cloud droplet by the end of the simulation (green dashed line). **Also notice that a second mode appears in the CDSD due to reactivation of aerosols after about 2 hours (see Figure 2d).** Thus the CDSD broadens after each cycle as the larger droplets become larger and the smaller droplets either remain similarly sized or become smaller ~~(see Figure 2d)~~. All these features are consistent with Korolev (1995) (see Fig. 5 in his paper).

Korolev (1995) analytically investigate the narrowing and broadening of cloud droplet size distribution during condensation when solute and curvature effects are considered. He considers a cloud parcel oscillating vertically in simple harmonic motion.

Results show that the CDSD is irreversible if solute and curvature effects are considered. Irreversibility of the CDSD will not only promote the growth of large droplets, but it will also lead to the evaporation, or even deactivation of small cloud droplets, and thus broaden the CDSD. However, the relative roles of the solute effect, curvature effect, deactivation and reactivation on the broadening of droplet size distributions have not been investigated.

To explore the relative roles of different factors in this CDSD broadening mechanism, three more cases are tested here. For the first case, we turn off both the solute and curvature effects for all cloud droplets after $700\ s$; this is the time when the cloud parcel first reaches $50\ m$ above cloud base and is just below the oscillation layer. **Specifically, we set $S_{sat} = 1$ for all droplets.** The result is shown in Figure 3a. For this case, the CDSD repeats for each cycle, consistent with Korolev et al. (2013), and the total cloud droplet number concentration (n) is constant (red solid line in Figure 3d). For the second case, we only turn off the curvature effect but retain the solute effect. **Specifically, we ignore the exponential term in Eq. 2 such that $S_{sat} = a_s$.** The result in Figure 3b shows that the largest droplet (with the most solute) can grow after each cycle while the smallest droplet size (with the least solute amount) **associated with a moving bin** does not change much after each cycle. However ~~the size that largest droplet~~ **the largest droplet size that a bin** can reach is much smaller than that in the control case. Because the saturated water vapor pressure over a droplet formed on larger aerosol is lower than that formed on smaller aerosol due to the solute effect, the larger droplet grows faster than the smaller droplet in the updraft region, and it evaporates slower in the downdraft region. For this case, the solute effect alone cannot explain the larger cloud droplets in the control case. In addition, n is also a constant and droplet deactivation does not occur (green dashed line in Figure 3d). In the third case, we consider both curvature and solute effects, but we do not allow droplet reactivation. This means that once the droplet ~~totally evaporates~~ **deactivates** it cannot be activated again. The result in Figure 3c shows that the growth of the largest cloud droplet is similar to the control case, but the size of **the** smallest cloud droplet **associated with a bin** also increases after each cycle. The reason for this CDSD broadening is the Ostwald ripening effect, where large droplets grow at the expense of small ones. Past studies have concluded that the ripening effect is typically slow and inefficient for droplet growth (Wood et al., 2002). But the vertical oscillations near cloud base that are considered here allow for droplet deactivation and result the decrease of n with time (see Figure 3d), as in the control case. Thus, the typically inefficient Ostwald ripening is amplified through the resulting deactivation of the smallest droplets. An early suggestion of this behavior is shown in Fig. 8 of Hagen (1979). The only difference between the control and this simulation is that n for the control case increases near the end of the simulation because of droplet reactivation (see Figure 3d). It should be mentioned that the step changes in n in Figure 3d are a result of using a discretized bin method to represent the continuous spectrum. A downward step in n means droplet deactivation, and an upwards step in n means droplet reactivation. Deactivation and reactivation can also be seen from the CDSD qualitatively: droplet deactivation occurs when the peak value of CDSD decreases (from red to blue as shown in Figure 2d), while droplet reactivation occurs when a subset of smaller cloud droplets appears.

From Figures 3 a and b, we can see that the solute effect contributes part of the CDSD broadening compared with the control case. But the solute effect alone is not enough to explain the growth of the largest cloud droplet. Droplet deactivation, which is related to the curvature effect, plays a crucial role here (see Figure 3c). Because the oscillations occur within the cloud region, 50 m above cloud base, droplet deactivation is surprising to us. There are two related questions: (1) Why and which droplet will deactivate? (2) Why is droplet deactivation related to the CDSD broadening?

The reason for the droplet deactivation is mainly because the cloud parcel experiences upwards and downwards oscillations. In the downdraft region, the air is subsaturated, which supports droplet evaporation. In addition, the saturated water vapor pressures over polydisperse droplets are different via both the solute and curvature effects. Smaller droplets with less solute and larger radii of curvature have higher saturated water vapor pressures, and thus evaporate faster than larger droplets in the downdraft region. Therefore, smaller droplets will evaporate first in the downdraft region.

The reason why droplet deactivation is related to the CDSD broadening can be explained in two ways. From the thermodynamic point of view, the liquid water mixing ratio is roughly a constant at a given height for each cycle (see Figure 2a). As the n decreases due to the droplet deactivation, we can expect that on average droplet size will be larger because the same amount of water will be redistributed on fewer cloud droplets. From the ~~kinematic~~ **kinetic** point of view, quasi-steady state supersaturation (s_{qs}) will become larger after each cycle due to droplet deactivation, as shown in Figure 2b. s_{qs}, the environmental supersaturation in quasi-steady state, is inversely proportional to the integral of mean droplet size $\overline{r}$ and droplet number concentration (n), $s_{qs} \propto (\overline{r}n)^{-1}$ (Lamb and Verlinde, 2011). Here the decrease in n due to droplet deactivation is much greater than the change of $\overline{r}$; therefore, s_{qs} will increase with decreasing n. This means that larger droplets grow even faster in the updraft region, and smaller droplets evaporate even faster in the downdraft region – beyond the solute effect alone. Conversely, an increase in s_{qs} will enhance droplet deactivation for smaller droplets, and it will also reinforce the growth of larger droplets in a positive feedback.

