

## Response to referee 1

We would like to thank the reviewer for their valuable comments. We have reworked the paper to address the relevant issues where necessary. The reviewer comments are written in italics, our response in normal type and changes to the manuscript in bold.

### General comments

*(1) To analyze and characterize the chemical composition and crystallographic structure and its features more effort has to be done as realized in the present study. This fact is also explicitly stated by the authors. However, this study only present a starting point as the feldspar characterization method used in the present study is imprecise. Hence the ice nucleation ability of the different feldspars could not be related to e.g. special chemical or crystallographic features such as intracrystalline defect or any other intrinsic property. This would be of great interest. The mineralogical composition is given in Table 1 and for the dominant feldspar phase in Table 2. I am wondering if any information of the general composition (e.g. any component but feldspar) or crystal purity is available or measureable. One main result from the present study is that the ice nucleation ability varies for feldspar except for K-feldspar. What is the reason for that? Does it depend also on the source regions for the different feldspars? Would it be necessary for modellers to account for such an effect? This should be first discussed and second stated more clearly in the manuscript.*

We think the paper is very important since it is the first survey across the feldspar group and it indicates that specific feldspars nucleate ice more efficiently than others. This means we are now in a much better position to do much more focused and detail orientated future studies where we look at specific properties of the select feldspars which nucleate ice effectively. Much of what the referee suggests in terms of trying to find what intrinsic property controls nucleation is sensible, but we view a detailed study as a next step. Nevertheless, the present results do indicate that the strongest ice nucleation is limited to the alkali feldspars and we hypothesise in the paper that the nucleating ability is related to microtexture. We are currently working on this hypothesis, but characterising microtexture and doing controlled experiments is a major study and is very much the topic of a future paper.

As the referee suggests, there are other properties of the feldspars which could be of interest. We have incorporated the space and point groups as well as the source locations of the minerals into tables 1 and 2 to try better display the known information for the studied minerals.

(2) *The authors try to explain the observed freezing behaviour of pure feldspars and feldspars aged in water using the concept of “active sites”. It is unclear how exactly an active site is defined in the context it is used in the present study. Rather it seems that an active site is used as a construct with which almost anything can be explained if it is not related to a property of an ice nucleating particle, which should be determined from an independent measurement, see general comment (1). The concept of active sites has to be introduced and motivated earlier in the paper and caution is required when conclusions are drawn. On page 5 line 14-17, the authors distinguish between 3 different kinds of active site. Is it known for example whether the active site type (i) is similar for all feldspars? Or do they differ? This is not clear to me. The study includes many interesting indications what these special properties are or at least how they change for different types of feldspars or due to aging in water, but a clear comprehensive explanation is missing.*

We have expanded our discussion of active sites in the experimental section:

**To allow comparison of the ability of different materials to nucleate ice, the number of active sites is normalised to the surface area available for nucleation. This yields the ice nucleation active site density,  $n_s(T)$ .  $n_s(T)$  is the number of ice nucleating sites that become active per surface area on cooling from 0°C to temperature  $T$  and can be calculated using (Connolly et al., 2009):**

$$\frac{n(T)}{N} = 1 - \exp(-n_s(T)A) \quad (1)$$

Where  $n(T)$  is the number of droplets frozen at temperature  $T$ ,  $N$  is the total number of droplets in the experiment and  $A$  is the surface area of nucleator per droplet.

Active sites may be related to imperfections in a crystal structure, such as cracks or defects, or may be related to the presence of quantities of other more active materials located in specific locations at a surface. While the fundamental nature of sites is not clear, and may be different for different materials,  $n_s$  is a pragmatic parameter which allows us to empirically define the ice nucleating efficiency of a range of materials (Vali, 2014).

