

Reviewer comments in bold and authors' response in regular typeface.

General comments:

This article reports that coating two types of mineral dust particles with a secondary organic aerosol proxy produced by dark ozonolysis of alpha-pinene made little difference to their immersion mode ice nucleating ability between 253 K and 233 K. No systematic differences in ice nucleating abilities of the dusts were observed with various atmospherically plausible coating thicknesses. Measurements were conducted simultaneously on three well established cloud chamber instruments with broadly consistent results. The study is relevant to the scope of ACP, presents useful and interesting new data and is scientifically sound. The paper is mostly well written. I have a few minor comments, but am quite happy for the paper to be published once these are addressed.

We thank the reviewer for the positive comments and address the minor comments below.

Minor comments:

The conclusion 'SOA coatings did not affect the immersion ice nucleation ability of the dust particles in the temperature range 253 to 235 K irrespective of coating thickness (3 – 60 nm)' and similar statements in the abstract seem too strong. While most of the data does support the statement the very reasonable uncertainties in measured INAS suggest to me that there could be some effect that it is not possible to discern with certainty from the data. The data are certainly suggestive and surprising, I would have thought that SOA coating would make a measurable difference to the INAS of the dusts, but I do not think this study allows the conclusion that SOA will not 'impede or enhance the ice nucleation ability by immersion mode of mineral dust in the mixed phase cloud regime'. The authors effectively acknowledge as much in their discussion but this subtlety is at least partially lost in the conclusions and abstract. Additionally, it is entirely conceivable that different results could be obtained if different dust samples were used. I do not think that two soil samples can be claimed to represent all desert dusts. To summarize, I think it should be made clearer that the study covers only a fairly narrow set of circumstances and that this topic likely needs further investigation. I do not think this detracts at all from the usefulness and interest of the study.

This is a valid point and we now refer to this point both in the Abstract and Conclusions sections. In the abstract, we specifically removed the reference to the atmosphere and the mixed-phase cloud regime. In addition, despite the uncertainties from *INAS* densities, even for the *AF* results, it is clear that the coatings do not make a difference. As such, we specify that the lack of impeding or enhancing is specifically for the SOA coatings used in this study ([page 1 line 31-32](#)).

However, we include now in the paper a comprehensive discussion of how coatings impair deposition mode ice nucleation but do not impair (or only partially impair) immersion mode ice nucleation. The results from this work, combined with previous numerous studies of organic and inorganic coatings on a variety of dust particles suggest that organic coatings and inorganic coatings (depending on temperature regime) will not impair immersion mode ice nucleation activity. We now note this in the conclusions section ([page 17 line 27 – page 18 line 3](#)). We also acknowledge that we use a limited number of samples in this study in the conclusions section ([page 17 lines 27-28](#)).

On a related note, it is increasingly clear that the ice nucleating ability of a mixed mineral dust depends on its composition, at least potentially (Harrison et al., 2016; Peckhaus et al.,

2016). While the information is in the literature as stated I think there should be a table reporting mineralogy of the two samples.

This is indeed a reasonable suggestion, however the mineralogy is only available qualitatively for our SD sample in Linke et al. (2006, called Cairo2 in their paper). Here we give the percentages for our AD sample, but for the SD sample, we can only say that the clays and dolomite dominate the composition compared to feldspars, as was reported in (Linke et al. 2006). In addition the AD sample has its complete mineralogy reported with mass percentages in Boose et al. (2016, called Taklamakan in that paper). Since we do not have the mass percentages for the SD sample, we refrain from constructing a table to report the mineralogy, and instead discuss it in the text (page 16 line 28 to page 17 line 6).

Similarly, it is stated in the conclusions that fit to data in this work ‘yield a parameterization for desert dusts’, which seems a very general statement for measurements conducted on two samples. Also, it does not seem to me surprising that the INAS spectrum fits reasonably to that of Niemand et al. (2012) when the measured samples are two of the five or so used in Niemand et al. I would suggest removing both these statements.

We agree with the reviewer’s comment that we have only presented two samples in this work. As such we remove the statement about the parameterization and instead say we obtained a fit for the desert dusts used in this work (page 18 line 12).

Furthermore given the technique for ice nucleation used in Niemand et al. (2012), was also one of the three techniques used in this work, we also removed the statement “..compared well (to within a factor of 5) in the temperature range 254 – 232 K to a previously proposed parameterization for desert dust..” from the conclusions section.

