We thank Reviewer T. Zolles for his constructive comments. We reproduce reviewer comments in blue in the following. Amended versions of the paper are given in *italics* for new sections and smaller red text for the original text. We have numbered the reviewer comments for clarity.

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 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) **Sampling and sample treatment** The decision to sample and compare air-born and ground samples is very well justified. As was stated the mineralogical composition between the used <2.5 µm fraction and the bulk (<32) that was analyzed with XRD may vary, but the general findings are very likely not influenced as neither mineral will be totally absent. This may influence the correlations in 3.3. Nevertheless, natural dusts and minerals are rather often found to have organic and biological material absorbed on their surface. It cannot be excluded that some are left on the dust particle surface. Gently heat treatment could have been used to destroy all organic material.*

We agree with the reviewer's comment (see also response to SC by Russ Schnell). We had performed parallel measurements of particle fluorescence as a proxy of biological material, but due to a failure of the xenon lamps these data were not useable. Unfortunately due to the small sample amounts, we are limited in repeating or performing additional measurements. We were able to repeat some of the deposition/condensation measurements with heat treatment as suggested by Reviewer Zolles. We will include the results of these 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 at 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 as droplets as was done in the present work. 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 1 shows the condensation mode n_s at 240 and 242 K. 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. Only 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 minerals with low ice nucleation activity. 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 the warmer temperatures in the manuscript (> 250 K) but unfortunately we cannot do 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).

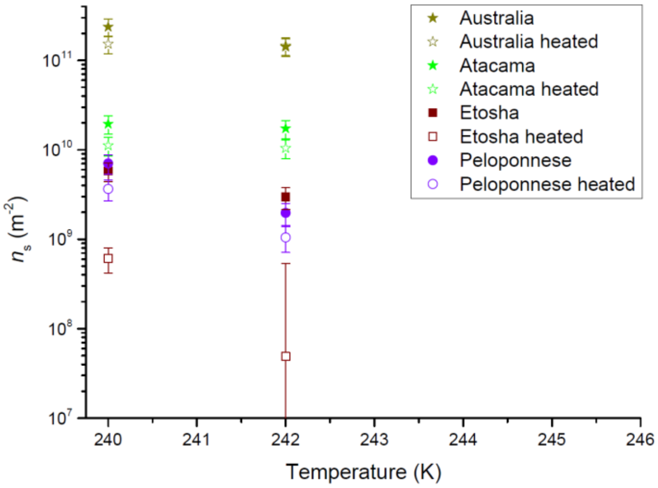

Figure 1: Condensation mode n_s for selected heat treated samples.

In the current 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,.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.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.27-28 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.*”

2) Secondly the dusts did undergo different treatments to produce sufficiently small grains. The surface samples were sieved or milled and with the Israel and Atacama sample two fractions were created. It is clear that the Australia and Morocco samples had to be milled due to not small enough dust fraction, but the authors give no reason why not of all other samples a milled and a sieved fraction was created. There is no clear explanation why in particular the Atacama and Israel samples were chosen. To study the effect of milling a few samples are probably sufficient, but this may lead to an increased uncertainty in the correlations in 3.3. If certain sample sites have an increased importance due to a milled and a sieved fraction.

We agree that it would be preferable if all ground-collected samples could have been treated the same way. Unfortunately, this was not possible since the volume – fraction of particles < 32 µm was typically only on the order of 1/1000 or less of the initial, pre-preparation sample. Only the Israel sample had a large enough mass fraction of particles smaller than 32 µm to allow the direct comparison of a milled sieved fraction to the unmilled sieved fraction. The Atacama sample also had a small fraction of < 32 µm but enough to do a comparison between a milled non-sieved fraction and an unmilled sieved fraction. For none of the other samples the amount of particles < 32 µm was large enough to allow further comparisons.

We have added on p.5,l.27-28 of the revised manuscript:
“*The sub-32 µm size fraction of the other samples was too small to investigate the milling effect.*”

We have repeated the correlations in section 3.3. including only either the sieved or milled Atacama and Israel samples. The trends stayed consistent with those presented in section 3.3. Therefore, an overrepresentation of these two sampling sites does not affect the conclusions presented in the paper.

*3) **Mineralogy analysis** The authors give a good description of the shortcomings of the analysis method as well as that the composition may not be valid for the particle sizes used in the freezing experiment.*
Page 4/30: Similarly, the milling of the Israel sample likely interfered with the preferred direction of the minor components in the sieved samples, leading to an observed reduction of these mineral fractions (e.g. illite, kaolinite, plagioclase) in the milled compared to the sieved sample.
There is a reduction in every relative fraction apart from the very soft calcite which, as the authors state, is probably increasing due to its softness upon milling. Why are these minor components of particular interest? What is the potential explanation of the observation?

For the ice nucleating behavior of the dust samples all components are of interest due to the unknown mixing state of the particles. In theory it could be that a minor component by weight is present on all particles and responsible for the ice nucleation. The minor components are mentioned here in regard to the preferred orientation during XRD to explain the differences between the Israel milled and sieved sample. In theory the mineralogy should be the same because the milled Israel sample was derived from milling the sieved Israel sample.

