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

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Reply by J. A. Curry and V. I. Khvorostyanov to Interactive Comment by VTJP Phillips on "Assessment of parameterizations of heterogeneous ice nucleation in cloud and climate models" by J. A. Curry and V. I. Khvorostyanov

In this comment, Phillips raises many of the same points raised by DeMott. The two main points are the model used in PDA08 to evaluate the ice nucleation parameterizations, and the ice nucleation parameterization developed by PDA08. We thought these points were adequately made in our paper (CK10), but Phillips continues to fail to understand our points. We will once again attempt to clarify the points we are making

surrounding these two issues.

As we already explained in CK10, the KC scheme differs from the previous classical nucleation theory of freezing by introducing dependence on the water saturation ratio Sw, which becomes less than 1 due to Bergeron process, leads to a strong increase of the critical energy of nucleation, and creates a very strong negative feedback that limits crystal concentration. Since Phillips used in his evaluation of KC scheme Sw=1 in his "model", this scheme is reduced to the classical theory based on J. J. Thomson (1888) expression for critical radius, contains only temperature dependence, and this scheme has nothing in common with KC scheme. When Phillips fixed Sw=1, as it is done in PDA08, Fig. 10, it is not the KC scheme any more. We supposed that this was clearly explained in CK10. However, Phillips continues to call his incorrect interpretation of this scheme in PDA08 as "KC". This erroneous application of the KC scheme and Phillips' continued defense of his erroneous application defies logic and science and is counterproductive. However, it is not only the KC scheme that is falsely represented in Figure 10 of PDA08, but also the schemes by Lohmann and Diehl (2006, LD06; based on Diehl and Wurzler, 2004, DW04), and Liu and Penner (2005) as well. Further, PDA08 extrapolated Meyers et al. (1992, MDC92) far beyond the region of its validity. All of these ice nucleation schemes assume presence of the natural feedbacks, Bergeron-Findeisen mechanism first of all, that reduces Sw well below 1 and substantially reduces concentrations of nucleated crystals. When Sw is kept equal to 1 down to –70 C, as in Fig. 10 in PDA08, this is in violation of the first and second laws of thermodynamics. PDA08 provided an erroneous representation of the T-dependence in all of these schemes and incorrect estimates of ice nucleation in general because of using his unphysical model "with exact water saturation artificially imposed at all temperatures". That is, Phillips artificially prescribed water saturation Sw = 1 or RHW=100 % in all processes with all high crystal concentrations and down to –70 °C (Fig. 10 in PDA08 and the text around). Thus, Phillips allowed crystal nucleation but prohibited crystal growth even at ice supersaturation of 30-70 % and more, neglected the supersaturation equation and prohibited droplet evaporation, ignored the Bergeron-

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Findeisen process and conserved mixed cloud with RHW= 100 % at low temperatures down to –70 C, well below the threshold of homogeneous nucleation, ∼-38 C, where full glaciation takes a few tens seconds and humidity relaxes to saturated over ice, which is much lower than 100 % RHW. Thereby, PDA08 excluded from their "adiabatic model" fundamental processes of cloud physics, in particular the powerful negative feedback by humidity that limits crystal concentrations and decrease them by several orders of magnitude. Ice nucleation schemes incorporated into this physically unrealistic model produce unrealistic ice concentrations; this incorrect methodology cannot invalidate in any way the KC and other nucleation schemes.

We summarize here the problems with the empirical parameterizations developed by Phillips et al. (2008), which are outlined in our CK10 paper and in our reply to DeMott: 1) The empirical parameterizations are based on real measurements, but are extended into unphysical areas on the T-Sw plane, where humidity is below critical and critical radius is negative. This is applied to MDC92 and to PDA08. 2) Parameterization by Phillips et al. (2008, PDA08) substantially underestimates crystal concentrations and yields almost liquid cloud down to –35 °C and lower in sharp conflict with cloud climatology established over several decades. 3) The deficiencies of PDA08 can be caused by substantial underestimates in the CFD chambers: the processing time in CFDC is 7-15 sec (PDA08), while real crystal nucleation takes much greater time from a few minutes to a few hours, as indicated by numerous parcel models, including parcel simulations in Eidhammer et al. (2009, EDK09). Thus, the derivation of parameterization in PDA08 is in conflict with its application in EDK09. 4) The independently developed and different parameterizations, empirical by Diehl and Wurzler (2004, DW04) and related similar parameterizations by Lohmann and Diehl (2006, LD06) and theoretical parameterizations by KC00-09 yield similar comparable crystal concentrations in parcel model simulations by EDK09, while PDA08 yields much smaller crystal concentrations which may indicate its insufficient efficiency.

