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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Assessment of parameterizations of heterogeneous ice nucleation in cloud and climate models" *by* J. A. Curry and V. I. Khvorostyanov

Anonymous Referee #2

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In this paper the authors use parcel model simulations to compare empirical parameterizations of ice nuclei concentration against predictions of the "extended" classical nucleation theory formulation developed by the authors (KC) for a number of cloud formation conditions. In doing this, the authors address some criticism raised to the KC approach in the works of Eidhammer, et al. [EDK09, 2009] and Phillips, et al. [PDA08, 2008]. They also discuss applications of the KC formulation in cloud resolving models.

GENERAL COMMENTS

When comparing empirical correlations against nucleation theory there is an issue of thermodynamic/kinetic interpretation of the ice nucleation process. The implicit assumption behind an empirical correlation of ice nuclei (IN) concentration derived from cloud chamber measurements is that the production of ice crystals is controlled by thermodynamics. In this framework, once the conditions associated with the freezing of an ice nucleus are reached, it freezes instantaneously. On the other hand, in the kinetic framework of nucleation theory, ice crystal concentration (Nc) depends on the residence time of the IN at some given conditions. This is because Nc is obtained by integration of the nucleation rate over a period of time. There is evidence that the temporal dependency of nucleation rate itself is second order [Vali, 2008]; Nc is however time-dependent.

In a cloud the time that aerosol particles spent at some conditions of Sw and T is controlled by the updraft velocity. Since updraft also controls Sw, nucleation and supersaturation cannot be separated. There are other disadvantages to the parcel model approach - i.e., negative feedbacks between ice crystal growth and supersaturation cannot be removed, uncertainty in the parameters that control the diffusional growth of ice crystals, and approximations defining the aerosol characteristics (insoluble fraction, composition, contact angles) - that add uncertainty to the assessment. This put into question the use of a parcel model to compare empirical correlations against nucleation theories, mainly because one is always bound to assume some dynamical conditions. In EDK09 and PDA08 high updraft velocities (above 1 m/s) were assumed, reflective of the idea that the CFDC measurements represent a thermodynamic limit of IN, which however remains to be proven. This in turn led EDK09 to propose the use of empirical constraints in theoretical models; however if compared at the right conditions (i.e., those inside the cloud chamber) such constraints may not be needed.

In the present manuscript, the conditions selected by the authors for the comparison between the KC approach, empirical correlations, and observations seem arbitrary. Particularly, Figures 4 and 5 suggest that by selecting the “right” updraft one may be able to reproduce any set of observations. However there is no point in doing that unless the updraft velocity represents the conditions at which the observations were taken. I think if some common ground between empirical and theoretical approaches can be found, this paper can be turned into a very important contribution to cloud physics. However in its present form, the authors' assessment do not represent a "clean" (i.e., without the effects of parcel model assumptions and made at the appropriate dynamic conditions) comparison of heterogeneous ice nuclei parameterizations. In my specific comments, I suggest some ways in which these issues can be addressed.

The authors also deal with many different topics in this work. They compare the results of their approach against climatological data, calculate thermodynamic constraints to ice nucleation, develop an ice crystal formation parameterization, and model Artic clouds. I consider all of these subjects relevant. However, in condensing them in a single work I feel the authors oversimplified their discussion in each section. To develop those topics appropriately would excessively extent the length of this manuscript. I recommend addressing these topics in separate works.

SPECIFIC COMMENTS

SECTION 1

Page 2671

Lines 9-12. It is true that the authors have rewritten the expressions of classical nucleation theory in terms of atmospheric relevant variables but in no way have they modified or extended any of CNT conceptual tenets. The dependency of nucleation rate on supersaturation is probably the most basic concept in nucleation theory, as expressed in the nucleation theorem [Kashchiev, 2000]. The existence of misfit strain, and active sites was proposed some time ago. The statement should be rewritten reflecting the authors' actual contributions.

Lines 13-15. How would the expressions developed by the authors help to improve CNT itself? It would be good to give an example.

Page 2672

Line 8. Give some examples of such applications of MDC92.

Page 2673

Lines 14-22. This statement is misleading. It suggests nothing happened over the last 100 years in nucleation theory and that supersaturation was never considered. The dependence on supersaturation (seen as the difference in chemical potential between old and new phases) is basic to nucleation theory [Kashchiev, 2000]. In ice nucleation it was assumed for a long time that the entropy of mixing was negligible and therefore no dependency on concentration/activity. It is great that the authors included such dependencies in CNT expressions written in terms of atmospherically relevant variables. However these statements should be rewritten reflecting the authors' actual contributions.

Page 2674

Equations 4-7. CNT predictions are very sensitive to the values of the physicochemical properties of the ice germ. It is not fair for a reader to have to track 4 or 5 papers to get the value of a single parameter. Please provide the specific values for all of the parameters in Eqs. 4 to 7 (or at least the specific references where they are taken from).

Page 2676

Line 10. Do the authors consider the dependency of mis, and ΔFact on relative humidity in this work?

SECTION 2

Page 2677

The authors suggest rcr<0 as a criteria for thermodynamic validity. This assumption however faces somes issues of CNT:

- Before rcr <0, there must be a region where rcr is small enough to be composed of

ACPD

10, C1123–C1131, 2010

just a few water molecules. However for this germ the capillarity assumption of CNT is no longer valid and CNT assessments are inaccurate. By extension, the “thermodynamically valid” rcr cannot be calculated using CNT. It is likely that rcr would tend to zero in a different fashion than predicted by CNT.

- rcr<0 as a thermodynamic constraint suggests that stable ice germs composed of a single water molecule should exist. For that case nucleation would always be thermodynamically favorable, which is contrary to observations. Even if they exist, CNT is not adequate to calculate the nucleation work of such single-molecule germs.