One question relevant to precipitation initiation is how fast can the largest cloud droplet grow in an oscillating parcel compared with droplets in an ascending-only parcel? **For the latter case, the cloud parcel ascends at a vertical velocity of** 0.5 $m\ s^{-1}$ **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.** The mean (yellow dashed line) and largest/smallest (upper/lower gray dashed lines) cloud droplets in an ascending-only cloud parcel are also shown in Figure 2d. It can be seen that the size of the largest cloud droplet **in a moving bin** at cloud top in each cycle of the oscillating parcel (blue color bar) is similar to that in the ascending-only parcel (upper gray line). This is quite surprising because when the parcel reaches 1200 m for the first time (i.e., the top of the oscillation cycle), the largest cloud droplet radius is 9.07 μm (see Table 1 and Figure 2c); however after several cycles, the largest cloud droplet radius is 17.3 μm, still at 1200 m. The size is similar to the largest droplet size **associated with a moving bin** in an ascending-only parcel at a height of about 6000 m. This means that the largest cloud droplet size **for a bin** in an oscillating parcel at 1200 m is much larger than calculated from a traditional cloud parcel model (ascent only), and hence shows "superadiabatic" growth. ~~It should be mentioned that cloud droplets in the ascending-only parcel at 6000 m are supercooled (around 248 K), but we~~

~~ignore ice nucleation in this study.~~ In addition, the size of the smallest cloud droplet **for a bin** and the mean droplet size are larger in an ascending-only parcel. Differences between the mean droplet sizes increases after each cycle, especially at the end of the simulation due to the reactivation of numerous small droplets. Therefore, the relative dispersion, which is the ratio of the standard deviation to the mean of a droplet size distribution, also increases after each cycle, and is much larger than in an ascending-only cloud parcel. ~~Broadening of the CDSD might quicken precipitation processes, and the increase of the relative dispersion is relevant to the albedo effect and increase albedo susceptibility.~~

3.2 Sensitivity studies

In this subsection, we investigate effects of several factors on the CDSD in the adiabatic parcel model with vertical oscillations. These factors include variations in the total aerosol number concentration, updraft velocity, and thickness of the ~~recycling~~ **recirculation** layer.

3.2.1 Effect of total 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~~, $10^2\ cm^{-3}$ and $10^4\ cm^{-3}$, 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** ($10^2\ cm^{-3}$) **and polluted clouds** ($10^4\ cm^{-3}$)**, which are consistent with previous studies (e.g., Xue and Feingold, 2004; Chen et al., 2018).** The results show that the CDSD for the relatively clean case ($10^2\ cm^{-3}$) behaves similarly to the solute effect alone (compare Figures 3b and 4b) – there is neither droplet deactivation nor reactivation. The CDSD broadening is due to the ripening effect alone, which is not as efficient as when it is accompanied by deactivation as in the control case. For the relatively polluted case ($10^4\ cm^{-3}$), both droplet deactivation and reactivation occur (see Figure 4d). The largest cloud droplet acts similarly as that in the control case, while the smallest cloud droplet is larger 1.5 h into the simulation but then begins to become smaller compared with the control case. We interpret these observations as follows. For the clean case, all aerosols are activated, and all droplets are able to grow to a relatively large size, making them unlikely to deactivate. However for polluted case, not all CCN are activated, there are therefore some smaller droplets that cannot grow very large and they will evaporate first in the downdraft region. **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).**

3.2.2 Effect of vertical velocity

~~The effect of vertical velocity on the CDSD is investigated next.~~ **Two vertical velocities ($0.1\ m\ s^{-1}$ and $1.0\ m\ s^{-1}$) are used to test their effects on CDSD broadening. These values are chosen based on observations that updraft in stratocumulus clouds is on the order of $0.1\ m\ s^{-1}$ and is cumulus clouds is on the order of $1.0\ m\ s^{-1}$ (Ditas et al., 2012; Katzwinkel et al., 2014).** For a relative low velocity of $\pm 0.1\ m\ s^{-1}$, the cloud parcel only experiences one and a half cycles within three hours (see Figure 5a). The parcel reaches cloud base around 1 hour, significantly later than the control case due to the small velocity (see Figure 5a). However, the largest cloud droplet size ultimately becomes similar to that in the control case, and we also see the cloud droplet number concentration decrease due to droplet deactivation. No droplet reactivation occurs because the small velocity generates a low supersaturation in the updraft region, which is unfavorable for droplet reactivation. For a relative high velocity of $\pm 1.0\ m\ s^{-1}$, the cloud parcel can cycle more times within three hours (see Figure 5c). The parcel reaches cloud base faster than the control case (see Figure 5c). **Here we keep the thickness of the recirculation layer constant. Therefore, larger vertical velocity results in a higher oscillation frequency.** Both droplet deactivation and reactivation occur in this case, and the largest and smallest cloud droplets behave similarly to the control case.

3.2.3 Effect of the thickness of ~~cycling~~ 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 ~~cycling layers~~ **recirculation layer depths** are ~~also~~ tested, $150\ m$ **and** $350\ m$**,** ~~here~~ **to investigate the effect of eddy size on CDSD broadening.** For a ~~recycling~~ **recirculation** layer of $150\ m$, which is $100\ m$ thinner than the control case, the parcel experiences more cycles within three hours (see Figure 6a). The total cloud droplet number concentration decreases with time due to droplet deactivation, but no droplet reactivation occurs (see Figure 6b). Therefore the largest cloud droplet is similar to the control case, but the smaller cloud droplet is larger than **in** the control case. For a ~~cycling~~ **recirculation** layer of $350\ m$, the parcel can penetrate the cloud base each cycle (see Figure 6c). In this case, all cloud droplets are deactivated below cloud base and reactivated again when the cloud parcel is supersaturated in the next ascending branch. Therefore the CDSD is repeated and no broadening occurs.

3.3 Discussion

We have studied the effects of total aerosol number concentration, updraft velocity, and thickness of the recirculation layer on CDSD 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 CDSD 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 CDSD, two more sensitivity

studies are conducted. One case is to set mass accommodation coefficient (α_m) to 0.06 based on Shaw and Lamb (1999). It is expected that a smaller value of α_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.

Table 1 summarizes the microphysical properties at cloud top for different cases. When the cloud parcel first reaches about $1200\ m$, the largest cloud droplet radius **associated with a moving bin** (r_{max}) is $9.1\ \mu m$ (case 0). If the cloud parcel continues rising for three hours as for the ascending-only case, $r_{max} = 17\ \mu m$ at $6000\ m$. However if the parcel experiences ~~cycling~~ **recirculation** within cloud region, r_{max} can also be around $17\ \mu m$ as long as deactivation occurs, except for the low N_a case (see Table 1). ~~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 CDSD 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.**

From the above, we see that droplet deactivation and droplet reactivation play crucially important roles in CDSD broadening in this study. Deactivation of smaller droplets is important for the growth of larger cloud droplets (e.g., see Figures 2d, 3c, 4d, 5b,d and 6b). Droplet deactivation occurs in the descending branch for smaller droplets due to both the curvature and solute effects (Ostwald ripening). The evaporation of smaller cloud droplets with less solute makes water vapor available for the growth of other larger cloud droplets. On average, the largest cloud droplet size **for a moving bin** increases with time after each cycle.