Specific comments are provided below in the point-by-point reply. For clarity, we cite

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below comments of the reviewer and our reply follows.

From Phillips's General Comment, p. C387: "In nature, if freezing of insoluble aerosol particles were almost a "Heaviside" function, with all of them freezing somewhere near -10 or -20 deg C in a water-saturated cloud, as predicted by the current KC scheme, then this would have been discovered by the community many decades ago."

Reply #2. Here again Phillips demonstrates lack of understanding of elementary physical processes. If aerosol particles are insoluble, they cannot freeze. KC scheme described freezing of mixed, partially soluble particles in condensation-freezing mode, and ice formation insoluble particles occurs via deposition mode. Thus, if Phillips considered ice nucleation on insoluble dust and soot, his applications of KC theory don't make sense.

This issue of the "Heaviside" function is interesting and deserves some discussion.

a) We explained already in CK10 and here above that PDA08 provide an incorrect and misleading representation of the freezing in KC theory. It is not a "Heaviside" function by any means. This "Heaviside" function near –10 or –20 C as Vaughan suggests, occurs only in the incorrect "adiabatic model" used by Phillips. Even in a water saturated cloud, the KC scheme predicts not the "Heaviside" step function, but the temperature dependence smoothed over some T-range. Figs. 2 and 3 in CK10 show that freezing with alpha(T) with water saturation occurs over a range of about 15 degrees, not as the Heaviside function. Fig. 3 shows that cloud glaciation predicted by KC scheme proceeds over a temperature range of 15 C with both choices of alpha. The slopes of the curves are comparable to those derived from a large number of observations and currently used in GCM parameterizations (NCAR CAM3 and ECMWF. As Phillips writes "then this would have been discovered by the community many decades ago." Yes, these KC curves are already close to what "has been discovered by the community". We realize that the KC slopes are comparable but still steeper than the observed slope. But the results shown in Figs. 2, 3 of CK10 were obtained with a single aerosol fraction

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and fixed values of contact angle and all the other parameters. Of course, there should be averaging over all of these parameters in an aerosol population. It is not a big problem to introduce several averaging procedures in the KC scheme and integration over several IN types, as explained in detail in our reply to DeMott's comments.

b) There is also another issue regarding the analysis of crystal concentrations Nc(T) both in PDA08 and EDK09. The data for a single chamber or field experiment are not given either in PDA08 or in EDK09, nor in their previous papers. All of the data on the temperature dependencies of Nc(T), like Fig. 1 in PDA08 or Fig. 6 in EDK09, are given for many or many tens or hundreds experiments, and represent a statistical ensemble of measurements. Thus, the temperature of freezing is always averaged over the large ensemble of initial conditions, different aerosol populations and their different properties, etc. This yields additional smoothing relative to individual freezing event and masks the real individual dependence. However, in Fig. 10 in PDA08 and in Fig. 6 in EDK09, the authors compare the results of a single model run with such ensemble-averaged data on Nc(T). This methodology cannot serve for verification of the KC or DW04 or other theory and PDA08 parameterizations. However, a comparison in Figs. 5, 6 in CK10 is compared correctly in this respect: ensemble of runs with an ensemble of experiments.

c) It looks like Phillips does not like steep "Heaviside" function for Nc(T) dependence. But what should this dependence be in a single experiment, that is, for a single nucleation event? In his comment, Phillips did not discuss this because he cannot say over what temperature range ice nucleation actually occurs, half of degree or 20 degrees, and how this temperature range depends on the initial conditions. Phillips blames KC theory for an apparent Heaviside function, but does not present any arguments what this function should be. Perhaps expansion chambers can give the temperature dependence for a single nucleation event, but CFDC, to our knowledge, cannot. CFDC can give T-dependence for many experiments, but not for a single event, thus cannot say what is the width of this "Heaviside" function. In any case, the data on these details

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are absent in PDA08 and EDK09, and the width of the temperature range in a single nucleation is unknown, and apparently this information cannot be provided from CFDC data.