This description is site specific and does not include time dependence. The role of time dependence in ice nucleation has recently been extensively discussed (Vali, 2014; Vali et al., 2014; Vali, 2008; Herbert et al., 2014; Wright et al., 2013). For feldspar (at least for BCS 376 microcline) it is thought that the time dependence of nucleation is relatively weak and that the particle to particle, or active site to active site, variability is much

more important (Herbert et al., 2014). The implication of this is that specific sites on the surface of most nucleators, including feldspars, nucleate ice more efficiently than the majority of the surface. As this study is aimed at comparing and assessing the relative ice nucleating abilities of different feldspars we have not determined the time dependence of observed ice nucleation in this work, although this would be an interesting topic for future study.

We acknowledge the referee's comments on whether the active site type (i) is similar for all feldspars and on the absence of 'a clear comprehensive explanation' with reference to the types of site. This is a problem for the entire field of ice nucleation, there is no clear, comprehensive and generally agreed upon explanation for why any substance should nucleate ice. We have made some intriguing observations which move us in the direction of a more comprehensive understanding. In the text we already discuss the different characteristics of sites on different feldspars samples. In addition we have expanded on the discussion in the conclusions section with the following lines:

**'It is possible that the sites of type *i* are present on the typical K-feldspars, but we do not observe them because ice nucleates on more active sites. Whether these different sites are all related to similar features on the surfaces or if they are each related to different types of features is not known. Nevertheless, it appears that feldspars are characterised by a range of site types with varying stability and activity.'**

*(3) At the moment without further tests, I am not convinced that the "hyper-activity" of one of the microcline and albite samples is real. I think the contribution from to biological ice nucleator contaminants can not be ruled out completely. The feldspar suspensions were left at room temperature (p.6 I.22). Under such conditions biological activity is not suppressed. The relatively gentle treatment in 100°C water for 15 min might destroy proteins (p.9 I.24-26) but not ice active polysaccharides or other organic/biological substances ice active at quite high temperature (Pummer et al., 2012; Tobo et al., 2014; O'Sullivan et al., 2014). Additional treatments with H<sub>2</sub>O<sub>2</sub>, H<sub>2</sub>SO<sub>4</sub>, etc. could be helpful as only heating is not always enough.*

As the referee states the treatment of the sample at 100°C would destroy proteins but potentially not other organic substances. However the only biological substances we know to be active at the high temperatures displayed for the TUD#3 microcline and Amelia albite are proteins. This leads us to think that the activity is inherent to the feldspars. Additional treatments with H<sub>2</sub>O<sub>2</sub> or H<sub>2</sub>SO<sub>4</sub> might be helpful but may also influence ice nucleation by feldspars which would make any results difficult to interpret. We agree that we should be more cautious in our statements and have modified the text accordingly:

**‘Certain biological nucleators have been observed to retain their ice nucleating activity despite heat treatment of this type (Pummer et al., 2012; O'Sullivan et al., 2014; Tobo et al., 2014) however, to the best of our knowledge, no biological species has been observed to nucleate ice at such warm temperatures after heat treatment. Additionally, grinding of Amelia albite which had been stored as a powder for many years increases its ice nucleating potential, which is consistent with exposing fresh surfaces with features which decay away on contact with water. This behaviour is not consistent with biological nucleators, unless the biological entity is within the Amelia albite particles and is somehow dispersed through the particle population during grinding. While we cannot exclude the possibility that some unknown biological species is present on microcline TUD#3 and Amelia albite it seems more likely that the minerals themselves are responsible for the observed ice nucleation activity.’**

*(4) Figure 3 and 4 has very poor quality and presents similar statements. The symbols are partly not visible. One can not distinguish between the different times of aging. The measurement uncertainties are missing, which must be known to judge if it is a real trend or just uncertainty of the experiment. This is most obvious in Fig.3 panel b) and c). I suggest to combine these figures.*

We have combined both plots into a single figure with multiple panels. We think both panels are needed to emphasise the nature of the decay in activity over time. It is noted that the figures are difficult to see due to resizing for the discussions paper. This should improve in the final copy. In addition, we have used stronger colours for the points and error bars as well as expanding the x-axis.