Preferred orientation of minerals can lead to an over- or underestimation of some minerals, which we suggest (p.7,l.10-14 of the revised manuscript) explains the difference between the milled and unmilled Israel sample.

4) Page 7/14: Natural mechanical weathering thus likely has enhanced the clay mineral and calcite content in the smaller particle fraction whereas feldspars and quartz tend to be found in the larger size fraction. Is the same expected for the sieved and milled samples in this study? In respect to the above mentioned part, it would be important to conduct milling on all ground samples.

The milling breaks up hard minerals such as quartz and feldspar and grinds them to smaller sizes. Therefore it changes the size distribution and size dependent mineralogy of the milled sample compared to the sieved sample. In this regards, the milled samples may show a different size dependent mineralogy compared to unprocessed dust. However, all unmilled samples, including the airborne ones, contain quartz and (except the Etosha (Namib) sample) also feldspar. Furthermore, as described on p.4,l.6-14 of the revised manuscript, quartz and feldspar are commonly found in airborne dust samples. Thus, the milled samples differ from the sieved samples but contain the same minerals found also in sieved or airborne samples.

Milling of all ground samples would allow a better overlap of particle sizes accessible with XRD and IMCA-ZINC. This would help answer one of our scientific questions, i.e. the correlation between mineralogical composition and ice nucleation activity. However, the studied samples would probably be less atmospherically relevant than the ones derived sieving the surface-collected samples.

5) Page 7/20: Hence, the Atacama and Israel milled, Australia, Crete, Peloponnese and Tenerife samples consisted mainly of particles smaller than 2.5 µm and the mineralogy is representative for the particles on which ice nucleation was studied.
I suggest adding the word milled to Australia here also: Hence Atacama, Israel and Australia milled as well as the airborne samples . . . to clarify and emphasize the origins.

We have deleted this sentence to avoid repetition. Instead, we rephrased the sentence on p.7,l. 28-30 of the original manuscript (now p. 8, l. 8-10):

"In summary, the identified mineralogical composition is well representative for the particle size fraction used for IN experiments on the Atacama milled and sieved, Israel milled, Australia, Crete, Peloponnese and Tenerife samples."

"*In summary, the identified mineralogical composition is well representative for the particle size fraction used for ice nucleation experiments on the Atacama milled and sieved, sieved Etosha, Israel milled, milled Australia, and the airborne Crete, Peloponnese and Tenerife samples.*"

6) The authors found a bimodal fit for all samples. As a none-expert in the field of particle shapes figure 3 seems to me that this could be a result of the two used measurement techniques? Is there an explanation for the two modes based on hardness or mineral composition?

We did not find a combination of shape factor and density that is realistic for dust particles which would lead to a size distribution of the dust samples with only one mode (p.9,l.14-15 of the original manuscript). It is reasonable that there is a very broad peak with two, sometimes even three shoulders related to the high inhomogeneity of the samples with respect to hardness and fracture. The majority of the mineral composition tends to be soft (e.g. calcite and clays), giving smaller particles, or hard (e.g. quartz and feldspar) resulting in larger particles.

We have added on p.10,l.2-3 (revised manuscript): "*They are likely related to the high inhomogeneity of the samples with respect to hardness and fracture.*"

7) The whole section is very well written and the authors highlight new results.
Page 10/28: As a consequence of the large mean surface area of the Great Basin sample its ns is shifted to the lowest values.
A shift can be misunderstood, maybe just: "shows the lowest ns values. "

Agreed. We have rephrased p.10,l.27-29:
"The Israel milled and the Peloponnese sample show a low IN activity. Overall, the airborne Saharan samples belong to the lower half of all ns curves. As a consequence of the large mean surface area of the Great Basin sample its ns is shifted to the lowest values."

now p.10,l.32-33 of the revised manuscript:
"*The n_s of the Great Basin sample, which has one of the highest FFs, is amongst the lowest, due to its coarse particle sizes.*"

and p.11,l.6-7 of the revised manuscript:
"*The Israel milled, the Great Basin, and the Peloponnese sample show a low ice nucleation activity.*"

8) ***Role of mineralogy***
The first part is very well written and points out the difficulties with single minerals in case of micas, muscovite and ankerite. The authors again emphasis well that surface collected samples may not be

representative for the atmosphere. In the following paragraphs the study tries to correlate ns with the mineral fractions. The aim of this is clear and the general findings may still hold true, but the statistical significance of the reported correlations is questionable. Firstly, the Pearson correlation coefficients (R) should not be used as the only statistical measure indicating the correlation. It is furthermore not clear from the text and table 5 which minerals were used at which temperature for obtaining the correlation coefficient. This information should be added to the manuscript. Including more statistical measures is probably out of scope of this study, but the regressions and plots yielding to the correlations should be added to the supplement.

