

# ***Interactive comment on “Ice nucleation activity of silicates and aluminosilicates in pure water and aqueous solutions. Part 2 – Quartz and amorphous silica” by Anand Kumar et al.***

**Anonymous Referee #2**

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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

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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.

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.

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?

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?

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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.

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.

Abdelmonem, A., et al. (2017). Surface-Charge-Induced Orientation of Interfacial Water Suppresses Heterogeneous Ice Nucleation on  $\alpha$ -Alumina (0001), *Atmos. Chem. Phys.* 17(12): 7827.

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