Atmos. Chem. Phys. Discuss., 9, C3299–C3303, 2009
www.atmos-chem-phys-discuss.net/9/C3299/2009/

Atmospheric Chemistry and Physics Discussions

ACPD

9, C3299–C3303, 2009

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

A. Nenes

nenes@eas.gatech.edu

Received and published: 24 July 2009

We thank the referee his/her comments.

While this study represents a step forward than their earlier studies (Barahona and Nenes, 2008; 2009) and also earlier work from theoretical point of view, I feel uncomfortable with the tones the authors uses to describe their work throughout their abstract and summary and conclusions, e.g., "any" size distribution and chemical composition; "any" form; "excellent" performance'; "extremely" fast";

addresses "all" the shortcomings of previous approaches.

The statements were meant to characterize the new framework, when examined against existing parcel-based mechanistic parameterizations. We never implied that all the issues of treating ice clouds in GCMs are addressed. The statements have been removed to avoid similar misinterpretation.

The important part of their parameterization framework is the heterogeneous nucleation spectrum function. One source is based on the earlier works which were derived from the observations: Meyers et al (1992) and Phillips et al. (2008). Meyers et al. formulation was derived from surface measurements. Extrapolation to upper troposphere regime with lower temperature and lower aerosol concentrations has been shown to greatly overestimate IN concentrations; Although Phillips et al. formulation claims to be used in ice cloud regime (T$<$-35 C), most data used to derive the formulation were obtained in several field campaigns in the mixed-phase cloud regime (T$>$-35 C). Another source for the spectrum function used in this study is from the CNT. However, CNT approach is fraught with uncertainty because there are a number of unconstrained parameters (e.g., contact angle). The simplicity in this theory fails to explain the complex heterogeneous ice nucleation process. Marcolli et al. (2007) observed the quantitative agreement with measured heterogeneous immersion freezing temperatures by assuming a distribution of contact angle among the dust particles.

The nucleation function is without doubt a fundamentally important input to the parameterization framework. The IN spectra presented here are not meant to be an exhaustive and complete review of published data; they are just examples to illustrate how flexible the framework is. More complete and constrained formulations can (and will) be used as they become available. The limitations of the CNT formulation have also been discussed (in section 2.2 of the revised paper).

We would like to add that a comprehensive IN spectrum is not the only important part

ACPD

9, C3299–C3303, 2009

of a physically-based parameterization; the other is correctly accounting for the supersaturation dynamics (as expressed by the original cloud parcel equations) that drives ice nucleation. By explicitly separating the nucleation function from the subsequent ice growth, an accurate and analytical solution to the parcel equations is possible (and derived), that accounts for the combined effects of homogeneous and heterogeneous freezing.

Specific Comments

1. Page 9. Values for ef,j, sh,j, and θj used in this study (section 4.1, Table 1). I don't see θj in Table 1.

The specific values are now listed in Table 2.

2. Page 11. Line 5 from bottom. Does s0' (freezing threshold) here and also below have the same meaning as sh,j? if so, please use the same symbol.

No, they do not. We have clarified their difference as follows: "$s_{h,j}$ is associated with the onset of large nucleation rates at which the aerosol freezing fraction reaches a maximum."

3. Page 12. Equation (11). Is Dc(si- s0') a function of Dc, or it multiplies Dc? The same is true in Equation (12).

We thank the reviewer for pointing this out. It means "a function of". The meaning of Eqs. (11) and (12) have been clarified in the revised paper.

4. Page 13. Equations (15), (16), and also below. Should s be si? Why do you change s in equation (15) to Δs in equation (16) and also in below?

s is used as an integration variable. s_i (or s_{max}) is the upper limit of the convolution integral

5. The title of section 3.2 is to determine smax. Then how and where is smax determined in section 3.2. by Equation (15)? I am not clear how you determine

ACPD

9, C3299–C3303, 2009

Interactive Comment

smax there?

We have simplified the derivation in section 3; the determination of s_{max} should be much clearer now.

6. Page 19. Lines 2-3. Do you use Equation (34) together with nucleation spectrum Nhet in Table 1 to determine smax?

Yes. It is mentioned at the end of the paragraph (Eq. 34 is now Eq. 26 in section 3.2).

7. Page 21. Equation (38) I don't know why you want to apply your equation (34) to T>235 K (mixed-phase cloud regime). In this regime, cloud microphysics and dynamics are more complicated than the ice nucleation under the dynamical framework in this study.

Although T<235 K defines the onset of homogenous nucleation, it is still possible to have pure ice clouds at higher T via heterogeneous nucleation. Thus, Eq. (38) (now Eq. (30)) is still applicable, provided that the relative humidity with respect to water remains below 100%. This is now mentioned after Eq. (38) (now Eq. (30)).

Also what do you mean fc<0? If there is only homogeneous nucleation it is fc=0.

$f_c \leq 0$ means that $N_{het}(s_{hom}) > N_{lim}$ and the growth of heterogeneously frozen crystals is enough to prevent homogeneous freezing, i.e., $s_{max} < s_{hom}$. $f_c < 0$ indicates that only *heterogeneous* nucleation takes place.

8. Page 25. Comparison with existing schemes. As you note, LP parameterization used αd = 0.1 and also assumed sh,j (threshold RHi)=1.2. Both of these will predict a higher ice number from heterogeneous nucleation and also limiting IN number and updraft velocity than that from this study which uses αd = 0.5 and sh,j=1.3. Thus I would like the authors to present the sensitivity of their results to different αd = 0.1 and sh,j

Good point. We have added new curve to Figs. 6 and 7 (now Figs. 5 and 6) showing

ACPD
9, C3299–C3303, 2009

Interactive Comment

the sensitivity of our comparison to the values of α_d, $s_{h,i}, e_{f,j}$, which is also discussed in section 4.2.

9. Why does smax from LP exceeds shom when V$>$0.2 m/s in Figure 7?

Excellent point and we apologize for this oversight. The expression provided in LP05 for s_{max} is only valid for heterogeneous nucleation and should not be applied for homogeneous nucleation. We have corrected the plot by setting s_{max} = min(s_{hom}, s_{max}) for LP05.

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

ACPD

9, C3299–C3303, 2009

Interactive Comment