Phillips' detailed comments.

Phillips. Yes, there was advice in this email on 16 April that the KC scheme ought to be run in a cloud model that predicts "the water supersaturation". I could not follow this Khvorostyanov's advice as I was comparing various schemes partly with laboratory data obtained by fixing the humidity at water saturation, for my 2008 paper.

Reply #3. See our Reply # 1. Once humidity is fixed at water saturation, it is not KC model, by no means. So, Phillips could talk about 'classical model", but not about KC, calling it KC scheme at fixed water saturation is incorrect.

Phillips. Another reason I did not follow Vitaly's advice is that I believe any scheme for heterogeneous ice nucleation ought to predict the ice concentration and freezing fraction for any situation, including one where the humidity is fixed (e.g. to water saturation). This is the situation in laboratory experiments to observe heterogeneous ice nucleation, such as at AIDA where, for some data, water saturation is artificially imposed. It is also the situation in natural mixed-phase clouds. The humidity is maintained approximately at water saturation by the presence of supercooled liquid in such clouds.

Reply #4. Water saturation can be maintained in the chambers and natural clouds for limited conditions, when supersaturation generation exceeds or equal to supersaturation production. This is already difficult at T below –20 to –30 C, as cloud climatology shows. But Phillips continued this situation down to –70 C, which produces results that are virtually impossible in nature.

Phillips. The claim on page 2685 of the present paper that the "KC scheme ... was constructed in PDA08 for the first time" is false. I applied the code from Vitaly

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Khvorostyanov without making any changes to it, in an adiabatic parcel for my 2008 paper (Phillips et al. 2008).

Reply #5. The claim in CK10 is not false. The code from Vitaly was not changed, but Vitaly's advice on the requirement to calculate supersaturation (cited above) was not followed. Once again, with fixed water saturation, this calculation is just classical theory (with temperature dependence only), not KC (with both temperature and supersaturation dependence). Of course, the curve called KC in Fig. 10 in PDA08, as explained above, was constructed for the first time, because was based on the absence of an appropriate parcel model, and all other schemes in this Fig. 10 are falsified as well. Phillips' persistent attempts to falsify this and to call it KC would deserve a better application. If this persistence was applied to make or borrow a good parcel model with good microphysics for nucleation studies, this would bring good results.

Phillips. This parcel necessarily had no microphysics, except for heterogeneous ice nucleation, in view of my direct comparison ..."

Reply #6. This is the key reason of why Phillips' representation of the cloud processes is incorrect. All who develop parameterization (Kärcher and Lohmann, 2002, 2003; Gierens, 2003; Khvorostyanov and Curry, 2004, 2005; Liu and Penner, 2005; Barahona and Nenes, 2008, 2009 and others) use parcel models to verify their new parameterizations in an interactive thermodynamic regime. Phillips developed a parameterization based on incomplete lab data, as explained in CK10 and in reply to DeMott, but could not verify it. If Phillips applied a parcel model with a correct microphysics of mixed cloud phase, he would discover how unrealistic his current "adiabatic model" is and the results shown in Fig. 10 in PDA08.

Phillips. Eidhammer et al. implemented the KC scheme independently of my effort, and in contrast with my 2008 paper, they did include the response of supersaturation...

Reply #7. Yes, and this immediately showed poor performance of PDA08 scheme that gave liquid cloud at –35 C in conflict with climatology. KC and DW schemes performed

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much better.

Phillips, p. C389. The claim on page 2685 of the present paper, that my 2008 paper implements the KC scheme "without the Sw-dependence" is misleading. In fact, both Phillips et al. (2008) and Eidhammer et al. (2009) implemented the KC scheme by including the Sw input to it.

Reply #8. Once again, Sw was strictly fixed to 1 in PDA08, as he just admitted, but he continues to claim that our statement is misleading that "Phillips implements the KC scheme without the Sw-dependence". This fundamental misunderstanding by Phillips has led to an incorrect representation of all of the other schemes tested in PDA08 in Figure 10.

Phillips' other comments and remarks are answered in our reply to referee DeMott.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 2669, 2010.