

Atmos. Chem. Phys. Discuss., author comment AC1 https://doi.org/10.5194/acp-2022-449-AC1, 2022 © Author(s) 2022. This work is distributed under the Creative Commons Attribution 4.0 License.

Reply on RC1

Nikou Hamzehpour et al.

Author comment on "The Urmia playa as a source of airborne dust and ice-nucleating particles – Part 2: Unraveling the relationship between soil dust composition and ice nucleation activity" by Nikou Hamzehpour et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-449-AC1, 2022

We thank Reviewer 1 for his/her thoughtful comments. We reproduce the reviewer's comments in black and our responses in blue. Line numbers refer to the revised manuscript.

Review of Hamzehpour et al.

This manuscript reports the outcomes of a range of experiments aimed at determining which components of a range of soil samples are responsible for their ice nucleation activity. The manuscript concludes, reasonably I think, that bio-organic matter is probably responsible for the ice nucleation activity of unaltered samples. It is found that removing carbonates and salts from the dust increases the ice nucleation activity of both the mineral and organic fractions of the samples. It is an interesting and thorough study and the paper is well written. I have a few comments which the authors may wish to consider, however I support publication.

Comments

The conclusions are based on quite small temperature shifts. I think it is important that there is some discussion of the temperature uncertainty of the two measurements reported from the DSC, as mentioned by Referee 2.

Reply: DSC has in fact a high precision. To clarify this, we add the following sentence to the manuscript on line 239:

"The average precision in T_{het} is ±0.2 K. Uncertainties in F_{het} are on average ±0.05, but may be much larger when heterogeneous freezing signals are weak or overlap (forming a shoulder) with the homogeneous freezing signal."

Page 19 line 438- Whale (2022) recently showed that $MgCl_2$ has a similar effect on ice nucleation by feldspar to monovalent cations.

Reply: Thanks for pointing this paper out. We added this reference to the text on line 445:

"These findings are supported by Whale et al. (2022) who recently showed that $MgCl_2$ has a similar effect on ice nucleation by feldspar as monovalent cations."

It seems relevant that the ice nucleation activities of both alkali feldspars (Harrison et al., 2016) and quartzes (Harrison et al., 2019) are known to vary a great deal. Something of why this is known for feldspars (Whale et al., 2017; Kiselev et al., 2021) however, as far as I am aware, there is no good explanation at all for the quite broad range of ice nucleation activities observed for quartz samples. It may turn out to be incorrect to treat all 'quartz' and all 'feldspar' as having the same responses to solution environment and the treatments laid out here. The 'reference' experiments are certainly interesting and worthwhile but there seems to me no guarantee at this point that the quartz and feldspar in the natural samples will behave similarly with regard to salt and pH environment.

Reply: The sensitivities of quartz and feldspars to ions and pH are indeed not the same, as we have discussed in Kumar et al. (2019a; 2019b). The decrease in IN activity of quartz at elevated pH and in the presence of ions can be traced back to the increased dissolution rate of quartz under basic conditions and with increasing ionic strength. During quartz dissolution, the best nucleation sites, which we assume to be high-energy sites that can be introduced through milling, are lost. Because we were aware of these sensitivities from our previous work, we wanted to quantify how large the effect is under the pH and ionic strength conditions covered by the LUP samples.

We discuss the sensitivities of feldspars and quartz to pH and ions in the manuscript from lines 440–451. We also refer to Kumar et al. (2019a), where the IN activity of quartz is discussed in detail.

For instance the work of Perkins et al. (2020) and Yun et al. (2020) showed unexplained enhancements of ice nucleation by feldspar in the presence of K^+ ions, which hasn't been observed in other studies, suggesting that there may be more than one mechanism responsible for ice nucleation by feldspars. In a similar vein, I would not be entirely shocked if it turned out hydrogen peroxide treatment damaged the ice nucleating ability of minerals. A recent study by Daily et al (2022) showed that heat treatment can slightly impair the ice nucleating ability of feldspar for instance, which probably wouldn't have been expected.

Reply: We are aware of Daily et al. (2022). This is indeed an important study helping to assign changes in IN activity after heat treatments to the components responsible for them. A similar study for H_2O_2 digestion would be welcome. Yet, our assignment of IN activity to organic matter is not only based on H_2O_2 digestion, but also on the significant correlation that we find between organic matter content and IN activity of the samples.

In our work, we dried the samples at 65° C (dry heating). In Daily et al. (2022) temperatures above 250°C were required to damage IN activity. In wet heating (e.g. while removing carbonates or organics), we kept temperature below 80°C, while in Daily et al. (2022) samples needed to be heated above 90°C to damage IN activity.

We will add the following text to the revised manuscript on line 213:

"Based on the study by Daily et al. (2022), samples needed to be heated above 90°C to damage IN activity of the minerals. Therefore, we do not expect a negative effect on the IN activity of the mineral components due to the heating required to remove organic matter, yet, we cannot exclude it."

I don't think these concerns undermine the study, but I do think it would be a good idea to make clear that the complexity of the problem is such that there are unknowns, and that the various chemical treatments employed could have unintended impacts on observed ice nucleation.

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