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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Numerical simulations of homogeneous freezing processes in the aerosol chamber AIDA" *by* W. Haag et al.

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**General comments:**

This paper provides numerical simulations of the unique chamber measurements provided by the study of Möhler et al. The combined set of physical and numerical simulations provides important validation of our understanding of cloud formation at low temperatures. The paper is mostly well written and is a very worthwhile contribution to ACP. I stumble on just a few issues. The major one is that although understanding of homogeneous freezing of liquid aerosol solutions has been advanced in recent years, there remains a body of observations that indicate that the water activity-based parameterization of homogeneous freezing captures only the essence of the picture and may miss some significant details. Also, Tabazadeh et al. [2002, J. Phys. Chem. A, 106



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(43), 10238] and Djikaev et al. [2002, J. Phys. Chem. A, 106 (43), 10247] have published studies supporting that homogeneous freezing may not be primarily a volume nucleation phenomenon, as has been assumed, but a surface phenomenon. I suspect that there will be much discussion of this idea. I believe that there is evidence that some discrepancies in estimated nucleation rates for experiments done with solutions of supermicron emulsions (primary basis of the Koop et al. [2000] parameterization of homogeneous freezing) versus free-flowing aerosol particles are indeed reconciled if surface to volume ratios are considered. This is not the forum for presenting such data. Nevertheless, the point is that the simple and singular relation between homogeneous freezing and water activity is still being scrutinized. Can it nevertheless be used to reasonably simulate homogeneous freezing in cirrus? This paper suggests that it can, at least for more vigorous updraft conditions that allow relatively large fractions of the population to freeze. My second comment is a suggestion for the future utility of applications of the physical/numerical simulation methodologies developed, namely a forward exercise of the simulation methodology for determining ice nucleation rates in solution droplets. In particular, it would be interesting to see what nucleation rates are determined for numerically predicted compositions of monodisperse droplets in the chamber. That is, the model would not assume a nucleation rate, but would simply provide the basis for determining the soluble composition of particles at the point of freezing. Since the nucleation rates are typically higher in the AIDA simulations than in emulsion studies, such experiments would provide the most stringent test of existing understanding and could provide new insights.

**Specific comments:**

1) Introduction, Page 1469, line 13: Please clarify what is meant by "higher freezing thresholds", as in what quantity is higher?

2) Introduction, Page 1469, lines 16-19: There exists some contention in the scientific community with regard to this point. For this reason, the term "threshold relative humidities" requires definition. For example, the values given by the study quoted were



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based on the freezing temperatures of large fractions of 1 to 10 micron droplets. In accepting the water activity parameterization at face value, it should be acknowledged that the parameterization is based primarily on studies of larger drops, not aerosol particles (i.e., the types of studies of which the authors are critical on page 1470, lines 9-12).

3) Page 1470, lines 12-16: The cooling rates in the AIDA chamber are only "typical" of a certain population of upper tropospheric clouds formed by more vigorous vertical motions. These cooling rates are probably most characteristic of the higher values found in cirrus clouds, the most widespread of UT clouds, although clearly the community lacks for accurate and comprehensive measurement of vertical motions and cooling rates.

4) Page 1470, lines 20-24: It may be worth mentioning here that the hygrometer is used in an "ex-situ" mode for this study. Therefore, rather than assessing the mixing ratio for an integrated path across the chamber, sampling is from a point position in the upper part of the chamber.

5) Page 1471, lines 1-2: More explicitly than stated here, the average air temperature begins approximately 1 degree warmer than the wall temperature. It would help to state this in order to later understand why the base simulation begins at 90

6) Page 1472, line 9: What is meant by "We drive the simulations"? How? Are certain values reinitialized at periodic intervals? What values (temperature only?) and at what intervals?

7) Page 1474: In the description of the procedure for growing and evaporating ice crystals, it seems that the implicit account of ice water losses should "reduce", not "remove" the uncertainties in the simulated time history of relative humidity. I do not understand the sentence at lines 26-27. Does it mean that losses may occur from all ice crystal sizes?



