

General Comment

The manuscript I was asked to evaluate is dedicated to the experimental study of immersion freezing behavior of several α -quartz samples in pure water droplets. The authors highlight the variability of the ice nucleation activity over different freshly milled quartz samples and investigate the short term as well as long term aging effects due to exposure to air and water. They further propose active site density parameterizations for several minerals and discuss the dominance of K-feldspar in the ice nucleation particle population in desert dusts. While I support the publication, I do have few remarks that the authors should address while preparing the final version of the manuscript.

Major comments:

Line 387 It should rather be “...parameterisation is representative of freshly milled quartz dust”. Airborne dust is eroded and gets exposed to air and even water. The authors should add a discussion about how representative is this for airborne dust. Proposing such parameterization - especially for ‘freshly milled’ quartz – is a bit of a stretch for now, given that we barely understand the surface due to limited work done and the high variability in its ice nucleation efficiency reported so far (including this work).

It should be clearly stated in the abstract that the study is on “freshly milled quartz” particles (also throughout the text) and should include a comment on the atmospheric relevance of such fresh surfaces when drawing comparisons with atmospheric dusts.

There are instances in Discussion section about cautiously using plagioclase and albite parameterization, yet this feature does not translate in the figures. In general, the devised parameterizations are a result of over-simplification and multiple assumptions which, though well-fitting, might not give a comprehensive view.

Given that members of K-feldspars show huge variations in their ice nucleation ability, with only microcline as a standout for most part (Harrison et al., 2016; Kaufmann et al., 2016; Welti et al., 2019), would the authors say that the K-feldspar parameterization proposed here is actually more representative for just microcline? In addition, following the information given in line 419-420, surprisingly recent data on natural dust mineralogy (Boose et al., 2016; Kaufmann et al., 2016) was not considered which may have painted a different picture.

Figure 2 & Methodology: It is unclear in the methodology whether the suspensions were tested just once or multiple times and why no uncertainties are shown in Fraction Frozen curves.

Figure 3: There are no error bars for freezing data of quartz samples undergone long-term aging in either water or exposed to air. Are these single suspension measurements? This should be made clear in the methodology section.

Minor comments

Line 129 Stone sample milling: average particle size range information of the samples could be helpful. I assume from the methodology that the stones were “hand-milled”. Given that quartz is such a hard mineral, one might still end up with powders consisting of large particles which contribute more to the mass fraction when preparing suspensions. I would highly encourage the authors to consider adding a discussion on size of mineral particles typically found at/near source regions versus the size range of particles probed in the experiments and its atmospheric relevance.

Lines 139-141 Glass vials as suspension storage containers: Glass is a source of Si and other ionic contamination. If the suspension reaches supersaturation in Si-concentration with respect to quartz

surface, it is likely that the quartz surface will start to grow over longer time scales and affect the ice nucleation ability of quartz (Kumar et al., 2019a). This process is governed by several factors eg. Si-concentration, particle surface exposed, growth rate, etc. (Baughman, 1991), which does not seem to lead to similar deteriorating IN efficiency in 3 quartz samples tested here in this study. It would be good if the authors could comment on this or add a discussion.

Line 248-286 Could it be that the indifferent IN ability in cases of Bombay Chalcedony and Smoky quartz during aging in water compared to Atkinson quartz be due to lower particle surface area exposed (low BET hence slower aging effect) for former 2 samples? Would have been interesting to see the long-term aging of the other chalcedony (Grape) which has similar BET value as Atkinson quartz.

Line 293 what exactly is meant by “dangling OH groups”? Highly hydroxylated quartz surfaces are dominated by vicinal and germinal silanols (Muster et al. (2001) and references therein) which then tend to create network of H-bonds with each other (Musso et al., 2011; 2012), therefore, not really free and “dangling” per se.

Technical comments:

Since the manuscript exclusively talks about ice nucleating ability of various minerals, I would suggest the term “material/materials” be replaced by “mineral/minerals” throughout the manuscript (eg. line 19, 171, 207 etc.)

Line 24-28 This is a long sentence. This can be split into 2 to better convey the meaning, eg. 2nd sentence could be "the ice nucleation particle population in desert dust aerosol is dominated by K-feldspars rather than quartz (or other minerals)."

Line 41 “type of aerosol particles” instead of “aerosol types”

Line 43 “Field observations of ice crystal residuals” instead of “Observations of aerosol at the centre of ice crystals”

Line 44 Cite the accepted ACP version of Eriksen Hammer et al. (2018)

Line 46-49 May consider splitting this into “Atmospheric mineral dusts are composed of several components. Clay is the major component of airborne mineral dust and is sufficiently small that its atmospheric lifetime is relatively long. Hence recent ice nucleation studies have focused on the clay group of minerals.”

Should be ‘INPs’ in place of ‘ice nucleants’ (line 51) and ‘ice nucleating material’ (line 71)

Line 67 ‘repeated’ instead of ‘repeat’

Line 70 remove ‘quite’

Line 74 “over time when suspended in water or exposed to air” would be better in place of ‘to time spent in water or air’

Line 104 TiO₂ is mentioned twice. Remove one of them

Line 139 this line gives the impression that the samples undergoing BET tests were used to make suspensions, which I believe is not the case. Consider removing “After BET analysis”

Line 176-177 delete ‘specific’ from line 176 and add it before “surface area” in line 177

Line 293-295 give references after ‘configuration’

Line 332 “It may be these microtextural differences that lead to the observed variability in ice-nucleating ability”

Line 377-379 Consider re-phrasing the lines to “However, we constrained the polynomial fits because the unconstrained fits poorly represented the data at the warmest and coldest ends.” for easy understanding.

Line 415 “its exceptionally high ice nucleation ability” in place of ‘it exhibiting exceptional behaviour’
Line 447 can also add Kumar et al. (2018); Kumar et al. (2019b) in the references in regards to aging of K-feldspar microcline

Line 471-472 Fig 8b shows parameterisation for desert dust by Niemand et al. (2012) in blue dashed lines and the K-feldspar parameterisation proposed in this study by solid red lines

Lines 506-510 Weathering in solutes has already been addressed by Whale et al. (2018) and 3 Parts series from Kumar et al. and should be addressed as part of this paper rather than referring to future work

Line 642, 666 and 673 - cite the accepted ACP version of these papers: Kumar et al. (2019a), Peckhaus et al. (2016), Pinti et al. (2012)

Figure 2 add suspension concentration in caption

Figure 5 Niedermeier et al. (2015) - correct author name in figure legend. Also, the curves are difficult to read, especially with orange-red tones, maybe use other dark colors. Size of symbols in legend needs to be increased for better readability

Figure 8 Check references to Boose et al 2016 (for 'a' and/or 'b') for data used in the figure. Symbol size in legend should be increased. In line 826 “...the natural mineral variability of freshly milled quartz highlighted...”

Line 795-796 equation formatting needs improvement

References

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