

We thank Reviewer 2 for their very constructive comments. We reproduce reviewer comments in blue in the following. Amended versions of the paper are given in *italics* for new sections and red text for the original text.

Boose et al. reports measurements of the effectiveness with which a range of natural dust samples from around the world to nucleate ice from supersaturated water vapour. Various analytical techniques were used to determine the composition of the samples and to attempt to establish what components are responsible for observed ice nucleation, leading to the conclusion that quartz and feldspar content is responsible for the ice nucleation observed in most samples. In one sample organic substances were found to be largely responsible while in another removal of organics actually enhanced ice nucleation effectiveness. The conclusions of the paper are interesting and relevant to the scope of ACP, highlighting the substantial complexity in ice nucleation by natural samples. The paper is mostly well written, sensibly structured, and is entirely suitable for publication in ACP, after a few minor corrections

### Minor comments

I have a few issues with the identification of feldspar phases via powder XRD in this work. Alkali feldspar structure is a complex topic and it is not clear to me that powder diffraction is adequate to certain about which polymorph is present in dust samples of the type characterised here. Indeed in part 1 of this study the authors state 'Where microcline and orthoclase are present in the same sample, their individual fraction could not be distinguished reliably' in the caption to the relevant table. It is not clear to me what has changed in the present study.

The analysis of XRD used in the present study is the same as used in part 1 (Boose et al. 2016c). Nothing has changed and the same uncertainty applies here as well.

The difference between orthoclase and microcline is essentially the degree of order of the aluminosilicate framework. There is not necessarily a hard line between orthoclase and microcline. Parsons et al (American Mineralogist (2015) 100 (5-6), 1277-1303) and references therein discuss some of the relevant issues. Additionally the differences between even very pure samples of the two minerals in powder XRD are subtle. It must be more difficult to be certain of phase when mixed dusts with very low feldspar contents are under investigation. In summary, if the statements in the paper regarding the relative amounts of orthoclase and microcline are to be kept a much better description of the powder XRD procedure, and justification the differentiation between orthoclase and microcline is needed. I do not think this is particularly key to the paper, because, as the authors say, there is not any difference between the feldspars as characterised anyway. I would suggest treating the topic as it was treated in part 1 of this study.

Relatedly, Pg 8 line 12 states that microcline is the more active K-feldspar polymorph without citation. I think this statement should be changed, or its origin cited and discussed. The superior ice nucleating ability of microcline seems to be assumed throughout the paper. I would note that Whale et al. (Phys. Chem. Chem. Phys., (2017), 19, 31186) which used pure feldspars '...found no correlation between ice-nucleating efficiency and the crystal structures or the chemical compositions...' of ice nucleation active feldspars, albeit in rather different conditions.

We thank the reviewer for these valuable comments. We have decided to take out the sentences which suggest microcline being more active than orthoclase. We had based this statement on earlier studies by Augustin-Bauditz et al. (2014), GRL (doi: 10.1002/2014GL06131) and Harrison et al. (2016), ACP (doi: 10.5194/acp-16-10927-2016) but admit that the study by Whale et al. (2017) proves this to not hold in general, which we had overlooked.

We have made the following changes to the manuscript:

Pg.2, ln.27: *Amongst the K-feldspars, microcline has a remarkable ice nucleation ability at temperatures up to 271 K (Harrison et al., 2016). However, Kaufmann et al. (2016) found microcline only in one out of eight dust samples collected in potential atmospheric dust source regions in South America, Asia and Africa and microcline was only present in one out of four airborne Saharan dust samples studied by Boose et al. (2016c).*

Was changed to:

Pg. 2, ln.27: *While Kaufmann et al. (2016) found K-feldspar only in one out of eight dust samples collected in potential atmospheric dust source regions in South America, Asia and Africa, we observed K-feldspar to be present in all but one sample from a collection of 21 samples from deserts around the world (Boose et al., 2016c).*

Pg.8, ln.9: *Microcline was found in four out of five investigated Izaña samples (4-5 wt%) and orthoclase (4-5 wt%) is present in one Izaña and the other three Saharan samples. Comparing the bulk mineralogy to the surface-dependent ice nucleation ability has the caveat to introduce uncertainty. Within this uncertainty there is no detectable effect from the presence of the more ice nucleation active microcline K-feldspar over orthoclase for the airborne samples at the studied temperatures.*

was changed to:

