

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

As major changes, we have

- renamed the “Namib sample” to “Etosha sample” throughout the manuscript to be more consistent with Kaufmann et al. 2016
- replaced “IN” with “ice nucleation” throughout the manuscript to avoid confusion with “ice nuclei”
- deleted p.9,l. 24-29 because the size distribution of the Australia sample was re-measured using an SMPS and APS. The mean surface area was very close to the original one (within 3%). The n_s values in the revised manuscript include the updated surface area but have changed insignificantly
- binned the IMCA-ZINC FF and n_s data into 1 K bin for visual clarity in Fig. 4
- split up the original Table 5 into Tables 5 and 6 in the revised manuscript
- re-calculated Tables 5 (and 6) after we realized that it was incorrect to correlate the mineralogical fraction with n_s . Instead we correlated the fractions now to $\ln(n_s)$. The trends did not change but the R values changed (typically by 0.03-0.1)
- added sample numbers in Tables 1 and 4 which we refer to in Tables 5 and 6
- added scatter plots (Figure 1 and 2) to the supplementary material corresponding to the correlation analysis in section 3.3
- added Figure 3 to the supplementary material, showing the correlation of freezing temperatures with mineralogical fraction which were taken from Kaufmann et al. 2016

1. Kaufmann et al. vs. Boose et al.: One of the major conclusions from this study (that is, mineralogy matters for dust IN) goes in the opposite direction of that from the study done by Kaufmann et al. (2016, AMTD; K16 hereafter), which points out the similar freezing behavior amongst multiple surface dust samples despite the difference/variation in mineralogy. Does this difference appear due to the technical issue/limitation (i.e., IMCA-ZINC-IODE vs. DSC) or totally something different? I am missing such discussion in this manuscript. Please elaborate to the extent of the community-wide scale. Providing a solid answer with respect to this point would be potentially important for modelers to consider if they require a computationally expensive mineralogy-resolved parameterization for their future simulation works or not.

We do not agree with the reviewer’s statement that the results of our study go in the opposite direction of the study by Kaufmann et al. (K16 hereafter) which is in ACP, not AMTD. On p. 11197 of K16, the authors clearly state: “*However, mineralogical composition does matter.*”

Furthermore, in the abstract of K16: “*In summary, the mineralogical composition can explain the observed freezing behaviour of 5 of the investigated 12 natural dust samples, and partly for 6 samples, leaving the freezing efficiency of only 1 sample not easily explained in terms of its mineral reference components.*”

However, the abstract of K16 also states:

“All natural dusts, except for the Antarctica and ATD samples, froze in a remarkably narrow temperature range with the heterogeneously frozen fraction reaching 10% between 244 and 250 K, 25% between 242 and 246 K, and 50% between 239 and 244 K.”

This statement is not in conflict with our results. The maximum temperature range found by K16 for the natural dust samples excluding the Antarctica and ATD samples is 6 K (frozen water fraction reaching 10 %) while the maximum T -range at the same n_s in our data is 10 K. The larger spread is probably due to the larger number of dust samples and a greater variety in the dust mineralogy in our study, particularly in the quartz content (K16: 1-26 wt%, our study: 1-91 wt%), the K-feldspar content (K16: 0-10 wt%, our study: 0-30 wt%) and the sum of all feldspars (K16: 0-23 wt%, our study: 0-65 wt%).

In addition, the resolution of the latent heat release peaks in the differential scanning calorimetry (DSC) may result in masking of the variability in freezing. In the DSC a droplet can contain more than one particle while IMCA-ZINC works on a single particle basis. If several particles are contained in a droplet, the particle with the most active site will initiate freezing. K16 studied droplets of about 2 μm which contain only a few particles. This curtails the effects but could still cause some of the variability not being observed with the DSC compared to the IMCA-ZINC.

Both studies conclude that as a first approximation, a parameterization like the one by Niemand et al. 2012 is applicable but a more sophisticated parameterization should rely on the mineralogical composition of dust (p.11198 in K16 and p.17,l.21-23 in our revised manuscript).

To clarify this point we have rephrased the following sentence on p.3,l.27-33 of the revised manuscript:

“A recent study by Kaufmann et al. (2016) investigated the ice nucleation ability of surface-collected samples from eight different arid regions worldwide and several single-mineral reference samples using differential scanning calorimetry. The authors found at maximum a 6 K spread in freezing temperatures of emulsion experiments amongst surface-collected samples from different atmospheric-dust source regions. They confirmed the exceptional freezing ability of microcline but found only a minor fraction (4 wt%) in one of the samples from the dust source regions studied. Their samples contained quartz fractions between 1 and 26 wt%, K-feldspar fractions between 0 and 10 wt%, and plagioclase fractions between 0 and 22 wt%.”

Furthermore, we show in Figure 1 of this document correlation plots of T_{het} at 10 and 50 % frozen water fraction of 0.5 and 1 wt% suspensions with the quartz, calcite, plagioclase, microcline and smectite fraction, all taken from K16. The highest correlation coefficient is found for microcline ($R = 0.93, 0.85$ and 0.93). The correlation with microcline however also suffers from a low number of samples. Microcline is followed by quartz ($R = 0.75, 0.72$, and 0.79) and plagioclase ($R = 0.53, 0.27$, and 0.4) as best predictors for ice nucleation activity (here in terms of T_{het} at 10 and 50 % frozen water fraction). These results show that our method works well and the conclusions are supported by K16.

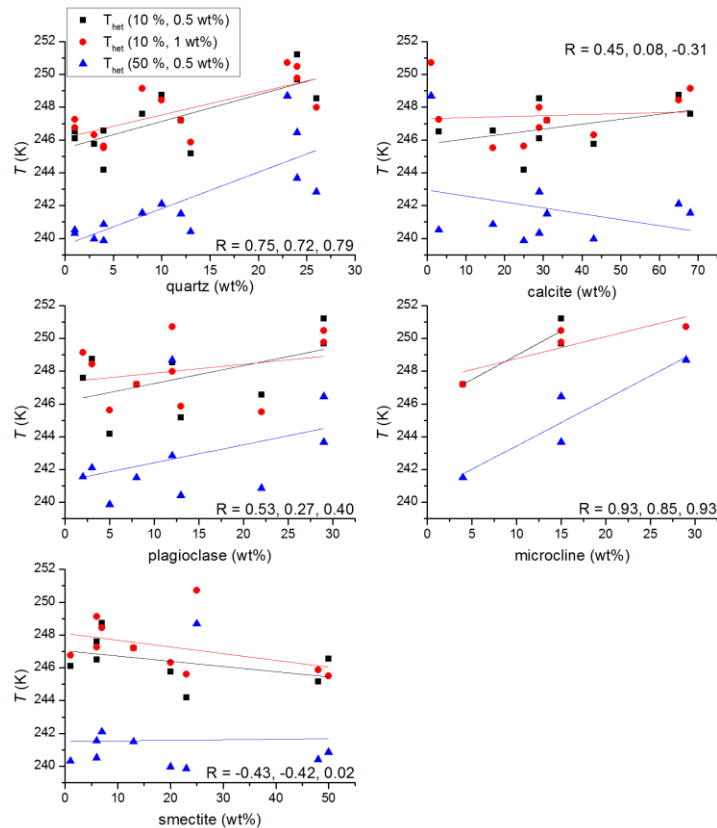


Figure 1: Correlation of freezing temperature of 10 and 50 % frozen water fraction of a 0.5 and 1 wt% solution with six different minerals for the data from Kaufmann et al. 2016

Along similar lines, K16 suggests that microcline-containing particles are not abundant in the atmosphere and, hence, may have overall small contributions to atmospheric ice nucleation (IN) and glaciation (e.g., P20 L22-25 of K16). Do the authors agree with that statement? The reviewer thinks that microcline is in general an important dust composition that can contribute to efficient IN of material composites. Anyhow, it is somehow puzzling for the reviewer to see that two papers from the same institute report opposite things...

