

Interactive comment on "On the Limits of Köhler Activation Theory: How do Collision and Coalescence Affect the Activation of Aerosols?" by Fabian Hoffmann

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Received and published: 30 March 2017

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 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

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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.

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.

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?
- **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

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the big aerosol particles will not reach their equilibrium in the necessary time and therefore will not become formally activated.

- **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)?
- **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?
- 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 μ m 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)?
- **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.

- 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?
- **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?

Technical corrections

- line 23 and 39: I would not use the word *even* when describing opposite behavior(?).
- **line 195**: When saying activation you mean collectional activation? Maybe it should be explicitly stated?
- **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.

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