

We thank Russ Schnell for his interactive comment. 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

*Mineral aerosol particles in the atmosphere and on the surface of soils rarely, if ever, do not contain associated biological material. As is well known, biological materials are the warmest IN observed in nature. Desert dusts are most readily generated in playas that are deposited by water that has concentrated small mineral particles and associated biological material into lower level areas. Desert dusts that travel appreciable distances rarely are sourced from the tops of sand (mineral) dunes. In this paper, it may be disingenuous to ascribe the measured IN content solely on the mineral content of the particles. The authors may consider heating the desert dusts in a furnace and boiling samples before and after the immersion IN freezing tests to remove the organics. If the sample IN contents are unchanged, the authors and their audience may be somewhat assured that the mineral components of the samples are the IN source and not organic passengers. Russ Schnell, NOAA, Boulder, CO*

At the comparably low investigated temperatures ( $\leq 253$  K), the role of biological particles is expected to be rather secondary (O’Sullivan et al. 2015) since some minerals are known to be efficient INP at these temperatures (e.g. Atkinson et al. 2013) and the concentration of dust particles compared to biological material is likely high (e.g. Fig. 19 of Murray et al. 2012). The focus of our study was therefore mineralogical composition of natural (airborne and surface-collected) dust samples and its role for ice nucleation. The small post-processing amount of most samples (particularly of the airborne ones) does

not allow a systematic study on the effect of heating of the samples on the ice nucleation properties with the same instrumentation (there is simply not enough dust left to repeat the measurements with heated samples). However, we were able to heat some of the samples at 300°C (573 K) for 10h and perform measurements in the condensation mode with the portable ice nucleation chamber, PINC (Chou et al. 2011), at a temperature of 240 K and 242 K (Fig. 1 below). We show here the results at  $RH_w = 102\text{-}105$  (240 K) and 100-103% (242 K), the highest measured  $RH_w$ . The measurements are not directly comparable to the immersion freezing results obtained with IMCA-ZINC since these  $RH_w$  values do not guarantee complete droplet activation prior to freezing during the residence time of the particles in PINC (about 5 s). Nevertheless, they give an indication of the effect heating has on the ice nucleation ability of the particles.

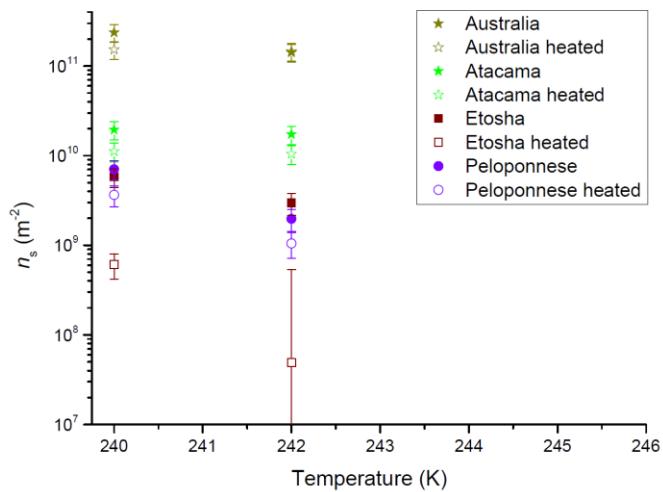


Figure 1: Condensation mode  $n_s$  for selected heat treated samples.

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. 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).

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, keeping its ice nucleating ability and lead to an increased ice-nucleating ability of the dust (Schnell, 1977; Conen et al., 2011; O'Sullivan et al., 2016)."*

We have rephrased p.5,15-16: "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)."

to now p.5,l.28-20 of the revised manuscript:

*"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.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"*

We have replaced p.14, l.17-19:

"The results suggest that potential coating of the particles only plays a secondary role for the immersion freezing ability of the mineral dusts as the majority of the results can be explained by the mineralogy as has also been observed by Kaufmann et al. (2016)."

with now p.15, l.10-13 of the revised manuscript:

*"While the correlations do not exclude an influence of non-mineral material, the results suggest that potential coatings or mixing of the particles only play a secondary role for the immersion freezing ability at the studied temperatures of the mineral dusts. The majority of the results can be explained by the mineralogy as also observed by Kaufmann et al. (2016). "*

## 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.

Murray, B. J., O'Sullivan, D., Atkinson, J. D., and Webb, M. E.: Ice nucleation by particles immersed in supercooled cloud droplets, *Chem. Soc. Rev.*, 41, 6519, doi:10.1039/c2cs35200a, 2012.

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