**Review of “Ice nucleation properties of K-feldspar polymorphs and plagioclase feldspars“ by André Welti and co-authors for Atmospheric Chemistry and Physics**

General comments:

Welti et al. present a study about the immersion freezing behavior of a variety of different feldspar samples which builds on recent investigations by Augustin-Bauditz et al. (2014), Peckhaus et al. (2016), Harrison et al. (2016), and Whale et al. (2017). The samples were chosen carefully to provide a variety of crystal structures, chemical compositions, and ordering of the crystal lattice. They include five polymorphs of K-feldspar and four plagioclase feldspars. The immersion freezing experiments were performed with droplets containing single, size-selected particles and care was taken to minimize the amount of multiply-charged particles in the sample aerosol. Furthermore, the study includes X-ray fluorescence measurements giving information about bulk chemical composition and scanning electron microscopy images of particle morphology.

What differentiates the present paper from the earlier studies is the discussion of the effect of particle size and the degree of order in the crystal network on the ice nucleation efficiency of the samples. The authors’ conclusions concerning these factors are generally comprehensible and well substantiated by the presented results. However, in some cases, which are pointed out in the specific comments, I am missing a more precise explanation. The figures are mostly clear, but I would like to suggest some alternatives for presenting the data (see specific comments). Language-wise, the paper is concisely written but some minor adjustments would increase readability (see technical corrections).

Overall, the paper is interesting, understandable, and fits within the scope of Atmospheric Chemistry and Physics. I recommend publication after minor editing.

Specific comments:

1) Size dependence of ice nucleation behavior

The authors often refer to the “pronounced size dependence of ice nucleation activity” as if this is a rarely observed feature. Normally, the ice nucleation behavior scales with the surface area of the immersed particles, meaning that the efficiency increases with increasing particle size. The authors should clarify to what extent the size dependent ice nucleation behavior of their samples deviates from the standard. In my opinion, this is best done by calculating the ice nucleation active surface site density $n_s(T)$ for differently sized particles. In contrast to the chosen $T_{50}$ approach, this method would have the benefit of providing an overview over the whole investigated temperature range. I hence suggest to replace Fig. 5 with a multi-panel figure (like Fig. 3) showing $n_s(T)$ of the different particle sizes for all investigated samples (see, e.g., Fig. 5 in Hartmann et al., 2016).

Furthermore, the authors state that “microcline exhibited immersion freezing even for 50 nm particles” whereas for orthoclase “ice nucleation requires active sites present on 400-800 nm sized particles” and relate this observation directly to the effect of these particles on atmospheric ice nucleation. Concerning the potential of these species as atmospheric INP, one must always combine their efficiency and their abundance. Larger orthoclase particles might be needed to trigger ice nucleation as efficiently as smaller microcline particles, but maybe many more orthoclase particles are emitted into the atmosphere? Besides, the fact that ice nucleation was not observed for 100 or 200 nm orthoclase particles is related to the detection limit of the instrument. If the authors had investigated more droplets, they would eventually have observed ice nucleation triggered by the small orthoclase particles. This should be made clear in the manuscript.

2) Difference to earlier studies

This refers to P3L14-18, where I think the authors should clarify the innovation of their study more. Like this, it sounds as if they might expect an effect of methodology on the results, as the other studies were performed with droplets containing numerous particles each. Please state that by using single particles, you are focusing on a different temperature range than the other studies (except Augustin-Bauditz et al., 2014).

3) Multiply-charged particles

The authors should be more precise concerning the amount of multiply-charged particles in the cases where the CPMA was not used. This issue could be addressed by including actually measured size distributions in Fig. 2 instead of the schematic ones. Alternatively, the authors could include the following statement on P4L28-29: "The use of the CPMA for the selection of larger particles (400 nm, 800 nm) was not necessary as the fraction of larger particles was reduced to … % by the cyclones and the impactor upstream of the DMA."

4) Figure 4

I see more benefit from one figure showing *FF* over *T* for 800 nm particles of all samples. This would be more suited for comparing the ice nucleation signatures of the different feldspars than just showing the range in which freezing occurred. Error bars could be omitted (because they are already shown in Fig. 3) and symbol size reduced for clarity.

Technical corrections:

