Response to Anonymous Referee #1

The manuscript discusses the process of activation of cloud droplets on big aerosol particles. It checks for what aerosol size range the process of activation of cloud droplets can be explained by collisions between aerosol particles. It also checks the importance of the process of activation via collection compared to activation via diffusion of water vapor. The study is done using an LES setup combined with Lagrangian (i.e. particle tracking) representation of cloud microphysics. In the discussed simulations both aerosol particles and cloud droplets are represented using the Lagrangian approach, which allows to numerically resolve the activation process.

As shown in the manuscript, the studied process of activation by collection is very rare and affects mostly big aerosol particles entrained above the cloud base. As discussed in the summary of the manuscript, the studied process can be safely neglected, or even more, it might already be implicitly covered in some of the activation parametrization schemes. The presented study is therefore more theoretical and shows, in my understanding, in what aerosol size range the term *activation* as understood by the Köhler theory has any meaning.

The manuscript is well written and my further comments are both few and minor.

Thank you very much for your comments which helped to clarify the manuscript.

General comments

The manuscript defines three scenarios of activation of an aerosol particle by collision (lines 135-143):

1. coalescence of two inactivated aerosol particles resulting directly or after some diffusional growth in activated particle,
2. coalescence of an inactivated aerosol particle and activated aerosol particle that leads to an inactivated particle that activates due to diffusion,
3. coalescence of an inactivated aerosol particle and activated aerosol particle that leads to an activated particle. This scenario is considered an activation via collection only when the critical radius of the created particle is bigger than the initial wet radius of the colliding activated aerosol.

The first scenario is straightforward, but in my opinion the second and the third scenario deserve more explanation why they are considered an activation via collection. Indeed, from the point of view of the colliding inactivated aerosol particle, it can be said that the activated aerosol particle with which it collided got annihilated and in turn the aerosol in question got activated after some additional diffusional growth.

However, from the point of view of the colliding activated particle it can be said that the activated aerosol particle scavenged the inactivated particle and thanks to diffusion of water vapor remained activated (i.e. the activated particle remains activated and the inactivated particle is annihilated).

In general, counting and labeling activation events that happen due to collision is more difficult because there are two initial particles and one resulting activated aerosol particle, whereas the traditional Köhler theory activation results in one-to-one correspondence between an activated aerosol particle and the created cloud droplet. Could you clarify which colliding particles are considered activated and which annihilated?

Could you consider adding some sketch or maybe a plot using Köhler curves that exemplifies how the considered scenarios work? It could help to clarify which particles are labeled as annihilated, activated and inactivated and to showcase the typical dry and wet radius sizes of the particles colliding in all scenarios.

