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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Technical note: A numerical test-bed for detailed ice nucleation studies in the AIDA cloud simulation chamber" *by* R. J. Cotton et al.

**Anonymous Referee #1**

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General Comments:

This is an interesting study in which a microphysical model is proposed for comparing to AIDA chamber experiments. It will very be useful for future AIDA studies. It is suitable for ACP and I would recommend publication after careful consideration of the specific comments below. My specific comments largely reflect three more critical points. I believe that there could be better definition of and justification for assumptions made about ice nucleation and crystal growth in the model. For example, I would have expected some studies of the relative insensitivity of the results to assumptions on mass accommodation coefficient and capacitance factor for ice growth. I also ex-

press concern about how well crystal mass growth rate is constrained by the total water measurement and about the level of certainty that can be applied to the mechanistic interpretations of ice formation that are presented. The paper could probably be made shorter through more careful organization and removal of some non-essential discussion material.

Specific Comments:

The comments are listed in order of appearance.

1) Abstract, line 8: What is a “size-resolving parcel model?” Please clarify this jargon.

2) Introduction: I find that this section could be clearer in introducing the topic of the paper.

a. First paragraph, page 9484, line 22: In the absence of consideration of secondary ice formation processes, I believe it must remain a hypothesis that the “initial” ice formation mechanism might determine the concentration and mean size of ice particles, evolution of other microphysical processes and precipitation.

b. Page 9485, lines 2 to 3: Heterogeneous ice nucleation has always been considered the first process for ice formation in clouds warmer than about -38C, has it not?

c. Page 9485, lines 7 to 9: Jensen et al. (1998) implied, by way of model sensitivity studies only, that heterogeneous nucleation can initiate ice in cold wave clouds. However, their paper emphasized homogeneous freezing as the dominant factor. DeMott et al. (1998) showed that the contributions of heterogeneous nucleation, based on measurements of ice nuclei, appeared consistent with characteristics of ice formation in one of the specific cloud cases also examined by Jensen et al.

DeMott P.J., D.C. Rogers, S.M. Kreidenweis, Y. Chen. C.H. Twohy, D. Baumgardner, A.J. Heymsfield, and K.R. Chan, The role of heterogeneous freezing nucleation in upper tropospheric clouds: Inferences from SUCCESS, Geophys. Res. Lett., 25, 1387-1390, 1998.



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d. Page 9486, lines 10 to 13: Many recent papers allude to the results of Durant and Shaw (2005), but as here, this reference may not be appropriate or its application straightforward without further clarification or assumption. In order to be appropriate to explaining the INTACC results, it is also necessary that the basic population of ice nuclei within drops is greatly enhanced in the situation of contact from inside-out. If this process only occurs for particles that are otherwise immersion-freezing nuclei, then the results of Durant and Shaw would suggest that the available population of such contact freezing nuclei is no greater than the available immersion-freezing nuclei at a temperature 4 degrees colder. To my knowledge, no one has ever measured immersion-freezing nuclei in concentrations approaching ice crystal concentrations found in evaporating cloud regions. Thus, in the worst case, I find the allusion to Durant and Shaw as a rather empty piece of the puzzle. In the best case, it is also necessary to say that there exists a transient or latent population of contact freezing nuclei that greatly exceeds numbers of other IN.

e. Just to be clear, on pages 9486 to 9487, perhaps it should be mentioned that the microphysical model treats the 84 cubic meter chamber as a single “box” with one temperature and one ice particle population. These assumptions should be justified, by reference or otherwise.

3) Section 2

a. Page 9487, lines 20 to 21: Are ice particles also as well-mixed as the thermal environment of AIDA? In other words, is it certain that sampling at one location represents what is happening to ice crystals throughout the volume? Could there be extended growth lifetimes for some crystals versus others?

b. Page 9488, line 6: What are the heat sources in the chamber? I think this is not mentioned until later in the paper.

c. Page 9488, lines 11 to 13: Is there a reference for the strength of updrafts in the “convective” regions of cirrus generating cells? One does not often think of cirrus as



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convective. I would guess that 1 to 2 m/s is tops.

4) Section 4

a. Section 4.1, page 9490, lines 9 to 11: Since all ice crystals probably do not enter the hygrometer tube efficiently, under what circumstances is total water measured in this manner considered reliable at all? This becomes a critical issue in later discussion and my most significant concern with the paper as written.

b. Section 4.4, page 9491, lines 6 to 7: How are ice crystals sampled from the chamber into the SID sampling volume? Is there a reference for this method? If there is a sample tube, does flow through this tube impact sampling in any manner? Finally, as regards the 1.4 correction factor for presumed spherical ice, how does one know that these particles are in fact ice crystals? Is the baseline value that seems always present subtracted and might this baseline change if unactivated liquid particles are present? After all, CCN up to 2 microns are present in the dry aerosol.