We already addressed this point in the original manuscript (p.12, l.10-12):

“We find microcline in at least three of the surface-collected (Great Basin and both Israel) and also in the airborne Tenerife sample. In the samples from Australia and Morocco both phases orthoclase and microcline seem to be present.”

and on p.14, l. 6-10:

“We find microcline in some of our natural dust samples and also in one of the airborne samples. However, we cannot confirm a superior role of microcline over orthoclase for ice nucleation as in most samples microcline was found to be less than 8 wt%. The Great Basin sample contained 30 wt% microcline in the bulk sample but likely significantly less in the size fraction < 2.5 μm . The highly polyminerall nature of our samples and the effect of differences in the size distributions prevent a further investigation of the role of microcline.”

We have merged the two parts above in the revised manuscript (now p.13,l.10-13):

“We find less than 8 wt% microcline in the sieved and milled Israel and the airborne Tenerife samples. In the samples from Australia and Morocco both K-feldspars orthoclase and microcline seem to be present. The surface-collected Great Basin sample contained 30 wt% microcline in the bulk sample but likely much less in the size fraction $< 2.5 \mu\text{m}$. ”

We have added a paragraph on the role of microcline in the conclusions (p.16,l.14-19 of the revised manuscript):

“Microcline was found in one airborne sample (6 wt%) and in surface-collected samples from four different locations. We could not confirm a superior role of microcline over orthoclase, in-part because a differentiation of the two minerals was often difficult and partly because their content was too low in the size range investigated for ice nucleation to cause an effect detectable with the IMCA-ZINC experiment. We cannot therefore conclude that microcline is generally atmospherically not relevant because even in low amounts of a few percent it could glaciate clouds at temperatures warmer than 253 K.”

We cannot confirm or negate a superior role of microcline over orthoclase due to the described limitations in the ice nucleation and mineralogy measurements in terms of concentration and temperature. We therefore abstain from any further speculation on the role of microcline in the atmosphere.

2. n_s , geo vs. n_s , BET: As the authors may be aware, there are two sub-metrics in n_s , namely geometric-based n_s and BET based n_s (Hiranuma et al., 2015, ACP; H15 hereafter). It appears to me that both n_s metrics co-exist in Fig. 5 - Atkinson, Murray and Broadley seem BET-based, while Niemand and all of your data are geometric-based. This is an apples-to-oranges comparison. Note that the difference in these two n_s metrics could be more than an order magnitude (e.g., for illite NX in H15), which would have a substantial effect on your stats presented in Table 5 & P13 L5-33. I strongly urge the authors to select either one metric (the conversion method is introduced in H15), re-evaluate your stats and re-visit your statement in P1 L21-23. Your conclusion may change.

Figure 5 is a comparison between our results (all in $n_{s,\text{geo}}$) to literature curves to give an overview on the ice nucleation ability of mineralogical components. Furthermore, we do not use the literature values in our statistical analysis, so the conversion of the literature data to $n_{s,\text{geo}}$ doesn't have any effect on Table 5 (and now also Table 6) or the interpretation. Keeping this in mind, we agree with the reviewer, that a conversion of all n_s curves into the same metric is a fairer comparison to the literature, despite the uncertainties inherent to such a conversion, particularly where values needed for such a conversion aren't reported. To acknowledge this uncertainty, we do not show single $n_{s,\text{geo}}$ curves in the revised Fig. 5 but instead the area between $n_{s,\text{BET}}$ and the calculated $n_{s,\text{geo}}$ curves.

To calculate $n_{s,\text{geo}}$ curves we have done the following:

-In the case of Atkinson et al. 2013 (A13 in the following), we have converted the K-feldspar $n_{s,\text{BET}}$ curve into $n_{s,\text{geo}}$ using a factor of 3.5 as given in the Supplementary Material of A13.

-In the case of Broadley et al. 2012 (B12 in the following), we have converted the illite $n_{s,\text{BET}}$ curve following Hiranuma et al. 2015, using the specific surface area of $104.2 \text{ m}^2/\text{g}$ given in B12 and a ratio of

the total surface area to total mass of $6.54 \text{ m}^2/\text{g}$ given in Hiranuma et al. 2015 for the same illite NX sample.

- For the Murray et al. 2011 (M11 in the following) case, we converted the kaolinite $n_{s,\text{BET}}$ curve into $n_{s,\text{geo}}$, following Hiranuma et al. 2015, using a specific surface area of $11.8 \text{ m}^2/\text{g}$ (M11), a density of $2.63 \text{ g}/\text{cm}^3$ (M11) and a mean mass-weighted diameter of 674 nm (Hudson et al. 2008). This yielded a correction factor of 3.49. It should be kept in mind that taking the mean mass-weighted diameter from another study leads to a high uncertainty, given that the size distribution might well be different from M11.

- For the newly added quartz curves we could not infer the total surface area to total mass ratio. Given that quartz is somewhat similar to feldspar however, we use the same factor of 3.5 also for quartz. This comes with high uncertainty as well, as the size distribution and also the particle shape of quartz are likely to be different from K-feldspar. The area shown in the plot covers the $n_{s,\text{geo}}$ curves calculated from the $n_{s,\text{BET}}$ curves from Zolles et al. 2015 and that calculated from the $n_{s,\text{BET}}$ curve for quartz from A13 and like the other areas stretches from the original $n_{s,\text{BET}}$ curves to the calculated $n_{s,\text{geo}}$ curves.

We have added the following paragraph on p.11, l.7-22 of the revised manuscript:

“For comparison, the n_s fits for K-feldspar from Atkinson et al. (2013), for kaolinite KGb-1b from Murray et al. (2011), for Illite NX from Broadley et al. (2012), data for quartz from Atkinson et al. (2013) and Zolles et al. (2015) and the n_s parameterization curve from Niemand et al. (2012) are shown. The K-feldspar, kaolinite, illite and quartz curves and data points were provided as $n_{s,\text{BET}}$, i.e. the surface area of the particles was measured with the Brunauer, Emmett and Teller (BET) nitrogen adsorption method (Brunauer et al., 1938). This method yields typically a higher surface area than that based on volume equivalent diameter. The literature $n_{s,\text{BET}}$ was converted to n_s using a conversion factor of 3.5 in case of K-feldspar as given in the supplementary material of Atkinson et al. (2013). For illite, we followed Hiranuma et al. (2015), using a specific surface area (SSA) of $104.2 \text{ m}^2 \text{ g}^{-1}$ (Broadley et al., 2012) and a ratio of total surface area to total mass of $6.54 \text{ m}^2 \text{ g}^{-1}$ (Hiranuma et al., 2015). Similarly, for kaolinite we used $\text{SSA} = 11.8 \text{ m}^2 \text{ g}^{-1}$, a density of 2.63 g cm^{-3} and a mean mass-weighted diameter of 674 nm (Hudson et al., 2008), yielding a correction factor of 3.49. For the same $n_{s,\text{BET}}$ values, the three quartz samples from Zolles et al. (2015) were active over a range of 10 K. No SSA values were provided therefore we used also a conversion factor of 3.5 given that feldspar is somewhat similar to quartz. This comes with a very high uncertainty, as the size distribution and particle shape of quartz are likely to differ from the K-feldspar of Atkinson et al. (2013). The K-feldspar, kaolinite, illite and quartz n_s areas cover the range from the $n_{s,\text{BET}}$ as provided in the literature and the calculated n_s to show the uncertainty inherent to the conversion. It can be seen that all desert dust samples fall between the K-feldspar and the clay mineral and quartz fits at all temperatures.”

