

Interactive comment on “Deposition freezing on mineral dust particles: a case against classical nucleation theory with the assumption of a single contact angle” by M. J. Wheeler and A. K. Bertram

Anonymous Referee #2

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In this paper, Wheeler and Bertram discuss results from laboratory ice nucleation experiments with the clay minerals kaolinite and illite in the light of different formulations for heterogeneous ice nucleation rates or aerosol surface densities of ice-active sites. The paper contributes to the ongoing debate about the nature of heterogeneous ice nucleation and the best way of parameterising the formation rate of ice crystals in cloud and climate models. One of the major conclusions is that the lab results cannot be approximated or fitted with the classical formulation for heterogeneous nucleation rates assuming just a single contact angle α (the so-called single alpha model). Previous work which came to similar conclusions is adequately discussed and cited. The paper is well structured and written, presents interesting results, fits well in the

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scope of ACP, and can therefore be considered for publication in ACP after revision according to the comments listed below.

Comments and questions

My major concerns are related to the definition of the onset condition and the calculation of the ice-active particle fractions for comparison with the different formulations of heterogeneous ice nucleation.

Heterogeneous nucleation rates and ice-active particle fractions are calculated for the so-called onset conditions defined as the time when the first ice crystal was detected in the humidity scans of the experiment. From a statistical point of view it is the worst case to calculate the rate of a stochastic process from just one event. I understand that the experiment was repeated a number of times, and an average was taken for the actual analysis, but would still like to know the accuracy of J_{het} . It appears to me that Eq. 5 with one crystal nucleated only gives a lower limit for J_{het} . Only a few seconds later, more ice crystals could have been nucleated, or the one could have been nucleated in a shorter time interval which seems to be chosen somehow arbitrarily.

The experimental rate J_{het} is then compared to the single alpha model. Furthermore, an average particle diameter is used to calculate the total number of particles in the sample and from that the ice-active particle fraction at onset conditions as 1 over the total number of particles. This is an important approximation in the analysis and interpretation of the experimental results, because any of the formulations used in the manuscript rely, as far as I can see, on the assumption that all particles have the same freezing probability at a given set of contact angle, T and RH_i . I think the analysis would only be accurate for monodisperse particle samples. Therefore I would like to ask the authors to provide some more information about the range of particle sizes on the substrate, and to give at least an estimated uncertainty for the ice-active particle fraction and the mean particle diameter used e.g. in Eqs. 7, 8, 9 and 17. The approximations used in the fit equations should more clearly be mentioned and explained in

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the final manuscript, also at the end of the conclusion section.

Minor points

Title and p. 21172, l. 24: I agree to the comment by Denis Niedermeier that the term "deposition freezing" should be replaced by "deposition nucleation", according to the definition by Vali, G. (1985).

p.21176, l.20: How did the size from the laser scattering experiment compare to the projected area diameter determined with the optical microscope?

p.21178, l.8-12: I think, ice activity spectra as shown in Figs. 2c and 3c are only useful for predicting freezing in the atmosphere if measured or defined for at least size distributions of particles relevant for the atmosphere. This is e.g. discussed by Möhler et al. (2006).

p.21187, l.4: I would recommend to state here, that the single alpha model seems not appropriate to accurately predict heterogeneous ice nucleation by mineral dust particles. The current statement implies that the single alpha model is accepted as the classical or "natural" formulation, which I think is not the case.

References

Möhler, O., Field, P.R., Connolly, P., Benz, S., Saathoff, H., Schnaiter, M., Wagner, R., Cotton, R., Krämer, M., Mangold, A., and Heymsfield, A. J.: Efficiency of the deposition mode ice nucleation on mineral dust particles, *Atmos. Chem. Phys.*, 6, 3007-3021, 2006.

Vali, G.: Nucleation Terminology, *Bull. Am. Met. Soc.*, 66(11), 1426-1427, 1985.

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