Specific comments

*p.1 I.16 & p.2 I.3 Specify what is meant by “soil dust”. In the context of this study it seems that you mean mineral dust. Otherwise you need to clarify this statement.*

The term soil dusts has been removed from the text to prevent any confusion.

*p.1 I.22 Considering point 1 and 2 of my major concerns I don't agree that the feldspar samples are “well-characterized”. They are only characterized with respect to their “macroscopic crystal structure” and BET surface area, but other probably decisive properties such as intra-crystalline defects are not considered at all.*

We have replaced the phrase “well characterised” with simply “characterised”.

*p.5 I.1-3 It might be true that the knowledge about the chemical composition of airborne mineral dust is limited. However, to be fair some studies investigating the composition of atmospheric mineral dust can be mentioned for example Glaccum and Prospero (1980), Kandler et al. (2007, 2009) to name a few.*

We have now cited these papers in the relevant section and the text now reads:

**‘There is limited information about the composition of airborne atmospheric mineral dusts (Glaccum and Prospero, 1980;Kandler et al., 2007;Kandler et al., 2009); where mineralogy is reported the breakdown of the feldspar family has only been done in a limited way.’**

*p.5 I.24-27 and in all figures The labelling of the different feldspar samples might be precise and traceably, but totally confusing for the reader. I suggest to simplify the nomenclature in a way that it will be transparent for the reader to whom the paper is addressed. Probably a table in the Appendix could be useful for the precise description.*

We have attempted to use naming conventions compatible with the geology literature. Our samples are named in the same fashion as they were in the studies which initially characterised them, where possible. Where we have introduced new samples we have followed the same naming conventions. We understand that the

number of samples and their naming can be cumbersome but we feel that it is important to retain consistency with previous work. We would therefore prefer to use the current nomenclature.

*p.5 l.28-30, p.6 l.1-2 Please give a motivation for this treatment of the sample*

We have inserted:

**‘As these two samples are chemically identical, differing only in that one is amorphous and the other crystalline, comparison of the ice nucleating efficiency of the two samples has the potential to reveal information about the impact of feldspar crystal structure on ice nucleating efficiency.’**

*p. 6 l.10-14 In that context, Zolles et al, (2015) claimed that grinding could lead to a disclosing of active sites and even an enhancement of available active sites. This should be addressed in the manuscript as well.*

The activity increase seen by Zolles *et al*, (2015) has now been mentioned:

**Zolles *et al*. (2015) have suggested that grinding can lead to active sites being revealed, or the enhancement of existing active sites. It was shown in Whale *et al*. (2015) that differently ground samples of BCS 376 microcline nucleate ice similarly. In contrast Hiranuma *et al*. (2014) show that ground hematite nucleates ice more efficiently (normalised to surface area) than cubic hematite. The evidence suggests that the ice nucleating efficiencies of different materials respond differently to grinding processes.**

*p.7 l.3 Specify what is meant by “quantity”. Is this quantity related to mass or mass concentration?*

This has now been adjusted to:

**“Briefly, droplets of an aqueous suspension, containing a known mass concentration of feldspar particles are pipetted onto a hydrophobic coated glass slide.”**

*p.7 l.5-7 I do not understand why using a small dry nitrogen flow should prevent frozen droplets from affecting their neighbour liquid droplets?*

This has been further clarified in the text as follows:

**“This slide is placed on a temperature controlled stage and cooled from room temperature at a rate of 5 °C min<sup>-1</sup> to 0 °C and then at 1 °C min<sup>-1</sup> until all droplets are frozen. Dry nitrogen is flowed over the droplets at 0.2 l min<sup>-1</sup> to prevent frozen droplets from affecting neighbouring liquid droplets. Whale et al. (2015) demonstrated that a dry nitrogen flow prevents condensation and frost accumulating on the glass slide so ice from a frozen droplet cannot trigger freezing in neighbouring droplets.”**