- CNT itself is not thermodynamically consistent. This is because the work of germ formation predicted by CNT does not approach zero at the spinodal point [Kashchiev, 2000]. Therefore it cannot be used to test the thermodynamic consistency of experimental measurements.

Please explain how these issues can be addressed when applying Eqs. 10 and 11.

SECTION 3

Equation 12. This equation has a qualitative rather than quantitative justification. Sensitivity to its parameters should be studied.

Page 2680.

Lines 23-24. All the references provided use a form of CNT (except Kärcher and Lohmann [2003], which is qualitatively similar to CNT), and, are theoretical studies. Is there a more logical way to justify this statement rather than comparison against other theoretical approaches?

Page 2681

Lines 5-8. This is a confusing statement. What do the authors mean by underestimation of heterogeneous nucleation? What is a crystallization effect and why is it expected to be noticeable?

ACPD

10, C1123–C1131, 2010

Interactive Comment

Lines 9-16. Why is this behavior more “realistic” than obtained with PDA08? What is the logical basis for these statements? How will this change if different conditions are assumed for the simulations?

Page 2682

Lines 11-19. The authors compare single runs of an idealized parcel model against data taken from thousands of aircraft measurements. There are more processes involved in cloud formation than what can be represented in a parcel model with idealized dynamics. After all, nucleation is only one of many factors defining the phase state of a cloud. Agreement with observations can be fortuitous. To make this compariosn the authors should at least run their model for a significant number of climatologically relevant conditions.

SECTION 4

Page 2684

Equation 13. The conditions for the formulation of this parameterization are not clear. Is Nc sensitive to the characteristics of the aerosol? What is the sensitivity of Nc to the values of m and α used? What is the error of this expression with respect to the parcel model simulations? How the results of this expression compare against other published parameterizations [e.g., Kärcher and Lohmann, 2003; Liu and Penner, 2005; Barahona and Nenes, 2009]?

Line 11. Were the initial conditions of the parcel model always the same in generating this parameterization? If so, it is unlikely that many parcels would be lifted at constant updraft for several hours from warm temperatures (T -10 C) to very cold ones (T - 60 C). In such a case, Eq. 13 represents an exception rather than a general representation of cloud formation.

Lines 20. As the dynamics of mixed-phase and pure ice clouds are considerably different, it is unlikely that a simple parameterization based on idealized parcel model

ACPD
10, C1123–C1131, 2010

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Interactive Discussion
Discussion Paper

simulations can reproduce all mixed-phase and ice cloud regimes. Equation 13 needs more assessment on this respect.

Line 22. It is not enough for a parameterization to reproduce observations at some conditions. Those conditions have to be those at which measurements were taken. In particular, for the exercise of Figure 5, the parameterization should be integrated over the spectra of updrafts and temperatures of cloud formation (assuming that the residence time of the particles inside the instruments are close to those in the cloud) relevant for the atmosphere.

Page 2685.

Lines 1-3. Was the PDA-KC line generated by the authors or copied from the PDA08 paper?

Lines 5-10. How can the PDA-KC curve be constructed without any parcel model simulations? Equations 8 and 9 clearly show a temporal dependency of the crystal concentration that cannot be eliminated by fixing Sw = 1. How is this dependency resolved in PDA08? Inspection of the PDA08 paper suggest the assumption w = 1 m/s and constant Sw. This assumption reflects a thermodynamic interpretation of the CFDC data (which remains to be proven). Why the authors do not discuss this issue?

Lines 15-18. By extension one can conclude that whenever Sw = 1, KC would produce unrealistic values of Nc. Please rephrase.

Page 2686.

Lines 1-5. I think residence time (i..e, the time the particles remain at some Sw and T conditions) rather than Sw dependency is the real issue here. In a parcel model they are unseparable due to the negative feedback. However, one can always make a thought experiment (or even a real one) in which T and Sw , and all other variables are maintained constant, and ice crystals are removed soon after they are produced; then ask: What would be the number of crystals produced?

ACPD
10, C1123–C1131, 2010

SECTION 5

This section may overload this work; it is also structured as a separate work. While I see its relevance it does not seem to complement any of the other sections in this manuscript. I'd encourage the authors to submit it as a separate work. Thus, a deeper and thorough discussion and comparison with other models, along with further model validation, can be included.

REFERENCES

Barahona, D., and A. Nenes (2009), Parameterizing the competition between homogeneous and heterogeneous freezing in cirrus cloud formation. Monodisperse ice nuclei, Atmos. Chem. Phys., 9, 369-381.

Eidhammer, T., P. J. DeMott, and S. M. Kreidenweis (2009), A comparison of heterogeneous ice nucleation parameterizations using a parcel model framework, J. Geophys. Res., 114, doi:10.1029/2008JD011095.

Kärcher, B., and U. Lohmann (2003), A parameterization of cirrus cloud formation: Heterogeneous freezing, J. Geophys. Res., 108, 4402, doi:4410.1029/2002JD003220.

Kashchiev, D. (2000), Nucleation: Basic theory with applications, Butterworth-Heinemann, Oxford.

Liu, X., and J. E. Penner (2005), Ice nucleation parameterization for global models, Meteorol. Z., 14, 499-514.

Phillips, V. T. J., P. J. DeMott, and C. Andronache (2008), An empirical parameterization of heterogeneous ice nucleation for multiple chemical species of aerosol, J. Atmos. Sci., 65, 2757-2783, doi:2710.1175/2007JAS2546.2751.

Vali, G. (2008), Repeatability and randomness in heterogeneous freezing nucleation, Atmos. Chem. Phys., 8, 5017-5031.

ACPD

10, C1123–C1131, 2010

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ACPD

10, C1123–C1131, 2010