Response to referee 2

We thank the referee for their valuable comments. We have reworked the paper to address the relevant issues where necessary. The reviewer comments are shown in italics, our response in normal type and changes in the manuscript in bold enclosed in inverted commas.

General comments

(1) The information provided on the different mineralogical compositions of the feldspar minerals is good, and Figure 1 summarizes this well. I was surprised to not also see more details provided regarding the crystal lattice structure, symmetry, and space group of the different mineral phases. These properties are often referred to in the text to try to understand the observed ice nucleation properties, but without a table or figure summarizing this information it hard to understand this important aspect. Please add as much detail regarding the other known properties of these feldspar minerals in a well organized table or similar.

We agree that this is valuable information and the crystal lattice structure, point and space groups of the different feldspars has been added to the tables.

(2) Introduction: It seems that the recent effort by Perlwitz et al. to incorporate better representations of the variable mineralogical composition of dust into global models should be referred to here. A major challenge regarding understanding and predicting the ice nucleation properties of atmospheric mineral particles is that we do not have a good understanding of the distribution, abundance, and transport of the different mineral types in the atmosphere.

Perlwitz *et al*. has now been mentioned in the introduction in a new statement:

'This is an important finding as it has been demonstrated that feldspar is a common component of aerosolised mineral dusts (Glaccum and Prospero, 1980;Kandler et al., 2009;Kandler et al., 2011;Atkinson et al., 2013;Perlwitz et al., 2015)'

(3) On a related note, it is important to also have some discussion of the size of atmospheric feldspar mineral particles. What aerosol size mode are these typically found in? This is crucial to predict their transport, lifetime, and deposition. One of the reasons that the clay minerals have been focused on for so long is that they tend to be present in the smaller atmospheric mineral particle sizes, and thus have longer lifetimes.

We have added the following text after the comments on the importance of feldspar for ice nucleation:

‘Feldspar particles in the atmosphere tend to be larger than clay particles and so will have shorter lifetimes in the atmosphere, however aerosol modelling work has suggested that feldspar particles can account for many observations of INP concentrations around the world (Atkinson et al., 2013)’

(4) Although you focus on immersion freezing here – without actually stating the heterogeneous ice nucleation mode you measure here (please clarify this so it is clear to the reader), it was odd that this paper on the depositional ice nucleation properties of Feldspar was not cited:

Yakobi-Hancock, J. D.; Ladino, L. A.; Abbatt, J. P. D. Feldspar minerals as efficient deposition ice nuclei. Atmos. Chem. Phys. 2013, 13, 11175–11185, doi:10.5194/acp-13-11175-2013.

We have now made clear in the last paragraph of the introduction and in the experimental section that we make use of heterogeneous immersion freezing:

‘Here this technique is used to make heterogeneous immersion mode nucleation experiments.’

Also, we have now cited the suggested work in the introduction:

‘Work conducted below water saturation using a continuous flow diffusion chamber has also concluded that feldspars, particularly orthoclase feldspars, nucleate ice at low relative humidity in the deposition mode than other common dust minerals (Yakobi-Hancock et al., 2013).’

(5) Some discussion of the vulnerability of feldspar minerals to chemical attack by e.g. sulfuric acid should be included. This is quite important to understand the actual contribution of feldspar minerals to atmospheric ice nucleation, and also provides some insights into the nature of the ice active sites. Wex et al. (2014), already cited here, discuss some of these aspects. I believe it is well known in the mineralogy community that these feldspar minerals can be readily converted to amorphous clay structure through reaction with sulfuric acid.

Greater detail into the weathering has been added to both the stability of active sites and the conclusions section to address this issue.

‘This result is in agreement with the fact that albite weathers faster than microcline in soils as Na^+ is more readily substituted for hydrogen than K^+ (Busenberg and Clemency, 1976; Blum, 1994).’

In the conclusions:

‘If the high energy defects along exsolution boundaries are responsible for higher ice nucleation activity of K-feldspars then this may offer an insight into acid passivation of ice nucleating ability observed in laboratory studies (Wex et al., 2014). Berner and Holdren (1979) suggest that the acid mediated weathering of feldspar occurs in multiple stages and suggest dissolution of feldspars is concentrated at high surface energy sites such as dislocations and crystal defects, sites which may be related to ice nucleation. More work is needed to explore the significance of exsolution, microtexture and the impact of weathering on feldspars with respect to ice nucleation activity.’

(6) Page 6: Why were those 3 mineral samples selected out of the 15 to perform the extended time in water experiments on? It would be valuable to conduct these tests on a larger number of the minerals, since the behavior seems quite variable between minerals. At the least some justification for the 3 sample chosen could be given.

These experiments were of an opportunistic nature as a rapid decay was noticed in both the Amelia albite and TUD#3 microcline between repeat runs. The justification of these samples being chosen has been added to section 5.2 stability of active sites.