Pg. 8, ln.21: *Differentiating between microcline and orthoclase and comparing the bulk mineralogy to the surface-dependent ice nucleation ability, introduces uncertainty. Within this uncertainty, there is no detectable effect from the presence of microcline versus orthoclase in the airborne samples at the studied temperatures. This is in line with the findings by Whale et al. (2017) that there is no correlation between ice nucleating ability and the level of ordering in the aluminosilicate framework, which determines if orthoclase (less ordered) or microcline (more ordered) is present.*

And we deleted the following from the initial manuscript:

Pg. 12, ln. 30: *Furthermore, we observed that the ice nucleation activity of airborne Saharan dust at these temperatures is not higher when microcline is present in the samples compared to when orthoclase is present at typical amounts of 4-5%. If one assumes that both feldspars are equally active at these low temperatures, this result is to be expected.*

Naming of samples is not entirely consistent through the manuscript I think, this should be checked.

We changed “**native**” to “**unheated**” on originally Pg. 9, ln 4 and Pg. 12, ln12

Furthermore, we have changed the naming of the Izaña201x\_x samples into Tenerife201x\_x and have numbered the 2014 samples from 1 to 3 to be consistent with part 1 of this study:

Izaña2013\_2 → Tenerife2013  
Izaña2014\_2 → Tenerife2014\_1  
Izaña2014\_3 → Tenerife2014\_2  
Izaña2014\_5 → Tenerife2014\_3

### Specific comments

Abstract line 7 to 9- sentence starting ‘in this study,....’ does not read well.

We have changed the sentence to:

Pg.1, ln.7: *In this study, the influence of semi-volatile organic compounds and the presence of crystal water on the ice nucleation behavior of desert aerosol is investigated.*

Abstract line 9- between 238 and 242K

Changed

Abstract line 15 reads poorly, missing comma after ‘diminished’?

Comma was added after ‘diminished’

Pg 2 Line 2- delete ‘already’

Done

Pg 2 Line 25- ACP version of Paramonov et al. is available

Reference was changed to ACP version

Pg 3 line 32- clumsy wording, probably delete ‘showing’

We replaced ‘showing’ with ‘found’. The sentence now reads:

Pg.3, ln.31: *At  $T \leq 245$  K the best correlation of the ice nucleation activity was found for the bulk quartz plus feldspar content in the dust samples while the fraction of clays was negatively correlated with the ice nucleation activity.*

Pg 3 line 10- typo ‘selected’

Corrected

Pb 3 line 33- clumsy sentence structure

Pg 3 line 35- ‘over’ not the right work I think.

The respective sentence

Quartz alone has been found to show various immersion mode ice nucleation activities in laboratory studies, ranging from being active at temperatures comparable to microcline (Zolles et al., 2015), over being active below feldspar temperatures but above those of clay (Atkinson et al., 2013), to showing ice nucleation activity at temperatures comparable or lower than those of clays (Kaufmann et al., 2016).

has been replaced by:

Pg.3, ln.34: *Quartz alone has been found to show various immersion mode ice nucleation activities in laboratory studies. Zolles et al. (2015) found quartz being active at temperatures comparable to microcline, while Atkinson et al. (2013) measured ice nucleation activity below feldspar temperatures but above those of clay. Kaufmann et al. (2016), on the other hand, only observed ice nucleation activity at temperatures comparable to or lower than those of clays.*

Pg 3 line 33 onwards- I note that there is interesting work on the topic of ice nucleation by quartz under review in ACPD at current (<https://www.atmos-chem-physdiscuss.net/acp-2018-1020/>), which may shed some light on the complexities of ice nucleation by various silicas.

We have added the following sentence on p. 4, ln. 2:

*These differences in ice nucleation ability can be related to the history of the quartz samples and different ways of pre-processing them (Zolles et al., 2015). Milling quartz samples leads to a break-up of Si-O-Si bridges on the surface, leading to the formation of Si-OH and Si-O-OH in the presence of water vapor, which increases the ice nucleation activity of the quartz particles (Kumar et al., 2018b).*

Further, we discuss our results now in the light of the study by Kumar et al. (2018b) in the Conclusions.