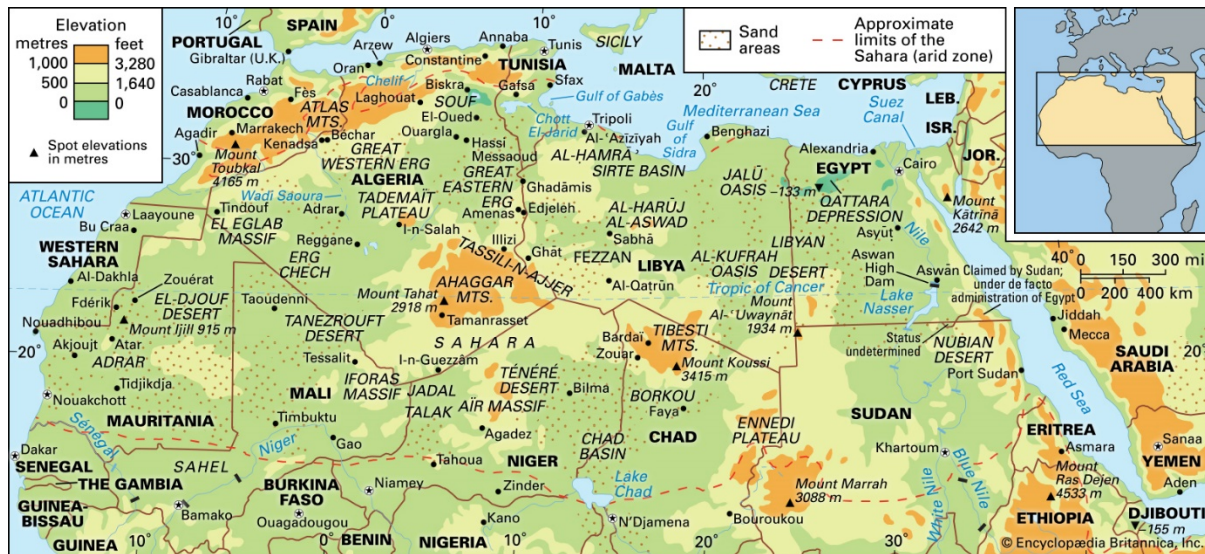
I am a little curious as to why this study was conducted on soil samples dug up from underground or collected from the surface. I would have thought either more directly atmospherically relevant samples or ‘pure’ mineral samples would be of greater interest. Possibly the authors think these samples are of substantial atmospheric relevance but I think this needs to be more thoroughly explained and justified if so. Finally, I realise it’s the established name for this sample but is it really reasonable to call dust collected north of Cairo ‘Saharan’?

Obtaining samples directly from airborne dust requires long collection times with high volume cyclone samplers necessitating proximity to the source for ground based sampling as was done for example in (Boose et al. 2016). Even in this work, the sampling conducted directly in a dust storm/event required sampling for over 24 hours and yielded enough sample on the order of 1-2 g to conduct one day of ice nucleation measurements. It would therefore not be possible to run multiple experiments with the three different instruments and various coating experiments because of the small sample size. We now clarify this on page 6 lines 17-19.

These samples were chosen because the Saharan and Asian regions are known to be the largest contributors to airborne atmospheric mineral dust from arid and semi-arid regions (Tang et al. 2016), we now clarify this point to motivate our sample choice better on page 6 lines 10-11.

We appreciate the reviewer’s concern over naming of the sample. Depending on the source, all of Egypt is considered to be part of the Sahara, except the Nile valley, Nile delta and the region close to the Mediterranean coast. But even these descriptions depend on source. For example, the Encyclopedia Britannica reports that all of Egypt is part of Sahara and that Cairo and the

regions north of it are also within the borders of the Sahara arid zone. However, the around Cairo may not be a sand area (see figure below).



1. Borders of the Saharan Region (taken from <https://www.britannica.com/place/Sahara-desert-Africa/media/516375/200>)

Recognizing it might be slightly sparse, I think the authors may want to consider a figure showing the absence of impact of thickness of coating on ice nucleation effectiveness. Currently, the reader is forced to refer back to table 1 to figure it out, which doesn't aid readability.

This is a good idea. We have prepared a new figure to also show the impact of coating thickness (or lack thereof) on the ice nucleation properties on both the AD and SD samples. These have been added as Figure 6 in the revised manuscript and we refer to these on page 11 line 22, page 12 lines 20, 27 page 13 line 4, page 17 line 26. We retain the other figures as well in order to refer to the discussion on specific experiment numbers as well as to allow a reader to refer to the specific coating thickness reported in table 1.

Specific comments:

Pg 15 Line 29- considered to be 'in' reasonable. What does reasonable mean? Some sort of quantitative description might be helpful, and perhaps a comment on how data produced by the different instrument types should be interpreted.

We changed the word "reasonable" to "good" and clarified this means within overlapping uncertainties (page 18 line 9). A comment on how the data from different instruments is to be interpreted is already provided in the conclusions section on page 18 lines 3-9.