*p.7 l.7 How large are the droplets and do they vary in size? How many feldspar particles are immersed in the droplets and how does this number vary between different droplets. Statements to clarify these quantities are required. These are necessary information the reader needs to assess the reasonability of the approach of determining the uncertainty in ns.*

The droplets are of  $1 \pm 0.025 \mu\text{l}$  volume, we have inserted the following (the uncertainty is small because we use an electronic pipette with low uncertainty):

**‘Briefly,  $1 \pm 0.025 \mu\text{l}$  droplets of an aqueous suspension, containing a known mass concentration of feldspar particles are pipetted onto a hydrophobic coated glass slide.’**

We comment on the number of particles per droplet in the next comment

*p.8 l.2-16 As mentioned in the last section further information of the distribution of particles (hence potential active sites) over the droplet population are needed. I do not understand the procedure to derive the uncertainty in ns. Especially I could not follow how the two distributions are combined. Further, is the assumption ” that each droplet contains a*

*representative surface area distribution” justified? Maybe this is only true for rather high particle mass concentrations.*

We have inserted the following to address the question of the number of particles per droplet:

**‘By assuming that the BET surface area of the feldspar powders is made up of monodisperse particles it can be estimated that droplets containing 1 wt% of feldspar will each contain around  $10^6$  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. In contrast, it has been suggested that ice nucleation data could be explained by variability of nucleator surface area through the droplet population (Alpert and Knopf, 2016). Our assumption that each droplet contains a representative surface area is supported by our previous work where we show that  $n_s$  derived from experiments with a range of feldspar concentrations are consistent with one another (Atkinson et al., 2013) (Atkinson et al., 2013; Whale et al., 2015). 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  $n_s$  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. However, the fact that  $n_s$  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; Whale et al., 2015). 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.**

With regard to the description of the way the uncertainty in  $n_s$  is calculated we have changed the description of the method in an effort to make it clearer. It now reads:

**‘In order to estimate the uncertainty in  $n_s(T)$  due to the randomness of the distribution of the active sites in droplet freezing experiments, we conducted Monte Carlo simulations. Wright and Petters (2013) previously adopted a similar approach to simulate the distribution of active sites in droplet freezing experiments. In these**



simulations, we generate a list of possible values for the number of active sites per droplet ( $\mu$ ). The theoretical relationship between the fraction of droplets frozen and  $\lambda$  can be derived from the Poisson distribution:

$$\frac{n(T)}{N} = 1 - \exp(-\mu) \quad (2)$$

The simulation works in the following manner. First, we take a value of  $\mu$  and we simulate a corresponding random distribution of active sites through the droplet population for an experiment. Every droplet containing one or more active sites is then considered to be frozen. In this way, we can obtain a simulated value of the fraction frozen for a certain value of  $\mu$ . Repeating this process many times and for all the possible values of  $\mu$ , we obtain a distribution of possible values of  $\mu$  that can explain each value of the observed fraction frozen. This resulting distribution is neither Gaussian nor symmetric, so in order to propagate the uncertainty to  $n_s(T)$  values, we take the following steps. First, we generate random values of  $\mu$  following the corresponding previously simulated distribution for each value of the fraction frozen. Then, we simulate random values of  $A$  following a Gaussian distribution centred on the value derived from the specific surface area per droplet with the standard deviation derived from the uncertainty in droplet volume and specific surface area. We assume that each droplet contains a representative surface area distribution as discussed above. This process results in two distributions, one for  $A$  and one for  $\mu$ , with these distributions we can calculate the resultant distribution of  $n_s(T)$  values, and from that distribution, we obtain the 95% confidence interval.'