Pg. 12, ln.27: *It remains unknown why laboratory studies show various ice nucleation activities of pure quartz particles (Atkinson et al., 2013; Zolles et al., 2015; Kaufmann et al., 2016), relating to the question of how ice nucleation occurs on dust in general and on quartz in particular.*

Was changed to:

Pg. 14, ln. 2: *According to a recent study by Kumar et al. (2018b), the variation in quartz ice nucleation ability found in laboratory studies (Atkinson et al., 2013; Zolles et al., 2015; Kaufmann et al., 2016) and the superior ice nucleation ability of the quartz-rich samples from Australia and Morocco in this study and its partner paper, may be explained by the pre-processing of the samples. Milling of quartz samples, as done in our study, increases the ice nucleation ability of quartz by creating Si-O• and Si• radical sites which can then react with water vapor (Kumar et al., 2018b). However, milling may not be the only reason for formation of the silanol (Si-OH) groups on the surface of quartz, because exposure to water molecules in ambient humidity could also result in passively converting surface siloxane groups (Si-O-Si) to silanol groups (Boehm, 1966; Wang et al., 2018). As such quartz samples may still exhibit high ice nucleation activity in the absence of milling due to the particles chemical history. Thus, it remains an open question, how much quartz contributes to the ice nucleation ability of (unmilled) atmospheric dust.*

Pg 4 line 7- while RH is increased

Changed

Pg 8 line 15- Some justification for using the conditions stated might aid the reader.

The original sentence:

*This comparison showed that the immersion mode ice nucleation activity at  $T \leq 245$  K correlates best*

with the quartz and quartz+feldspar fractions of the dust samples, while at T = 253 K it correlates best with the K-feldspar fraction alone (Boose et al., 2016c).

was changed to:

Pg. 8, ln.27: *This comparison showed that the immersion mode ice nucleation activity correlates best with the K-feldspar fraction alone at T = 253 K, a temperature where only feldspar minerals are found to have significant ice nucleation activity (Atkinson et al. 2013). At T ≤ 245 K, the ice nucleation activity correlates best with the quartz and quartz + feldspar fractions of the dust samples (Boose et al., 2016c). At these lower temperatures, quartz and to a lower degree also clay minerals were found to nucleate ice efficiently (Atkinson et al. 2013).*

Pg 11 line 15- I'm not sure what is meant by the sentence 'However, it was not...'. I suggest clarifying this.

We have deleted the sentence. We just wanted to stress that ammonium sulfate alone is not responsible for ice nucleation of dust. However, this is likely obvious for the reader and the sentence rather confusing.

Pg 9 line 15- I am not sure what is meant by 'hardly' in this context? This sentence could be clearer.

We believe the reviewer is referring to pg. 9 ln. 17 of the initial manuscript. We have now changed this sentence from

*However, the complexity of the dust samples in this study, which consist of several minerals and likely also various other components, cause the spikes to widen and hardly discrete steps to be observed.*

to now pg. 10 ln. 5:

*However, the complexity of the dust samples in this study, which consist of several minerals and likely also various other components, cause the spikes to widen thus reducing the possibility to observe discrete steps.*

Pg 11 line 12- remove comma after 'both'

Done

Pg 12 line 6- brackets around '2006' after 'Laird'

Done

Pg 12 line 10- sentence starting 'Thus, according to our observations...' is clumsy

The respective sentence

*Thus, according to our observations, the collapse of the smectite lattice should, if at all, only influence the Izaña2014\_2 sample and then be related to an increase in ice nucleation ability.*

was changed to:

Pg.13, ln.4: *Thus, the collapse of the smectite lattice should only influence the Tenerife2014\_1 sample. In case it has an influence, this would be related to an increase in ice nucleation ability.*

Figure 2- It is not obvious what the \* in 0.63\* refers to without referring to final table.

We added a sentence in the caption of Fig. 2 for clarification:

*The asterisk in the legend indicates that the correlation is significant at the 0.05 level.*