Pg 3 line 15- Ammonium sulphate has been observed to enhance ice nucleation of mineral dusts recently (Kumar et al., 2018; Whale et al., 2018). It may be appropriate to note this here.

We have now added that in addition to suppression of ice nucleation, enhancements are also observed for immersion freezing and cited the suggested studies and more (see page 4 line 5-13).

Pg 4 line 23- Why is immersion freezing being mimicked? Is the process not immersion freezing?

Here we meant to say “simulate” the atmospheric process of immersion freezing in the instruments we use. We now clarified this (see page 5 line 22).

P 7 line 31- Are convective clouds not natural?

We have corrected this. We meant to say “covering a range of weakly to strongly convective wave clouds” (page 9 line 6)

Pg 10 Line 20- Why does the number of large particles change reported AF? A bit more discussion may help the reader.

We agree, we now explained this further, by stating that larger particles are more effective INPs thus contribute to the INP population and will influence the *AF* if excluded from being sampled (see page 11 line 32 to page 12 line 3).

Technical comments:

Section 4.4 follows section 3.3 currently, this should presumably be section 3.4.

Thanks for catching that, now corrected.

Pg 1 Line 15- The first sentence of the abstract make it seem as if there were two sets of experiments conducted, which was not the case, I would suggest revising this.

Two sets of experiments were indeed conducted, one with coated and uncoated AD, the second with coated and uncoated SD. As such, we leave the sentence as is.

Pg 2 line 28- ‘inferred’ should be ‘by inference’ or similar I think

We changed ‘inferred’ to ‘by inference’ (page 2 line 28)

Pg 3 line 20- I would use ‘INPs’ instead of INP. I would suggest checking that INP and INPS are used properly throughout.

The change to INPs was done (now page 4 line 23). We also checked the whole manuscript and made a number of corrections to “INPs” and “an INP”

Pg 7 line 33- pg 8 line 1- ‘...homogeneous temperature control of below ± 0.3 K.’ is clumsy.

We now changed this to read “*homogeneous temperature control $< \pm 0.3$ K*” (page 9, line 8)

Pg 8 line 29-32- contribution to aerosol number maybe?

It should be contribution to surface area, not aerosol number like suggested by the reviewer. However, the sentence was a little confusing, and thus we have clarified this to read “*However, the contribution to the surface area from the particles above $1 \mu\text{m}$ (see Figure 2) is significant enough (see Figure 3) to have to account for the impactors used upstream of PINC and CSU-CFDC in determining INAS densities*” (see page 10 line 5-7)

Pg 10 line 16- ‘Way above uncertainty’ is a bit loose.

We changed this to ‘much greater than the maximum uncertainty in *AF* (28%)..’ (page 11 line 28-29).

Pg 11 line 17- missing word after ‘indicated’.

The word “by” was missing. Now corrected (page 12 line 33)

Pg 12 line 8- Favouring immersion freezing over what?

Favouring immersion freezing compared to deposition nucleation where coatings suppress the ice nucleation activity of dust INPs. This clarification has now been added ([now page 14 line 5-7](#))

Pg 13 line 7- maybe mention the origin of this factor of 3.

The origin of the factor 3 has been discussed already in the introduction ([page 5, line 23-28](#)) and in the results and discussion section ([page 10, lines 20-25](#)) and thus at the said location ([now page 15 line 11](#)) we refer the reader to these sections rather than repeating the origin of the factor 3 yet again.

Pg 15 line 20-21 ‘appreciable’ is very vague.

We have now clarified that by appreciable we mean tens of nm ([page 17 line 26](#))

Pg 16 line 6-8- This sentence is poorly written.

We have now clarified this sentence ([page 18 lines 15-17](#))

Reference list- Ullrich et al. 2017 and several others lack journal names and Megahead should be Megahed I believe. I suggest checking the list carefully.

Thanks for carefully checking this. Megahed now corrected and all references to have been checked to include journal names.

References

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Whale, T. F., Holden, M. A., Wilson, Theodore W., O’Sullivan, D., and Murray, B. J.: The enhancement and suppression of immersion mode heterogeneous ice-nucleation by solutes, *Chemical Science*, 9, 4142-4151, 10.1039/C7SC05421A, 2018.

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Boose, Y., et al. (2016), 'Heterogeneous ice nucleation on dust particles sourced from nine deserts worldwide – Part 1: Immersion freezing', *Atmos. Chem. Phys.*, 16 (23), 15075-95.

Linke, C., et al. (2006), 'Optical properties and mineralogical composition of different Saharan mineral dust samples: a laboratory study', *Atmospheric Chemistry and Physics*, 6, 3315-23.

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