Response to Shin-ichiro Shima

I would like to recommend this paper to be published but after major revisions.

This is an interesting paper introducing a new mechanism of cloud droplet activation named "collectional activation". The author investigated its contribution theoretically, then numerically using an LCM. Note also that this analysis could only be possible if using an LCM. One of the conclusion is that the impact is small because it seldom occurs compared to conventional "diffusional activation", but I think the community still needs to be aware of such possibility.

However, there exist at least one major issue in this manuscript. Unfortunately, the determination criterion of "collectional activation" the author introduced is not appropriate. Please see the attached note "Possible_collectional_activation_scenario.pdf". You can see that r>r_crit is not a rigorous criterion to determine "collectional activation". I strongly suggest the author to examine all the materials minutely, keeping the above fact in mind, I am still not fully sure how big the revision could be, but because all the analyses are based on the above criterion, this correction could affect the paper substantially, though it probably do not change the main conclusion significantly.

Please also see other major/minor comments annotated in the attached pdf.

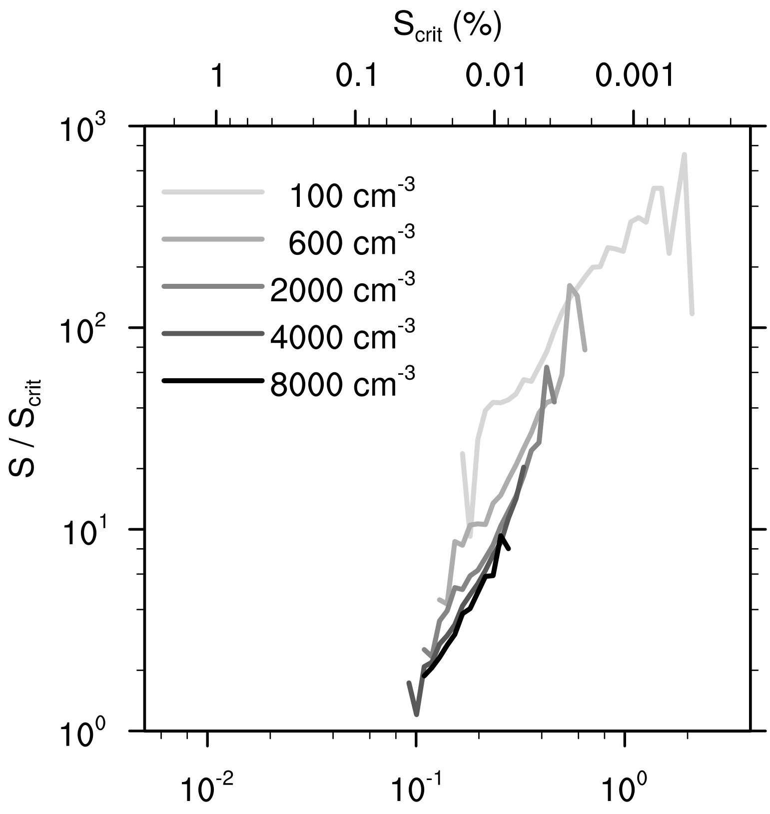
I am very thankful for the reviewer's comments which helped to clarify the paper in various aspects. However, I do not agree with his major comment on the appropriateness of the applied criterion for the detection of collectional activations, which will be outlined in this general response. More detailed answers will follow below.

The reviewer argues that the applied criterion to determine if an aerosol is activated or not, i.e., to distinguish between aerosols and cloud droplets, by comparing their radius against their respective critical radius (r vs. r_{crit}), is not adequate. In the present manuscript, I consider a particle as activated if it has grown beyond its critical radius ($r > r_{crit}$), a criterion which has been used and applied by various authors before (e.g., Rogers and Yau, 1989; Chuang et al., 1997; Khain et al., 2000; Boucher 2015; Hoffmann et al., 2015). Additionally, I request that the supersaturation enables further diffusional growth in the moment of activation to establish equivalence of diffusional and collisional activation (see line 119 - 123). Accordingly, the reviewer's collectional activation scenarios (ii) to (iv) are already considered in this study, which has been clarified and explained in more detail in the revised version of the manuscript (line 119 - 135 and comment 8 below). I only disagree with the reviewer's scenario (i). The reviewer argues that all particles which experience a supersaturation that exceeds the critical supersaturation ($S > S_{crit}$) should be considered as activated irrespective of their radius.