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 CDSD observed in previous studies might have the problem of instrumental broadening due to low instrument resolution or long-distance averaging of the sampling volume (Brenguier et al., 2011; Devenish et al., 2012). A broad CDSD 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).

We note that deactivation is suppressed for a thin recirculation layer $\Delta H = 150\ m$ 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 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.**

One interesting result is that the size of the largest cloud droplet **associated with a moving bin** within each cycle is similar to that in the ascending-only parcel (i.e., approximately within one micrometer), as shown in Figure 8. The general trends approximately follow ~~a linear mass~~ **the** growth rate that is independent of aerosol number concentration, vertical velocity and the thickness of the oscillation layer, as long as deactivation occurs. This suggests that the growth of the largest cloud droplets strongly depends on the amount of time such droplets remain in the cloud **(residence time of cloud droplets)**, rather than the temporal variability of supersaturation in updrafts and downdrafts. The reason is that the environmental (i.e., the in-cloud) saturation ratio (S_e) is buffered by the equilibrium saturation ratio (S_{sat}) over smaller droplets. Figure 9 shows the changes of S_e and S_{sat} over two droplets (same used as in Figure 2c) in the control case. Instead of being symmetric around 1 for the pure water case (ignoring solute and curvature effects), S_e in the oscillating parcel is symmetric around S_{sat} over the small cloud droplets. For example before 1.5 hours, droplets formed on $r_a = 51nm$ are the smallest cloud droplets in the population, and the average S_e (gray line) during one oscillation is roughly symmetric around the blue line (Figure 9). The fact that S_e is buffered by S_{sat} over small cloud droplets is mainly because the number concentration of the smallest cloud droplet (36 cm^{-3} in the control case) is much larger than that of large cloud droplet ($1.8 \times 10^{-9}\ cm^{-3}$). When those small droplets ~~totally evaporate~~ **deactivate** (between 1.5 to 2.5 hours), S_{sat} (blue line) for those deactivated droplets is the same as S_e (gray line). During this period, S_e is symmetric around S_{sat} over the remaining small droplets (larger than the droplets formed on $r_a = 51\ nm$ but smaller than for $r_a = 503\ nm$). When the droplets formed on $r_a = 51\ nm$ are reactivated (after 2.5 hours), S_e is symmetric around $S_{sat}(r_a = 51\ nm)$ again until they are deactivated. It should be mentioned that number concentration of those reactivated droplets increases steady after each cycle after 2.0 hours (See Figure 3d). By the end of the simulation, the number concentration of the reactivated droplets is similar to that of the remaining large droplets (about $150\ cm^{-3}$). Therefore, the effect of those reactivated droplets on the environmental saturation ratio becomes stronger after 2.0 hours (see Figure 9).

This symmetric property of S_e can be also explained using the quasi-steady supersaturation s_{qs}. For pure water droplets, $s_{qs} \sim \frac{w}{nr}$ (Lamb and Verlinde, 2011). **This can be obtained from the analytical expression of supersaturation in an adiabatic cloud parcel:** $\frac{dS_e}{dt} = Aw - Bnr(S_e - 1)$**, where A and B are parameters depending on thermodynamic properties (Korolev and Mazin, 2003).** A symmetric distribution of w around 0 will generate a symmetric distribution of s_{qs} around 0 (i.e., S_e around 1). If ~~the~~ curvature and solute effects are considered, s_{qs} will be symmetric around s_k given the same condition of w, because $\frac{dS_e}{dt} = Aw - Bnr(S_e - S_{sat})$ **and thus** $s_{qs} \sim \frac{w}{nr} + s_k$, where $s_k = S_{sat} - 1$ is the equilibrium supersaturation ratio over a mono-disperse droplet. **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. Therefore S_e is symmetric around S_{sat} over smaller droplets before they ~~totally evaporate~~ **deactivate** in the oscillating parcel. $\overline{S_e} - S_{sat}$ controls the growth of a large droplet and it is positive on average. That is why the large droplets can grow after each cycle. In addition, the influence of S_e fluctuations on droplet growth is small if S_{sat} over a large droplet is much lower than S_e and its fluctuations. The extreme examples of this phenomenon are when droplets form on GCCN in warm clouds (Jensen and Nugent, 2017) or ice particles form in mixed phase clouds. Therefore, the growth of the large droplet here is dominated by its in-cloud lifetime. **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).**

However if all cloud droplets are deactivated, CDSD broadening does not occur (see Figure 6d). Without droplet deactivation, the CDSD can also broaden due just to the solute effect, as is the case when the curvature effect is ignored (Figure 3b) or when the total aerosol number concentration is low (Figure 4b). CDSD broadening due to the ripening effect without droplet deactivation is not as significant as it is with droplet deactivation, but it also might be important after several hours as suggested by Wood et al. (2002).

Droplet reactivation usually occurs in the updraft region after several cycles, and those reactivated droplets will be deactivated again in the downdraft region. Formation of smaller cloud droplets can broaden the CDSD at smaller sizes, decrease the mean cloud droplet size, and increase the relative dispersion. Meanwhile, the generation of new cloud droplets also suppresses the growth of larger cloud droplets (see Figure 2d).

4 Conclusions and atmospheric implications

In this study, we investigate the condensation growth of cloud droplets in an adiabatic parcel with vertical oscillations based on a ~~Lagrangian bin-microphysics~~ **moving-bin** cloud parcel model where cloud droplets are formed on polydisperse, submicrometer aerosol particles. Both the solute and curvature effects are considered for all cloud droplets before and after activation during the whole simulation. The CDSD can also broaden by condensation growth due to Ostwald ripening together with droplet deactivation and reactivation, which is consistent with the results of Korolev (1995). Droplet deactivation occurs in the descending branch due to the combination of the solute and curvature effects. Deactivation of smaller droplets makes water vapor available for other larger droplets, and thus broadens the CDSD at larger sizes. The growth of the largest cloud droplet **in a vertically oscillating cloud parcel** approximately follows ~~a linear mass~~ **the** growth rate **in an ascending-only cloud parcel** after each cycle, and it is independent of aerosol number concentration, vertical velocity, and the thickness of the oscillation layer, as long as deactivation occurs. The size of the largest cloud droplet strongly depends on the time that droplet remains in the cloud rather than on the variability of the in-cloud supersaturation. This is because the environmental air is buffered by the large number of smaller cloud droplets: the environmental saturation ratio in an oscillating parcel is symmetric around the equilibrium saturation ratio over smaller cloud droplets. The ~~linear mass~~ growth rate **for the largest cloud droplets** can be used to roughly estimate the ~~right upper boundary~~ **large-size upper boundary** of the CDSD, at least in this study. Droplet reactivation usually occurs after a few cycles. These cloud droplets are activated in the ascending branch, and deactivated in the descending branch. They are usually very small (less than $5\ \mu m$) and thus broaden the CDSD at smaller sizes. The mean cloud droplet size significantly decreases when reactivation occurs, which leads to an increase in relative dispersion. On the other hand, those newly-formed cloud droplets compete against other cloud droplets for water vapor, thus suppressing the growth of larger cloud droplets.