*p.10 l. 5-9 I do not agree completely. I agree that flat increase in ns indicates a diversity of ice nucleation properties. However, the steeper slopes of ns or analogous quantities at higher temperature are also predicted by Classical Nucleation Theory assuming only one contact angle. In other words, even when similar ice nucleation properties (one contact angle) are assumed, the slopes become steeper at higher temperature. As a conclusion, the same effect can be explained also by a different hypothesis.*

Good point, the following has been added to the paper.

**‘The smaller diversity in the sites active at warmer temperatures may explain the observed steep slopes in  $n_s$ , however it should be noted that Classical Nucleation Theory also predicts steeper slopes at higher temperatures, assuming a single contact angle.’**

*p.11 l.19-20 It seems that in Fig.3 the variation might also be explained by the measurement uncertainty? There is no trend in one direction with increasing time, or is the legend incorrectly labelled? This must be clarified.*

The error bars have now been shown more clearly to justify that the overall trend in of both Amelia albite and TUD#3 are not simply an artefact of experimental error. Note that there is some variability for the TUD#3, but the overall trend over 16 months is clear.

*p.11 l. 25-28 Additionally, the study of Marcolli et al. (2007) and Hartmann et al. (2016) can be mentioned.*

The papers have now been cited.

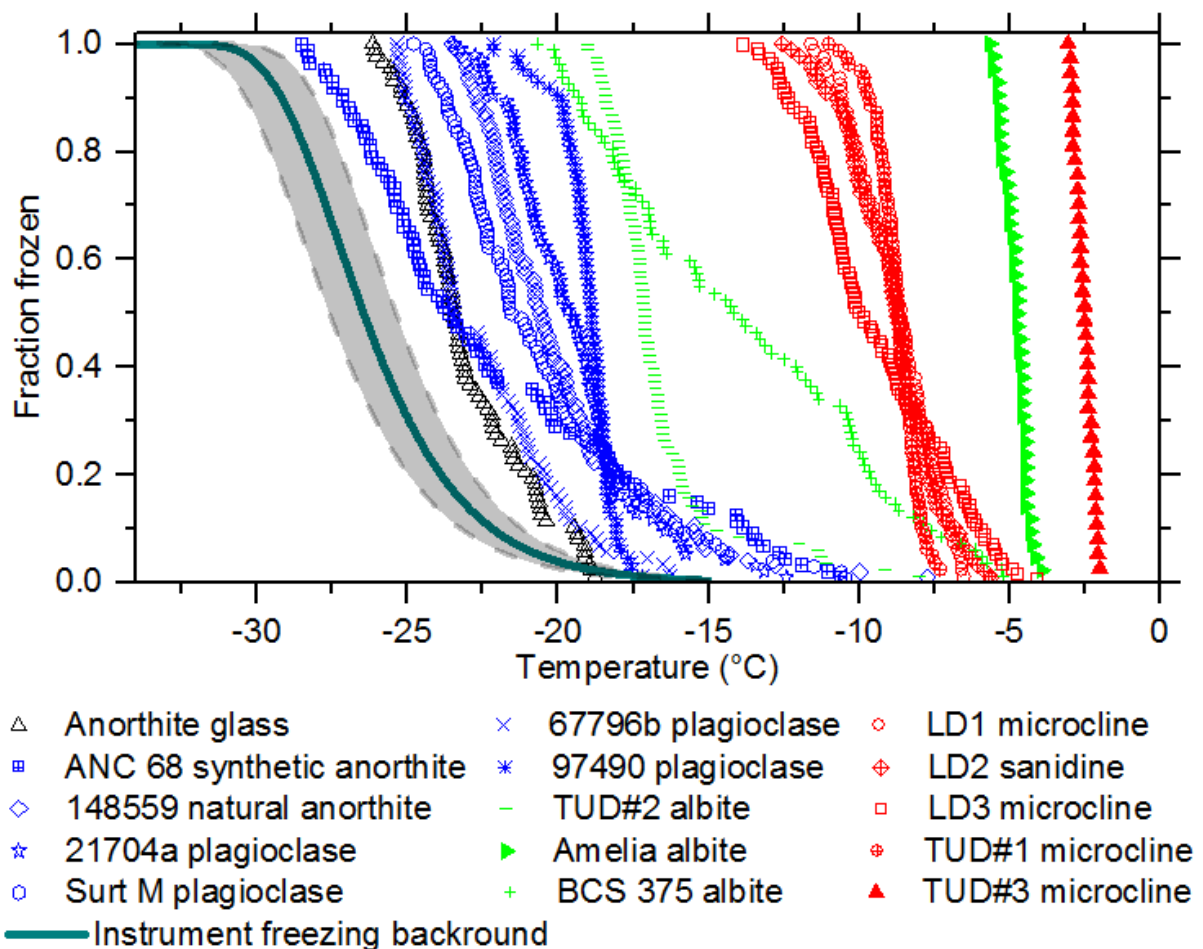
*p.13 l.26-32 Larger  $n_s$  values always imply lower available particle surface area relevant for heterogeneous ice nucleation or lower number of active sites when similar ice nucleating materials are analyzed. The mass concentration and size of a droplet containing particles of different sizes (particle distribution) is not the essential quantity, but the total surface area of the particles. I feel that in both experiments (Atkinson et al., 2013 and Zolles et al., 2015) this quantity is not determined with sufficient reliability. Consequently, this is not a conclusive argument. If  $n_s$  is carefully derived from the experimental data, this effect should be already considered. Error bars could clarify the uncertainty in  $n_s$  derived from different experiments.*