3. Your n_s vs. Niemand’s n_s : The parameterization described in Niemand et al. (2012; N12 hereafter) is essentially based on the observed ice number normalized to the measured “total” surface area (see Eqn. 1-4 in N12), while your parameterization introduces another approximated parameter, the weighted mean aerosol surface area ($A_{w,w}$; P6 L11). Are these two n_s values apples-to-apples? The performance comparison of IMCA-ZINC-IODE to the AIDA chamber, where the N12 study was conducted, with a

reference IN material seems available in H15. The authors may extend the discussion given in H15 a bit more to justify the use of $A_{ve,w}$ in its first appearance.

The weighted mean aerosol surface area is the area corresponding to the mean of the surface area distribution. We chose to use this value because it is impossible in our case to use the total surface area, due to the fact that the frozen fraction FF is always larger than 0.1. Therefore Equation 2 of Niemand et al. 2012 cannot be approximated and we would have to consider each size bin separately. This on the other hand is not possible because we don't know which particles acted as INPs. Therefore we use the mean of the surface area distribution (which we call weighted mean aerosol surface area), which is the most comparable value to the Niemand et al. 2012 parameterization.

We agree with the reviewer that the wording "weighted mean aerosol surface area" which we used to distinguish the area from the area calculated by the mean diameter of the number concentration distribution, was confusing. We are now simply referring to it as "mean particle surface area".

Furthermore, we have rephrased p.6,l.9-11

"The surface area-weighted mean diameter ($d_{ve,w}$) was calculated from the resulting fit for each sample (see Table 1) as well as the surface area corresponding to this weighted average diameter ($A_{ve,w}$)."

to now p.6,l.22-23 of the revised manuscript:

"The mean particle surface area ($A_{ve,w}$) was calculated from the resulting fit for each sample (see Table 1) as well as the corresponding surface area-weighted mean diameter ($d_{ve,w}$)."

In addition, the general discussion of the ns concept seems scattered over the multiple sections and somehow cumbersome (i.e., P11 L3-6, P9 L 4-7 and P10 L12-16). For clarity, I suggest the authors to briefly describe the following two things within one section (any):

1) Three assumptions of the ns parameterization, which is relevant to the immersion freezing characterization, include i) the probability of ice nucleation is proportional to the available surface area of immersed aerosols, ii) ice nucleation active sites are uniformly distributed over individual particle surfaces and iii) ice nucleation occurs at specific site in a deterministic manner (predominantly T dependent).

We have added on p.9,l.9-12 of the revised manuscript:

"Earlier studies have shown that the probability of a particle to act as INP scales with the surface area of the particle immersed in a droplet (Archuleta et al., 2005; Welti et al., 2009; Kanji and Abbatt, 2010). So-called ice-active sites (Vali, 1966) are assumed on the surface of an INP in the deterministic concept (Langham and Mason, 1958). The probability of such a site to be present on a particle increases with the surface area."

And rephrased p.9,l.4-5

"It should be kept in mind that the assumption that ns stays constant with particle size most likely has limitations for complex polymineral samples such as desert dust particles."

to now p.9,l.21-22 of the revised manuscript:

“The assumption that active sites are uniformly distributed over individual particle surfaces, and therefore that n_s stays constant with particle size, most likely has limitations for complex polymineral samples such as desert dust particles. “

2) The use of the weighted mean aerosol surface area ($A_{ve,w}$) is something unique in this study (and different from N12).

As described above, we have rephrased p.6,l.9-11 to now p.6,l.22-23 of the revised manuscript to avoid any confusion:

“The surface area-weighted mean diameter ($d_{ve,w}$) was calculated from the resulting fit for each sample(see Table 1) as well as the surface area corresponding to this weighted average diameter ($A_{ve,w}$).”

“The mean particle surface area ($A_{ve,w}$) was calculated from the resulting fit for each sample (see Table 1) as well as the corresponding surface area-weighted mean diameter ($d_{ve,w}$).”

Accordingly, I suggest revising the conclusion (P 16 L15-17).

As described above, the way we calculated n_s is the method most comparable to N12. Therefore, we leave the conclusions unchanged.

4. The effect of milling: The discussion regarding the effect of milling is misleading. Specifically, the reviewer’s concerns are as follows:

P1 L18-20: This statement is misleading as it sounds like the milling process generally deteriorates IN efficiency of any composite materials. In fact, this statement contradicts to the IN results of the Atacama samples ($n_{s,milled} > n_{s,sieved}$) presented in Sect. 3.4. (i.e., P14 L29-30). I suggest rephrasing this sentence to be more specific (adding the “may” word would not help).

We have rephrased the sentence, to now (p.1, l.18-21 new manuscript):

“Furthermore, we find that under certain conditions milling can lead to a decrease in the ice nucleation ability of polymineral samples, due to the different hardness and cleavage of individual mineral phases causing an increase of minerals with low ice nucleation ability in the atmospherically relevant size fraction.”

P15 L11-13: Without any evidence of the alternation in defect densities on the Isralel dust surface or calcite surface per milling, this statement (morphology vs. mineralogy) seems too ambitious/strong. It is likely the micro pores on the surface (Subramanyam et al., 2016, Appl. Mater. Interfaces, doi:10.1021/acsami.6b01133) that enhance the IN activity of particles due to the inverse Kelvin effect (Marcolli et al., 2014, ACP). If calcite etc. is breaking up but maintaining smooth surface, there is no reason for its fragments to enhance IN activity. The authors may be aware, but the nature of active sites

is still uncertain and under investigation. Concerning these points, I suggest the authors to soften the tone of this statement.

We believe that we have made clear that this statement is valid only for the Israel sample and we do not generalize this finding (p.15, l11-12 of the original manuscript: “Thus, we conclude for the Israel sample that a morphology effect is small in comparison to the change in mineralogical composition caused by the milling in the analyzed size range.”). Furthermore, we have stated that we were not able to investigate properties like the defect density, surface irregularities etc. and base our conclusion on the finding of earlier studies which all found an increase in ice nucleation activity with an increase in defect density etc. Since the Israel sample lost ice nucleation activity with milling, an increase in defect density therefore is unlikely (except if defects are not really active sites). At least its effect on ice nucleation activity must be small. Hence, the change in mineralogy, which is expected for the sub 2.5 μm particles, likely explains the increase in ice nucleation activity. We have modified the respective sentence to make it more clear that we only refer to the Israel sample in this case. p.16, l.4-6 of the revised manuscript:

“As milling reduced the ice nucleation activity for the Israel sample, we conclude for this specific sample that any morphology effect is small in comparison to the change in mineralogical composition of the analyzed size range caused by the milling.”

5. Airborne vs. Surface: To me, the essence of this paper is summarized in P10 L9-10. This single statement and the method to reach this point would be worth a paper. The authors have already provided a great conclusion (P16 L8-10 & P1 L9-10). Focus your story along with this line. I have some questions regarding airborne dust vs. surface-collected dust as follows:

We thank the reviewer for the remarks above.

P11 L9-10: Interesting. The authors are right - according to Table 4, airborne samples in general seem having larger α ($0.48 \pm 0.07 \text{ K}^{-1}$) as compared to the average of the rest ($0.38 \pm 0.16 \text{ K}^{-1}$), suggesting high T dependency of the airborne samples. But, this is not all about T dependency. This may be rather an indication that the airborne particles miss certain active sites that can be activated at high T (owing to the difference in mineralogy??). The authors may discuss and clarify this point. This observation seems important.

