Interactive comment on "Comparing contact and immersion freezing from continuous flow diffusion chambers" *by* Baban Nagare et al.

Response to Anonymous Referee #3

We would like to thank the reviewer for careful reading the manuscript and the suggestions of improvement of readability. The responses to the comments and questions are given below in italic.

My major comment is that main conclusions of the paper are not visible. Some revision is needed to enhance the readability, and also it is necessary to clarify what are the major conclusions of the paper. It is also not clear why this study is important, and what the atmospheric implications are. Some more discussion is needed to understand why AgI, ATD and Kaolinite particles were used, why natural dust or soil dust particles were not chosen as these are more atmospherically relevant. This is nice study, overall contact freezing is not well understood, but main message is buried. Below some comments may help to revise this paper further.

It is important to know whether contact freezing is more efficient than immersion freezing for parameterization in atmospheric models and for the microphysical understanding of the different heterogeneous ice nucleation processes. We emphasize this now more in the abstract by adding on line 4:

"To date, direct comparisons of contact and immersion freezing with the same INP, for similar residence times and concentrations are lacking."

We preferred to first study INPs that have been investigated before. This is the case for AgI, ATD, and kaolinite. Moreover, these samples represent different types of ice nuclei. ATD is a mixture of different minerals, while AgI and kaolinite contain one main component. AgI induces ice nucleation at a rather high temperature and shows a close lattice match with ice, while kaolinite does not have a close lattice match and induces freezing at much lower temperature. In future this study could be extended to investigate natural mineral dust or soil dust samples.

-What is the typical size of supercooled droplets observed in mixed phase clouds? How often 80 um droplets are observed. Atmospheric relevance of droplet size should be discussed.

For heterogeneous nucleation, droplet size is not important. The relevant quantity is the surface of the INP, which is in our experiments in the atmospherically relevant range.

-Following two sentences (i and ii) needs to be elaborated. Bulk liquid water properties are different from individual water droplet properties. Please define what you mean by sprinkling. Do particles were size-selected, how many particles were used, what is the temperature of the liquid water, do water is pure or distilled or regular lab supply grade, how long this experiment was performed, do all particles sediment, and how this observation was made (visual observation, microscope).

(i) "When we sprinkled ATD on a water surface, most particles immediately immersed and sank to the bottom. This suggests that when ATD particles collide with water droplets, the particles become immediately immersed such that in immersion freezing and contact freezing experiments the immersion mode is probed."

(ii) "When we sprinkled kaolinite powder on water, we observed that some particles floated on the surface while others became totally immersed and sank to the bottom."

We give the requested information in the revised manuscript in the new Section 2.3. These were very simple experiments to confirm the wetting behavior predicted by evaluating the contact angles between water and the particles. There should be no difference between a droplet surface and a bulk water surface as long as the Kelvin effect is not important, which is the case for droplets larger 1 µm, i.e. all cloud droplets.

We add to the observations for kaolinite the timescale:"… some floated on the surface for hours while others became totally immersed and sank to the bottom within seconds."

-It is mentioned that "A particle on the surface can induce ice nucleation in the immersion mode with the part immersed in water or in contact mode with the part exposed to air." How this can be assumed, what is the basis for this?

We refer here to the contact freezing process occurring when a particle adheres to the surface of a droplet. In the ACPD version of the manuscript we referred to this process as contact freezing inside-out. In the revised manuscript we change the terminology to "adhesion freezing" because naming it "contact freezing inside-out" was criticized by reviewers 2. If the part of the particle that is exposed to the surface is less efficient at *nucleating ice than the part of the particle immersed in the droplet, the freezing efficiency should still equal approximately the one observed for cases when the particle is totally immersed in water.*

-Section 5.6: It is not clear what results are discussed. This section looks like reading a literature review. There is only one sentence (The immersion and contact freezing studies compiled in Fig. 6 suggest that contact freezing is more efficient than immersion freezing with an onset temperature that is about 3 K higher), which describes the results, but there is no discussion. I suggest use present results to discuss the figure 6, but not previous results (as they have different instrument platform to study Kaolinite properties). For example XRD analysis of Kaolinite particles differ from group to group because of the XRD instrument sensitivity issues, and also impurities within the Kaolinite samples. Note that Kaolinite from different vendors have different properties, also shown by Wex et al (http://www.atmos-chem-phys.net/14/5529/2014/acp-14-5529-2014.pdf) who shown ice nucleating properties are sensitive to the particles procured from different vendors.

We are aware of the different qualities of kaolinite depending on the vendor. For our experiment we used Fluka kaolinite from Sigma Aldrich (K-SA). We therefore compare to studies, which also used Fluka kaolinite. This is the case for Wex et al. (2014) and Tobo et al. (2012). This comparison is therefore justified. It is well known that Fluka kaolinite is not pure. This is why the composition determined by XRD is important. The information given in this section is relevant for the interpretation of the results. Reviewer 1 even asked us to "place our results more clearly in the context of previous work".

-Section 5.6: Second paragraph. How this is applicable to the present study. This material is not relevant, if yes please discuss how. As mentioned above this reads like a literature review.

We need information about the morphology and surfaces of kaolinite to discuss whether kaolinite particles adhere to the surface of the droplet or whether they are immersed. A discussion of previous literature is needed. Reviewer 1 even asked for a more profound discussion of kaolinite and suggested inclusion of more previous work.

-Please see Section 5.5 too. Discuss the present results. There is lot of discussion on previous studies, but how they are related to this study. It is not clear why these studies are discussed. I suggest move this material to Intro section to increase the readability.

In this section the results for ATD are discussed and put in context with previous studies on ATD. Such a discussion is necessary.

-Last three sentences from Conclusion section (page 14, line 8-11). Do authors performed any experiments to conclude this, or these are the conclusions from previous studies. If later then I suggest move this to intro section.

These are the conclusions of the present study. We make this clearer in the revised manuscript by writing: "Our experiments and calculations…"

-Can majority of Section 5.2 (except page 17, line 17-23) and Section 5.3 be moved to Intro section? They do not discuss any results.

We moved Section 5.2 to the introduction and Section 5.3 to an appendix.

-It may be a good idea to combine section 4 and 5. Section 5, for dust particles, has lot of discussion concerning previous studies and may help to increase the readability.

We prefer to keep the results and the discussion of the results apart.