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Atmospheric Chemistry and Physics Discussions

Interactive comment on "On the ice nucleation spectrum" *by* D. Barahona

Anonymous Referee #2

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This work is an important contribution to the description of heterogeneous ice nucleation. The description of the surface properties of ice nuclei is the most challenging part in heterogeneous ice nucleation, and various attempts have been made to incorporate the aerosol surface into classical nucleation theory. The present work offers a concise, pragmatic formalism to the community, which is likely to become used in atmospheric modeling studies. Based on the developed formalism, the assessment of the influence of different parameters on the resulting frozen fraction in deposition nucleation contributes to the understanding and to a feeling for this ice nucleation mechanism. I also appreciate the excellent structure and the thoroughness of the paper. Hence, I recommend this manuscript for publication. However, as the emphasis of this work is on the mathematical description of the respective freezing mechanism and the physical significance of the developed formalism, derivations should be comprehensible to the reader, which in my opinion is not always the case.

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Interactive Discussion

Discussion Paper

General comments:

1. The concept of the nucleation time should be explained better in order to clearly differentiate it from the integration time t or the duration of an experiment, delta t_exp. In chapter 2 about general theory, it is not a priori clear that the introduction of a nucleation time scale and hence the approximation in equation (8) is justified, given the fact that J_hom usually depends strongly on T and S. The justification of such an approach rather seems to be the result of the calculations in 3.1.1 and 3.2.2.

2. In 3.2.3, it should be clearly stated that the dependence of the deposition ice nucleation spectra on different influencing parameters, as shown in Fig. 3, relies on the assumption that the nucleation rate coefficient J_het is constant throughout the surface of an individual ice nucleus. In the general introduction in section 2.1.2, two different approaches are presented, one of which uses a surface distribution of active nucleation sites, and the other uses a nucleation rate coefficient. The derivations in section 3 are based on the latter approach, whereas the former is not considered further. This choice should be mentioned and motivated, since the choice of either assumption to describe heterogeneous nucleation can influence e.g. the results presented in Fig. 3.

3. In chapter 2.1.2, it seems that the second approach (with rho_s describing the IN surface) refers to the singular hypothesis about heterogeneous ice nucleation. However, the concept of preferential sites for ice nucleation on the surface of an IN can also be combined with the stochastic concept of a nucleation rate coefficient (e.g. Marcolli et al., 2007). Although adapting the formalism for deposition nucleation to further concepts involving active nucleation sites might be beyond the scope of this paper, it would be interesting if using such descriptions would significantly change the dependency of f_f on T, S, etc.

4. On line 59-60 it is stated that existing models of describing the surface properties of a population of IN rely on idealized pictures of the particle surface structure. In what respect can the formalism presented here be considered as less idealized? In fact, the

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quantity "xi“ used in the NPDF seems to be based on the assumption of a constant contact angle throughout the surface of an IN. If this is true, then this formalism does not conceptionally differ from some of the concepts used in the stated literature. If this is not true, then more explanation is necessary to prevent misunderstanding.

Specific comments:

Line 69: Add “approximately” to “singular behavior”. If the ice nuclei followed strictly singular behavior, then fluctuations in the ice embryo size would lead to negligible spread in freezing temperatures, which is not the case according to Vali (2008).

Line 114: How exactly is n_c in eq. (6) defined? Shouldn’t the left side of eq. (6) also be a differential?

Line 124: I think if the upper limit of integration in eq. (7) is t, then the variable in the integral should be different, for example t’. This also holds for more equations of the same kind.

Line 136. Please give a precise reference (including the equation number in Pruppacher and Klett (1997)) for rho_s. Does rho_s represent the surface density of sites that nucleate ice at specific conditions (T,S) according to the singular hypothesis, or is it a function of e.g. contact angle, as introduced by Marcolli et al. (2007)? Furthermore, I suppose that rho_s should also be written instead of rho_as as in eq. (9, 10, 11) and several other places.

Line 146: Is the proportionality in eq. (11) valid independently from the functional form of the NPDF?

Line 181: The derivation of equation (20) from eq. (18) and (19) is not clear, and should become comprehensible to the reader. More steps would probably help, and the cooling rate gamma has not been introduced up to this point.

Line 221: The sentence “The NPDF allows a finite probability. . .” does not make sense to me. Explain better.

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Line 241: The sentence “To assure physical consistency. . .” needs further explanation, and probably a reference to eq. (22).

Line 275: On the right hand side of eq. (34), where has the prefactor 1/sqrt(f) gone that is present in eq. (26)? Furthermore, it should be mentioned that Δg_g has to be evaluated at f=1.

Line 277: In eq. (35), f should be a function of theta, but the derivative should be evaluated at the characteristic theta. I guess this would be mathematically more rigorous.

Line 279: The step from eq. (35) and (36) is not comprehensible to the reader. Give more details.

Line 340: Is it justified to speak about a “previously unidentified behavior of homogeneous freezing”? I think it is immediately clear from CNT that the presence of small droplets limits the frozen fraction.

Line 362: Along the lines of general comment number 2, one might add here that since an alternative description of deposition nucleation using a distribution of active sites would probably be closer to the singular hypothesis than the formalism developed in the present manuscript (assuming a constant contact angle on individual IN), the observed temporal dependence of the frozen fraction might be considered as an upper limit for temporal effects.

Technical comments:

Line 174: The subscript of J should be “hom” instead of “het”. This also applies to the left hand side of eq. (17).

Line 197: Insert “of” in front of “mixed-phase”.

Line 204: There is a double “and”.

Line 235: Add “ice germ” in front of “surface”.

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Line 268: in the calculation of n_g, r_g should be raised to the third power.

Line 363: Replace “increases” by “decreases”.

Figure 2: Shouldn’t gamma have a negative sign?

NB: All references refer to references given in the manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 29601, 2011.

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