5) Section 5

a. Page 9492, lines 21 to 26: In line with the last part of previous comment, I wonder if it is truly valid to state the general assumption that the CCN are only important for warmer temperature expansions. Are unactivated drops possible in simulations, even at sizes above 1 micron?

b. Section 5.2, page 9494, lines 1 to 13: The method selected to parameterize the wall vapor flux is the method using the MBW hygrometer as an accurate measure of the total water content. This raises the critical issue of particle sizes entering the MBW. The paper presently states that the total water is only 100% accurate for 7 micron and smaller ice crystals. How often does that describe the chamber ice distributions during critical measurement periods? I would expect not very often, except below say -50C and close to nucleation time periods. One especially notes larger crystals in Figures 5 and 6.



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C: Section 5.3, page 9494, lines 18 to 19: If most of the aerosol particles are larger than 0.1 microns, why are ice particles initiated at 0.1 microns? Does it make a difference if they are initiated at their actual sizes at nucleation? What is assumed regarding crystal capacitance and water vapour accommodation coefficient? It is not stated anywhere.

6) Section 6

a. Page 9495: Defining nucleation regimes seems an artificial construct that may not have explicit meaning. The figures suggest that nucleation could follow more than two modes and may in fact represent a continuum of behavior in regimes II and III.

b. Page 9495, lines 27 to 28 and continuing on page 9496: The condensate mixing ratio depends both on the model microphysics and the assumption made on the validity of the total water measurement. It seems possible that the agreement with the TDL could be a convoluted and fortuitous result.

c. Page 9497, section 6.2: This paragraph touts the agreement between model and measured RH, but one wonders if this is expected given that ice crystals larger than 10 microns, not efficiently sampled by the MBW, are already present at the point that the second nucleation mode is observed. Could the actual RH be higher? Also, since the separation between nucleation modes becomes more evident in secondary expansions, does this imply that the most efficient IN are lost from the total aerosol (sedimentation and other losses)? Has this been considered and does the model account for it at all?

d. Page 9498, section 6.3: Temperature regime III is said to contain two nucleation modes. Yet the first mode is similar in magnitude to the continued rise in the ice signal between modes in temperature regime II. This is what I mean by the artificiality of the multiple “mode” construct. It looks like it could be a continuum behavior.

e. Page 9498, section 6.4: The fact that ice formation appears to require water saturation/liquid cloud conditions really says little about the exact ice formation mechanism,



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even whether or not it rules out deposition, as suggested in the last sentence of this paragraph. It seems enough to simply say that water condensation appears to be involved based on the conditions of activation.

f. Pages 9498-9499, section 6.5: I have a major comment on this section. One must question the validity of the line in Fig. 8 indicating the "critical ice saturation for freezing of ice in aqueous ammonium sulfate drops" for defining heterogeneous freezing conditions. This line is specific to the freezing of 50% of 20-50 micron pure solution drops by specific clay minerals of large sizes. I note that Archuleta et al. (2005) found similar freezing conditions for rather small metal oxide particles, so dust size may not be critical. Nevertheless, it should probably be stated that this line may not be well anchored for the specific dusts used, nor do you know if the freezing conditions might be quite different for the small (if any, since it is not inferred in any way) amount of soluble matter present on mineral dusts used in the present study. The consequence is that I believe one has little justification in concluding that the second ice nucleation mode is "probably" immersion freezing rather than homogeneous freezing. Is there any ability to conclude so when considering the validity of applying the Zuberi et al. results and within the stated RH uncertainty of measurements in this study? Also, is this not a central topic of the Field et al. paper already published?

Archuleta, C.A., P.J. DeMott, and S.M. Kreidenweis, Ice nucleation by surrogates for atmospheric mineral dusts and mineral dust/sulfate particles at cirrus temperatures. Atmos. Chem. Phys., 5, 2617-2634, 2005.

f. Page 9500, section 6.6: Although interesting, I find this discussion and Fig. 9 rather unconvincing for distinguishing heterogeneous and homogeneous ice formation mechanisms. For this reason, I think this section could be pared. The fact that observed ice formation spreads over a broader range of conditions than expected for freezing particles as if they were solutes could be an artifact of assuming that crystals are perfectly well mixed in the chamber (versus some settling), the existence of lower and upper size thresholds for observation of crystals, and the possibility that a range of soluble



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contents could be spread across a range of dust particle sizes.

7) Figures 5 and 6: Reflecting earlier comments, although there are two distinct modes of ice formation in Figure 5, the signal is not really constant between them right? It is no more constant than for the “first” mode identified in Figure 6.

8) The SID size threshold is variously stated or used in different places in the text and figure captions as anywhere from 1 to 4 microns. Please correct for consistency.

9) Figure 8: I think a line is shown for water saturation conditions, but it is not identified. The caption should also note that the dashed line is immersion freezing for micron-sized kaolinite particles in 20 to 50 micron ammonium sulfate drops. The caption mention of the homogeneous freezing lines should reiterate that these are for two specific “dry” particle sizes (0.5 and 5 microns).

10) Figure 9: The caption mentions variations of C but does not say what the mass accommodation coefficient was fixed at for ice mass growth calculation.

Technical Corrections:

Page 3, sentence 4: Replace “were” with “where”

Page 15, line 3: remove “of” after “growing”.

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Interactive comment on Atmos. Chem. Phys. Discuss., 6, 9483, 2006.