‘TUD #3 microcline and Amelia albite were chosen for this experiment as they contained highly active sites, represented two different types of feldspar and were the only feldspars observed to exhibit this rapid decay in activity. BCS 376 microcline was also included in this activity decay experiment as it had provided consistent data over repeated runs and served as a standard in the Atkinson *et al*. (2013) paper which could therefore be tested.’

(7) Page 6, line 31: Citing 8 of the authors own publications that uses the same (and rather simple) experimental method seems like excessive and unnecessary self-citation, especially when Whale et al. (2015a) already provides a detailed discussion of the method. Please restrict these to the most necessary and relevant citations.

Only the most relevant references have now been cited.

Page 7, line 19: More of the recently published experimental work that has explored the role of time-dependent freezing should be cited, such as:

Wright, T. P.; Petters, M. D. The role of time in heterogeneous freezing nucleation. J.Geophys. Res. Atmos. 2013, 118, 3731–3743, doi:10.1002/jgrd.50365.

Wright and Petters has now been cited also.

(8) Some discussion of the similarity of the Monte Carlo approach to estimate the uncertainty of the ns values to other work should be presented. Is this the first time these authors have used this approach, or that anyone has used a similar approach? Wright & Petters (JGR, 2013; cited above) also used a Monte Carlo approach to analyze and interpret their droplet freezing data. Please discuss this. As it is presented it reads as if this is a completely new approach.

We have inserted the following:

'Wright and Petters (2013) previously adopted a similar approach to simulate the distribution of active sites in droplet freezing experiments.'

(9) Page 8, line 14: "We assume that each droplet contains a representative surface area distribution." Please clarify what "representative" means. Is the goal to account for the non-uniform distribution of particle number and surface area in each droplet?

We have addressed this issue in response to referee 1. We added:

'By assuming that the BET surface area of the feldspar powders is made up of monodisperse particles it can be estimated that the droplets will each contain around 106 particles. While there will be a distribution of particle sizes we assume that there are enough particles per droplet that the uncertainty in surface area per droplet due to the distribution of particles through the droplets is negligible. This assumption is supported by our previous work where we show that ns derived from experiments with a range of feldspar concentrations are consistent with one another (Atkinson et al., 2013). If the particles were distributed through the droplets in such a way that some droplet contained a much larger surface area of feldspar than others we would expect the slope of ns with temperature to be artificially shallow. The slope would be artificially shallow because droplets containing more than the average feldspar surface area would tend to freeze at higher temperatures and vice versa. This would mean that ns data derived from experiments with different feldspar concentrations would be inconsistent with one another. However, the fact that ns data for droplets made from suspensions made up with a wide range of different feldspar concentrations all line up shows that the droplet to droplet variability in feldspar surface area is minor (Atkinson et al. 2013). Hence, the droplet to droplet variability in feldspar surface area is neglected and the uncertainty in surface area per droplet in these experiments is estimated from the uncertainties in weighing, pipetting and specific surface area of the feldspars.'

(10) Figure 3 is hard to read at the presented size. The symbols are too small and faint.

We recognise that the figure is hard to read in current form. It was rescaled for the discussion paper. This should hopefully be corrected in the final paper. We have also made an effort to improve the clarity of the error bars.

(11) Figure 4 is begging for some error bars or other measurement of the uncertainties, so it can be determined what degree of the observed changes in median freezing temperature are significant and above the experimental uncertainties. It seems that only the Amelia albite sample displayed any significant change.

We have added temperature error bars to both figures and attempted to improve the clarity of both panels of what is now figure 4.

(12) Figure 5: Some annotations/captions added directly to the figure pointing out what data is plotted where would improve the clarity of this paper. It is difficult to have to keep going back to the figure legend to decode what each dataset is from.

We think that labelling data sets would be quite confusing and add a lot of clutter to the plot. But, we have grouped the like minerals in the key.

Technical corrections

Page 3, line 19: "experiments"

This has now been changed.

Page 7, line 5: Missing a space, should be "C min-1"

This has been corrected.

Page 9, line 30: "(nucleation rate) vs." Versus what?

The typo 'vs' has been removed.

A space in-between the number and "degreeC" is often missing, such as throughout pages 12 & 13.

Spaces have been added in the relevant places.

Page 14, line 18: "regimen this study". Word is missing?

The text now reads as:

"Within the microliter regime in this study we have observed some variability amongst the K-feldspars (see Figure 2), but no difference between sanidine and the 4 out of 5 microclines which fall around the line defined by Atkinson et al. (2013)."

Page 15, line 13: "sites"

This has been changed as suggested.

Page 15, line 15: "that are stable"

This correction has been made in the text.

References

Atkinson, J. D., Murray, B. J., Woodhouse, M. T., Whale, T. F., Baustian, K. J., Carslaw, K. S., Dobbie, S., O'Sullivan, D., and Malkin, T. L.: The importance of feldspar for ice nucleation by mineral dust in mixed-phase clouds, Nature, 498, 355-358, 10.1038/nature12278, 2013.

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