We agree with the reviewer that a steep n_s curve, having a high α value in Table 4 can, in the active site concept, be described as missing certain active sites which activate at warm temperature. These two aspects (warm T active sites and T -dependency) do not contradict each other but are descriptions of the same observations. We have added on p.11, l.32-33 of the revised manuscript:

“This can be seen as an indication of active sites, which activate at warmer temperatures, being more frequent on the surface-collected samples compared to the airborne samples.”

P12 L26-27: This is another important statement. Elaborate a bit further by discussing the atmospheric relevancy of the minerals uniquely found in the airborne samples. Put clear emphasis if your bulk measurements at least suggest the atmospheric relevance of quartz/K-feldspar. According to P7 L14-15

and P7 L20-22, I have a feeling that there would be reduced amount of quartz/K-feldspar in air. Any comments? This argument seems the core research question of this manuscript.

All airborne samples stem from North Africa whereas the surface-collected samples are from sources all over the globe. Hence, the direct comparison of airborne to surface-collected samples might be biased by location dependent differences in mineralogy. This is why we don't focus on this comparison alone.

We have discussed in the original manuscript that harder minerals such as feldspar and quartz are more likely to be found in larger grain sizes whereas softer minerals such as calcite and clays tend to be found more often in the small grain sizes. (P.3, L.30-31). Furthermore, we have described in the introduction the mineralogical composition of airborne dust samples over the North Pacific, Morocco and Israel, all indicating that quartz and feldspar are commonly found in atmospheric dust particles (p.3,l.34-p.4,l.6).

To stress the atmospheric relevance of quartz further, we have added on p.3,l.26-27 of the revised manuscript:

“Quartz is commonly (5-50 wt%) found in atmospherically transported Saharan dust samples (Avila et al., 1997; Caquineau et al., 1998; Alastuey et al., 2005; Kandler et al., 2009).”

Our analysis of the six samples where the mineralogy analysis was directly representative of the particles on which we measured ice nucleation, i.e. which were in the size range smaller than 2.5 μm and therefore also likely to be lifted by wind, suggested also that feldspars plus quartz are the best predictors of ice nucleation activity. Out of these six samples, three were airborne. If the analysis is done on the airborne samples alone the same trend is seen.

We further stress the importance of feldspar and quartz even in low concentrations on p. 15, l.9-10:

“However, our results indicate that feldspar or quartz present in the bulk dust will dominate its freezing behavior down to 238 K.”

and in the conclusions on p.16, l.21-28:

“Keeping in mind that quartz is ubiquitous in atmospheric desert dust, this suggests that quartz plays a more prominent role for atmospheric ice nucleation than previously thought. The clay mineral (illite and kaolinite) and calcite content of the dust samples negatively correlated with n_s at all studied temperatures, suggesting a minor importance of these minerals for the ice nucleation activity of natural dust samples in the immersion mode, especially if quartz or feldspar are present. Atkinson et al. (2013) suggested that the global mineral dust INP concentration down to a temperature of about 240 K is dominated by feldspar. At temperatures between the homogeneous freezing limit and 240 K, where quartz is an active INP, it dominates the total INP concentration as it is much more abundant than feldspar. Our experiments on natural dust confirm this suggestion.”

and on p.17, 1-4:

“For all desert dust samples we found a high correlation of the ice nucleation activity of particles smaller than 2.5 μm with the quartz content of the dust samples. This shows that despite the dominance of the

clay minerals in the small size fraction, quartz is an important atmospheric INP component and also found in the particle size fraction with the longest atmospheric residence time. ”

Table 3: It is really bothering me that the Tenerife sample (6% microcline!) is not showing any superb IN behavior as compared to other airborne dusts. The authors said that the mineralogy inferred by XRD is representative of the aerosolized Tenerife sample (P7 L28-30). The authors disregard the contribution of atmospheric processes (P14 L17-19). Then, what is limiting the IN of this particular dust?

Mineralogy analysis is a bulk measurement, hence we cannot infer the mixing state and therefore don't know, if a) 6% of all particles are pure microcline particles or b) all particles contain 6% microcline or c) an intermediate mixing state. In case a): even if all microcline particles would activate as INP, this would not reach the lower detection limit of IMCA (10 %). In case b): it may well be that the 6% microcline aren't on the surface of each single dust particle but covered by less active minerals (e.g. Zhu et al 2006). For case c) both effects could play a role.

6 % microcline is clearly at the lower end of K-feldspar content in our samples. An effect on ice nucleation from the 6 % of microcline should be detectable at warmer temperatures but unfortunately no FRIDGE measurements are available for this sample. The microcline fraction is included in the K-feldspar fraction used for the correlations in section 3.3.

6. Table 5 (P13 L5-P14 L24): This part includes a number of flaws (i.e., ns, geo vs. ns, BET) and needs substantial improvements. In fact, with given limitations/assumptions in P14 L6-24, I am not convinced that these 'relative' correlations add much meaning to the manuscript. Do the authors really require this part to draw their conclusion? This whole statistics part of the manuscript could be deleted?

The conversion of the literature $n_{s, \text{BET}}$ to $n_{s, \text{geo}}$ has no effect on the correlations because the whole analysis was consistently done using $n_{s, \text{geo}}$. We believe that this approach of directly correlating the mineralogical components with the ice nucleation ability (n_s) is a novel approach and the results are valuable. We have added the correlation plots in the supplementary material and have added if the correlations are statistically significant in Table 5 and the new Table 6. We agree that the method has limitations due to the low number of samples and the comparison of a volume property (mineralogy) to a surface property (ice nucleation ability). Therefore, the correlations are often statistically not significant. Both limitations have already been discussed in the original version of the manuscript. Despite these limitations, the results shed new light on the importance of mineralogy and in particular of quartz for ice nucleation of atmospheric dust. We believe this part is clearly required to draw the conclusion that quartz seems to play a more important role for ice nucleation of natural desert dust samples at the investigated temperatures than believed so far and much more than do clays or any other minerals apart from feldspar. This has been suggested by Atkinson et al. 2013 based on mineralogical maps and n_s of single minerals. Our study confirms this now for airborne and surface-collected natural dust samples. Since single mineral studies of the ice nucleation ability of quartz have yielded inconsistent results and focus has been placed on the role of clays and in recent years on feldspar, this is an important new finding. We have added on p.16, l.25-28 of the revised manuscript:

“Atkinson et al. (2013) suggested that the global mineral dust INP concentration down to a temperature

of about 240 K is dominated by feldspar. At temperatures between the homogeneous freezing limit and 240 K, where quartz is an active INP, it dominates the total INP concentration as it is much more abundant than feldspar. Our experiments on natural dust confirm this suggestion. ”

Again, I generally agree that quartz and microcline are IN active and may have potential importance in the atmospheric IN. What would be more valuable to see is if there is any ‘absolute’ relation between mineralogy and IN. For example, the authors may explore if the natural dust n_s (or FF) can be optimized/predicted by its composition and associated individual n_s scaled to the surface fraction (SF) of each component ($x_1, x_2 \dots$ to x_i) [i.e., $n_{s,dust} = (n_{s,x1} \times SF_{x1}) + (n_{s,x2} \times SF_{x2}) + \dots + (n_{s,xi} \times SF_{xi})$]. Note that H15 attempted, but no success. Give more in-depth thoughts regarding the role of particular mineral IN.

