Responses to Reviewer #1

We thank the reviewer for the careful reading of the manuscript and the suggestions for improvement. Our point by point responses are given below in italic.

Summary

The authors investigated the relationship between the immersion freezing behavior of diverse natural dusts from the ground as well as reference mineral samples and their mineralogical compositions (that are based on XRD). The immersion freezing measurements were conducted using a DSC, in which bulk powders were emulsified and homogenized in a mixture of mineral oil and lanolin. The authors evaluated the immersion freezing behavior in two metrics, f_{ice} (ice nucleation active particle fraction) and $T_{\mathrm{het,xx\%}}$ (freezing temperature, T, of given frozen fraction ranging from 10 to 50%), in the sub-zero temperatures above ~236.5 K.

The major finding of this work is that a majority of surface dust samples exhibit similar freezing behavior despite the difference/variation in mineralogy. In turn, the authors suggest the atmospheric dust freezing to be potentially represented in global models in a simple manner (P20 L11-12). Notable exceptions are the samples with microcline that are known as a highly efficient immersion freezing component of dust. Nonetheless, the authors imply that microcline-containing particles are not abundant in the atmosphere and, hence, may have overall small contributions to atmospheric ice nucleation (IN) and glaciation (e.g., P20 L22-25).

General comments

The authors conducted very careful and dedicated experimental works. The manuscript is well organized and carefully written to derive a delicate conclusion (i.e., P2 L1-4; P21 L22). The research topic is an important addition to ACP. The authors are knowledgeable in the subject and perceptive about the importance of mineralogy-resolved IN study as they clearly state the necessity of further investigations (e.g., analysis of airborne dust mineralogy and associated modeling simulation works, P20 L25-26; P21 L2-4). I support publication of this manuscript in ACP after the following comments are properly addressed.

Major comments

P22 L9-11: The authors suggest that there is only negligible amount of microcline in natural dusts, such that atmospheric IN triggered by microcline may be negligible. The authors did a great job in justifying and documenting their mineralogy results of bulk powder with manual rock interpretations (Sect. 5.2.1). That said, the authors report the XRD diffraction "accuracy" of ± 15% based on the comparison of two XRD analyses for the very same commercial standard material (i.e., ATD) in P10 L11 and Appendix B. Concerning given XRD accuracy, I feel that the statement in P22 L9-11 is a bit too aggressive. As microcline can be K-feldspar (and is not a rare feldspar), we can at least assume it is there in general. The authors may consider softening the tone.

Our conclusion for microcline was just based on the results from dust samples of this study. Very recently, a study by Boose et al. appeared in ACPD. They found microcline in several samples. We will cite this work in the ACP version and rephrase the text accordingly:

"Boose et al. (2016) found microcline present in one out of four investigated airborne dust samples originating from the Sahara and in three out of eight surface-collected dust samples. If microcline particles are indeed common in atmospheric dusts they could be relevant for cloud glaciation at temperatures above 260 K."

P11 L21-22: Regarding sanidine, the results reported in *Harrison et al.* (2016, ACPD) suggest no difference between sanidine and several microcline samples in terms of IN. If an atmospherically representative sanidine was IN active, the discussion regarding the microcline-scaling IN activity (e.g., P1 L30-31) would be misleading? More or less, I just wonder if it would change any of the authors' conclusions or not.

It does not change our conclusions because Harrison et al. performed their experiments with microliter

droplets containing many particles while in the emulsion droplets, there are only one or a few. We refer to this in the revised manuscript by adding the following text:
"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."

While the authors report the results of their DSC freezing experiments carried out with the homogenized samples (P4 L10-11), it seems their XRD composition measurements are based on the bulk powder samples without any pretreatment (P5 L24). The size distribution characterization (SMPS/APS as well as EM) may have been performed with the sieved-aerosolized samples (P5 L18-26).
The homogenizer was used to produce the emulsions, not to homogenize the dusts. The vigorous stirring of the homogenizer might reduce aggregation. The XRD measurements were performed with the sieved samples. Freezing experiments, size distribution characterization and XRD measurements have all been performed with the sieved samples.
Since the authors combine these three independent results afterwards for their data analyses and interpretations, the reviewer suggests the authors to clarify the followings:

- Sample homogenization may do more than just emulsification; e.g., promoting the particle breakup, altering the abundance of certain components, changing the size distribution and scratching the surface of particles? Any comments? As the authors might be aware, alternations in size and composition, especially for a composite material, are often inherently related (as they discuss some in Sect. 6.4; P19 L5-13).