We note that there are additional factors that might affect droplet growth that are not treated in this study. For example, we do not consider the sedimentation of cloud droplets in this study, similar to Korolev et al. (2013) and Jensen and Nugent (2017). This is a reasonable assumption for an updraft velocity of $0.5\ m\ s^{-1}$ or above, but ignoring sedimentation in the low velocity case ($0.1\ m\ s^{-1}$) will limit the accuracy of our results. ~~Entrainment and mixing are also ignored here.~~ In addition, we do not consider the collision coalescence between droplets. Although CDSD broadening is favorable for collision processes, it might be interesting to determine how this broadening will accelerate rain formation.

We have used idealized simulations to find this new CDSD broadening mechanism, so it is reasonable to ask if this mechanism is likely to occur in nature. To answer this question, we investigate the two necessary conditions for this mechanism and address whether these conditions exist in real clouds. The first necessary condition is that droplets form on polydisperse aerosol particles with different solute effects. This is a very general occurrence in the atmosphere due to the complexity of aerosol size and composition (Murphy et al., 1998; Khain et al., 2000). The second necessary condition is that a cloud experiences upward and downward oscillations. This is also a general occurrence in natural clouds due to turbulence and circulations that can become established within a cloud layer (Wood, 2012). Therefore we expect that this mechanism of CDSD broadening is possible in the real clouds.

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). How important this mechanism is to CDSD broadening in real clouds compared with other mechanisms is worth future investigation, but is beyond the scope of this study.

There is an implication of this mechanism for the cloud modeling community. Most of the bulk and bin microphysical schemes only consider the curvature and solute effects during the activation process based on Köhler theory. Cloud droplets are assumed to be pure water after they are activated. 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., Andrejczuk et al., 2010; Ovchinnikov and Easter, 2010; Lebo and Seinfeld, 2011). 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 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).** Large eddy simulations with a similar microphysical treatment would be useful to investigate how important this mechanism is to CDSD broadening in more realistic clouds.

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Table 1. Microphysical properties at cloud top for different cases: r_{max} is the largest cloud droplet radius **in a moving bin**, r_{min} is the smallest cloud droplet radius **in a bin**, $\bar{r}$ is the mean cloud droplet size, σ is the standard deviation of droplet radius, and $\sigma/\bar{r}$ is the relative dispersion. Case 0 is when the cloud parcel reaches the cloud top for the first time with the same setup as the control case (shown as black circle in Figure 3). For other cases, results represent the parcel at cloud top for the last time after 3 hours simulation; the example of the control case is shown as the green circle in Figure 3.

	r_{max} (μm)	r_{min} (μm)	$\bar{r}$ (μm)	σ (μm)	$\frac{\sigma}{\bar{r}}$	deactivation	reactivation
case 0	9.1	4.2	5.8	0.5	0.088	no	no
ascending only	17	12	13	0.55	0.041	no	no
control	17	6.1	7.5	1.6	**0.22**	**yes**	**yes**
$\alpha_m = 0.06$	17	5.1	7.0	1.9	**0.27**	**yes**	**yes**
N_{bin}=200	17	5.9	7.5	1.6	**0.22**	**yes**	**yes**
pure water	7.8	5.9	6.0	0.086	0.014	no	no
only solute effect	13	5.8	6.0	0.21	0.035	no	no
without reactivation	18	7.9	10	1.1	0.11	**yes**	no
low N_a	16	9.6	11	0.40	0.036	no	no
high N_a	17	3.1	4.7	1.5	**0.32**	**yes**	**yes**
low w	13	7.7	8.8	0.60	0.068	**yes**	no
high w	17	4.6	5.3	1.0	**0.19**	**yes**	**yes**
thin ΔH	17	6.2	8.5	1.4	**0.16**	**yes**	**yes**
thick ΔH	9.0	4.1	5.8	0.50	0.087	no	**yes**

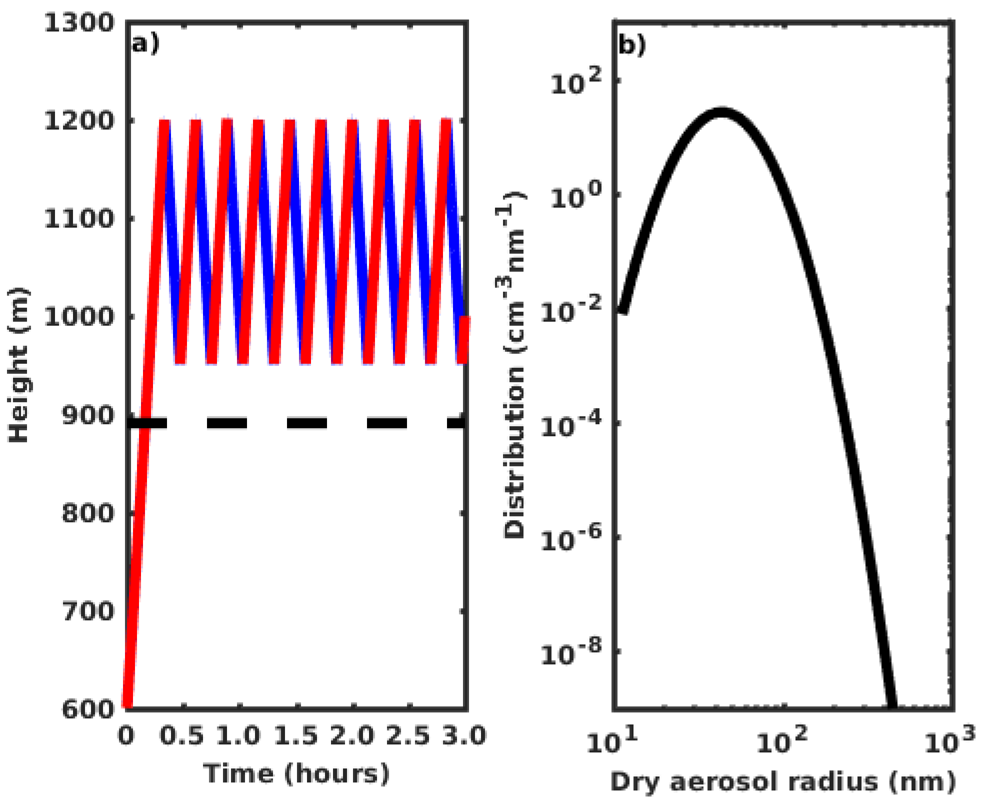

Figure 1. a) Trajectory of cloud parcel with upward and downward oscillations. Velocity is constant and is $0.5\ m\ s^{-1}$ for the ascending parcel and $-0.5\ m\ s^{-1}$ for the descending parcel. The dashed line is the cloud base, and the red and blue lines represent ascending and descending parcels. b) Initial dry aerosol size distribution. The total aerosol number concentration is $1000\ cm^{-3}$.