We have indicated now with an asterix which *R* - values are significant at the 0.05 level for each correlation in Table 5 and the new Table 6. We have added the most important scatter plots yielding the correlations as Figure 1 to the supplement. These are the correlations of n_s at 243 K, 245 K and 253 K with quartz, calcite, K-feldspars and kaolinite, respectively.
Furthermore, we have added the following lines at p.14,l.6-9 of the revised manuscript:

"*Table 5 shows the Pearson correlation coefficients (R) between the mineral fractions and the n_s at five temperatures. Only a few of the correlations are statistically significant, owing to the low number of samples. Nevertheless, the overview of the correlation coefficients gives an idea of the effect of certain minerals on n_s. The related scatter plots are given in Figure 1 of the supplementary material.*"

To clarify which samples were used at which temperature we have added sample numbers in Tables 1 and 4 and provide these sample numbers in Table 5 and the new Table 6 as "samples included". We have split the original Table 5 in two: the new Table 5 contains the former first five columns and the new Table 6 the former last three columns. Furthermore, we clarify on p.14, l.2-5 of the revised manuscript:

"*For each temperature, all samples which showed a FF between 0.1 and 0.9 were used for the correlations. The Etosha sample was excluded from the correlations with feldspars, quartz and clays, as it does not contain any significant amount of these minerals. However, it was included for the comparison with calcite.*"

9) In respect to the content I find the term anti-correlation of ns for illite and other clays rather confusing, independent of the fact that the values of |R|<0.5 have a low significance. Anti-correlation could be understood as an anti-freezing behavior. By increasing the content of example illite you reduce the ns at the given temperature, but in fact this is depending on the other mineral components.

Rather than being relative to the fundamental process of freezing, the mineralogy correlation coefficients calculated are relative to the mean mineralogy of the samples. This means that if a sample has increased levels of (presumably) ice nucleation inactive minerals such as illite and calcite, it will have a correspondingly lower concentration of the (presumably) ice nucleation active quartz and feldspars. This produces the anticorrelation between these minerals and n_s. To avoid any confusion we have replaced "anticorrelation" with "*negative correlation*" and "anticorrelated" with "*negatively correlated*".

10) The study reports that at 253K there is a good correlation between ns and the Kfeldspar content, while 245K the correlation with quartz alone is better than with feldspars and quartz. The question arising is: how much of the total ns is still available at 245K to be explained by quartz if it had been already partly by K-feldspar at the higher temperature? By including a different amount of samples in the correlations at every temperature the relations are harder to identify. In my perspective it is necessary to also have a look at the correlations for the same 3 samples that were used at 253K alone at 245K and if they are better explained by quartz alone or joined by quartz and feldspar. The authors do conduct a manual selection of which samples are included to obtain the correlation coefficient. I therefore want to emphasize once more to state which samples are taken for which correlation and to add this correlation plots to the supplement.

In response to comment 8, we added sample numbers to Tables 1 and 4 and provide the sample numbers for each correlation (Table 5 and Table 6).
We abstained from presenting correlations at T < 253 K for only three samples due to the even lower significance. However, we have included them now as Figure 2 in the supplement. The correlations for only Taklamakan, Egypt, and Atacama milled at $T < 250$ K are consistent with the correlations given for all relevant samples. Since none of the three samples contains any illite and only one sample contains kaolinite, the addition of clays to the correlation is meaningless. At 245 K the sum of quartz and all feldspar is the best predictor for n_s ($R = 0.97$) compared to the K-feldspars ($R = 0.68$) or the sum of all feldspars ($R = 0.85$). At T=240 K and 243 K, the sum of quartz and feldspar correlates best with n_s. At T = 238 K finally, quartz alone leads to the best correlation ($R = 0.72$) with n_s.
We have added on p.14, l.23-26 of the revised manuscript:
"*To exclude a bias from varying numbers of samples at different temperatures, we have repeated the correlations at 245, 243, 240 and 238 K for the Atacama milled, Egypt and Taklamakan samples only. The corresponding scatter plots are provided as Figure 2 of the supplementary material and confirm the observations that quartz plus feldspar yield the best correlations at T < 245 K.*"

11) Minor adjustments: In figure 5 the great basin sample for me does not appear in the same color in the plot and the legend below 150

Corrected.

12) In table two what is the order of the minerals. It does not seem to be milled, sieved or airborne neither are they alphabetically sorted. Do you want to have the Israel samples on the same page?

We have sorted the samples now alphabetically in all tables.

References:

Boose, Y., et al.: Heterogeneous ice nucleation on dust particles sourced from 9 deserts worldwide - Part 2: Deposition and condensation freezing, in prep., 2016

Chou, C., Stetzer, O., Weingartner, E., Jurányi, Z., Kanji, Z. A., and Lohmann, U.: Ice nuclei properties within a Saharan dust event at the Jungfraujoch in the Swiss Alps, Atmos. Chem. Phys., 11, 4725–4738, doi:10.5194/acp-11-4725-2011, 2011.

Kaufmann, L., Marcolli, C., Hofer, J., Pinti, V., Hoyle, C. R., and Peter, T.: Ice nucleation efficiency of natural dust samples in the immersion mode, Atmos. Chem. Phys., 16, 11177-11206, doi:10.5194/acp-16-11177-2016, 2016