

We thank Reviewer 2 for his/her constructive comments. We reproduce reviewer's comments in *blue* and our responses in black.

*The manuscript I was asked to evaluate is the second part of the publication series dedicated to the experimental study of freezing behavior of several types of mineral dust particles immersed in the droplets of pure water and weak inorganic solutions. The motivation behind this work is not only the persisting need to understand the heterogeneous nucleation of ice on the molecular level, but also a demand to establish a model framework that would incorporate the original ice nucleating (IN) properties of aerosol particles, their chemical ageing under atmospheric conditions, and interaction with water and water vapor in tropospheric clouds. This complete paper series and the part 2 in particular is undoubtedly one of the most comprehensive experimental studies of various effects arising from the interaction of water, solute, and mineral surfaces that have been published recently. While I fully support the publication, I do have a few critical remarks that the authors might want to address while preparing the final version of the manuscript.*

*Abstract, lines 10-11: "We performed immersion freezing experiments and relate the reported contradictory behavior to the influence of milling, and to the aging time and conditions since milling." It is not clear to me what contradictory behavior is meant here. Is that some behavior contradictory to what has been published previously or any inconsistency within your own results? In any case, this is too much information for one sentence in an abstract. Please reformulate the abstract in a more concise way.*

We agree that the meaning of "contradictory behavior" is not clear. We therefore re-formulate: "We performed immersion freezing experiments and relate the observed variability in IN activity to the influence of milling, the aging time and to the exposure conditions since milling." (lines 12 – 13)

*The milling as a factor controlling ice nucleating properties of quartz is mentioned already in the second sentence of the abstract and then discussed throughout the text. It creates an impression that effect of milling on IN activity of quartz is the main objective of the study, and that all quartz samples have been milled by the authors. Only later it becomes clear that all samples have been milled by the manufacturer and thus any commercially available samples of crystalline quartz are called "milled". Perhaps this should be made clear at the beginning.*

This is a valid point. We add a sentence to the abstract to make clear that most quartz samples are obtained by milling (page 1, lines 8 – 9): "Since most studies so far reported IN activities of commercial quartz dusts that were milled already by the manufacturer, IN active samples prevailed."

*Lines 173-175. Why the freezing and the melting temperatures on the DSC thermograms are treated in a different way: as a leading edge for freezing peak, and peak maximum for melting? How narrow are melting peaks, would that make any difference if the melting temperature would be measured at the leading edge? Could you include a typical melting thermograms into the supporting material?*

We chose the heterogeneous freezing onset to characterize the freezing temperature because it is a very well defined parameter easily evaluated from the thermograms. Moreover, the maxima of heterogeneous freezing is not detectable in cases where the heterogeneous freezing peak is only a shoulder on the homogeneous freezing peak. Moreover, the maximum observed in the DSC thermograms is not the maximum of freezing events, but the maximum of heat flow. Therefore, we consider the freezing onset as the more characteristic

quantity for evaluating heterogeneous freezing. Furthermore, reporting the onset is consistent with previous studies on homogeneous ice nucleation such as Koop et al. (2000).

While melting of ice occurs at one temperature, ice within a freeze concentrated solution shows eutectic melting yielding a melting peak with no well-defined onset. Since ice remains in thermodynamic equilibrium with the freeze concentrated solution, it melts gradually, such that the melting signal constantly increases until it peaks when ice completely melts. Therefore, the peak of the melting curve can be used to evaluate the water activity of the solution.

We added melting thermograms for pure water and 5 wt%  $(\text{NH}_4)_2\text{SO}_4$  to the Supplementary Material (Figure S30).

*I don't quite understand the need for SMPS/APS measurements of aero-dispersed samples since they were not used for any DSC studies. The dispersion method always introduces some distortion into the initial particle size distribution in the powder, due to the intrinsic size selectivity. Large particles don't make it around the bends of the tubing, the small ones would be lost via diffusion deposition, there are electrostatic charging effects etc. Is there any physical reason for bimodal size distribution?*

The SMPS/APS measurements were carried out to obtain a rough estimate of the particle size distribution and to verify the size distribution information provided by the manufacturer. In addition, scanning electron microscopy was conducted on quartz samples. It ensured the presence of enough micrometer or smaller size particles to fill most water/solution droplets with at least one particle. We do not know the reason for the bimodal size distribution of the quartz sample. It might depend on the sample-handling procedure adopted by the manufacturer.

*What density and aerodynamic shape factor have been used for calculation of volume equivalent diameter of SMPS and APS data? I have a feeling that the SMPS/DMA data is an unnecessary piece of information that can be easily omitted from the manuscript.*

Density  $2.6 \text{ g cm}^{-3}$  and shape factor of 1 have been used for the calculation of volume equivalent diameter from SMPS and APS data. The coarseness of the sample needs to be roughly known since it influences the fraction of droplets in the emulsion that contain at least one dust particle.

*Finally, I am curious if the PZC-based discussion of surface charge role for the IN properties of quartz (section 4.5.4) makes much sense given the variability of the hydroxyl group number density across different crystalline faces of quartz (as you state yourself in lines 502 - 503). Since quartz does not have cleavage planes, the PZC measured with the conventional method is an effective value that does not reflect local anomalies. For ice nucleation, any surface patch larger than the size of a critical nucleus that would have the right surface charge would trigger the nucleation of ice. I, therefore, don't see any contradiction between your results and the work of Abdelmonem, (2017) and suggest that you remove the generalizing sentence (line 550) from the manuscript.*

This is a good point. We are also skeptical whether there is a general effect of surface charge or whether the effect of surface charge depends on the specific mineral surface. We therefore remove the generalizing sentence as suggested by the reviewer.

## References

Koop, T., Luo, B., Tsias, A., and Peter, T.: Water activity as the determinant for homogeneous ice nucleation in aqueous solutions, Nature, 406, 611-614, 2000.