Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC1
https://doi.org/10.5194/acp-2021-806-RC1, 2021

Review of "The impact of (bio-)organic substances on the ice nucleation activity of the K-feldspar microcline in aqueous solutions" by Kristian Klumpp et al.

Anonymous Referee #1

Referee comment on "The impact of (bio-)organic substances on the ice nucleation activity of the K-feldspar microcline in aqueous solutions" by Kristian Klumpp et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-806-RC1, 2021

The paper reports on alteration of immersion freezing on microcline by several organic solutions. It is demonstrated that dissolution at the surface reduces the ice activity slowly and permanently, while complexation and adsorption reduce the ice activity rapidly and are reversible.
The paper is well written, experimental procedure and results are adequately rationalized. Suggestions for improvements are listed below. Once these are addressed the paper should be published in ACP.

Major comments:

- By using the onset freezing temperature T_{het} the effects on the few, most ice active microcline particles are highlighted. Onset freezing temperatures might not be the most relevant ice nucleation characteristic of a substances (as you mention on line 95f). Consider using the mean freezing temperature or the temperature of e.g., 10% frozen fraction as indicator for the bulk surface reaction to the treatments. Would using the mean freezing temperature for the analysis change the conclusions?
- There are two reasons why the reported frozen fractions F_{het} change through the treatments. The number of freezing droplets changes and the range of freezing temperatures broadens to below the homogeneous freezing temperature. The first reason is due to the removal of activity, and the second due to a reduction of activity to various degree that leads to a broadening of the freezing temperature spectrum. Instead of discussing active sites (e.g., line 20ff), I propose to plot some, selected freezing temperature spectrum (frozen fraction as function of temperature) to show and discuss the effect of treatments on the entire population of freezing droplets.
- Please discuss the implications of the observed sensitivity of microcline to solutes on microclines assumed great importance for immersion freezing in the atmosphere. Is it not justified, or will cloud droplet activation prior to immersion freezing reverse most of the organic alterations?

Specific comments:

- The uncertainty in F_{het} that is shown in Figs. 3,4,7,8 seems to underrepresent the observed variation between experiments. Looking at the data in the supplement, the frozen fraction in repetitions of an experiments differ often by 10-20%. Consider showing the range of frozen fraction for all datapoints as vertical lines instead of averages.
- Repeat the experiments marked with a black star in Figs. 3,4,7 to have at least one duplication.
- Section 2.2., line 174 ff. Explain why a readjustment of the procedure was necessary and how it affected the measurement.
- Line 184: clarify if every experiment was repeated or not. Other than stated here, the caption of Fig. 3 notes that some measurements were not repeated (marked with black stars).
- Line 191: justify why onset temperatures are chosen to characterize freezing, but the peak maximum for melting.
- Line 243: The oxalic acid onset temperature data seem in disagreement to the results for neutralized oxalic acid shown in Fig. 4, which show a strong effect. The effects of the treatment could be shown clearer in a temperature spectrum of the frozen fraction.
- Line 248: Instead of speculating about active sites, it could be stated that the treatment broadened the temperature range of droplet freezing and shifted it to lower temperatures. Both effects causing a larger fraction of droplets to freeze homogeneously.
- Line 282f: Looking at the shift of the peak heat flow in Fig. 5a), a decrease in frozen fraction seems often accompanied by a decrease in the median freezing temperature similar to the 2K reported in Peckhaus et al. 2016. Please check if the shift in median temperature does occur and is only not present in the onset temperature.
- Line 394ff: Can you propose a method to test your suggestion that reduced ice formation after heat or H_2O_2 treatment is due to removing ammonia? Heat sensitivity of samples from remote locations could be counterevidence to your suggestion.
- Fig. 1d): Explain how dissolution reduces the ice formation activity. Does it come to a reprecipitation and coating of the microcline particles with a secondary mineral after dissolution?

Technical comments:

- «Impact» shouldn’t be capitalized in the title.
- Line 124: redundant “(“ in citation.
- Line 383: redundant “;” in citations.
- Line 405: redundant “)” after citations.