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Atmospheric Chemistry and Physics Discussions

**ACPD**

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Interactive Comment

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

**Anonymous Referee #2**

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## 1 General

The paper presents a generalisation of earlier work by the same authors (Baharona and Nenes 2008, 2009) aiming at an analytical method for computation of the crystal number density formed heterogeneously and homogeneously during a single nucleation event in an uplifting air parcel. The generalisation uses a "nucleation spectrum" which I think is a clever idea.

Although I believe that the derivations and results in the paper are mainly correct (and the paper is suitable for the journal), I suggest a major revision of its presentation.

## 2 Major comments

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

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.

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.

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?

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



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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 $s_{max}$ is reached.

## 3 Specific comments

pg. 10959, l. 9: unclear sentence.

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

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

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

Eq. 10: On first reading it is unclear what you mean with neglecting of non–continuum effects and where the $\Gamma_2$ is gone. You could refer to Appendix B here.

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

Eq. 11: I was expecting an explicit formulation of $D_c(s_i - s_0\prime)$ but that's probably not possible?

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).

pg. 10977, l. 23: you should say that homogeneous nucleation was switched off, otherwise $s_{max} = 1$ is hardly possible.

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



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Final sentence of summary and conclusions: Tone down (see above).

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



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