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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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This study presents a dynamical framework for ice cloud formation which accounts for homogeneous nucleation, heterogeneous nucleation (based on current empirical IN formulations available in the literature or on the classical nucleation theory CNT), and competition between the two mechanisms. 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 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.'s 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.

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.

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.

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

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?

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 smax there?

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

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

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 and sh,j.

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

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