This is a good point, we were trying to make clear the differences between the experimental procedures used in different papers but wrote the section poorly. We agree that the total surface area of the particles in a droplet is the important quantity, but wanted to highlight the differences between the experiments. We have improved this discussion with the insertion of:

“In principle,  $n_s$  should be independent of droplet volume and particle concentration, but differences between instruments and methods have been reported (Hiranuma et al., 2015). Additionally, Zolles et al. (2015) estimated the surface area of their feldspar particles using a combination of SEM images and the BET surface area of quartz. This leads to an unspecified uncertainty in their  $n_s$  values.”

*p.23 Fig.2 For the observed freezing at low temperature, it is necessary to show the limit of the measurement due to pure water freezing.*

We have inserted a new figure showing droplet fractions frozen for all experiments, along with the freezing temperatures for pure water showing that the experiments we have conducted are not interfered with by the background freezing of the instrument, although one of the plagioclase runs gets quite close to this limit.



**Figure 1: Droplet fraction frozen as a function of temperature for 1 wt% suspensions of ground powders of various feldspar samples. The K-feldspars are coloured red, the plagioclase feldspars are coloured blue, the albites are coloured green the feldspar glass is coloured black and the background freezing is coloured cyan. A fit to the**

background freezing of pure MilliQ water in the  $\mu$ l-NIPI instrument used by Umo et al. (2015) is also included. The shaded area around this fit shows 95% confidence intervals for the fit. It can be seen that all the feldspar samples tested nucleate ice more efficiently than the background freezing of the instrument.

*p.23 l.12 I do not agree that  $n_s$  values can be simply subtracted without introducing further unnecessary uncertainty. An internal mixture of different ice nucleating particles has to be accounted for.*

We do not simply subtract  $n_s$  values and have removed the confusing statement. The process is described in Umo et al. (2015).  $n_s$  values are not subtracted directly.  $K$  values are calculated for the background freezing and subtracted from  $K$  values for the experiment. The resulting  $K_{\text{het}}$  values are then converted into  $n_s$  values.

#### Technical corrections

*p.3 l.16-18 The citation list in brackets has to be sorted in the order beginning from the oldest to the recent publications. This needs to be changed in the whole manuscript.*

This has been corrected.

*p.16 l.2 . . . higher temperatures . . .*

A correction has been made.

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