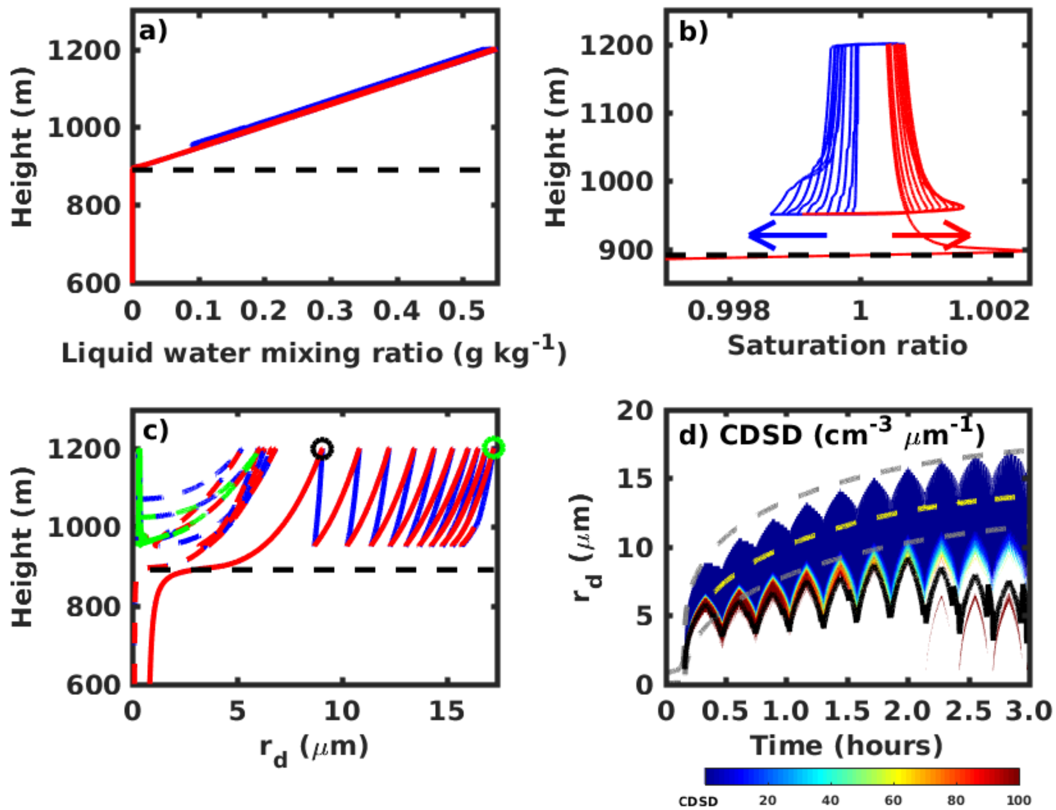

Figure 2. Thermodynamical and microphysical properties of an adiabatic cloud parcel with upward and downward oscillations. a) Liquid water mixing ratio changes with height. b) Cloud parcel saturation ratio changes with height. Arrows in b represent the evolution of saturation ratio profile with time. c) Radii changes of two selected cloud droplets with height. The solid line is for the largest cloud droplet that formed on a dry aerosol with radius of $503\ nm$, and the dashed line is for droplet that formed on an aerosol of $51\ nm$. The red and blue lines in a-c represent ascending and descending parcels, and the black dashed line indicates cloud base height. The green dashed line indicates the reactivation of that bin. The black and green circles are referred to in the text. d) Cloud droplet size distribution changes with time. The black line represents the mean cloud droplet radius change with time. The yellow dashed line is the change in mean droplet size for the ascending-only cloud parcel with a constant velocity of $0.5\ m\ s^{-1}$, and the upper and lower dashed gray lines represent the largest and smallest cloud droplets in the ascending-only parcel.

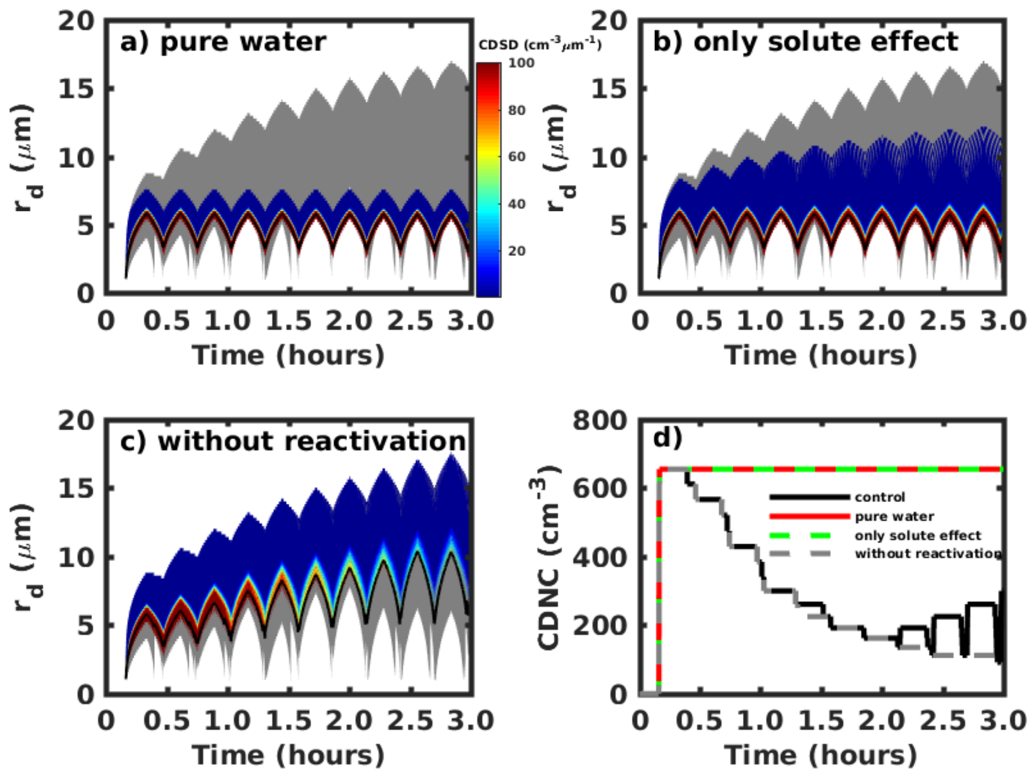

Figure 3. a) Cloud droplet size distribution (CDSD) changes with time without solute or curvature effects. b) CDSC changes with time with the solute effect but without the curvature effect. c) CDSD changes with time including both solute and curvature effects but where droplet reactivation is not considered. d) Total cloud droplet number concentration (n) 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.

