

Review of manuscript number acp-2017-499 by Hande and Hoose 2017

General Comments:

Hande and Hoose present a study in which simulations of different cloud types in high resolution (LES simulations) with a variety of ice nucleation parameterizations representing different heterogeneous freezing modes and homogeneous freezing are used to elucidate the contribution of each freezing mode. This topic is of interest to the readers of ACP and the paper is generally well written. I would recommend the paper for publication in ACP after the following comments have been addressed.

I think the authors could do a better job of explaining the nuances of some observed aspects of their simulations. They are not over a page limit, and I think the paper can benefit from more explanations.

As it stands now, even though well written, it sounds more like a report with a few places in the manuscript where they apply an analysis of what the results mean. For example the authors make reference to steady state being achieved but without specifying with respect to which parameter?

Also, when there are effects of changing aerosol concentration, these are rightfully stated, but I think the authors could go one step further and explain why this would be expected to have influences (+ve or -ve biases) on the precip amount or total water content. On one hand, I understand that this is assumed knowledge, but on the other it would make the paper more round and complete.

Specific comments below:

Page 1

Line 1: maybe in parenthesis specify the modes contact, immersion and deposition so that it is clear that no other mode is being considered for instance evaporation freezing/PCF in presence of an active site etc.

Line 6: thermodynamical should read thermodynamic

Line 8: "little" should be replaced with "only a small"

Lines 11-12: here can you draw a connection between large aerosol variation and the mode of ice nucleation that would be dominant?

Line 15: I would say increasingly probable for temperatures lower than -35 C

Line 18: delete "in order"

Line 21: water vapour deposits directly to ice – you don't specify ice, you just mention deposition of water vapour which doesn't include a phase transition.

Line 25: "have long" should read "has long"

Page 2

Line 4: if talking about recent reviews then *Kanji et al.* [2017] is the most recent one that discusses the same topics mentioned in this paragraph.

Line 13-20 can be one paragraph. No need to have two paragraphs

Line 33 “ delete “to this”

Line 34, doesn't the study by [*Spichtinger and Cziczo, 2010*] deserve mention here, since they looked into the competition between homogeneous freezing and heterogeneous ice nucleation (deposition nucleation)

Page 3

Line 5: thermodynamic conditions

Line 10: supersaturation is one word and everywhere else in the paper

Line 15-18 should be in the introduction – doesn't fit in the model description section

Line 30-31: dust size distribution at Jungfraujoch from 0.1 – 100 μm , I find that a little hard to believe. Do you have better reference for that? There are a lot of papers published on aerosol and INP properties at the Jungfraujoch that may give you a representative size distribution. I didn't think 100 μm dust particles would make it from North Africa to central Switzerland, or at least not in any significant proportion.

Line 32-33: If dust is not removed by precipitation in the model, which should be one of the key removal processes – how does this affect ice nucleation in further time steps, i.e. dust that was at lower altitudes that did not activate (because T is not low enough) but also did not get removed by precipitation could then be available to be lofted or for convective uplift for next time steps? Wouldn't this positively bias the role of dust as INP in the model runs?

Page 4

Line 8-11. The authors state that contact freezing of rain drops is not considered but in the every same sentence say that rain drops collect many particles through collision-coalescence processes. But why should this be a reason for contact freezing not to be considered. Perhaps the collision processes could lead to freezing rather than coalescence? Also, in a recent study it was shown that a deliquesced surface of an aerosol particle colliding with a droplet can also induce contact freezing [*Niehaus and Cantrell, 2015*], therefore potentially enhancing the contribution of contact freezing

Line 14-15: this is 50% by number I assume? Please specify

Line 17: Depletion of immersed aerosols is not taken in to account in these simulations? Does this mean that aerosols that get immersed in cloud droplets are still available for CCN activation in further time steps? How does this impact the results obtained? Given that 50% of your aerosol (by number I assume) is immersed, how would this influence the outcome of the simulations if they are assumed to be available for CCN activation/immersion freezing in subsequent time steps?

Line 19: Replace “so” with “therefore” so is colloquial

Line 30: Has CAPE been defined before?

Line 9: x-hour simulation? Perhaps I misunderstood something?

Page 5

Line 28-30. I am confused by the wording and reasoning in this sentence. The smaller size and droplet number inhibit INP formation? Perhaps the authors meant “reduce the effectiveness of contact freezing” because of fewer collisions? More INPs would be expected to be active at colder temperatures, but it is fathomable that if collisions do not occur in the first place, then the role of contact freezing would be limited. Perhaps clarify, but the wording “formation of INP” sounds incorrect to me.

