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Atmospheric Chemistry and Physics
Discussions

# *Interactive comment on* "Ice nucleation activity of silicates and aluminosilicates in pure water and aqueous solutions. Part 3 – Aluminosilicates" *by* Anand Kumar et al.

**Anonymous Referee #2**

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General comments

Kumar et al report on the ice nucleation activity of a range of aluminosilicate minerals and on the impact of various solute molecules on this activity. It is concluded that exchange of native cations with protons probably plays a critical role in the ice nucleating ability of aluminosilicates and that this explains why alkali salts inhibit ice nucleation by some aluminosiliciates. Similarly, it is concluded that that NH3 and NH4+ adsorb to the surface of feldspars rather than exchanging with cations and from there interact with water in a way that promotes ice nucleation, and that dissolution of feldspar surfaces and formation of an amorphous surface layer inhibits ice nucleation by feldspars.

The paper also summarises the more numerous findings of the entire 3 part series of papers, concluding that relevant factors for the IN activity of silicates and aluminosilicates more generally include adsorption and ion exchange with the mineral surfaces and changes to surface structures induced by dissolution and growth of mineral surfaces exposed to water. The authors are sensibly tentative about their conclusions and suggest that molecular dynamics simulations and surface science techniques may allow them to be tested more thoroughly.

The paper contains many results and reasonable interpretations. I am sure it will be of substantial interest to the audience of ACP, and possibly to those interested in heterogeneous nucleation more generally.

While I support publication, I note that the paper is long and complicated. While mostly well written, it is very hard to read and digest. It would certainly be beneficial if its bulk were reduced and the information it contains summarised more concisely. While I appreciate the difficulty in being more concise when dealing with so many different results I would strongly recommend that some effort be made to address this issue before publication. I also have some more specific scientific comments that the authors may wish to consider.

Specific comments

I have some thoughts on the interpretation of results concerning the feldspars. Feldspar mineralogy is complex compared to that of the other minerals investigated, is clearly of substantial relevance to the topic in question but is not, to my mind, dealt with thoroughly in this manuscript.

Characterisation of the feldspar samples is slightly lacking. What is the chemical composition of the three samples used? That the plagioclase sample is named 'andesine' implies a composition but no precise composition is given. The sanidine and microcline samples used could have any composition between 37% K and 100% K from what has been presented. Kauffman et al. (2016) does not contain this information either, as far as I can tell and also does not have an ‘andesine’ sample. I do not think it is reasonable to assume that the ice nucleating ability of the samples used are representative of their crystallographic structures. Harrison et al. (2016) and Whale et al. (2017) observe substantial variability in the ice nucleation effectiveness between crystallographically similar feldspars. Notably, Harrison et al. tested a sanidine sample that nucleated ice with similar effectiveness to most microclines and Whale et al. confirmed that it was sanidine using Raman spectroscopy. The argument is put forward here that the sorts of features that Whale et al. hypothesise cause ice nucleation in larger water droplets are not likely to be present in the smaller droplets used for this study. I am not sure about this. It is quite clear that different alkali feldspars have different structures across multiple scales, including the nanometer scale (Parsons et al., 2015). This is relevant because a) nanoscale features could easily be present on the ‘submicrometer’ particles used in this paper and b) nanoscale features are most probably on a scale similar to the critical ice nucleus.

It is a little hard to pick out but it appears that the authors think that the plagioclase and sanidine feldspar they have tested are less active than microcline because the sites present on these feldspars are dissolved away effectively immediately on contact with water. Feldspar dissolution is clearly a complex topic. I do not feel I am qualified to comment on the authors’ interpretation of this literature but I would note that it does not appear to be a well settled and understood subject from what I have read. It seems likely that feldspar-to-feldspar variability in dissolution rate is associated with more than just crystal structure and stoichiometry.

Still it is helpful that opposing hypotheses for the differing ice nucleation abilities of feldspars now exist, as it is probably straightforward to test which is more consistent with experiments. It would, for instance, be interesting to see if the LD2 sanidine of Harrison et al. and Whale et al. nucleates ice better than a micro-texturally pristine Eifel sanidine (which is what I expect has been tested in this study) in the smaller emulsion droplets.

The citation for the dissolution rates of feldspars is wrong. I think the authors have read the numbers used from graphs in ‘The mechanism of dissolution of the feldspars: Part I. Dissolution at conditions far from equilibrium’ but have cited part IV.

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