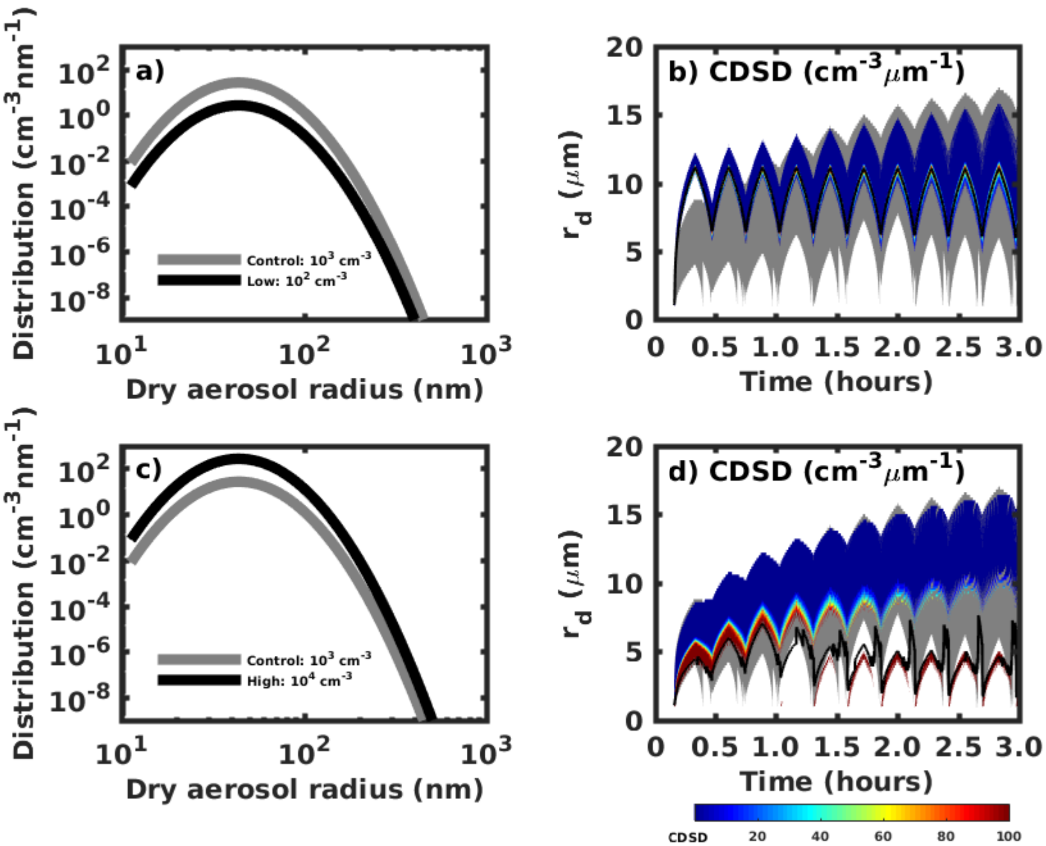

Figure 4. a) Aerosol size distribution for a low number concentration of 10^2 cm^{-3}. b) Cloud droplet size distribution changes with time for the low aerosol number concentration case. c) Aerosol size distribution for the high number concentration of 10^4 cm^{-3}. d) Cloud droplet size distribution changes with time for the high aerosol number concentration case. Gray lines in a and c represent the control case with a total aerosol number concentration of 10^3 cm^3, and gray regions in b and d are the range of the cloud droplet size spectrum for the control case.

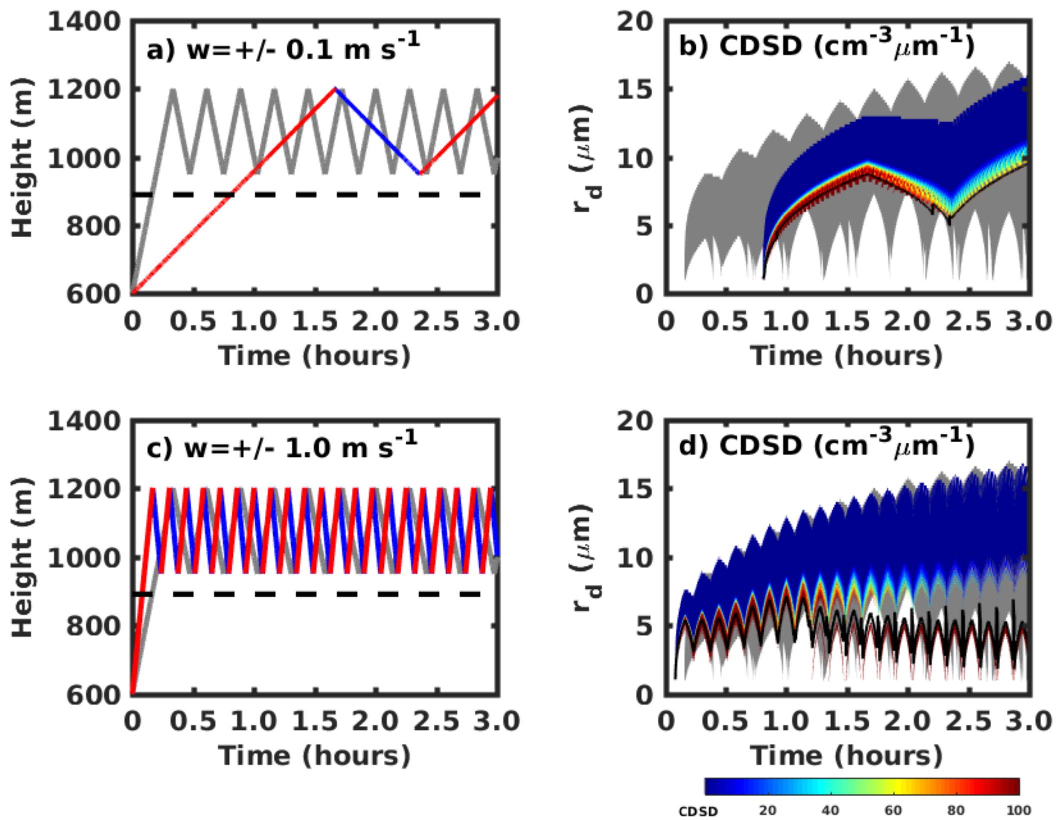

Figure 5. a) The height of cloud parcel changes with time for the low velocity case of $\pm\, 0.1\ ms^{-1}$. b) Cloud droplet size distribution changes with time for the low velocity case. c) The height of the cloud parcel changes with time for the velocity of $\pm\, 1.0\ ms^{-1}$. d) Cloud droplet size distribution changes with time for the high velocity case. Gray lines in a and c represent the control case with velocity of $\pm 0.5\ ms^{-1}$, and the gray regions in b and d are the range of cloud droplet spectrum for the control case.