  We acknowledge the uncertainty in the size distribution as one major uncertainty in the calculation of the fraction of active particles (Appendix A and especially section A1). We think that surface modifications due to scratching with the homogenizer might be important when applied to dry particles rather than particles mixed with oil and water.

  We discuss the influence of aggregation in the revised manuscript in Sect. 6.3 by adding the text:

  "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 aggregation in some cases. 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."

- Can the authors justify a consistency of the size distributions amongst these individual measurements, especially with respect to aerosolized particles vs. homogenized/emulsified particles? Otherwise, the assumption of the consistency should be clearly stated in text (e.g., P6 L2 and P14 L17-19). I do agree with the authors that sieving with a 32 µm grid helps represent the size distribution of airborne dusts.
  Possible inconsistencies are discussed in Appendix A. We extend this discussion in the revised version and write more about aggregation (see above).

- In terms of the particle size, my feeling is as follows; bulk powder > sieved-bulk > aerosolized (EM) > aerosolized (SMPS/APS) > homogenized. It seems that the authors use the aerosolized (SMPS/APS) data as a reference of particle size distributions in homogenized droplets. If so, wouldn't that means the authors may be overestimating V_p and f_{act} in eqn. 3 and eqn. 8, respectively (and underestimating n in eqn. 4)? Concerning aerosolized vs. homogenized, the f_{act} error may be even larger than the values given in Sect. A4? Currently, only EM vs. SMPS/APS is discussed in A1. If that is the case, the overall potential impact should be stated in text.
  We add a discussion of aggregation to the revised manuscript where we compare f_{act} obtained for different suspension concentrations, indicating a tendency of aggregation in the emulsion droplets (see above). Particles in the submicron size range are more frequently primary particles than aggregates. The size distribution from SMPS/APS peaks in the submicron region, therefore we think that primary particles dominate. In case of primary particles, the emulsification by the homogenizer should therefore not lead to a further breakup of particles.

Minor comments

P3 L31: For clarity, the authors may consider rephrasing "natural dust samples" to "surface dust samples"? The authors may consider modifying the title accordingly as well.
We prefer to keep "natural dust samples", because we think that with the applied pretreatment of only sieving, our surface-collected samples should represent well airborne samples.

P5 L18-20: How did the authors aerosolize the bulk powders? The method (incl. generator spec.) should be briefly described here.
The bulk powders were aerosolized in a Fluidized Bed Aerosol Generator (TSI Model 3400A). We add this information to the revised manuscript

P18 L12-14 & L26-28: What exactly the authors mean for "systematic errors"? I encourage the authors to extend the discussion in a bit more detail. The IN research community seems putting some efforts to tackle the issue regarding data diversity amongst many different techniques recently. The authors may at least cite proper papers.
We discriminate between random errors, which decrease by averaging repeated measurements and systematic errors, which remain, even if repeated measurements are averaged. Systematic errors need to be quantified by measuring a reference (calibration) sample with known measurement value or by comparing with a reference measurement performed with another instrument/technique. Unfortunately, in atmospheric sciences, such reference samples or reference techniques are usually not available and the systematic errors can only be guessed.

P20 L6-8: So what is the atmospheric implication of typical IN (that is, emulsion measurement results) vs. best IN (that is, based on bulk)? According to the Appendix A4 (P23 L30-31), using bulk may have some technical issues, correct? This point should be clarified in the main manuscript (e.g., either in Sect. 6.2.2 or Sect. 7).
Emulsion measurements give an information about the most probable freezing behavior/temperature. Bulk measurements give information about the "best" possible freezing behavior/temperature. Bulk temperatures are usually much higher than emulsion temperatures, but only few INPs are able to induce freezing at temperatures observed for the bulk measurements. To make this clearer, we add the following sentence to

section 6.2.2: "Comparison with freezing temperatures observed for emulsion samples show that experiments with suspensions containing a high number of particles do not represent the freezing behaviour of typical INPs in a sample."

P20 L10-12: These sentences seem speculative and seem not match with the focus of the current manuscript. Some parts are opinionated. I suggest rephrasing.
We deleted this sentence in the revised manuscript.

Specific & Technical comments
P1 L22: best particles/sites → best ice-nucleating particles/sites
Done.

P2 L12 and all "IN" hereafter: ice nuclei (IN) → ice-nucleating particles (INPs) according to *Vali et al.* (2015, ACP)?
We use INP in the revised manuscript.

