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Atmospheric Chemistry and Physics Discussions

# *Interactive comment on* "Modelling the effect of condensed-phase diffusion on the homogeneous nucleation of ice in supercooled water" *by* Kathryn Fowler et al.

**Anonymous Referee #2**

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The authors extend their previous development of a single-particle aerosol model, which they used to calculate aerosol-water vapor equilibration times for individual organic aerosol particles exposed to a step change in relative humidity. Here they use the new model to calculate the response of an aerosol size distribution that is initially at equilibrium with water vapor and then ascends until homogeneous freezing leads to an approximately steady state ice crystal number concentration. They consider a range of aerosol size distribution parameters, ascent rates, and initial parcel thermodynamic states. The methodology appears sound, although I am not an expert in all details, except that some additional information is required to make the results reproducible. The conclusions appear generally reasonable, but should be placed more quantitatively into



the context of the deficiencies in current models that motivated this work. There are grammatical errors that can be readily handled by a copy editor; I only note one below where the meaning was not clear to me. To the limits of my expertise, I would rate a revised manuscript as suitable for publication in ACP if it addresses the following questions.

General comments

1. Reproducibility

1a. Is the ascent adiabatic with respect to the model parcel? If so, parcel temperature must account for the latent heat of condensation. Please explain how environmental temperature and water vapor mixing ratio are calculated during parcel ascent.

1b. Somewhere before section 4, please report the size bin structure used in the model (such as smallest bin size, bin spacing, and total number of bins), as well as the embedded radius bin structure for the new model (such as whether it is the same for all particle size bins), and how these were chosen.

1c. D* and k* that appear in equation 2 are not called out in the text. Please clarify.

1d. Please add at least one example of shell molar ratios, including nucleation rates as a function of shell and size bin. The non-monotonic behaviors seen in Figure 5 and Appendix B leave me skeptical about whether the model is numerically converged, and this would help another group reproduce your results, in addition to aiding some explanations.

2. Statement of the problem

2a. In the introduction, please be more quantitative about what is meant by "low temperature" (page 2, line 5).

2b. What is the significance of the clouds being sub visible and how is that defined?

2c. Page 2, lines 6–7: Quantitatively by how much are numbers and supersaturations

lower and higher than expected, respectively? In other words, how much of a problem is this?

2d. Page 2, line 14: What is meant by the “magnitude” of "both the physical and chemical aerosol properties" in the TTL?

2e. Page 2, line 14: Does the reported composition differ in any known way from aerosol particles elsewhere in the tropical upper troposphere? Please provide context.

2f. Page 2, line 20: Are you referring specifically to the tropical "upper" troposphere? Please provide some quantitative or definitional indication of what zone you’re referring to, and how it may be distinguished from cirrus globally. I would also replace “were” with “are”.

2g. Page 2, line 28: Grammatically, ‘however’ should be ‘but’ or a new sentence begun. That said, I don’t understand how these clauses are oppositional, so I would just start the second clause as a new sentence with “Further. . .” Am I missing something?

3. Explanation and interpretation of results

3a. Page 11, line 1: This statement is not true for aerosol size distribution 2 as far as I can tell. Please note and explain that to the reader.

3b. page 11, line 8: I think the authors have the model output to evaluate this hypothesis without extensive additional calculations. Is there a reason that they stopped at suggesting it?

3c. Bottom of page 12 and Figure 6: I do not understand how the control model can give the same result if freezing is occurring at different temperatures and pressures along the trajectory owing to different initial supersaturation. Freezing rates should depend on temperature and pressure trajectory, right? What am I missing?

3d. Figure 6: Please report in the text which aerosol size distribution is used for these simulations.



3e. Also Figure 6: Does the trend change if the aerosol used changes or the cloud base temperature changes? As a reader, I can’t place this monotonic trend into the context of obviously non-monotonic trends discussed thus far.

3f. Bottom of page 13: Inside-out nucleation is based on the heterogeneous ice nucleus touching the air-water interface. Are you suggesting that the more hydrated core is touching the solution-air interface? That seems unlikely to me. What is keeping it less hydrated is the proximity to the surface itself. Otherwise I don’t see how this can be appropriately called inside-out nucleation, whether or not other authors have done so.

3g. Page 14, line 2: Isn’t this just a matter of changing the initial or boundary conditions on your simulations? That does not strike me as complicated model development. Or am I missing something? Please explain whether any equations used to integrate the model need to be changed to account for this, and if so, why.

4. Significance of results relative to stated problem

4a. Last sentence on page 15 comparing simulated to grossly observed concentrations and concluding that the new model is better: I think this is quite an overreaching statement. Both models seem to span the enormous, five orders of magnitude range in ice crystal number concentrations reported equally well to me. Or perhaps neither model can reproduce the lowest ice crystal concentrations? And isn’t the control simulation the only one that is generating the highest ice crystal number concentrations measured? I think the main point is just to show how the models differ and that both models reproduce values within the (huge) observed range, but the comparison with observations at this level is not distinguishing between these models meaningfully.

4b. Please discuss quantitatively whether differences in terminal supersaturation and ice number concentration in control versus new models are consistent with the model deficiencies discussed in the introduction, which have motivated this work. Quantitatively, specifically, what degree of ice supersaturation and number concentration respective excesses and deficiencies relative to expectations from control-type models

have been observed and are they roughly similar to what you are finding in your new versus old model as a function of temperature (which look to be an order of magnitude in number concentration and 10% in terminal supersaturation at the upper end)?

5. Minor suggestions

5a. Can Table A2 be placed after Table 1? I see no need to put so little material that is actually very relevant into an appendix. Or put the figure in the appendix and retain the table, which is more quantitatively understandable vis-a-vis past data sets for those experienced with aerosol measurements (i.e., one can read off a meaningful number concentration). Can the columns be labeled size distribution 1, 2 and 3 or made to otherwise match the column headings in Figure 5? References can be lowered to table line item. I had to keep flipping back and forth between appendix and text with figures to remember the number concentrations and basic differences.

5b. Figure 5: Rather than writing the enhancement and suppression regions, could the authors add some contours that show where the differences are larger than, say, 10% and 50% or similar relevant round numbers?

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