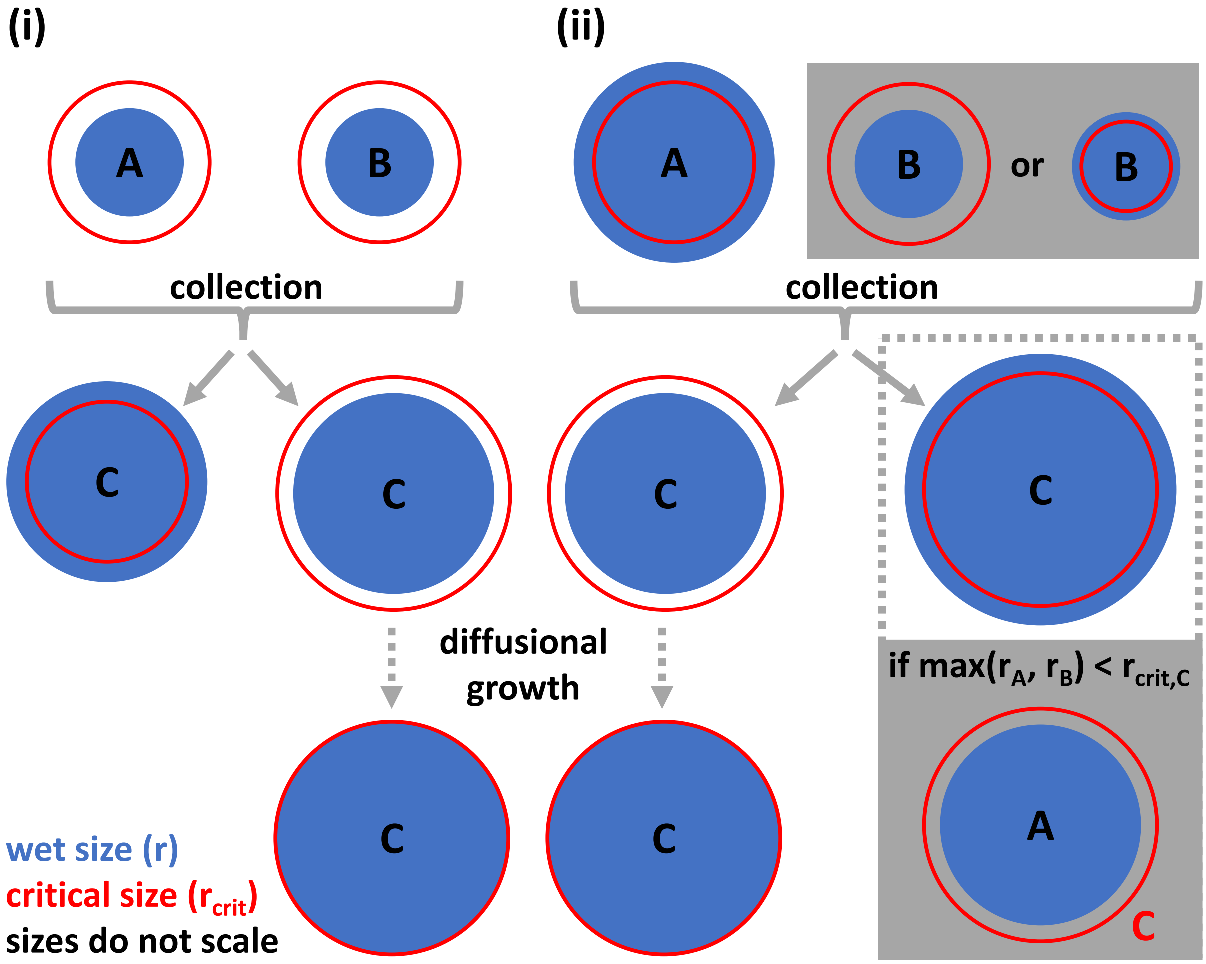
The identification of collectional mass growth is based on the comparison of the collectional mass growth $\Delta m|_{coll}$ to the diffusional $\Delta m|_{diff}$. The scenarios exemplify how this $\Delta m|_{coll}$ is able to exceed $\Delta m|_{diff}$. Accordingly, they are scenarios defined, they result from the collections I observed. And indeed, the first scenario is straight forward, but the other scenario might also lead to $\Delta m|_{coll} > \Delta m|_{diff}$, and need to be considered. I added a sketch to the manuscript (Fig. 3) which illustrates each scenario. The sketch displays the critical radius (red) as well as the wet radius (blue) of each particle during the process of collectional activation. A more in depth discussion of the relevant processes have been added to the text (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."



Specific comments

• **line 26**: As discussed in the Summary when referring to the work by Nenes et al. 2001, it is not necessary for a cloud droplet to become formally activated (i.e. reach its critical radius as defined by the Köhler theory) in order to grow in the cloudy environment and behave similar to the formally activated droplets. Could you consider adding such comment also in the introduction?

Yes (line 25 - 27): "Due to their large size, however, these particles may behave like regular cloud droplets inside the environment of a cloud although they are not formally activated (Nenes et al., 2001). Accordingly, Köhler activation theory is usually considered a weak concept for these particles."

• **line 32**: I think the question this article addresses is about "limits of traditional Köhler

activation theory". As discussed in the Summary and in the referred work of Chuang et al. 1997 and Nenes et al. 2001, the Köhler theory can be used to calculate the equilibrium saturation for big aerosol particles. The problem is that the big aerosol particles will not reach their equilibrium in the necessary time and therefore will not become formally activated.

You are perfectly right. I added the word "activation" to clarify this (line 34).

• **line 103**: Does it mean that the weighting factors for all super-droplets are constant? Does it affect the representation of collisions (compared to the tests presented in Unterstrasser et al. 2016)?

Initially, the weighting factors are the same, which might impede collections in a zero-dimensional setup as tested in Unterstrasser et al. (2017). Accordingly, the results should be considered as a lower estimate of the impact of collectional activation. However, as super-droplets experience collections, their weighting factor reduces resulting in a wide range of different weighting factors during the simulation. As discussed in Unterstrasser et al. (2017), this might facilitate collisions if more than one grid box is simulated, i.e., if super-droplets are allowed to interact with another ensemble of droplets when they move from one grid box to the next. The following addition has been made (line 314 - 315): "Additionally, the collection algorithm itself might underestimate collisions due to the initial distribution of weighting factors (Unterstrasser et al., 2017), and the determined influence of collectional activation should be considered as a lower estimate."

• **Figure 4**: I think the panels should be bigger (at least as big as those in Fig. 3). What is causing the spikes for maximum diffusion radius for the simulation with the lowest aerosol concentration? For convenience, would you consider adding a panel that shows the diffusional activation rate calculated basing on the simulations discussed here?

The size of the panels has been increased (Fig. 5). A panel of the diffusional activation has been added (Fig. 5 d), which caused some subsequent changes in the text (line 198 - 200).