P3 L7: *Augustin-Bauditz et al.* (2016, ACP doi:10.5194/acp-16-5531-2016) may be a good additional reference regarding the effect of biological materials on mineral dusts in immersion freezing behavior.
Thank you for pointing out this paper, we refer to it in the revised manuscript.

P3 L18: *Wang et al.* (2016, Nature Geosci. doi:10.1038/ngeo2705) may be a good ref to add for the composition transfer function from soil to airborne dust.
Thank you for pointing out this paper, we refer to it in the revised manuscript.

P3 L20: important → abundant or dominant?
We changed to "abundant"

P3 L26: define "large" quantitatively
We cite here the abstract of Atkinson et al. (2013). It was not more specific.

P3 L32: I disagree. The authors applied a number of mechanical processes. See my major comment. It seems heat and additional mixing may have been applied to a subset of samples (P22 L29)?
The only pretreatment of the mineral dust samples was sieving. P22 L29 refers to the mineral oil / lanolin mixture. This mixture does not include mineral dust particles. The mineral dust was added to the water. Subsequently, the oil and the aqueous phase were mixed together by vigorous stirring with a homogenizer leading to the emulsion.

P4 L6: → best available ice-nucleating particles/sites
Done.

P4 L8-9: I suggest defining the "lower average freezing temperature" here. The authors may consider moving P5 L5-6 to this part.
We reversed the order and now explain first bulk freezing measurements and then emulsion freezing experiments.

P5 L24: → …composition of the bulk powder samples was measured by XRD
We rephrased: "The mineralogical composition of the sieved natural dust samples and the milled reference samples was measured by X-ray diffraction (XRD).

P6 L8: Reference/explanation for 2.6 g/cm^3 is missing.
This value is taken from Möhler et al. (2006). We add this reference to the revised version.

P9 L10: → number of ice-nucleating particles
We changed to "INPs"

P9L14: The authors may explain the usefulness and implication of the D_{pl} parameter here.
We explain this parameter better in the revised manuscript.

P13 L5-6: Please clarify what the authors mean for "minor components". It seems quartz and muscovite are not that IN active according to the results given in Table 5. In general, kaolinite seems containing some K-feldspar (P13 L20-23), which may be responsible for their high IN as inferred in Table 5 as compared to other reference samples. The authors mean it as a minor component?

Indeed, the minor components can explain the ice-nucleation activity of the anorthite sample only partly. We therefore rephrase: "For the anorthite sample, in addition to plagioclase as the main component (59%), 12% quartz, 8% muscovite and 5% kaolinite were identified as most abundant minor components. These minor components can explain the ice nucleation activity reaching to a higher temperature compared with the other (Na,Ca)-feldspar samples only partly."

P19 L29-30: The word "should" is bothering. Any particular references?
Following reviewer #2, we deleted this sentence because it is speculative.

P21 L11: → comparable in size after the processing, such as sieving and aerosolization (the authors may consider making a similar statement in P14 L17-19 to clarify this point).
We add the following sentence on P14 L19: "We therefore consider the sieved ground-collected natural dust samples as comparable in size with airborne mineral dusts."

P22 L1: The influence of agglomeration alone on IN should be discussed in Sect. 6.4 with proper citation (e.g., *Emersic et al.*, 2016, ACP, and references therein). Otherwise, remove the agglomeration word.
We add a discussion of agglomeration at the beginning of Sect. 6.3:
"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 aggregation in some cases. 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."

P22 L11: analysis of dust samples → analysis of airborne dust samples
Done.

P22 L11: Would the analysis of ice residual particles may help (e.g., *Kupiszewski et al.*, 2015, AMT) as a future work?
EDX measurements on single particles may indeed help and are a good idea for future work.

P22 L16: largest → be more quantitative, put the uncertainty values with respect to f_{act}
"largest" is specified in the following text as an uncertainty by a factor 2.8 – 4.4.

P37 Table 5: Two different fonts are involved.
We correct this in the revised manuscript.

P38 Fig. 1: The x-axis should read "droplet diameter"?
Yes, indeed. Thank you for pointing this out.

P41 Caption: Oman and Qatar → Qatar and Oman
Corrected.

Appendix B: The source of the uncertainty may include the sample itself as well. For instance, ATD is a material composite, and the sample may not be completely homogeneous in terms of mineralogical distribution even within a same batch. The authors may consider briefly mentioning it.
This is a good point. We add the following sentence to Appendix B: "ATD is a material composite. There might be variations in exact composition between batches or even within a batch. This might be a reason for discrepancies in addition to the accuracy of the XRD evaluation."