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Interactive comment on "A laboratory investigation of the ice nucleation efficiency of three types of mineral and soil dust" *by* M. Paramonov et al.

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This paper provides a thorough investigation of the different IN active components on mineral and soil dust by using various treatments on the dust and then testing the INA pre and post treatment. It is a very nice paper that is well written and adds to the body of literature on soil dusts and their INA.

Response: The authors would like to thank the referee for reviewing the paper and for the praise!

Major comments

1) Page 5 line 14: More information is needed about the soil collection. What was used to collect it? How was it transported and stored? How many cm below the surface was collected? Was it all from one spot or was random sampling done over a gridded area and the samples combined to give a representative sample? Please give information about the specific sieve you used and the procedure. Was it sieved by hand or with a machine?

Response: The following sentences have been added to the main text on page 5: "All dusts were collected either right at or directly below the surface at each individual location. After collection the dusts were then stored in plastic bags in dark conditions at room temperature. To allow for ice nucleation experiments, the collected dust samples were sieved with a series of dry sieves to select only the particles below 45 μm in diameter (Retsch Vibratory Sieve Shaker AS 200)."

2) I very much enjoyed the discussion of the surface area uncertainties and how the washing and heating procedures can alter the surface area. However, it makes me wonder if the paper would benefit from also including ns,geo as a means to compare to other studies that use ns,geo and to see if the results hold (meaning Himalayan ns is highest, Iceland lowest etc.). It might be an interesting exercise, especially since you call out BET as potentially not being a good way to go about calculating ns. Adding ns,geo to the discussion would complete the thought exercise of which is the best way to present INA data.

Response: The authors completely agree with the referee comment and have, indeed, for a long time considered including ns,GEO in the manuscript. This, however, was not done for several reasons. First, ns,BET values of all untreated and treated dusts is one of the most unique aspects of the paper. As the analysis went along, it became apparent that ns,BET would become the focal point of the paper and result in the discussion related to its limitations and uncertainties. Second, since we have conducted experiments with monodisperse particles, ns,GEO values are computed directly from AF values, and they would show the same trends and results as did AF values (Fig. 1)

when it comes to comparing the INA of dusts to each other for the same size. The only clear benefit of the inclusion of ns,GEO values in the discussion would, as correctly pointed out by the referee, allow for comparison to other previously published studies. This feeds into the third reason for omitting ns,GEO values. The paper is already quite long, and the discussion of AF and ns,BET values is already rather detailed and labourious. Adding ns,GEO would require to significantly expand the manuscript further, add at least two new figures, modify almost every section of the manuscript and include ns,GEO in the discussion about which parameter is the best for describing the INA of any given species, something that has already been done by, e.g., Murray et. al. 2012 and Hiranuma et.al. 2015. I believe the significant expansion of the manuscript would not be worth it considering that the only important benefit of including ns,GEO values is the comparison to previously published studies, which, in the end, is not one of the main focal points of the paper anyway. No modifications have been made.

Minor comments

1) Page 5 lines 7- 12: It would be nice to include the latitude and longitude of these locations.

Response: The geographic coordinates have been added on page 5.

2) Page 9 lines 23-24: Instead of "The study" it would be clearer to combine with the previous sentence to read ": : :as presented in Ullrich et al. (2017), which investigated particles in polydisperse: : :". Or if you think that's too long of a sentence revise to say "The Ullrich et al study investigated: : :"

Response: For clarity, the second sentence now reads: "The mentioned study investigated. . ."

3) Page 9 lines 17-25: It is not necessary, but would aid the reader to have the Kanji et al 2011 and Ullrich et al. 2017 data on a plot with your data (maybe in the supplemental) to visualize the comparison you are describing here.

Response: Figure 1 modified to include data points from the two referenced studies. The text at the bottom pf page 9 and top of page 10 now reads: "Comparing the onset values to those published in a study by Kanji et al. (2011) reveals that in the 238−243 K temperature range the Iceland, China and Himalaya dusts are all more active than the Saharan (SD) and Canary Island (CID) dusts with the exception of 200 nm Iceland dust particles at 238 K (Fig. 1). At 233 K all examined dusts and particle sizes are more IN-active than the Canary Island dust (Kanji et al., 2011). The referenced study examined polydisperse particles with a mode size of 200−300 nm in diameter. The onset values presented also compare well to those of Asian desert dusts AD1 and AD2, and Saharan desert dust SD2 as presented in Ullrich et al. (2017) (Fig. 1). At the warmest temperature Saharan desert dust SD19 seems to be more ice-active than any of the dusts examined in the current study (Fig. 1). It should be kept in mind that, with the exception of dust AD2, the onset values of Ullrich et al. (2017) seen in Figure 1 were defined for AF higher than 0.001 assumed here.".

4) Page 10 5-7: This sentence is repetitive.

Response: This sentence serves as a reminder, since the discussion immediately thereafter elaborates further on the comparison of dusts and shows that which dust is most active depends on which quantity is examined (AF or ns). No modifications made.

5) Page 10 lines 22-26: "While it is not possible to directly determine the reasons behind this observed difference, it may be possible that large particles contain more soluble material blocking the active sites and/or that small particles may contain more IN-active material on their surface, e.g. bacteria or active minerals. Particles of approximately 200 nm in size, including mineral dust species, have previously been reported as constituting the majority of the INP found in the ice crystal residual size distributions (Mertes et al., 2007)." Is the activated fraction higher for the 200 nm particles as well?