This is an interesting suggestion. Please note that the method of correlating the sum of minerals that are assumed to contribute to the ice-nucleation activity at the investigated temperatures with n_s is a simplified version of the proposed method. However, we do not have surface specific mineralogical information, therefore no surface fraction of each component can be determined. As a first attempt one could assume surface fraction to equal volume fraction (see O’Sullivan et al. 2014). But given the uncertainty related to the surface-specific mineralogy, the conversion from $n_{s,BET}$ to $n_{s,geo}$, the polydisperse size distribution of our sample, the wide spread of n_s of minerals that are supposed to be the same within one single study (e.g. plagioclase and K-feldspar in Harrison et al. 2016, quartz in Zolles et al. 2015) or between studies (comparison provided in Harrison et al. 2016 of their own study and Zolles et al. 2015, Emersic et al. 2015 and Atkinson et al. 2013 for feldspars) and the failed attempt of H15 for a rather simple dust sample, we believe that the method proposed by the reviewer is too ambitious to give meaningful results for our dust samples at the moment. This however would be an excellent suggestion for the future, when monodisperse natural dusts are measured with the same instrument/method at the same temperatures as reference minerals and the surface fraction of each component can be determined with reasonable accuracy.

Specific comments/suggestions

Introduction

P2 L22: Briefly explain what the authors mean for ‘contradicting results’.

We have replaced

“The IN ability of soot (Brooks et al., 2014; Kulkarni et al., 2016) and secondary organic aerosol (Prenni et al., 2009; Ignatius et al., 2015) at heterogeneous freezing temperatures is still debated as contradicting results have been observed.”

with (p.2,L.21-25 of revised manuscript):

“The ice nucleation ability of soot (Brooks et al., 2014; Kulkarni et al., 2016) at heterogeneous freezing temperatures is still debated as contradicting results were observed, spanning from hardly any ice nucleation ability at $T > 236$ K (Kanji et al., 2011) to up to 3 % of soot particles active in the immersion mode (DeMott, 1990). Similarly, the reported freezing behavior of secondary organic aerosol particles varies from inefficient to comparably efficient (Möhler et al., 2008a; Prenni et al., 2009; Wang et al., 2012; Ladino et al., 2014; Ignatius et al., 2016).”

P2 L24: Briefly describe ‘certain minerals’.

“Recently, also certain minerals have been identified to nucleate ice at temperatures up to 271 K (Harrison et al., 2016).”

has been replaced by (p.2,l.27-28 revised manuscript):

“Recently, the K-feldspar microcline and the Na-feldspar albite, both minerals found in atmospheric dust, have been identified to nucleate ice at temperatures up to 271 K (Harrison et al., 2016).”

P2 L25: “For the implementation...” - I suggest starting a new paragraph here regarding the IN parameterizations. This way, the previous paragraph (L18-25) reads more like a general introduction of atmospheric INPs and their diversity (biological and non-biological).

done

P3 L19: The authors may mention that the abundance of quartz in atmospheric dusts is consistently high (i.e., ~10% in volume) in the size range of ~1 to 35 μm geometric diameter (see Table 1 of Kandler et al., 2009, Tellus; cited in this paper), which would add the atmospheric relevance and general importance of quartz. Such information could also fit in P4 L1-3.

We added (p.3,l.26-27 of the revised manuscript):

“Quartz is commonly (5-50 wt%) found in atmospherically transported Saharan dust samples (Avila et al., 1997; Caquineau et al., 1998; Alastuey et al., 2005; Kandler et al., 2009).”

and (p.3,l.32-33):

“Their samples contained quartz fractions between 1 and 26 wt%, K-feldspar fractions between 0 and 10 wt%, and plagioclase fractions between 0 and 22 wt%.”

Methods

P5 L26-27: Does the size distribution of particles in the chamber change over 3.5 hours (i.e., the filter sampling period; P8 L15-16)? Large particles settle down faster than the smaller ones, and the authors infer that certain mineral compositions are large in their sizes (e.g., P15 L33). Please clarify and discuss potential consequences, if any.

We state on p.6, l. 11-13 of the original manuscript (now p.6, l.14-16), and in Table 1, the mean surface area varied by 6-24 % over the course of a day of experiments (which lasted typically between 5-12h), except for the Great Basin sample (64 %). Since the 3.5h filter sampling period is lower, the variation is also smaller. Nevertheless, this maximum error has been used when calculating the uncertainty of n_s (see p.10, l.18-19 of original and p.10, l.21-22 or revised manuscript). Since the uncertainty resulting from this decrease in surface area is small compared to the uncertainty in the INP concentration, we do not discuss potential consequences. We have added on p.10,l.29-30 of the revised manuscript:

“The error bars in n_s are derived by error propagation from the error in FF and ($A_{ve,w}$) and are dominated by the error in FF.”

P5 L28: This background (~10%) seems high for the IN research. Any justification or measurements that this much background particles have no substantial contributions to heterogeneous freezing?

We agree that the value is comparably high. But since we only consider frozen fractions between 10-90% in the IMCA-ZINC experiments, we have no influence of the background in our analyses even if all background particles would act as INP and none of the sample particles were INPs. Furthermore, it is expected that the residual particles are of the smallest sizes and hence least ice nucleation active. Nevertheless, in case of the FRIDGE measurements there could be an influence even though the observed results suggest there was hardly any: The order of measurements was Egypt, then Atacama milled, then Etosha, and at last Taklamakan. The Atacama milled sample had an order of magnitude higher n_s than the other samples. So, the influence of residuals from the Egypt sample on the Atacama milled measurements (which would decrease the Atacama milled n_s) would be less than 1 % if the n_s of the Egypt sample residuals was the same as during the actual experiment. Due to their higher n_s , residual particles from the Atacama milled sample could have increased the observed n_s of the Etosha sample, even though less than 6 % of the particles were residuals. This could potentially explain why we found a comparably high ice nucleation activity which cannot be related to any of the known ice nucleation activities of the mineral components. As shown in Figure 2 of this document however, the ice nucleation activity at 240-242 K seems to be related to organic/biological material. This does not rule out a large influence of the Atacama milled residual particles on the Etosha sample in the FRIDGE measurements but makes it unlikely.

The Etosha and the Taklamakan sample finally show similar n_s in the FRIDGE data. This is in line with the results from the IMCA-ZINC measurements at lower temperature, suggesting that there is only negligible influence if any.

P5 L30: Briefly describe "coincidence effects" or cite proper papers.

We have added (p.6, l.12 of the revised manuscript)

"For the IMCA-ZINC measurements the particle concentration was diluted to about 60 cm^{-3} to avoid coincidence effects in the detector which occur if more than one particle is present in the laser beam of the detector Nicolet et al. 2010."

P7 L2: According to Table 3 of K16 (Kaufmann et al.), the Israel sample contains some sanidine. Moreover, sanidine in the natural surface samples seems non-negligible as 6 out of 12 natural surface samples examined in K16 shows the presence of sanidine. Was your sample totally different from K16? Or the the sample may not be completely homogeneous in terms of mineralogical distribution even within a same batch? Was the XRD data interpretation method somewhat different from K16? Please clarify.

The Israel and the Namib (Etosha) samples are the same as studied by K16 as we stated on p.5, l.7-8 (now p.5, l. 17 of the revised manuscript): *"The Israel sample and the Namib sample are from the same batch as those studied in Kaufmann et al. (2016)."*

The XRD data interpretation method was done slightly differently from K16. K16 used in addition to AutoQuan the software EVA, whereas we used AutoQuan and only for unidentified minerals used EVA. The Israel sample only contains about 2 % K-feldspar. Due to this low fraction and since the feldspar

analysis is one of the most challenging part of the mineralogical composition analysis of a polymineral sample, the different results are reasonable.

