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Atmospheric Chemistry and Physics
Discussions

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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In this study the authors carry deposition and condensation freezing experiments to study the ice nucleation activity of different soil samples collected around the world. The authors study the effect of size and temperature as well as of different treatments applied to the particles. It is found that the active site density correlates well with the Feldspar content of the samples pointing to a mineralogy control of the ice nucleation activity. However the effect of different heating, washing and chemical treatment is not consistent with such a hypothesis. This is an interesting and detailed study of ice nucleation of dust samples relevant for cloud formation. The authors emphasize the

limitations of the active site density approach and warn about the assumption of the mineralogy control of the ice nucleation activity of dust samples. The paper is well written and organized. I recommend its publication in ACP after some comments are addressed.

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

Comments

1) My only general comment deals with the slight but important differences between immersion and condensation freezing. It has been shown in cold stage studies of immersion freezing that slight modifications in the environment around the droplet have large effects on the measured active site density (like for example a droplet evaporating during the experiments). Condensation freezing is by definition a non-equilibrium measurement since the droplets are presumably changing their size during activation.

Response: The above is true for a cold stage study where the same particles experience an increasing or decreasing relative humidity, but in the current work, the aerosol particles are exposed to a constant RH given that their residence time in the chamber is 7 seconds and the rate of change of RHw is 2% min-1 implying that the aerosol particles see a change of $\sim$ RHw of 0.2% and as such should not be growing or changing size of the droplet significantly. This can be also confirmed from the data in Figure 2, where ns is shown as a function of RHw, and one notes that above RHw of 100%, the curve plateaus suggesting that there is no effect from having a larger droplet at higher saturation conditions, at least for the range of the supersaturated conditions studied here. However, this would be different for conditions below water saturation (see response below).

Even for deposition it is likely the water coverage of the particles changes during the ice nucleation measurements. So my question is: how does the water adsorption and eventual droplet activation process affect the ice nucleation measurements? For example by virtue of the Kelvin effect when exposed to a given RHw the 400 nm particles would activate more easily and tend to absorb water more rapidly than the 200 nm particles.

Response: If in fact "deposition nucleation" is occurring via liquid water formation, the above would be true that CCN activation could play a role, but this should only be important for conditions very close to water saturation. Let's consider two scenarios: Under the operational definition that deposition nucleation is occurring by vapour adsorbing onto the particle surface, to form a critical ice germ followed by bulk ice nucleation, the larger surface of the 400 nm particle plays a role by providing more surface for an active site to accommodate adsorption of the vapour that forms the critical ice germ. Given that our particles are largely insoluble, droplet activation is not expected until RHw is very close to or above water saturation. As such, it is not expected that eventual droplet activation would affect the ice nucleation measurements below water saturation. This is demonstrated by the plateauing of the ns curves for RHw > 100% (Fig. 2).

That being said, in a second scenario, if liquid water is being absorbed into the particles below water saturation because of the presence of soluble material, as is evident from the data in the paper (Table 3), and then this condensed water can cause freezing because of an active site present on the dust particle, then it would be the case that the increase in ns as a function of increasing RHw results from a form of hygroscopic uptake due to the soluble material, but not CCN activation, as activation into drops would only occur at supersaturated conditions. However, because of the hygroscopic uptake and resulting growth and increase in the ice active fraction (or ns) would be observed as more absorbed water results on the particles with increasing RHw resulting in a more dilute hygroscopic layer.

This could explain why they apparently look more active, when in reality they may have been exposed to a different thermodynamic environment. Washing may remove some of the soluble material hence the particles may condense and activate more easily, thus leading to an effect on cloud condensation nuclei activity that could be mistaken

by an effect on ice nucleation. The authors should comment on how this may affect their measurements.

Response: If indeed liquid water is involved in the deposition regime, the above would be true, but this would be due to water absorption and hygroscopic growth rather than CCN activation. This comment is acknowledged and now clarified this in the revised manuscript on page 8, lines 11-18, page 11, lines 6-8, page 16, lines 17-19.

Technical comments

1) Page 2, Line 17. Must be "higher" instead of "warmer" temperature.

Response: Corrected.

2) Page 5, Line 4. Deposition defined as RHw<100% seems ambiguous. I know this is standard practice, but a can dust particle adsorb several monolayers of water at RHw<100 %? How long does it take for a dust particle to reach equilibrium coverage when exposed to a given RHw? Is the residence time of the instrument long enough for it to happen?

Response: It is agreed that this is just an operational definition. The adsorption and absorption of water < RHw 100% is acknowledged. See response to the main comment above.

3) Page 8, Eq. 1. Please specify whether this is the surface area per particle or the total surface area in the population.

Response: Corrected. The sentence immediately prior to the equation now reads: "The ns per particle was calculated as follows:"

4) Page 9, Line 6. Is the increase in AF consistent with CNT predictions? Although CNT predicts an increase in AF with area it tends to be much more subtle than typically measured.

Response: CNT calculations were not carried out in this study, and the reference to

CNT has been removed. Instead, additional references have been added, all of which invoked CNT and performed calculations. The sentence on page 9 lines 17-18 now reads “Such size dependence is consistent with previous observations (Archuleta et al., 2005; Welti et al., 2009; Kanji et al., 2011)”.

5) Page 9, Line 10. This is unexpected since dust is assumed to be a good ice nucleating particle. Is the fact that the maximum AF reaches only 1% due to residence time or particle size? Would it be the higher/lower in the atmosphere?

Response: This is true and is addressed a bit more on page 10, lines 7-15 in the revised manuscript. There are several reasons why our maximum AF is $\sim$33% (China dust, 400 nm, coldest T, highest RHw). One explanation is, indeed, the residence time in the chamber, which is nominally seven seconds. Another explanation could be the deviation of particles from the laminar sample flow, although the lab experiments showed that this deviation affects $\sim$10% of particles (unpublished data). Yet another explanation could be that even for mineral dust, which is indeed a good INP, not all particles would activate into INP, highlighting the overall notion that INPs in the atmosphere are rare. It is very difficult to say how the AF in the atmosphere would be different from the lab experiments especially given that in the atmosphere particles are externally mixed, as such low AFs would be the norm. Additionally, lower AF would be expected in the atmosphere because the supply of supersaturated water vapour may not be continuous in the atmosphere as it is in PINC. Temperature in the atmosphere is also likely to change due to the release of latent heat of condensation.

6) Page 13, Line 8. Recent work has shown that the dust surface morphology may be more delicate that previously thought. For example, active sites may be susceptible to the addition of very low concentration of ammonium sulfate and other solutes. Thus the heating treatment and the H2O2 hydrolysis seem harsh. Maybe for next experiments enzymatic hydrolysis could be considered to better target organic material.

Response: Authors absolutely agree with and thank the reviewer for this suggestion.

7) Page 14, Lines 29-32 (also Page 16, Lines 19-22). The authors make this claim several times. However caution should be taken since immersion and condensation are different and in this particular case may not be completely comparable. The treatments may not only affect the ability of dust to act as ice nucleating particle but also its ability to act as a cloud condensation nuclei (CCN). See my general comment above.

Response: Please, see the response to the main comment and the parts of the text where additions have been made.

8) Page 25, Line 23. Is the higher area of Himalayan dust a result of less aggregation or higher porosity?

Response: The BET surface area of Himalaya dust is the smallest among all dusts (Table 4). Authors are not sure what this comment is referring to.

9) Page 20, Line 7-10. This is true as long as the treatments do not affect CCN activity.

Response: That’s correct.

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

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