Responses to reviewer #2

We thank the reviewer for the thoughtful comments. Our responses are given in italic below.

In this study, Kaufmann et al. use a DSC method to examine the nucleating behavior of a wide range of both natural dusts and reference minerals. It is found that the variability in freezing behavior for natural dusts is relatively small. The consequences of this finding is that for model studies, it may be sufficient to represent natural dusts with a single parameterisation, at least in the temperature ranges examined during this study. The difference in variability in the freezing behavior between natural dust samples and reference materials, which was found to be greater in the case of the latter, is also a key finding, is sure to be of interest to researchers in this area.

My main comments/questions on the paper surround the experimental procedure, and how the data is interpreted. Following clarification of these points, I would recommend the paper for publication in ACP.

Comments and Suggestions:

- It is not immediately obvious why the data from DSC measurements cannot be normalized to nucleation rates or ice active site densities. I can envisage some difficulties in doing this, but a statement on why nucleation rates or ice active site densities are not calculated would be of value to the reader.
  In DSC measurements the heat flow due to freezing is measured. The presented data was for emulsion measurements where a high number of droplets with different sizes and containing different numbers of particles freeze. Nucleation rates are not a direct result of the experiments. To derive nucleation rates and ice active site densities, a complete modeling of the DSC curves is needed as was done in Marcolli et al. (2007) for Arizona test dust. Such a modeling is a major task and was therefore not intended in this study, which focuses on the mineralogical composition. It could be the subject of a follow-up study.
- At this point in time, there are two other pertinent papers which are in peer review in ACPD (Harrison et al., 2016; Peckhaus et al., 2016), which are not considered here, but I would highlight that they are very relevant. For the final ACP version of this paper, if these related papers are accepted prior to this one, I would certainly include discussion of them.
  Thank you for pointing them out. We discuss the aspects of these paper that are relevant for our study in the ACP version:
  In Sect. 5.2.2: "Harrison et al. (2016) have recently performed freezing experiments with microliter droplets of aqueous suspensions of a ground sanidine sample. The observed freezing temperatures indicated a similarly high ice nucleation activity for this sanidine sample as for microclines. The mineralogical composition was investigated by Rietveld refinement of powder XRD patterns confirming sanidine as the dominant feldspar phase, however, without specifying minor components. Considering the high number of particles present in the microliter droplets, the results are not directly applicable to the freezing of emulsion droplets containing only one or a few particles. Nevertheless, we note a quite large variability in ice nucleation activity between sanidine samples, which does not seem to correlate with the mineralogical composition. Based on the emulsion experiments with DSC, we consider sanidine as ice

nucleation active but only at lower temperatures compared with microcline."
And also in Sect. 5.2.2: "Harrison et al. (2016) investigated three albite samples with very different ice nucleation activities. One of them showed similarly high freezing temperatures as the microcline samples but lost its activity over time while suspended in water. The other samples showed distinctly lower freezing temperatures than the microclines but slightly higher ones than the plagioclase samples that they also investigated."
In Sect. 6.4: "Peckhaus et al. (2016) investigated four milled feldspar minerals in freezing experiments. Bulk mineralogical composition determined by XRD revealed the three K-rich samples to consist mainly of microcline (76 – 80 %) with a minor (Na, Ca)-feldspar component (16 – 24 %). They used an environmental scanning electron microscope to record images of single particles and energy dispersive X-ray (EDX) to infer the mineralogical composition of the particles. The SEM images showed agglomerates consisting of several large particles with smaller particle fragments on their surface. The (Na,Ca)-feldspar sample exhibited large inter-particle variability in Na/Ca ratio. The K-rich feldspar particles contained varying amounts of sodium and also calcium. Only one K-rich feldspar contained some pure K-feldspar particles with no share of sodium. Interestingly, this sample showed the highest freezing temperatures."

- Throughout the paper, IN is used, instead of INPs. I would consider changing this as per (Vali et al., 2015)
  We use INP in the revised manuscript.

- P1L18: for clarity, I would add point out that the 2 um figure given here is from the number distribution.
  We reformulated: "For emulsion measurements, water droplets with a size distribution peaking at about 2µm,"

- P3L7-8 an L18-19: The references here don't all match with the statements made on how organic matter can influence ice nucleating activity, in particular, Baker 2005 and to a lesser extent maybe Hallar 2011; neither of these studies examined ice nucleation as far as I'm aware. Also, there are multiple more pertinent references here e.g. (Augustin-Bauditz et al., 2016; O'Sullivan et al., 2014; Tobo et al., 2014)
  We removed references that were not fitting and added the suggested references.

- P3L20: "important" is a very qualitative word- I suggest changing to something more concise.
  We replaced "important" by "abundant".