Of course, a supersaturation which exceeds the critical supersaturation ($S > S_{crit}$) will result in a radius which exceeds the critical radius ($r > r_{crit}$) at some point in time. And indeed, if the temporal dimension of particle growth and hence activation is neglected, both criterions are identical (see lines 20 - 27). But the time necessary for activation increases significantly for larger aerosols due to the kinetically limited transport of water molecules to the particle (Chuang et al. 1997; Hoffmann 2016). And if the supersaturation varies, as it

is the case in a real cloud due to entrainment/turbulence or simply due to the cloud's limited lifetime, the considered particle might not grow beyond its critical radius although the critical supersaturation has been exceeded for a certain period of time. Accordingly, the criterion of $r > r_{crit}$ is essential to decide if an activation has been completed or not.

Moreover, the critical supersaturations of the aerosols affected by collectional activation are so low that they are easily exceeded anywhere inside the cloud (cf. Fig. 5b). For the smallest aerosols affected by collectional activation (0.1 µm dry radius), the critical supersaturation is 0.03 % and decreases significantly for larger ones (e.g., 0.005 % for a radius of 0.4 µm, i.e., where the collectional fraction of activations becomes significant). The following figure shows the average supersaturation at the moment of collectional activation. Accordingly, the critical supersaturation is not restricting activation; it is exceeded several times by the supersaturations found in the simulated clouds.



Accordingly, the reviewer's criterion to consider all aerosols with $S > S_{crit}$ as activated makes no sense for the analysis carried out in this study. It would probably consider all aerosols larger than 0.1 µm as activated. And we would have no information if these aerosols succeed to grow beyond the critical radius for activation. (Which is probably not the case due to the kinetically limited transport of water vapor to the particle (e.g., Chuang et al., 1997; Nenes et al., 2001; Hoffmann et al. 2015).) Anyhow, the reviewer's questions shows perfectly the problems associated with Köhler activation theory at these large aerosol radii: It is simply not valid anymore. The critical supersaturation is easily exceeded, but the growth beyond the critical radius can be impeded by the naturally occurring variations of the supersaturation.

Further reviewer comments (I copied them in a chronological order from the reviewer's PDF annotations):

1. Major request.
This is not true for "collectional activation". Modify it appropriately.
See main response above.

2. Major request.
The discussion here is interesting and helpful to understand "collectional activation". However, r>r_crit is not a rigorous criterion for "collectional activation". Consider how to revise or justify the analysis.
See main response above.

3. Minor request.
To avoid confusion, you should explicitly mention that condensation/evaporation process is ignored in the theoretical analysis in this section.
Good point: "Moreover, all other microphysical processes, specifically diffusional growth, are neglected." (line 50)

4. Minor suggestion
To avoid confusion, you should clearly mention that those two red lines represent the critical radii, not the particle radius.
Good point: "For scenario B, an initially inactivated particle and an initially activated particle are examined (the critical radii are displayed in red by a continuous or dashed line, respectively)." (line 73 - 74)

5. Major question
Isn't this too big for calculating collision coalescence? Maybe it is okay for your method but have you checked the sensitivity to dt?
I didn't check the sensitivity to dt in this study, but a general study on the sensitivity of the collection algorithm to dt can be found in Unterstrasser et al. (2017). For a timestep of 1.0 s the results are reasonable. Accordingly, they should also be reasonable for a timestep of less or equal to 0.5 s. A reference to the study of Unterstrasser et al. (2017) is already given in line 356.

6. Major request
Please make it clear how you decide the initial dry aerosol radius. Uniform random sampling in log(dry_r) space? or any other?
Yes, as already stated two sentences above: "The dry aerosol radius is assigned to each super-droplet using a random generator which obeys a typical maritime aerosol distribution represented by the sum of three lognormal distributions (Jaenicke, 1993) (Fig. 2)." (line 109 - 110)

7. Major request
Not true for "collectional activation"
See main response above.

8. Major request.

Not true for "collectional activation". They can grow even when 0<S<S_crit if r>r_s.

This is covered in the study. The corresponding text has been clarified: "In this section, the applied methodology for untangling the contributions of diffusion and collection to the activation of aerosols is introduced. An aerosol becomes activated when it grows beyond its critical radius ($r > r_{crit}$). Moreover, activation requires the particle to be located in a volume of air with a sufficient supersaturation to enable unhindered diffusional growth. Depending on the microphysical process responsible for the final crossing of r_{crit}, different supersaturation allow unhindered diffusional growth.