We have replaced (p.7,l.2 of the old manuscript):

“No sanidine feldspar was found in any of the samples.”

with (p.7,l.15-17) of the new manuscript:

“In case of a low K-feldspar content of a few wt% it was not possible to determine if K-feldspar was present as microcline, orthoclase or sanidine or a mixture of the different phases. Values are given for the K-feldspar with the best Rietveld fit result.”

P7 L20-22: Doesn't this just mean that a majority of large particles (up to 32 μm) break up by the RBG milling? I mean that RBG may do more than just aerosol dispersion, correct? Long story short, is it really fair to assume that those aerosolized particles are identical to the sieved bulk used for XRD as the authors mention in P13 L20-23? Further clarification seems necessary. Accordingly, the authors may consider rephrasing the relevant text in the conclusion (i.e., P15 L29-32). 2.5 μm sounds like a magic number as it is right now.

The RBG may deagglomerates but does not break up or grind particles as the residence time is low and it is a stainless steel brush not a mill or balls. Even if particles would break up during generation, their mineralogical composition would remain the same. If the mineralogical composition is representative depends only on the fraction of particles that make it into the tank reservoir compared to those that are left behind in the cyclone. This fraction has been semi-quantified as described in the manuscript.

The 2.5 μm is simply the D_{50} of the cyclone used during aerosol generation (p.5, l26 of the original manuscript). There is nothing magical about this number, but it is the size cut-off we used consistently for particle generation.

P7 L28-30: For the reason given above, I am not sure if the authors can asset like this.

We hope we could convince the reviewer of the plausibility of the method.

Results & Discussion

P10 L3: The homogeneous freezing regime presented in this manuscript is based on CNT? Or anything different? I suggest adding proper reference(s) here at least.

The homogeneous freezing regime indicated in Figure 4. a) - c) is derived from CNT for 10 μm droplets using formulae and constants given in Ickes et al. 2015. Reference experiments on homogeneous freezing with the IMCA-ZINC setup can be found in Hoyle et al. 2011 Fig.5. We add a reference to Hoyle et al. 2011 and Ickes et al. 2015 to the figure caption.

P10 L9-10: I encourage the authors to clearly state that the same trend holds true for another metric, $n_s(T)$.

We have added on p.10,l.33-34 of the revised manuscript:

“Like their FF, the range of n_s of the Saharan samples is comparable to those of the non-Saharan ones (Fig. 4f).”

P11 L2-3: It looks to me that the Niemand parameterization falls in the middle of your 15 $n_s(T)$ spectra. Please clarify what “rather at the lower end” means quantitatively.

We have added on p.11,l.26 of the revised manuscript as subclause:

“(a factor of 3 to 4 below the average $n_s(T)$ of all measured curves, not shown).”

P11 L13-14: The word “overpredicts” implies that the Niemand parameterization (N12) is wrong. The authors may want to soften the tone and rephrase. Your assumption vs. assumption made in N12 would be discussed here.

We have rephrased the sentence to (p.12,l.1-3 of the revised manuscript):

“The parameterization from Niemand et al. (2012) predicts one to two orders of magnitude higher n_s than measured by FRIDGE. Only for the Atacama milled sample at $T = 251 - 256$ K the parameterization shows about 30 % higher values than the measurements.”

We believe that a parameterization cannot be right or wrong. At first it is simply representative for the samples which were used to define it. Even for those, there is an uncertainty related to the parameterization because of the scattering of the data points included. It must be tested if the parameterization also represents other samples and if not, why not. This is what is done in our manuscript.

P11 L11-20: I do not find the scientific significance of having FRIDGE data included in this manuscript. The authors briefly discuss about the FRIDGE results in this part and only a bit more afterward. The reviewer does not find that the FRIDGE results are complementing IMCA-ZINC-IODE, vice versa. In addition, no proper justification for why FRIDGE was conducted for a subset of samples is provided. Does the authors' conclusion change without the FRIDGE data?

The scientific significance of having FRIDGE data included is given on p.4, l.18-19 of the original manuscript:

“This allowed examination of immersion freezing at temperatures between 250 and 262 K, hence covering a wide range of heterogeneous freezing temperatures.”

We have rephrased it to p.4, l.31-32 of the revised manuscript:

“This allowed examination of immersion freezing at temperatures between 250 and 262 K, covering a wider range of heterogeneous freezing temperatures than would otherwise be possible with IMCA-ZINC alone.”

IMCA-ZINC is not sensitive enough to measure FF below 10 % reliably. Therefore, FRIDGE was used to extend the covered temperature range. This was not initially planned as a part of the dust measuring

campaign and for logistical reasons, the particle collection for FRIDGE was only done at the end of the campaign. Therefore only four samples were collected. We don't see a value of adding this information to the manuscript.

The authors' conclusions on K-feldspar determining mainly the ice-nucleation behavior of natural dust at $T > 250\text{K}$ (p.15, l.18-20 of the original manuscript) is entirely based on the FRIDGE data. We therefore see a high value in keeping the FRIDGE data.

Fig. 5: Adding quartz reference spectrum (Atkinson et al., 2013) would be nice.

We did not show a reference spectrum for quartz in the original manuscript because different authors found very different results even within a single study (Zolles et al. 2015, Atkinson et al. 2013). However, since we are now showing areas of n_s than lines, we have included an area of n_s for quartz.

Fig. 5 Cont'd: Having another panel depicting the highest-median-lowest spectra of airborne n_s vs. those of surface-collected n_s would add some clarity and strengthen the paper.

We agree, that's a great suggestion. We have added such a Figure (6a) in the revised manuscript. Furthermore, we have added the following paragraph on p.12,l.9-18:

“Figure 6a shows the median and minimum to maximum $n_s(T)$ range of the airborne and surface-collected samples. This illustrates that the n_s range of the airborne samples falls in the lower half of the n_s range of the surface-collected samples or even below. It shows that for immersion mode ice nucleation surface-collected dust samples are not representative for airborne dust samples, which all stem from North Africa, the world's largest source of atmospheric dust. This might be caused by a non-representative surface-dust collection, e.g. soil rather than dust is collected which has a different size distribution and composition, or dust from a location where threshold wind velocities for dust lifting are not reached. Another cause could be that atmospheric processes taking place during or after particle lofting may alter the particle surface and decrease the ice nucleation ability which has been suggested to occur in the field (Cziczo et al. 2013) and laboratory (Sullivan et al. 2010, Augustin-Bauditz et al. 2014). The potential effects of mineralogy on the ice nucleation activity at different temperatures is investigated in the following section.”

P12 L4-9: The discussion given here makes the review think something other than minerals (e.g., P5 L15-17) are competing for IN at given T range. The reviewer is aware that the focus of the current work is on mineralogy vs. IN (and no biological INP perspective at all). That said, the authors should extend the discussion regarding the potential bioaccessibility of dust surface (Augustin-Bauditz et al., 2016, ACP; O'Sullivan et al., 2016, ACP) and other IN species that might be present in soil (O'Sullivan et al., 2014, ACP; Tobo et al., 2014, ACP; Pummer et al., 2015, ACP). Such information would strengthen the manuscript.

We agree with the reviewer’s comment. We performed parallel measurements of particle fluorescence as a proxy of biological material, but due to technical reasons these data were not useable. We were able to repeat some of the deposition/condensation measurements, which will be included in the part 2 paper associated with this manuscript (Boose et al., 2016). The experiments were performed using the portable ice nucleation chamber PINC with the original samples as well as samples heated to 300°C (573 K) for 10h. PINC, which works like ZINC but is shorter, has a shorter residence time and sampled particles which were dry rather than pre-activated. Therefore, the results are not necessarily comparable to the immersion mode results from the current manuscript because full droplet activation prior to freezing is not guaranteed.

