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Interactive Comment

Interactive comment on “Ice nucleation by soil dusts: relative importance of mineral dust and biogenic components” *by* D. O’Sullivan et al.

Anonymous Referee #1

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In this manuscript, Daniel O’Sullivan and co-authors present and discuss the ice nucleation behavior of soil samples collected at four different locations in England. The motivation for this study was to provide a basis for quantifying the immersion mode freezing behavior of atmospheric dust particles related to fertile soil areas. The authors applied a special pre-treatment to the soil samples in order to extract the smaller particle fraction, and further treated with heat and hydrogen peroxide to differentiate between the mineral and organic content of the samples acting as ice nuclei. This is a relevant paper with a significant amount of new and important results. The topic fits well in the scope of ACP. Though I rate the manuscript to be excellent and to clearly deserve publication in ACP, I have major concerns about some statements and implications as detailed below. In brief, my major comments are related to an over-interpretation of the laboratory results in terms of atmospheric implications. Of course such laboratory work is very important to also improve the knowledge about the nature and source of ice nuclei in the atmosphere, but I'm willing to accept the manuscript for final publication only if the more speculative components and implications are significantly reduced in the revised version. The manuscript is clearly relevant and above average concerning scientific contents and quality even without speculative or weak interpretations. General remarks: My major comment and concern about the manuscript refers to the statements and assumptions about what can be expected to be the mineral contribution to the ice nucleation activity of the soil dust samples used for the present study (e.g. page 20276 (abstract), lines 14-17; page 20287, lines 21-24; page 20290, lines 4-9; page 20291, line 5; page 20293, lines 3-5;). Obviously the authors take such interpretation from the paper by Atkinson et al. (2013) which includes strong statements on the importance of feldspar as the source of ice nucleation in atmospheric dust based on selected laboratory studies and simplified or idealized modeling studies. But how can one be sure that it is also the feldspar that dominated the mineral component related ice nucleation properties of the soil dusts used in the present work? The samples are of different origin and are also treated and pre-processed in a completely different way (also see some more detailed comments below). Therefore I do not see a robust basis for any statements on what is “the dominant ice-nucleating component” or what is “the background mineral ice nucleation activity” in the soil samples used in the present study. Such formulations suggest that the nature and amount of ice nucleation in mineral aerosols is well understood, which by far is not the case to my understanding.

Major comments:

Please check the manuscript again for consistent use of the terms dust, mineral, biological, or organic material.

Page 20276, lines 19-21 (abstract): This conclusion is very speculative and should be modified to just stating that agricultural activities can significantly contribute to the atmospheric IN concentrations.

ACPD

13, C7792–C7798, 2013

And what do you mean with "particularly at temperatures above 258 K" here? A few lines above you stated that at lower temperature the IN activity of the investigated soils approaches those of other mineral samples. How can a minor contribution to the total burden than have a special role in global IN activity? This enhanced IN contribution can at max be suggested for the higher temperatures where additional contribution from biological material was measured.

On the other hand: was the IN activity in this T range really higher than published for e.g. desert dust material? And how relevant or representative are the four samples for soil dust in the atmosphere? Therefore be more careful with conclusions and implications, e.g. add some statements like "... if our laboratory studies are representative for the immersion freezing activity of aerosols originating form fertile soils ..".

Page 20280, lines 23/24: This sounds like you can quantitatively compare the particle fraction you extract in your laboratory with the one that would be found in the atmosphere if the wind is dispersing particles from the region you collected your samples. A suspension-sieve-settling process is still somewhat different from a wind-driven dispersion and atmospheric settling process, therefore "atmospherically-relevant" is a strong word in this context. Please modify the formulation in the way of more describing what was actually done and less interpreting your lab methods for atmospheric relevance without further explaining the limitations of such interpretations.

Page 20283, lines 18-21: What about the broad range of drop sizes in the pico and nano droplet spray experiments. Did they affect the interpretation of the freezing rate measurements? And can you be sure that the concentration of the suspensions is the same in the droplets of all sizes. This is a huge range of droplet sizes. What about surface tension or capillary effects to cause systematic shifts or separation of water and suspended material during aerosolization?

Page 20284, lines 14-19: I guess the pipette was operated in vertical orientation. How long did the drop formation step go on? Did you check if particle settling inside the pipette needle eventually enhanced the suspension concentration during drop generation?

Page 20287, lines 21/22: As mentioned above, I'm not in favor of such general statements and conclusions from selected lab and simplified modeling studies. Changing "the dominant ... component" to "a dominant ... component" or, even more appropriate in my view, to "a major ... component" would be more appropriate here. Well, of course you are correctly citing the Atkinson et al. (2013) work which includes such strong statements even in the abstract, so at this place it is up to you to change this formulation or leave it as is.

Page 20287, lines 22-24: Such important data treatment should be mentioned and discussed in the plain text and only shortly be mentioned in the figure caption (see also my comment for caption of Fig. 4).

Page 20290, lines 5/6: What is the background mineral ice nucleation activity? I really do not see a robust data base for such a statement with regard to the ice nucleation activity of the soil samples investigated in the present work. Each sample can be different, samples you compare here are treated much differently, and concerning the IN results for the nanoliter droplets shown in Fig. 4 I do not even see a systematic trend of the IN activity with the feldspar content: samples B and C seem to follow the postulated "feldspar fraction rule", but samples A and D do not, though they contain a similar feldspar amount as sample B.

