Review of "Ice nucleation efficiency of natural dust samples in the immersion mode" by Kaufmann et al.

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 $T_{het,xx\%}$ (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.

P11 L21-22: Regarding sanidine, the results reported in *Harrison et al*. (2016, ACPD) suggests 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.

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). 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).

- 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.
- 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.

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.

P5 L18-20: How did the authors aerosolize the bulk powders? The method (incl. generator spec.) should be briefly described here.

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.

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).

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.

Specific & Technical comments

P1 L22: best particles/sites → best ice-nucleating particles/sites

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

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.

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.

P3 L20: important → abundant or dominant?

P3 L26: define "large" quantitatively

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)?

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

P4 L8-9: I suggest defining the "lower average freezing temperature" here. The authors may consider moving P5 L5-6 to this part.

P5 L24: → …composition of the bulk powder samples was measured by XRD

P6 L8: Reference/explanation for 2.6 g/cm^3 is missing.

P9 L10: → number of ice-nucleating particles

P9L14: The authors may explain the usefulness and implication of the D_pl parameter here.

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?
P19 L29-30: The word "should" is bothering. Any particular references?
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)
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.
P22 L11: analysis of dust samples → analysis of airborne dust samples
P22 L11: Would the analysis of ice residual particles may help (e.g., *Kupiszewski et al*., 2015, AMT) as a future work?
P22 L16: largest → be more quantitative, put the uncertainty values with respect to f_{act}
P37 Table 5: Two different fonts are involved.
P38 Fig. 1: The x-axis should read "droplet diameter"?
P41 Caption: Oman and Qatar → Qatar and Oman
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.