Atmos. Chem. Phys. Discuss., 9, C3304–C3308, 2009
www.atmos-chem-phys-discuss.net/9/C3304/2009/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Parameterizing the competition between homogeneous and heterogeneous freezing in ice cloud formation – polydisperse ice nuclei" *by* D. Barahona and A. Nenes

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Received and published: 24 July 2009

We thank the referee for the thorough and thoughtful comments presented.

It is extremely difficult to follow the reasoning and the derivation throughout section 3 even after reading the two precursor papers.

We apologize for this, and have reworked the section for clarity.

1) It would be very helpful if the authors provide a roadmap telling the reader step by step what they aim at and for which purpose.

Great point. A paragraph has been added to section 3.2 summarizing the main aspects of the derivation.

2) In there derivations the authors introduce a number of ancillary parameters like $D_{c,char}$, etc. But often the argument comes that something is a strong function of s_i and so it is dominated by its value at $s_{i,max}$. In view of this, is it really necessary to introduce all these ancillary quantities? Can't you simply express everything just in terms of $s_{i,max}$? It might be that that would render the errors larger, but it might also be that you can hide all these complications with the ancillary quantities within suitable correction factors (or functions), introduced simply due to further insight or due to parcel model results. This would probably allow to disentangle the derivation substantially."

We agree that the derivation could be presented in a simpler manner, and thank the reviewer for encouraging us to rework section 3. This has been done, and presented in section 3.2 of the revised manuscript.

Fitting functions from parcel simulations are not necessary, especially since we have developed an analytical solution to the parcel equations, the final form of which is quite simple (despite the admittedly involved derivation).

3) Section 2.2 uses classical nucleation theory to derive the nucleation spectra. However, on pg. 10962 the authors give arguments in favour of the "singular" hypothesis. To my view (please correct me if I am wrong) the "singular" hypothesis does contradict the classical theory which is based on the "stochastic" hypothesis. Can you please clarify that?

CNT indeed has a temporal dependence, and the singular hypothesis does not. Application of each would give a nucleation spectrum, which could be used in the framework. The only issue is not having a temporal dependence, so that Eq.1 applies. Assuming that enough time is allowed for heterogeneous freezing during the IN measurement, the stochastic component of CNT is small, the CNT-based heterogeneous freezing spectra would be independent of time and consistent with Eq. 1.

We have included the above discussion in section 2.2.

4) Like Reviewer 1, I find that your statements are sometimes too bold. For instance on pg. 10960 you say that “these correlations are restricted to (largely unconstrained) assumptions ...”. I do not see that your paper does any contribution to further constrain the unsafe assumptions.

True, we do not address the uncertainties related to the values of these parameters, but this is beyond the scope of this work. Here we develop the framework and demonstrate its use; constraining the parameters requires assimilation of observations, and will be the goal of future work.

I do neither see that your paper addresses “all the limitations of previous approaches”. So please tone that down.

The statements were meant to characterize the new framework, when examined against existing parcel-based mechanistic parameterizations. We have removed the statements to avoid similar misinterpretation.

5) Finally I would like to see some more outlook on the possible application areas of the parameterisation and on its restrictions. For instance, in a cloud resolving model where one can resolve the nucleation phase using small time steps, one will probably not switch to your method. Although the results might become questionable when the first crystals formed heterogeneously have time to fall out from the grid box (from the parcel) before smax is reached.

We have included a paragraph with such discussion in the section 5 of the revised paper.

Specific Comments pg. 10959, l. 9: unclear sentence.

The sentence has been removed.

pg. 10962, ll. 15, 16: the density should not depend on the surface area.

The statement now reads: “The aerosol freezing fraction is then related to the density of active nucleation sites (which generally depends on particle history and chemical composition (Pruppacher and Klett, 1997; Abbatt et al., 2006)) and to the surface area and number concentration of the aerosol population.”

pg. 10965, l. 1: how can a constant depend on something?

The statement now reads: “. . . and C depends on the mean surface area of the j^{th} aerosol population, $\bar{\Omega}_j$.”

pg. 10965, l. 21: better write “because depletion balances the s_i increase due to cooling”.

Corrected

Eq. 10: On first reading it is unclear what you mean with neglecting of non–continuum effects and where the Γ_2 is gone. You could refer to Appendix B here.

The statement now reads “assuming negligible non-continuum effects on mass transfer; i.e., $\Gamma_1 >> \Gamma_2$ (Appendix B), and, $\frac{dD_c}{dt} \approx \frac{s_i}{\Gamma_1 D_c}$.......(Barahona and Nenes, 2008),”

pg. 10967, l. 22: the assumption that s_i is generally above 20% is not always fulfilled.

True. Significant growth however occurs only for large s_i values. We have changed the sentenced to “(s_{max} generally above 20%)” which is more accurate.

Eq. 11: I was expecting an explicit formulation of $D_c(s_i - s_o^{'})$ but that’s probably not possible?

Generally it is not, because $D_c\,(\Delta s)$ depends on $\frac{ds_i(t)}{dt}$ (Eq. (10)) which is not known in advance. As we show however, knowing the specific functional form of $D_c\,(\Delta s)$ (or for $n_s(s_i)$) is not required to solve Eq. (7).

pg. 10968, l. 15: No! they grow as long as $s_i > 0$.

pg. 10970, l. 16: this is a constant, but no "integration constant" (different meaning).

We thank the reviewer for these points. The statements have been deleted in the new derivation (see comment (2) above).

pg. 10977, l. 23: you should say that homogeneous nucleation was switched off, otherwise smax = 1 is hardly possible.

The statement now reads "…for pure heterogeneous freezing (i.e., homogeneous freezing switched off) and from 0.05 to 0.6…"

pg. 10978, l. 13: "a slight...". Corrected

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 10957, 2009.