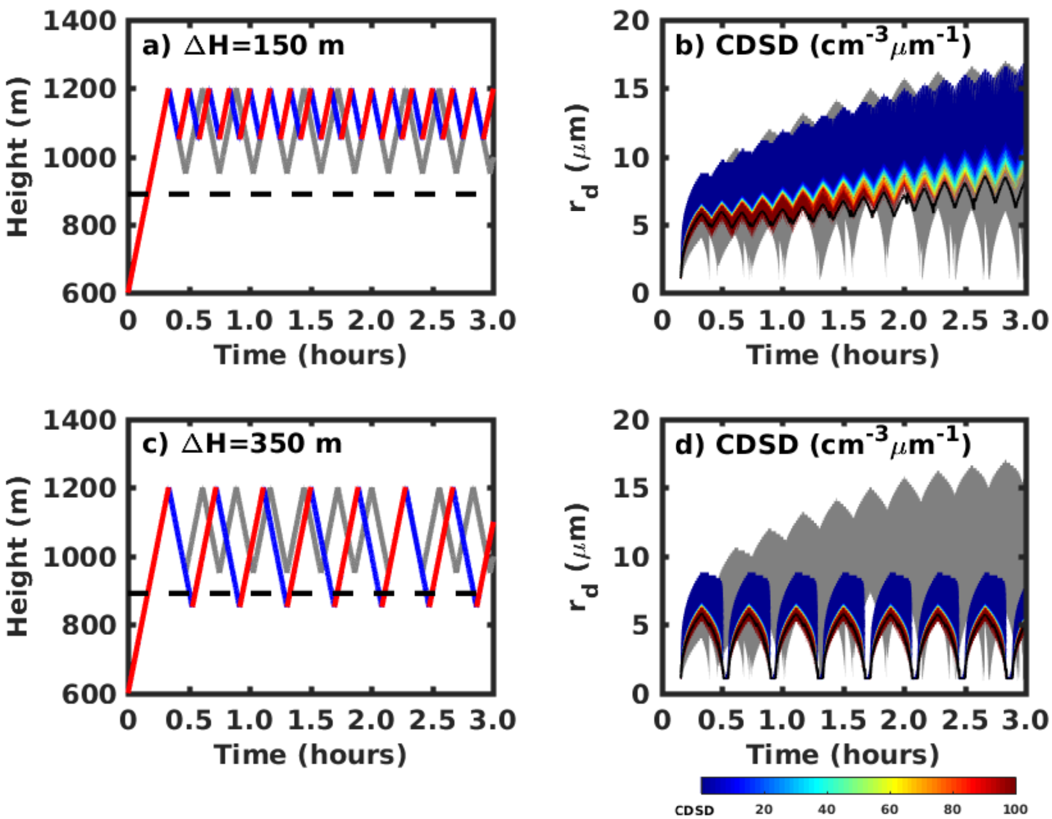

Figure 6. a) The height of cloud parcel changes with time for the thin ~~cycling~~ **recirculation** layer of 150 m. b) Cloud droplet size distribution changes with time for the thin ~~cycling~~ **recirculation** layer case. c) Aerosol size distribution for the thick ~~cycling~~ **recirculation** layer of 350 m. d) Cloud droplet size distribution changes with time for the thick ~~cycling~~ **recirculation** layer case. The gray lines in a and c represent the control case with ~~cycling~~ **recirculation** layer of 250 m, and the gray regions in b and d are the range of cloud droplet size spectrum for the control case.

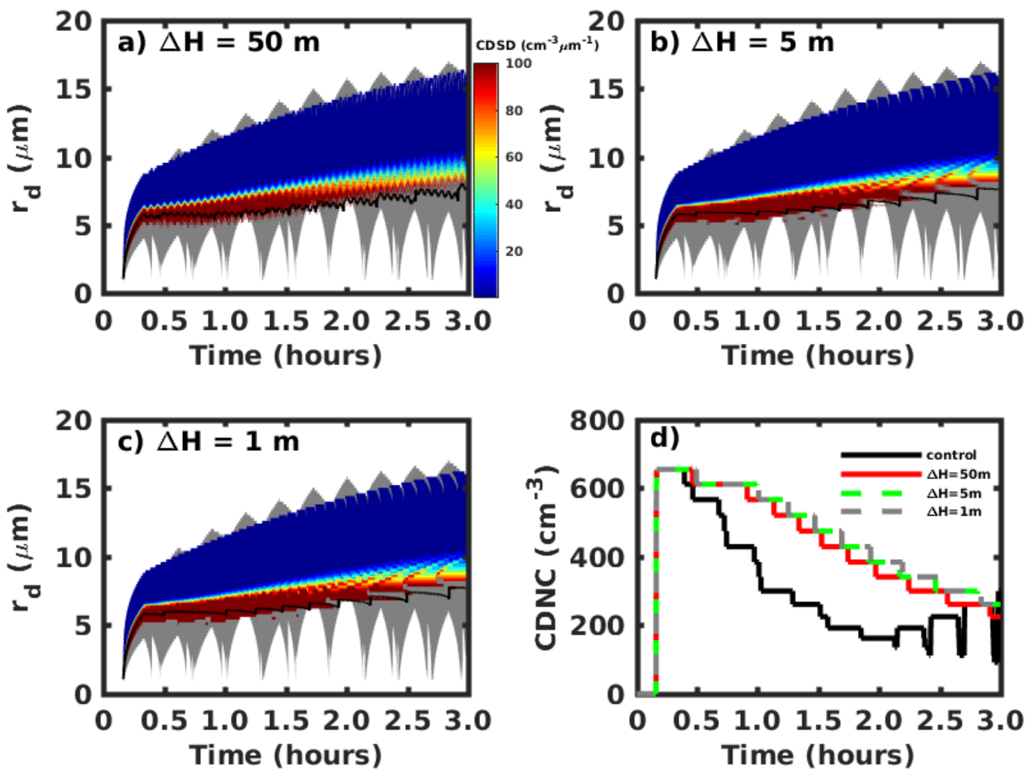

Figure 7. Cloud droplet size distribution (CDSD) changes with time for different thicknesses of recirculation layers: a) $\Delta H = 50\ m$, b) $\Delta H = 5\ m$, c) $\Delta H = 1\ m$. d) Total cloud droplet number concentration (n) changes with time for the different cases. The gray region in a-c represents the range of the droplet size spectrum for the control case, and the black lines represent the mean cloud droplet radius change with time.