P1L9: Replace "Na/Ca-rich feldspar" with plagioclase to be consistent with the title. The composition is explained below anyhow.
P1L11: Replace "are" with "were" in "Samples are selected…".
P1L18-20: This sentence would benefit from being split into two.
P1L24-25: Either omit the "s" at "temperatures" or at "depends".
P1L29: There is also contact freezing in which the contact causes nucleation, not an immersed particle.
P2L12: Within a sentence "e.g." should be preceded by a comma. A comma should also follow in case you are using American English. This also applies to "i.e.". Generally, check your manuscript for consistency with either British or American English. E.g., see "favouring" on P11L10 or "generalise" and "analysed" on P12L8 and L13.
P2L13: Less efficient in comparison to which other species?
P2L14: Change "for example" to "e.g.".
P2L22: Capital "X" in "x-ray".
P3L21: Mention that XRF is a bulk, not a single particle technique.
P3L28: Omit comma following "polymorphism" and add "s" to "occur".
P3L30-31: Add comma behind "sanidine". Be consistent using either "temperature" or "temperatures".
P4L1-2: I suggest to remove the brackets and structure the sentence as follows: "sanidine in volcanic and very high-temperature metamorphic rocks, orthoclase in … rocks and microcline in … rocks."
P4L2: "feldspar": This should be plural.
P4L3: Why is Table 3 referred to before Table 2 is mentioned? Should the labels be switched?
P4L3-10: I appreciate the discussion of the atmospheric relevance of the samples. However, I feel that the last sentence in this paragraph might better be shifted before "We note…" to introduce the reader to this topic.
P4L12: Change "are" to "were".
P4L21: Change "multiple charged" to "multiply-charged", also in the other instances. Also, "single charged" should become "singly-charged".
P5L12: Remove hyphen in "ice-layer".
P5L18: Insert hyphen in "in line".
Fig. 3: Please state how you derived the error bars.
P6L13: How were the 25 % derived? Which particle size are you referring to?

P6L22-25: Could you state the parameters of the amazonite contact angle distribution? Should amazonite be capitalized on P6L22?
P6L26: Here you could refer to the Fig. showing $n_s(T)$ which I suggested as a replacement for Fig. 5.
P7L7-9: This statement would be more convincing if you provided actual numbers for the remaining multiply-charged particles in the 400 and 500 nm aerosol.
P7L10-11: I advise not to use $T_{50}$ for comparison to other studies. In this regard, my suggestion from above, i.e., showing $n_s(T)$, would be helpful.
P7L23-25: "it remains unknown what particle property other than chemistry and crystallography or morphological features … could be active sites": This conclusion cannot be made at this point in the manuscript since you only discuss these factors in Sec. 5. Please reword. On P7L25, do you mean "as discussed"?
P8L8: "sanidine" should also be followed by a comma.
P8L11: Sometimes you use "(see Figure…)", sometimes only "(Figure…)". Be consistent.
P8L13-14: What is the difference between a defect-free and an ordered crystal? Please clarify.
P8L29: I think, it might be helpful to indicate the perthitic structures in Fig. 6, maybe with the help of overlaid boxes.
P9L29: "Contrary to the Pb content, …": Are you referring to microcline not quite fitting the linear relation in Fig. 7? This should be discussed in the previous paragraph.
P10L4: Move "(increase entropy)" behind "structuring of water".
P10L6-7: The explanations of kosmotropic and chaotropic in brackets should be moved to P10L2, where the terms are first mentioned.
P10L10-11: I suggest to move this statement towards the beginning of Sec. 5.2. Otherwise the reader might wonder for quite some time how valid your conclusions about the bulk chemical composition are for the investigated submicron particles.
P10L28: Insert comma between "cold" and "low".
P11L9: Missing bracket after "sanidine".
P11L25-26: Either change "temperatures" to "a temperature" or "that" to "those".

References:


Augustin-Bauditz, S., Wex, H., Kanter, S., Ebert, M., Niedermeier, D., Stolz, F., Prager, A., and Stratmann, F.: The immersion mode ice nucleation behavior of mineral dusts: A comparison of different pure and surface modified dusts, Geophys. Res. Lett., 41, 7375-7382, doi:10.1002/2014GL061317, 2014.

Harrison, A. D., Whale, T. F., Carpenter, M. A., Holden, M. A., Neve, L., O'Sullivan, D., Vergara Temprado, J., and Murray, B. J.: Not all feldspars are equal: a survey of ice nucleating properties across the feldspar group of minerals, Atmos. Chem. Phys., 16, 10927-10940, doi:10.5194/acp-16-10927-2016, 2016.

Hartmann, S., Wex, H., Clauss, T., Augustin-Bauditz, S., Niedermeier, D., Rösch, M., and Stratmann, F.: Immersion Freezing of Kaolinite: Scaling with Particle Surface Area, J. Atmos. Sci., 73, 263-278, doi:10.1175/JAS-D-15-0057.1, 2016.

Peckhaus, A., Kiselev, A., Hiron, T., Ebert, M., and Leisner, T.: A comparative study of K-rich and Na/Ca-rich feldspar ice-nucleating particles in a nanoliter droplet freezing assay, Atmos. Chem. Phys., 16, 11477-11496, doi:10.5194/acp-16-11477-2016, 2016.

Whale, T. F., Holden, M. A., Kulak, A. N., Kim, Y.-Y., Meldrum, F. C., Christenson, H. K., and Murray, B. J.: The role of phase separation and related topography in the exceptional ice-nucleating ability of alkali feldspars, Phys. Chem. Chem. Phys., 19, 31186-31193, doi:10.1039/C7CP04898J, 2017.