Thank you for the hint regarding the spikes in the 100 cm^{-3} simulation. They result from the recirculation of large particles (see Naumann and Seifert, 2016, doi: 10.1002/2016MS000631), which have grown by collection inside the cloud, then detrained from the cloud, evaporated smaller than their critical radius outside the cloud (i.e., deactivated), entrained into the cloud again, where they grew larger than the critical radius by diffusion (i.e., activated by diffusion). Since the algorithm for distinguishing between diffusional and collectional activation only considered the growth between deactivation and activation, they have been spuriously considered as diffusional activations. In total, only 2 x 10^{-4} % of all diffusional activation have been affected by this process. I was able to remove these false diffusional activations from the analysis of the 100 cm^{-3} simulation. No influence of recirculations has been found for simulations with a

higher aerosol concentration. The new profile for the 100 cm^{-3} simulation has been added to Fig. 5. The conclusions did not change.

• **Figure 7a and lines 207-214**: Figure 7a is difficult for me to read and understand. First, the lines are plotted on top of each other making it difficult to see the behavior of each simulation. Second, the description of what is on the axes and what is actually plotted is unclear to me. For example, in the simulation with 4000 aerosols in cm^{-3} for dry radius of 0.1 µm there are 0.2 collisions with inactivated aerosol particles and 0.8 collisions with activated aerosol particles to activate the aerosol particle. In the same time in the description it is stated that only one collision is needed to cause activation and that the collision occurs between an activated and inactivated particle. Could you clarify, or maybe provide some example? Third, are all aerosol particles counted twice in this plot? – Once as the aerosol particle that is going to be activated (i.e. the location on the x-axis) and once as colliding particles (i.e. the different lines shown on the plot)?

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

• **line 258-259**: "collectional activation affects predominantly particles that have been entrained above cloud base, i.e., activates aerosols that have not been able to activate by diffusion at cloud base (...)" Does this sentence mean that the aerosols in question were not activated at cloud base because they were never at the cloud base? If yes, then I think saying that those aerosols have not been able to activate at cloud base is misleading, because they were never there.

You are right. The sentence has been clarified to: "Moreover, collectional activation affects predominantly particles that have been entrained above cloud base, i.e., above the region of the cloud where the highest supersaturations occur. Accordingly, these particles experience systematically lower supersaturations which prevents diffusional activation." (line 280 - 282)

• **line 353**: Could you clarify what values of dissipation rate were used for the collision efficiency from the Wang and Grabowski 2009 paper? The efficiencies in this paper are provided for two dissipation rates (either 100 cm2/s3 or 400 cm2/s3). Was the closer one chosen? Or was a constant dissipation rate assumed when choosing the collision efficiency?

The kinetic energy has been determined in the sub-grid scale model of the LES and the efficiencies of Wang and Grabowski (2009) have been interpolated to that value (using the given data for 100 cm^2 s^{-3}, 400 cm^2 s^{-3}, as well as unity for a zero dissipation rate). This has been clarified to: "These turbulence effects are steered by the kinetic energy dissipation rate ϵ calculated in the LES subgrid-scale model (Riechelmann et al., 2012). The

parameterizations by Ayala et al. (2008) are a direct function of ϵ, while the tabulated values of the enhancement factor for the collision efficiency by Wang and Grabowski (2009) are interpolated to the present value of ϵ." (line 384 - 386)

• **line 356**: Would you consider Brownian motion of aerosol particles as another possibility for activation due to collisions? Would a collision kernel representing both Brownian motion of aerosol particles and turbulence effects be an interesting extension of this study?

Indeed, a collision kernel with Brownian motions and turbulence would be an interesting extension of this study. Especially for very small collected particles, the consideration of additional processes affecting the collection process might result into a larger fraction of collected particles (e.g., Ardon-Dryer et al., 2015, doi: 10.5194/acp-15-9159-2015). However, I would expect that Brownian motions would rather have no impact on collisional activation by facilitating the collection of aerosols with a negligible amount of liquid water but a comparably large fraction of aerosol mass. This would result in a faster increase of the critical radius than the wet radius and therefore inhibit collisional activation (as discussed in Section 2 of the manuscript). I added a short discussion to Section 6 (line 309 - 312): "Moreover, the collection kernel might not incorporate all processes relevant for collections among aerosols and droplets. For instance, Brownian diffusion might increase the collection of smaller particles (e.g., Ardon-Dryer et al., 2015) but might not lead to collectional activation since it will add predominantly aerosol mass and only a small amount of water (cf. Section 2)"

Technical corrections

- **line 23 and 39**: I would not use the word *even* when describing opposite behavior(?).

  Ok. The word "even" is not necessary there.

- **line 195**: When saying activation you mean collectional activation? Maybe it should be explicitly stated?

  Done.

- **line 326**: I think that the paper by Shima et al. 2009 should be referred here again when introducing the "all-or-nothing" representation of collisions for the Lagrangian microphysics.

  Good point. Done.