Due to the continuous character of diffusional growth, the supersaturation has to be larger than the critical supersaturation in the moment in which the critical radius is exceeded:

$$S > S_{crit} = S_{eq}(r_{crit}),$$

where S_{eq} is the equilibrium supersaturation calculated according to Köhler theory (see Eq. (A3)). This condition is automatically fulfilled in the case of diffusional growth due to the constraints of Köhler theory on the equilibrium supersaturation. If the critical radius is exceeded by collection, the radius after collection might be immediately larger than r_{crit} and, hence, the necessary supersaturation is allowed to be smaller to enable unhindered diffusional growth:

$$S > S_{eq}(r_{ac}),$$

where $r_{ac} > r_{crit}$ is the wet radius after collection. This criterion is not automatically fulfilled and checked additionally to establish the formal equivalence of both processes, i.e., enabling unhindered diffusional growth after activation. Note that the process of activation, i.e., the entire growth beyond r_{crit}, can be driven by diffusional growth or by accumulating liquid water due to collection or by a combination of both." (line 119 - 135)

9. Major question and suggestion.
In my point of view, the definition of the collectional activation employed here is too complicated and unnatural.
Is it really necessary to include
inact + inact -> inact -> act
inact + act -> inact -> act
inact + act -> act (exclude scavenging)
as collectional activation?
Aren't these very rare events that can be negligible?
Further, I think collectional deactivation should be also interesting.

This is just an idea, but in my opinion, it is better to separate the instantaneous activation/deactivation analysis and history analysis, to clarify the structure of the paper.

It sounds natural to me to define the activation/deactivation categories using only instantaneous information:
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diffusional activation
inact -> act
diffusional deactivation
act -> inact
collectional activation:
inact + inact -> act (only direct one)

collectional deactivation:
inact + act -> inact
act + act -> inact

For the first step, analyzing the instantaneous activation/deactivation characteristics, should be sufficient.
Then, in the next step, you can carry out history analysis, and indeed it is interesting and important,
However, doing both at once complicate the discussion.
Please consider my proposal.

Actually, there is only one way to cause a collectional activation in the current study: In the moment a particle grows larger than the critical radius, the integrated collectional mass growth needs to exceed the integrated diffusional mass growth ($\Delta m|_{coll} > \Delta m|_{diff}$). The various types of interactions have been added to exemplify the naturally occurring microphysical processes that lead to $\Delta m|_{coll} > \Delta m|_{diff}$. They have been illustrated in Fig. 3 and need to be considered in the interpretation of the results. The only unnatural intervention is the exclusion of scavenging or the collection of drops if $max(r_A, r_B) > r_{crit,C}$. This has been clarified by rewriting the whole paragraph (line 149-166):

"To identify a collectional activation, the integrated collectional mass growth $\Delta m|_{coll}$ is compared to the diffusional $\Delta m|_{diff}$ in the moment the particle grows beyond its critical radius. If the former exceeds the latter, $\Delta m|_{coll} > \Delta m|_{diff}$, this activation is considered as collectional. There are various microphysical interactions resulting in $\Delta m|_{coll} > \Delta m|_{diff}$, and its basic types are illustrated in Fig. 3. Note that also a combination or a repetition of these types is possible, i.e., multiple subsequent collections. In a collectional activation of type (i), the water mass growth by collection dominates, i.e., the coalescence of two previously inactivated aerosols A and B results directly or after some diffusional growth in an activated particle C. In a collectional activations of type (ii), the critical radius increases faster than wet radius, i.e., the coalescence of an already activated particle A with another activated or an inactivated particle B results in inactivated particle C, which activates after some diffusional growth. If the resulting particle is directly activated, this process is only considered a collectional activation if the largest wet radius of the two coalescing particles A and B is smaller than the critical radius of the newly produced particle C:

$$max(r_A, r_B) < r_{crit,C}.$$

This ensures that the combined water of particles A and B is necessary to activate particle C. If this is not the case, i.e., the water of particle A or B is able to activate particle C on its own, the latter process is considered a regular collection of cloud droplets or as scavenging and neglected in the following analysis. Moreover, the coalescence of two activated particles resulting in a collectional activation is mathematically possible but not found to play a role in the analyzed simulations. Note that only collectional activations of the first type are able to increase the number of activated aerosols, while the second type might have no or a negative impact on the total number of activated aerosols since the coalescence of at least one activated particle results in one activated particle."

10. Typo
d -> Delta
Done.

11. Typo
Done.

12. Minor request.
This is ambiguous. Do you mean when it will be activated by diffusion without further coalescence?
This has been clarified. See answer to comment 9.

