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Interactive comment on “Heterogeneous ice nucleation on atmospheric aerosols: a review of results from laboratory experiments” by C. Hoose and O. Möhler

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We thank Dr. Pummer and Dr. Grothe for their careful reading and for the important comments which complement our manuscript. The comments are included below in italics.

Congratulations to the authors! This is a well-written and timely review paper on ice nucleation summarizing and comparing the existing laboratory experiments on heterogeneous ice nucleation caused by different aerosol particles.

The authors correlate the higher nucleation temperatures of bioaerosols with the usu-

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ally larger sizes of biological particles. As we showed in our paper (Pummer et al., 2012), it is not the whole pollen grain being necessary for ice nucleation, but only some macromolecules, which are much smaller than the grain itself, and which can nucleate ice independently. Even the much larger bacterial IN clusters (Warren and Wolber, 1991) are relatively small in comparison with the mineral dust particles applied in most of the quoted studies. So in fact the biological ice nuclei are tendentially smaller than the dust particles.

Our statement is based on the fact that the atmospherically relevant dust particles are in the submicron size range, and most of the primary biological particles which have been studied so far are supermicron. The ice nucleating macromolecules which, as (Pummer et al., 2012) have shown, can be suspended from pollen grains, are a special case. However, as shown in Fig. 13, the empirically determined INAS density of the entire pollen grains (which may reflect the concentrations of the ice nucleating macromolecules per surface of the pollen grains) turns out to be very similar to the INAS density of mineral dusts. This is what we were referring to in the abstract. Two sentences have been reformulated to make this clearer: “At the same time, it is shown that INAS densities of some other biological aerosols, like certain pollen grains, fungal spores and diatoms, tend to be similar to those of dust. These particles may owe their high ice nucleation onsets to their large sizes.”

A hypothesis, which is also given in the discussion paper, says that the IN activity of dust is also not caused by the whole grain, but by certain active sites of the surface. Through the eyes of a chemist, we are particularly missing the chemical perspectives: Up to now, no investigation of the chemical nature of mineral dust ice nuclei has been carried out. The open questions are: What is the chemical nature of these active sites? Do they have the same composition as the bulk, or are they something else? Could they be external contaminants (e.g. biological), which are too small to be seen in typical phase composition measurements (e.g. XRD)? By which processes are these active

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sites formed, and how common are these processes in real life? It would be imbalanced to assign the IN activity to a certain type of crystalline phase, if it originates in fact from active sites, which could be of completely different chemical origin, and might not be necessarily present on all mineral dust particles of a kind. To gain more information about the mineral surface, one should initiate enhanced spectroscopic and microscopic surface characterizations of the nuclei, at least on some selected examples. Ample techniques might be scanning electron microscopy (SEM), Energy-dispersive X-ray spectroscopy (EDX), as well as vibration spectroscopy (IR, Raman, and neutrons), Low-energy electron diffraction (LEED) and X-ray photoelectron spectroscopy (XPS).

This is a good point. Such a discussion would go beyond the scope of this manuscript, but we have included this as an additional item into our recommendations: “Spectroscopic, microscopic and chemical characterization methods should be used to determine the nature and location of the ice nucleation active sites on various materials.”

In Fig. 5a data points from our paper (Pummer et al., 2012) are shown and labeled as onset temperatures, but in fact these are median freezing temperatures. This is important for consideration, because consequently the numeric values are relatively lower than those of studies where the onset freezing temperature is quoted (e.g. those by Diehl et al., 2002).

We have inserted a note into the caption of Fig. 3: “The data from Pummer et al. (2012) refer to median freezing temperatures.”

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References

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