Response: No, it is not. AF is higher for larger particles. This is mentioned on page

9, lines 16-17 and further addressed on page 20, lines 8-17 of the revised manuscript, where the authors pose a question as to which of the two parameters (AF or ns) is truly representative of INA of a given species.

6) Page 14 line 8: Figu. 4 change to Fig. 4

Response: Corrected.

7) Page 14 lines 9-10: "Figure 3 shows that in deposition nucleation mode the decrease in INA after heating is minimal and becomes more pronounced in condensation freezing mode" change to "Figure 3 shows that in the deposition nucleation mode the decrease in INA after heating is minimal and becomes more pronounced in the condensation freezing mode".

Response: Corrected.

8) Page 14: Throughout this discussion "the" should be added before the different ice nucleation modes. Example "the condensation mode".

Response: Definite article "the" inserted throughout the text, where appropriate.

9) Page 16: "The conductivity of supernatant water and, hence, the deduced amount of soluble material was highest for Iceland dust" Would different molecules or types (ie organic acids versus salts) affect the conductivity differently? Can you say 1:1 that the magnitude of the increase relates to the amount of soluble material? Or would salts increase the conductivity more than organic acids? I'm asking just out of pure ignorance to this type of measurement. It may help other readers who are not familiar if you add a sentence about this whether it would or wouldn't change the conductivity.

Response: Organic acids would be expected to be less conductive than salts, but this would be purely because they are weak acids and therefore partially dissociate, thus the ionic strength of the solution would be weaker than that of inorganic salts or acid solutions. Since we measure the conductivity, which should be proportional to the total ionic strength/content of the supernatant water, it is clear that the contribution of ions

to the supernatant water could come from both organics and inorganics. It could be that the water is composed of organic and inorganic ions but it would be impossible to distinguish if a lower conductivity would be due to presence of organic acids or a comparatively lower amount of inorganic ions. Similarly, we cannot speculate that a high conductivity is due to inorganic ions or a comparatively large amount of organic (acid) ions. As such, the use of the phrase "soluble material" is preferred to be inclusive of any potential contribution from both inorganic and organic compounds. Therefore, the electrolytic strength of the solution is expected to be directly proportional to the conductivity.

As such, it would serve no purpose to distinguish between the conductivity of organics vs. inorganics because the relative contribution of these species to the ionic content of the supernatant water is not known. Thus, no modifications to the revised manuscript are made in this regard.

10) Page 17: I really enjoyed the detailed discussion of the H2O2 procedure and how each step may impact the results and thus the implications. It was well thought out and clearly explained so the reader could follow along with the logic. Nicely done.

Response: Thank you!

11) Figure 8: I know it is written out in the figure caption, but it would be easier for the reader if the SEM images were labeled right on the image with Iceland, China, and Himalaya. Maybe on the upper right hand corner of each image. It would just be easier to compare them without having to remember the order.

Response: Figure 8 modified.

12) Page 20: "It was shown that at temperatures of 238–243 K, the ice nucleation activity of the untreated, surface collected soil dust in condensation freezing mode can be roughly approximated by one of the existing surrogates for the atmospheric mineral dust, such as illite NX, for example." If this is true, then why does one even bother

with the post-treatments? Also, I would not expect a 1:1 correlation for the amount of material removed and the decrease or increase in INA because it is not linear. It is based on active sites but those may not be evenly distributed on the particles and coatings covering active sites can be covering the whole particles or a blob on one side and so the mass of something removed will not always cover the same surface area. This is especially true when you are looking at bulk removal across all sizes and then trying to correlate that to size selected particles. I would not expect it to be linear or to have a simple relationship. This ties back to what you were saying about bulk measurements complicating matters and dust being known to have varying chemical composition with size. The discussion might benefit from a little bit more explanation about these complexities.

Response: The post-treatments were intended to investigate whether other, non-mineralogical compounds of the mineral and soil dust affect its INA and, as it turned out, they did. Hence, the subsequent discussion saying that mineralogy alone is not able to fully explain the observed INA. In the atmosphere, the mineral and soil dust particles are complex mixtures of various compounds, and their INA is governed by the INA of all individual species present on the particle surface. The statement mentioned by the referee serves to say that even though the dust samples examined here are complex mixtures of various compounds, their INA can still be roughly approximated by one of the existing surrogates for the atmospheric mineral dust. This demonstrates that individual compounds on the particle surface do affect its INA, but not significantly enough that they cannot be approximated by an existing surrogate.

As for the second issue raised, in several places in the manuscript it is mentioned that different individual heat-sensitive and soluble compounds must have different individual INA, and that is specifically due to the fact that the response of the dusts' INA to the removal of said species was not proportional to the amount of material removed. This is explicitly mentioned in both heating and washing sections. The point raised by the referee is also explained in detail in the washing section where not only the different

soluble species and their different individual INA may have affected the overall INA of dusts, but also the exposition of the underlying active sites that were otherwise blocked/covered by the soluble material may be important as well. The conclusion also addresses the point that the overall observed INA of dust has to be attributed to different INA of individual heat-sensitive and soluble species. The identification of individual compounds on the particles' surface and the examination of their individual INA are beyond the scope of this paper. No modifications have been made.

References:

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- 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, 6519–6554, 2012.

Thank you very much, again, for taking the time to read, comment and, therefore, improve the paper!

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