Figure 2 shows the condensation mode n_s at 240 and 242 K and at the highest RH_w sampled (100 – 105 %) to mimic immersion freezing (Hiranuma et al. 2015). Filled symbols refer to unheated, open symbols to heated samples. Heating had little to no effect on the ice nucleation activity of the Australia, the Atacama milled and Peloponnese samples. The Etosha sample lost most of its ice nucleation activity after heating, which is the sample for which we could not relate the comparably high ice nucleation activity to its mineralogy as it consisted mostly of calcite and dolomite, which have a low ice nucleation activity (Atkinson et al. 2013, Kaufmann et al. 2016), and ankerite, of which the ice nucleation activity is not known (Kaufmann et al. 2016). This is consistent with the manuscript conclusions that the ice nucleation activity is mainly caused by the mineralogy of the dust samples. The situation may be different at warmer temperatures but unfortunately we cannot repeat investigations at these conditions due to the small remaining sample size. The full data from these additional measurements and further analysis will be shown in the second paper of the series (Boose et al. 2016).

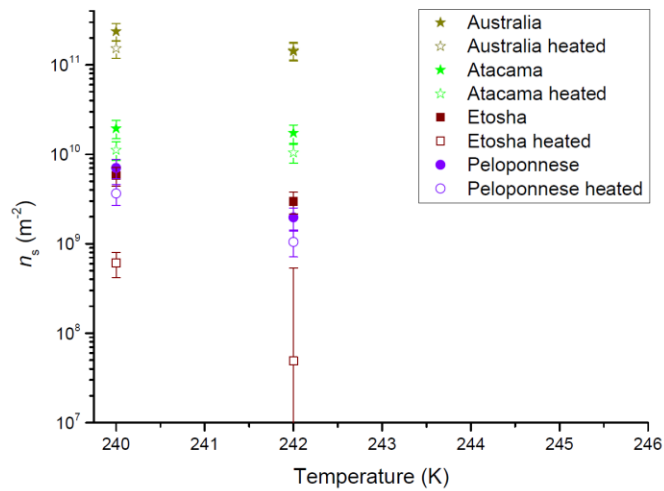


Figure 2: Condensation mode n_s for selected heat treated samples.

In the revised manuscript we have added a paragraph on non-mineral matter mixed with the dust and its potential effects on the ice nucleation ability of the dust (p.4,l.15-20 of the revised manuscript):

“Non-mineral matter, which can become internally or externally mixed with the mineral dust before or after emission, may affect the ice nucleating behavior of the dust. Sulfuric acid (Sullivan et al., 2010; Augustin-Bauditz et al., 2014) or secondary organic aerosol coating (Möhler et al., 2008a) has been observed to decrease the ice nucleating ability while exposure to ozone (Kanji et al., 2013) or the presence of ammonium sulfate (Boose et al., 2016b) has been suggested to improve it. Biological material can adsorb to mineral dust, enhancing its ice-nucleating ability (Schnell, 1977; Conen et al., 2011; O’Sullivan et al., 2016).”

We have rephrased p.5, l.15-16 (now p.5, l.28-30 of the revised manuscript):

“All natural dust samples are expected to be very heterogeneous, i.e. external and internal mixtures of different minerals and potentially biological material (Meola et al., 2015).”

“The composition of natural dust samples is presumed to be heterogeneous, i.e. external and internal mixtures of different minerals and potentially containing organic or biological material (Meola et al., 2015). ”

We have replaced p.12, l.8-9 (now p.13, l.7-9 of the revised manuscript):

“No study so far has investigated the IN behavior of pure ankerite. Thus it remains unclear what leads to the observed high IN activity at $T < 242$ K of the Namib sample.”

“Thus, the high ice nucleation activity at $T < 242$ K of the Etosha sample is not explainable by the known ice nucleation ability of its mineral components. To our knowledge, no study so far has investigated the ice nucleation behavior of pure ankerite.”

We have replaced p.14, l.22-23:

“If this is due to reasons other than mineralogy such as coating as suggested by Kaufmann et al. (2016) or if the present minerals ankerite, dolomite or muscovite can lead to a high IN activity at $T < 243$ K under certain circumstances is not known.”

with (now p.15, l.15-16 of the revised manuscript)

“Measurements in the condensation mode, which are the subject of part 2 of this study, suggest that the ice nucleation activity of this sample is in large part related to organic or biological material mixed with the dust.”

Conclusion

P15 L18-19: Which data infers the FF and $n_s(T)$ results at temperatures above 250 K? I do not see them in Fig. 4. FRIDGE?

The n_s of FRIDGE at $T > 250$ K is shown in Fig 5b). We have deleted “the FF” on p.15, l.19 (original manuscript). The sentence reads now (p.16, l.12-13 in the revised manuscript):

“The comparison showed that at temperatures above 250 K the highest n_s is related to the highest fraction of K-feldspars in the sample...”

P16 L2-4: Your data presented in Tables 2 and 3 (i.e., Atacama milled vs. sieved; Israele milled vs. sieved) seem contradicting to this statement. For instance, I do not see any increase in the quartz fraction.

It is correct that for the Israel and the Atacama sample, the milling did not lead to an increase in the quartz fraction. This has the following reasons: For the Israel sample, the milled and the sieved sample should have the same mineralogy because the batch which was already sieved to below 32 μm was further milled. In the case of the Atacama sample, were the original pre-processed sample was milled and compared to the < 32 μm sieved fraction, the quartz fraction actually is slightly lower in the milled compared to the sieved sample. However, one can see that the K-feldspar (orthoclase) fraction is 10.5 % higher than in the sieved sample. As feldspar is also hard and in addition due to its reactivity less common in the small particle fraction, the milling increased the feldspar fraction, resulting in a reduction of the quartz fraction.

We refer here mostly to the Morocco and Australia samples which had almost no particles in the size range < 32 μm and therefore needed to be milled. They consisted both mainly of quartz and feldspar.

P16 L6-8: This part seems contradicting to the previous statement (that is, P15 L33).

We have now changed the sentence on p.15,l.33:

“Quartz is a comparably hard mineral and thus less common in the smallest dust size fractions. Since we measured IN activity of particles smaller than 2.5 μm and found a high correlation with the quartz content of the dust samples we show that quartz is nevertheless an important atmospheric INP component ”

to p.17,l.1-4 of the revised manuscript:

“For all desert dust samples we found a high correlation of the ice nucleation activity of particles smaller than 2.5 μm with the quartz content of the dust samples. This shows that despite the dominance of the clay minerals in the small size fraction, quartz is an important atmospheric INP component and also found in the particle size fraction with the longest atmospheric residence time.”

With this change we now in the revised manuscript want to say that one could expect that quartz should be only found in the larger size fraction of dust particles because of its hardness (rather than stating that as a fact as previously done so). We then follow this with a discussion demonstrating that in our results quartz can be indeed found in the atmospheric size fraction and can be important to predicting ice nucleation behavior.

Technical comments/suggestions

P1 L1-2: “Traditionally, clay minerals were assumed to determine...” → “Since natural dusts are composite in nature, clay minerals were typically used as a proxy to determine...”

We prefer to leave the sentence as it is. We base this statement on early publications on ice nucleation, e.g. Kumai 1961. Furthermore clays were used as a proxy because clays can form a substantial component of natural dust.

P1 L14: "...activity in a given sample above 253 K that can be attributed to..."