Page 6

Line 1-3: is there a chance to discuss here or comment on whether homogeneous freezing was suppressed or its initiation was suppressed because of the formation of ice heterogeneously and potentially depleting water vapour. This is hard to deduce from the way the figures are presented. One could discuss size of ice crystals here as well. Perhaps this is lower in the manuscript under temporal distribution..

Line 5-10: So you have droplets available for collisions with INPs at RH below 80% how long do they survive and how concentrated are they if they are surviving as droplets at such low RH?

Line 17-20: I don't get the reasoning here. The RH being high in the mid troposphere should warrant deposition and homogeneous freezing taking place? I would imagine high is a relative term here. 60-70% is high compared to what? So 60-70% RHw is low for homogeneous freezing to take place, so not the result being referred to here is not surprising. Based on lab studies, I am not surprised that deposition nucleation is not contributing at such low RHs either. Generally RHw 70% at about 223 K is usually required for deposition nucleation (or pore condensation and freezing).

Page 7

Line 16-17: it is not clear to me how the authors come to this conclusion about the precipitation. Is this in reference to the total precipitation over the course of the simulation for both cases being compared, or comparing precipitation at a given time stamp? Or the precipitation at the end of the simulation?

Page 8

Line 1-10: I like the table with the relative contributions of each freezing mode. I assume this is the total contribution over the course of the simulation. However, it would be nice to see more interesting versions, for example you could consider just quantifying the relative

contributions up to the point of precipitation initiation – if you just consider the simulation until precipitation starts, could you say something about how much each mode contributes to initiation of precipitation?

Page 9

Line 15: Reference format?

Line 15-18: I assume you are talking about different aerosol species. In this work you have tested dust parameterizations, but one could easily predict how this would change if an organic aerosol parameterization for ice nucleation or soil dust one was used? This should simply make the ice nucleation more or less effective (i.e. lower N_s in the case of organics compared to dust for a set T for example). Couldn't a quick statement from a simulation be made about that in this paper. At least the contribution from soil dust would have been interesting to see here.

Line 23: homogeneous freezing accounts for 6% of INP concentrations? Do you mean ice crystals, I didn't think homogeneous freezing was associated with INPs?

Line 26: steady state being referred to in terms of which quantities?

Page 10

Line 3: "on" the dominant ice nucleation mode?

Figures:

Are Figures 1-6 for high or low dust concentrations?

Figure 1: nm or μm is more intuitive. I do acknowledge you want to stick to SI units.

Figure 3. Homogeneous freezing sets in at about 8km but you have quite a number of liquid droplets above this altitude, are these meant to be conc. solution droplets because your RH_w isn't that high? Can you please clarify the existence of liquid droplets here? Referring to the top 2 panels in the right column.

Figure 4: Similar to Figure 3: should there be any droplets when homogeneous freezing has kicked in? Also for the mean cloud drop radius a small comment, more ticks on the scale could be helpful.

Figure 5: In all these panels, is it possible to zoom into the orographic cloud more and reduce white space .. i.e. there is nothing to show for the altitudes above 3 km - it would give a more clear picture of the orographic cloud. The parameterization limit lines are shown in the Figures 3 and 4, so the reader can be referred to the same limits in Figure 5.

Figure 6: Same comment as Figure 5.

Figure 10. Is this figure referred to in the text? I don't think so. Also, why the differences in total water content for the high dust case and lower precip for the lower dust case? perhaps explain this a bit more in the text.

References:

- Kanji, Z. A., L. A. Ladino, H. Wex, Y. Boose, M. Burkert-Kohn, D. J. Cziczo, and M. Krämer (2017), Overview of Ice Nucleating Particles, *Meteorological Monographs*, 58, 1.1-1.33, doi:10.1175/AMSMONOGRAPHS-D-16-0006.1.
- Niehaus, J., and W. Cantrell (2015), Contact Freezing of Water by Salts, *J. Phys. Chem. Lett.*, 6(17), 3490-3495, doi:10.1021/acs.jpcllett.5b01531.
- Spichtinger, P., and D. J. Cziczo (2010), Impact of heterogeneous ice nuclei on homogeneous freezing events in cirrus clouds, *J. Geophys. Res.-Atmos.*, 115, D14208, doi:10.1029/2009jd012168.