Page 20291, line 5: Again speculation, see above.

Page 20293, lines 1-3: You may also add other literature results like the desert dust related curve by Niemand et al. (2012).

Page 20294, lines 15-18: Again, this is a very speculative statement. I agree that it is interesting to compare such rough estimates based on globally averaged aerosol concentrations from highly idealized and simplified model runs with field measurements of IN concentrations. But, on the other hand, one should not argue that such estimates can explain or "account for" actual measurements at certain locations and for certain time periods. To my opinion, this is not more than an order of magnitude estimate, if at all useful to get significant additional information about the relation between aerosol properties and ice nuclei concentration. A more thorough and meaningful study should compare model results at the point and time of measurement and should also include measurements of aerosol properties together with the IN measurements.

Page 20294, lines 19-29: This section also is rather speculative and does not include extra information other than repeating that a certain amount of primary ice nuclei is needed to trigger the ice multiplication process in clouds. Therefore, this section should be removed.

Page 20311, Fig. 4: This figure should be enlarged, in particular in the y-axis direction, in order to better identify agreement or disagreement between the different data sets and so better identify sample to sample variability.

Page 20313, Fig. 6: This figure should be larger in the final version. In the caption you mention that the Atkinson et al. (2913) data have been scaled down to simulate the mass fraction of the feldspar in the soil samples. I think, this is not a simulation, it is just a scaling factor which not even needs to be representative for the different IN activity in the respective sample. There are other factors like completely different pre-treatments of the feldspar and soil samples which may affect the IN activity even more than just the different feldspar content (see other comments above).

Minor points and typos:

Page 20276, lines 8/9 (abstract): What about mentioning the particle size range and concentration already in the abstract?

Page 20276, lines 13/14 (abstract): If possible indicate if the ice nucleation from biological material contributes all, a major fraction or just a minor fraction.

ACPD
13, C7792–C7798, 2013

Page 20276, lines 18 (abstract): "We conclude that, although only . . ."

Page 20276, lines 23-25: I wondered why you cited these recent papers here. The freezing behavior of pure water was investigated much earlier and was investigated by some more recent papers since then for several reasons. This, and also the statement of the following sentence is text book knowledge.

Page 20283, line 10: Please also mention respective range of drop diameters. A 1 μl droplet already has a diameter of more than 1 mm, I think.

Page 20285, line 28: I would prefer replacing "stochastic approach" with "nucleation rate approach", because this refers to what the approach really is, namely another approximation to measured freezing rates, just using rate equations from classical nucleation rate theory, which in turn of course assumes stochastic freezing behavior.

Page 20285, lines 28: ". . . that nucleation behaved like a time-independent process . . ."

Page 20286, lines 7/8: I preferred to see only those work cited here that gives direct evidence of time-dependent freezing going on in nature, like the one by Westbrook and Illingworth (2013), though, as far as I can see, also this work gives only indirect evidence on time-dependent freezing mechanism.

Page 20287, lines 1-3: You may more explicitly mention that in microliter experiments the droplets appeared to be of more similar or monodispers size, whereas in the nano and picoliter experiments the droplets appeared to be more polydispers.

Page 20289, line 5: Add year of publication

Page 20289, lines 10/11: And what about the uncertainty of the optical technique for a-spherical particles? Which equivalent diameter is actually measured? And which surface area is then available in a model to apply lab result for ns parameterization?

Page 20289, line 14: to which experiments do you refer to herer?

ACPD

13, C7792–C7798, 2013

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Page 20291, line 27: "... makes it difficult ..."

Page 20292, line 29: Why did you assume only 20% of the soil dust related aerosol particles to be ice-active?

Page 20315/316: Any reason for reversing the temperature scale in Figs. 8 and 9?

Page 20317, Fig. 9: Figure could be larger, caption should be shorter. In caption, publication year of Hoose et al. reference is missing. You may also mention in the caption for which pressure range you selected the model dust load as input for the IN calculations (I think you mentioned this in the text somewhere).

References:

Atkinson, J. D., Murray, B. J., Woodhouse, M. T., Carslaw, K. S., Whale, T. F., Baustian, K. J., Dobbie, S., O'Sullivan, 5 D., and Malkin, T. L.: The importance of feldspar for ice nucleation by mineral dust in mixed-phase clouds, Nature, 498, 355–358, doi:10.1038/nature12278, 2013.

Niemand, M., Mohler, O., Vogel, B., Vogel, H., Hoose, C., Connolly, P., Klein, H., Bingemer, H., DeMott, P., Skrotzki, J., and Leisner, T.: A particle-surface-area-based parameterization of immersion freezing on desert dust particles, J. Atmos. Sci., 69, 3077–3092, doi:10.1175/Jas-D-11-0249.1, 2012.

Westbrook, C. and Illingworth, A.: The formation of ice in a long-lived supercooled layer cloud, Q. J. Roy. Meteor. Soc., doi:10.1002/qj.2096, 2013.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 20275, 2013.