13. Minor request.
Same as above
This has been clarified. See answer to comment 9.

14. Typo
Done.

15. Typo
Done.

16. Minor suggestion.
Do diffusional activations also occur at high altitude? If so, wouldn't it be informative for readers to show also the vertical profile of the diffusional activation?
Yes, partly because of newly entrained aerosols or due to the kinetically limited activation of aerosols within the central updraft (see, e.g., Slawinska et al. 2012; Hoffmann et al. 2015 as stated in line 200). A vertical profile of the diffusional activation rate has been added (Fig. 5d).

17. Major question.
This is not trivial. Do you have any clear explanation why this does not happen? Is this just caused by the lack of aerosol particles of this size or is there any other mechanism to inhibit both diffusional and collectional activation?
For both activation types, the large critical radius inhibits activation for larger aerosols within the typical lifetime of the simulated clouds (about 15 min). The kinetically limited flow of water molecules slows down the diffusional activation at larger radii, e.g., more than 1000 s are necessary for the activation of an aerosol of 1 µm dry radius at 1 % supersaturation (Hoffmann 2016). Similarly, collectional activation is not able to produce the necessary radii in the available time since the droplets might be too small to cause intense collisions. Moreover, the critical radii might be too big for the simulated clouds to sustain them and they might fall out of the cloud before activation (the largest activated aerosol is 200 µm in wet radius, Fig. 7).

The questioned sentence has been extended (line 214-215): "Larger aerosols do not activate at all since their critical radius is too large to be exceeded by diffusion or collection."

18. Minor question.
If red is 0.8 and blue is 0.2, and 100 collectional activations occur, I understand that 80 activated and 20 inactivated aerosols are involved in these

100 collectional activation events. Is this correct?
If so, in Fig.7, red is always larger than blue, but this is puzzling.
At the section starting from L.135, it is declared that the following two processes are considered a collectional activation:
inact + inact -> act
inact + act -> act
It means, the number of activated aerosols involved in collectional activations must be always smaller than the number of inactivated aerosols
involved.
However, this is not the case in Fig.7.
Please make this point clear.
Maybe just the legend is opposite? That is, red is inactivated and blue is activated? or maybe you count
inact + inact -> act
as blue and
inact + act -> act
as red?

The whole figure has been changed to clarify the manuscript. Figure 8 shows the average number of collections necessary for the collectional activation of one aerosol. The number of collected activated or inactivated particles has been neglegted in this figure. All necessary information on how many activated aerosols have been involved in the analyzed collectional activations was contained in the former Fig. 7b (now Fig. 9).

19. Minor question.
Same question as above. How do you calculate the red and blue line for this case?
See last comment.

20. Minor request
It is difficult to follow the meaning of this sentence. In particular the last half. Do you mean "average entrainment height of all particles inside the cloud is the cloud base"?? Please give a clear and detailed explanation.
Yes. The sentence has been clarified to: "Since multiple collections are necessary for their activation (see Fig. 8), the lower average entrainment height is representative for the average entrainment height of all particles inside the cloud, which is the cloud base through which most particles enter the cloud (e.g., Hoffmann et al. 2015)." (line 263 - 265)

References:
Boucher, O. (2015). Aerosol–Cloud Interactions. In Atmospheric Aerosols (pp. 193-226). Springer Netherlands.

Chuang, P. Y., Charlson, R. J., and Seinfeld, J. H. (1997). Kinetic limitations on droplet formation in clouds. Nature, 390(6660), 594-596.

Hoffmann, F., Raasch, S., and Noh, Y. (2015). Entrainment of aerosols and their activation in a shallow cumulus cloud studied with a coupled LCM–LES approach. Atmospheric Research, 156, 43-57.

Hoffmann, F. (2016). The Effect of Spurious Cloud Edge Supersaturations in Lagrangian Cloud Models: An Analytical and Numerical Study. Monthly Weather Review, 144(1), 107-118.

Khain, A., Ovtchinnikov, M., Pinsky, M., Pokrovsky, A., and Krugliak, H. (2000). Notes on the state-of-the-art numerical modeling of cloud microphysics. Atmospheric Research, 55(3), 159-224.

Nenes, A., Ghan, S., Abdul-Razzak, H., Chuang, P. Y., and Seinfeld, J. H. (2001). Kinetic limitations on cloud droplet formation and impact on cloud albedo. Tellus B, 53(2), 133-149.

Slawinska, J., Grabowski, W. W., Pawlowska, H., and Morrison, H. (2012). Droplet activation and mixing in large-eddy simulation of a shallow cumulus field. Journal of the Atmospheric Sciences, 69(2), 444-462.

Yau, M.K. and Rogers, R.R. (1989). A short course in cloud physics. Elsevier.