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8) Page 1475, lines 13-15: It seems that the particle distribution in the chamber would be in equilibrium after some 200 minutes, but I wonder if any 5 micron and larger equilibrated particles would still be around after that amount of time. The companion paper by Möhler et al. uses 1.5 microns as an indication of ice particles in the chamber, so few if any sulfuric acid particles must be present at these sizes. Is it possible that the lognormal fit should be truncated to be truly representative of the aerosols present in the chamber?

9) Page 1476, lines 10-13: While the statement of the freezing conditions of different sized particles is excellent information, what is lacking here is a definition of the precise freezing conditions. That is, what is the definition of the "threshold value for homogeneous ice nucleation"? Is it where a certain fraction of aerosols have frozen or is it where a certain concentration of ice is predicted in the chamber? The measurements summarized in Koop et al. [2000] typically reported median freezing temperatures of droplet populations. On the other hand, the nucleation rate formulation derived by Koop et al. could be applied to predict some threshold freezing conditions as long as one realizes that the original data were never analyzed to determine where small fractions of drops freeze. Finally, as mentioned later in the paragraph, cooling rate must be a factor. The experimental cooling rate is mentioned later, but that mention needs to come earlier, along with a statement on the range of cooling rates for which the results will be strictly valid.

10) It would be interesting to know how well the prediction of larger ice particles agrees with observations from the ice particle counter. So, for example, what would the observations look like in the bottom panel of Figure 8?

11) Page 1481: In discussing wall effects on ice growth, it might be worth pointing out that temperature control of the chamber walls to approximate adiabatic cooling would reduce the artificial effects of water flux to growing ice crystals in future studies.

12) Page 1481: The statement of the value of the ice deposition coefficient used for



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simulations belongs earlier in the paper. This quantity is of course critical to the simulation of ice water growth. The consideration of uncertainty in deposition coefficient is discussed nicely later in the paper (section 6.3), although I have to note that since nucleation and ice growth are convoluted, it would be hard to know if both ice growth rate and nucleation rates are correctly simulated. It is necessary to believe that one factor is well known, selected in this case to be the nucleation rates.

13) Figure 11: It would be very useful to include estimates of uncertainty on the modeled results to account for the uncertainty of chamber RH measurements and how this is initialized. Error bars on a single point would be sufficient.

14) Page 1482-1483: The statement that the increased tendency for higher onset ice formation humidities than the parameterization curves is caused by the non-equilibrium composition of particles of D>2 microns is not consistent with Figure 8, which shows that most of the ice initially comes from particles smaller than 1 micron. I believe that the statement may still be correct, but the sizes may not. The other possibilities are that the cooling rate is at play and the nucleation rate parameterization J values are not correct for ice onset conditions. Could this not be the case?

15) The inclusion of sensitivity studies is excellent. These mostly deal with the impact on predicted ice number density. I would like to know how the uncertainty in initialization conditions affect the onset RHice as it is shown in Figure 11. This is the critical issue in my mind as to how appropriate the ice nucleation parameterization is for cirrus simulation and how well this study validates that.

16) Page 1489, lines 9-12: Again, I take issue with the blanket statement that the homogeneous ice nucleation parameterization can "safely be employed to simulate cirrus clouds". This statement belies the current state of agreement on the validity of that parameterization for the full range of cirrus conditions and nucleation rates, it provides no sense of the range of cooling or updraft rates for which the conclusion of satisfactory agreement is valid, and it does not consider unresolved factors such as the



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effect of organic aerosol components on freezing conditions.

**Technical corrections:**

Page 1478 and Figure 7: It would be helpful to state the basics of how ice concentrations were measured in the experimental study.

Section 5: The text should refer to Table 1 at the first mention of experiment numbers (e.g., B4-1). I did not know what these numbers meant initially.

Page 1483, lines 28-29: The first sentence of this paragraph should note that this discussion continues to refer to experiment C1-1 (e.g., sentence could end "in experiment C1-1").

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Interactive comment on Atmos. Chem. Phys. Discuss., 2, 1467, 2002.