We prefer to keep the wording "at 253 K" because we did our analysis of correlating n_s with mineralogy only at 253 K and not at warmer temperatures. Hence, "above 253 K" would be speculative.

P1 L17 and hereafter: Use the Italic font for T throughout the manuscript.

done

P2 L3: "determine" → "influence" - Besides primary ice nucleation, secondary ice processes can also contribute to the lifetime of clouds.

done

P2 L20: Pummer et al. missing as INM references

"Pummer et al. 2012" was added

P2 L21: DeMott et al. missing as soot IN references

"DeMott et al. 1990" was added.

P2 L32: ", respectively"

A comma was inserted.

P2 L32-P3 L1: Too many things packed in a single sentence. Split into two sentences.

done (p.3, l.2-4 of the revised manuscript)

P2 L33: between → amongst

done

P3 L13: "K-feldspars (microcline, orthoclase and sanidine) were..."

done

P3 L20-22: Redundant (P3 L4-6)

We have deleted the first half of the sentence (p.3,l.24-26 of the revised manuscript):

"It has been proposed that differences in the surface structure can lead to different IN abilities of quartz samples and it is suspected that functional groups on the surface of feldspars and quartz are responsible for their higher IN ability (Zolles et al., 2015)."

“It is suspected that functional groups on the surface of feldspars and quartz are responsible for their higher ice nucleation ability (Zolles et al., 2015) but it is unknown where the high variability stems from.”

P3 L27: Delete “hence”. Size and composition are inherently related. Is that what the authors want to say? If so, state so.

The sentence

“It has been observed that the size distribution and hence mineralogical composition of dust changes during its emission and transport compared to that on the surface (D’Almeida and Schütz, 1983; Murray et al., 2012; Knippertz and Stuut, 2014). The reason for this is a size dependent mineralogical composition caused by differences in the hardness, cleavage and shape of minerals.”

has been changed to (p.3, l.34 - p.4, l.2 of the revised manuscript):

“It has been observed that the size distribution of dust changes during its emission and transport compared to dust on the surface. This leads to variations in the mineralogical composition of the dust, (D’Almeida and Schütz, 1983; Murray et al., 2012; 35 Knippertz and Stuut, 2014) as the mineralogical composition is size dependent due to differences in the hardness, cleavage, shape and reactivity of minerals.”

P3 L31: No “hence”

done

P4 L29: “The GPS coordinates of our collection sites...”

done (p. 5, 7-8)

P5 L15-16: “The composition of all natural samples are presumably heterogeneous...”

done (p.5, l. 28-30)

P6 L10: “... sample (see ...)”

done (p.6, l. 23)

P6 L15: “... (sieving/milling)”

done (p.6, l. 29)

P8 L5: “The cloud droplets” → “The simulated cloud droplets” or “The activated droplets”

We have deleted “cloud” (p.8,l.16 of the revised manuscript).

P8 L15: “subsequent” → “independent”

done (p.8, l. 29)

Eqn. 4: Missing negative sign on the RHS of Eqn.4?

done (p. 9)

P9 L4-5: Awkward sentence. I suggest rephrasing.

The sentence

“It should be kept in mind that the assumption that n_s stays constant with particle size most likely has limitations for complex polymineral samples such as desert dust particles.”

has been rephrased to (p.9,l.21-22 of the revised manuscript):

“The assumption that active sites are uniformly distributed over individual particle surfaces, and therefore that n_s stays constant with particle size, most likely has limitations for complex polymineral samples such as desert dust particles.”

P9 L14-15: Awkward sentence. Simply say something like our measurements are valid with χ in the range of 1.1 to 1.6.

We have rephrased this and the preceding sentence

“Since one mode was detected in each instrument’s size range, a variation of the shape factor was tested to check if a better overlap of the two size distributions could be achieved. Within realistic limits ($1.1 \leq \chi \leq 1.6$) for shape factors of atmospheric dusts (Alexander, 2015) the two modes remained distinguishable and are thus assumed to be real.”

to (p.9,l.32-p.10,l.3 of the new manuscript):

“Since one mode was detected in each instrument's size range, the shape factor χ was optimized to give the best overlap of the two size distributions. For any shape factor within realistic limits for atmospheric dusts ($1.1 \leq \chi \leq 1.6$, Alexander 2015) the two modes remained distinguishable.”

P9 L21-22: Awkward sentence. Simply say ...because of the presence of predominantly large particles... or something similar.

done (p.10, l.9-10)

P 10 L12-13: “Due to the heterogeneous and possibly size-dependent particle compositions, a partial step function like activation spectrum could be...”

done (p.10, l.23-24)

P10 L27-28: two consecutive “overalls” are bothering.

The second “overall” was deleted. (p.11, l. 6)

P11 L2: “The comparison between the Niemand parameterization and our parameterizations based on...”

We have rephrased the sentence to (p.11,l.24-26 of the revised manuscript):

“For $T < 250\text{ K}$ the parameterization falls in the lower end of the range of n_s observed for our broader collection of global surface collected and airborne dust samples (a factor of 3 to 4 below the average $n_s(T)$ of all measured curves, not shown).”

P11 L5: “polydisperse nature” → “heterogeneous properties”

We have replaced “nature” with “size distribution” (p.11, l.28 of the revised manuscript)

P11 L23-24: This sentence does not fit in here. I suggest deleting.

We have added another sentence to create a smoother transition (p.12,l.20-21) of the revised manuscript:

“By analyzing the bulk mineralogy we investigate if the different natural dust's mineralogical composition explains the observed ice nucleation activity.”

P14 L34: The authors may want to remind the reader that $2.5\ \mu\text{m}$ is D_{50} of your cyclone.

We added “(the D_{50} cut-off of the particle generation system used)” after “ $2.5\ \mu\text{m}$ ” on p.16, l.5 of the revised manuscript.

P15 L10-13: “However... Thus...” - Awkward transition. Rephrase.

The sentences have been rephrased to (p.16, l.3-6 of the new manuscript):

“However, an increase in defect density and surface irregularities has been shown to increase the IN activity of monomineral or single compound samples. Thus, we conclude for the Israel sample that a morphology effect is small in comparison to the change in mineralogical composition caused by the milling in the analyzed size range.”

“An increase in defect density and surface irregularities has been shown to increase the ice nucleation activity of monomineral or single compound samples. As milling reduced the ice nucleation activity of the Israel sample, we conclude for this specific sample that any morphology effect is small in comparison to the change in mineralogical composition of the analyzed size range caused by the milling.”

P15 L22 above or below?

We have deleted the respective sentence.

P15 L27-28: I do not understand this sentence. Rephrase.

The sentence

“The size dependent enrichment of different minerals leading to differences in n_s highlights the interplay between IN ability and atmospheric relevance of certain mineral phases.”

was replaced by (now p.16, l.29):

“The variation in mineralogy with particle size leads to variations in n_s .”

P15 L29-32: Awkward sentence. Rephrase.

Sentence rephrased (p.16, l.33-p.17, l.1 of the revised manuscript):

“The focus of earlier IN studies on the clay mineral IN behavior is supported by the observation that three of the four airborne samples (Crete, Peloponnese and Tenerife), which had been long-range transported and were almost entirely in the size range < 2.5 μm, had the highest clay mineral fraction.”

“Three of the four airborne samples (Crete, Peloponnese and Tenerife) had the highest clay mineral content and were amongst the least ice nucleation-active samples.”

P16 L 12: “can not” → “cannot” (to be consistent with the rest of the manuscript)

done

P16 L 21-22: Redundant (P4 L22)

We deleted the sentence.

References:

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