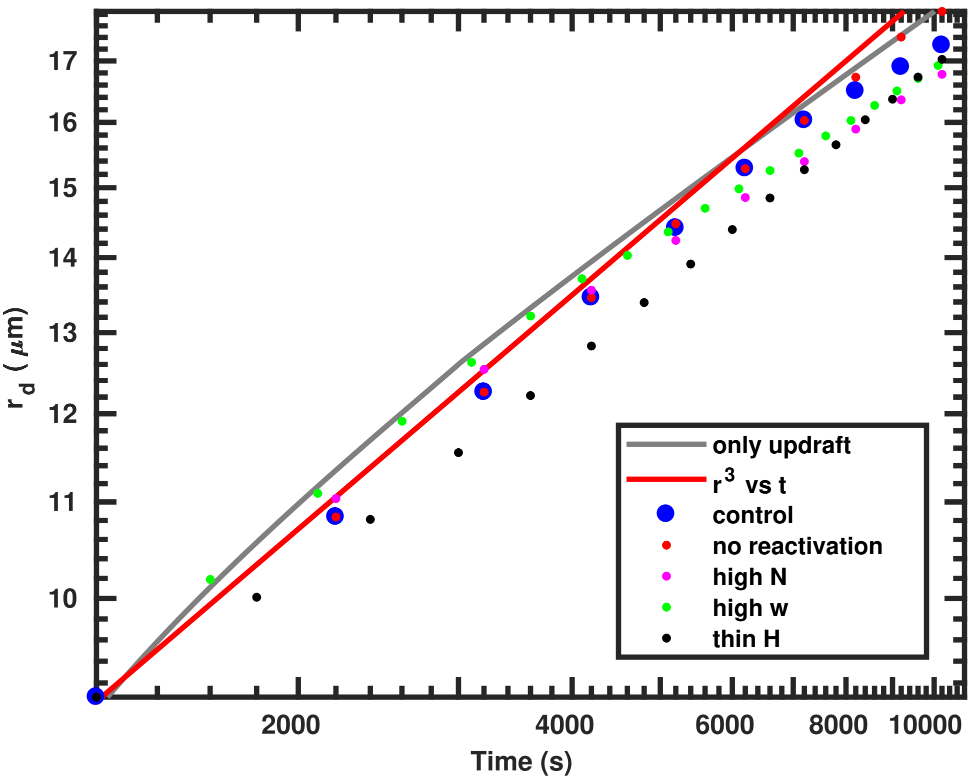

Figure 8. The largest cloud droplet size after each cycle is plotted for different cases discussed before: blue dots, control case; red dots, no reactivation case; pink dots, high number concentration case; green dots, high vertical velocity case; and black, thin oscillation layer case. The gray line is for the ascending-only case from Figure 4, and the red line represents the growth of a droplet ~~with a linear mass growth rate~~.

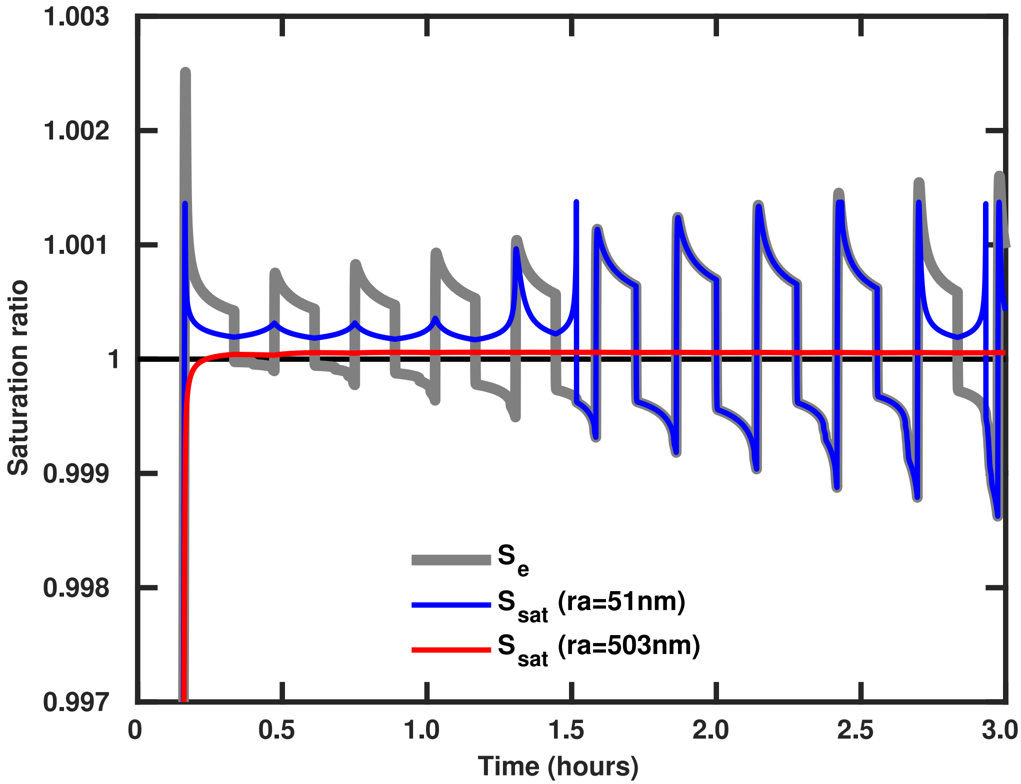

Figure 9. Changes of environmental saturation ratio (grey) and equilibrium saturation ratios over two droplets (red and blue) with time in an oscillating parcel. The blue line is for a droplet formed on a dry aerosol with radius of 53 nm and the red line is for a droplet formed on a dry aerosol with radius of 503 nm. The smaller cloud droplet (formed on a dry aerosol with radius of 53 nm) deactivates at approximately 1.5 hours and reactivates at approximately 2.5 hours.