- P4 experimental setup: Very high concentrations of dusts are used during some experiments, up to 50% (!). The authors refer to these as suspensions (by 50 wt %, I envisage this is more of a slurry than a "suspension"), but no indication is given on their stability. Emersic et al. (2015) suggest that aggregation, and surface area occlusion in droplets of 1 wt% is an issue for droplet freezing experiments- could this be an issue for these experiments at much higher concentrations? A discussion on these points is warranted, perhaps in the experimental section.

50 wt% is indeed a very high concentrations and was only used in cases where the DSC signal was too low to be detectable at a lower concentration. We add a discussion of aggregation to Sect. 6.3 of the revised manuscript:
"Depending on size and suspension concentration, droplets of the investigated emulsions may be empty or contain one or a few particles. Empty droplets as well as droplets containing only ice nucleation inactive particles contribute to the homogeneous freezing signal in the DSC curves. Tables 2 and 5 list in the second column D_{p1}, the average diameter of a droplet with 1 particle inside for 2 wt% suspensions, indicating that smaller particles are empty and larger ones contain one or a few particles. Assuming that all particles are able to nucleate ice, the heterogeneously frozen water volume fraction, p_{het}, can be calculated and compared with the measured one, $p_{het,lab}$. The ice nucleation active particle fractions were calculated for all concentrations and are given in Tables 2 and 5 for the 2 wt% suspensions. They range from f_{act} = 0.025 – 0.32 (Table 2) for the natural dust samples excluding ATD and from f_{act} = 0.0004 – 0.64 for the reference minerals (Table 5). Ideally, the derived active particle fractions should be independent of suspension concentration. If particles aggregated in suspension, the active particle fraction would be underestimated because the effective number of empty droplets would be larger than the one determined from the size distribution of the dry aerosol. Stronger aggregation is expected at higher concentration leading to an increasing low bias with increasing suspension concentration. To elucidate whether such a tendency is present, the ratios of f_{act} of 0.5 wt% and 2 wt% suspensions, $f_{act}(0.5)/ f_{act}(2)$, and the ratio of f_{act} of 2 wt% and 10 wt% suspensions, $f_{act}(2)/ f_{act}(10)$, are also listed in Tables 2 and 5. For most natural dust samples the ratios are > 1, indicating some aggregation. The reference samples give a less clear picture. The ratios show quite a large scatter with values between 0.5 and 2 and a tendency to values > 1, indicating some aggregation. Pinti et al. (2012) have discussed the possibility of aggregation for clay minerals concluding that kaolinites show quite strong aggregation mainly at low pH, no aggregation is expected for montmorillonites while no clear information could be obtained for illites. Emersic et al. (2015) hypothesized a possible influence of coagulation to explain the discrepancy between wet-suspension- and dry-dispersion-derived ice nucleation efficiency of mineral particles using kaolinite, NX-illite and a K-feldspar as examples. They showed aggregation for kaolinite using dynamic light-scattering but did not present corresponding data for illite and the K-feldspar."

- P5L 8-17 and Appendix A2: I have missed it elsewhere, but it would be useful to know here how many separate emulsions were examined in the determination of the droplet size distributions, and the total number of droplets examined. Also, this info should be added to the caption of figure 5.
  We add this information in the revised manuscript on page 5.

- P5L20: were these wet or dry sieved?
  The dry samples were sieved. We add this information by adding the sentence:
  "Sieving was performed with the dry samples."

- P6, section 3: If I understand correctly here, the authors are using size distributions measured by SMPS/APS, but are then using this information to estimate the number of particles in suspension droplets. The particle size distributions will be different in the suspension than from the aerosol phase due to aggregation. Will this not lead to significant errors in the calculation of the number of dust particles per droplet, and hence f_{act}?

We discussed uncertainties of the fraction of active INPs in Appendix A and concluded that a major uncertainty is indeed due to the uncertainties related to the size distribution. In the revised manuscript we discuss also the effect of aggregation (see response above).

- P19 L14-30: The authors attempt to explain the freezing behaviors of dusts which did not entirely fit with their hypothesis that mineralogical composition is the dominant factor accounting for this. Again, it would seem to me that recent papers in open discussion (Harrison et al., 2016; Peckhaus et al., 2016) are particularly pertinent to the discussion here.

  We add a discussion of these two paper to the revised manuscript, see the response to the comment above.

  P19L16-18. Do the authors have data to substantiate that in solution, the milled reference samples do not aggregate also?
  We added this discussion to the revised manuscript. See above.

- P19L29-30: Perhaps the amount of organic matter could be expected to be small, but the OM content of the dusts was not investigated here. Even trace amounts of organic matter could affect the nucleating abilities of the dusts. Either the authors should further add to arguments that the amounts of OM are too small to affect the freezing behavior, or drop this last sentence.
  Indeed, little is known about the content of organic and biological matter and therefore we drop the sentence.

- P20L12: This relates to my first comment above again: it would be useful to state why the thermogram data cannot be transformed into a parameterization which could be implemented in models.
  See the response above.

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