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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

J. A. Curry and V. I. Khvorostyanov

vitalykh@tdn.ru

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Reply by J. A. Curry and V. I. Khvorostyanov to Interactive comment by P. DeMott (Referee) on "Assessment of parameterizations of heterogeneous ice nucleation in cloud and climate models" by J. A. Curry and V. I. Khvorostyanov

Replies #1-3 to the general comment.

In his very lengthy review (19 pages in ACPD) of our paper in ACPD (hereafter referred to as CK10), the reviewer puts forth the perspective that a limited number of existing laboratory and field observations are sufficient to falsify the heterogeneous ice nucleation theory put forward by KC. We argue that existing observations are not even close

to being adequate to either verify or falsify this theory, and we further argued in CK10 that the CFDC measurements of ice nuclei are inadequately characterized in terms of their uncertainty and what is actually being measured. In the absence of adequate experimental data of IN measured concurrently with other aerosol properties and environmental conditions required to test this theory, we further utilize climatological observations of cloud phase as a function of temperature and also field measurements of cloud microphysical characteristics as further observational tests of the ice nucleation parameterizations. The referee's recommendation that Sections 3 and 5 be removed from the paper (comparison with climatological observations and field measurements of real clouds) seem contradictory to the reviewer's emphasis throughout the review on comparison with observations. It seems that the only apparent "problem" and "deficiency" of our simulation of MPACE is that it was successful, and without any special additional tuning and invoking sophisticated ice nucleation mechanisms. The reviewer provides no concrete scientific arguments as to why these simulations "are irrelevant as validations" and should be removed.

The apparent preferred parameterization of the reviewer is PDA08 in terms of fidelity to the laboratory observations. CK10 provides substantial critiques of this empirical parameterization and its application in parcel model simulations by EDK; surprisingly, the reviewer (a coauthor on both of these papers) does not provide any substantive response to CK10's criticisms of this parameterization.

We find in this lengthy review only a few remarks that are useful, including suggestions for rewording several statements and including aerosol variability that can be easily incorporated into KC scheme.

Our response to specific remarks made by the reviewer is provided below.

Specific Comments.

DeMott, p. C264, Abstract,

“DeMott, a. Line 9-13. Using theory to provide restrictions on empirical schemes is an interesting concept, but a strange one if the schemes are actually based on data collected in the “forbidden” regime. ... but the abstract is not clear on the fact that the thermodynamic restrictions are for a specific ice formation mechanism as quantified by theory not yet validated using specific ice nucleation data.”

Reply # 4. These limitations on parameterizations come only from the equation for the critical radius of freezing, which yields also critical humidity for this process. The only theory used for derivation of theses quantities in KC00-09 was general thermodynamics, namely, the entropy equation, still before using nucleation kinetics or aerosol properties. It would be astonishing if the first law of thermodynamics and the entropy equation were to be invalidated by observations. We of course encourage other scientists to attempt to refute these specific equations using laboratory measurements.

“DeMott, b. Lines 14-16: Clouds sometimes remaining mostly liquid to as low as –35 °C are facts based on documented cloud observations (see references later). The parameterizations reflect actual ice nucleation data.”

Reply # 5. Great remark. The climatology based on many tens thousands measurements yields a low frequency of liquid state at –35, less than 1 %, see PK97, the references in CK10. However, PDA08 predicts such a liquid state with probability 100 %, which is in conflict with all climatology, and illustrates substantial failure of PDA08 parameterization. KC, DW04 and LD06 perform much better than PDA08 in this respect as shown in CK10.

DeMott, c. Lines 17-18: Not applying the KC parameterization to the entire aerosol distribution is an approach that corrects for invalid assumptions in the simplified model, not a misapplication. A parameterization of ice initiation should only act on the relevant source nuclei population, by way of explicitly (e.g., some particles have no active sites) or implicitly limiting this population.

Reply # 6. Referee probably missed line numbers, but does not matter, referee makes

here a good point, we absolutely agree with this statement. Of course, not all aerosol has IN properties. But the KC scheme does not require integration over all aerosol; integration should be performed not over all aerosol particles but only over the appropriate fraction, or over several such fractions. This fraction should be hypothesized or measured.

DeMott, d. Lines 20-23: “This statement should be clear on which previous study is being referred to. It misrepresents what was done by EDK09, who restricted ice nucleation to the size range and compositions from which natural ice nuclei are known to come.”

Reply # 7. We meant here misapplication in Fan et al. (2009), not EDK09. What EDK09 did in this respect, by limiting relevant aerosol fraction, is a good development of KC scheme, we agree and did similar thing in this CK10 and extended further by introducing variable alpha(T), but it should be continued (not in this paper). Abstract will be corrected in this place.

1. Introduction.

DeMott, a. Page 2671, lines 11-15: KC have created a framework for describing ice formation (potentially in all of its dependencies) in clouds from a theoretical basis. The improvements mentioned here are needed before any further application is made to simulate cloud systems (see EDK09).

Reply # 7. The improvements may be needed in some situations or not in others, but simulations even with the current version of KC yield quite reasonable results in many cases. Note that improvements like size spectra of IN as a small fraction selected from the general aerosol population must be provided by measurements. Thus, desired improvements of the KC scheme is not a problem of this scheme, its easy, but is a problem of desperate lack of complete experimental analysis, which is the task of experimentalists (e.g. the referee), not ours.

DeMott, b. Page 2673, lines 3-8: It was not appropriate for this parameterization to appear published in an open access journal prior to my ability to first publish and describe it. ... I recommend removing DM10, and if the figure is retained, comparing instead to PDA08, which does seek to resolve all ice nucleation in the phase space shown.

Reply # 8. Yes, we will take care of this as we have explained to referee in private mailing. As to PDA08, it is based on MDC92 and should suffer the same problem in T-Sw plane. We could try PDA08 also if referee can help us with its FORTRAN codes as we helped the referee with KC codes.

DeMott, c. Page 2675, equation 8: The problem with this equation is that not all CCN and not all aerosols are potential ice nuclei. Use of such an equation necessitates differentiation of (and summation over) the variety of specific physical and chemical properties of particles at each size and over the distribution. It should not be applied absent such knowledge or without qualification.

Reply # 9. Yes, we agree, and this has already been done in CK10: we used only a small fraction of aerosol in several sections, not all. As to the knowledge, this should be provided by the experimentalists. Until such info is available, we can try various versions, as it was done in many MPACE models, and as it is done for 5 decades with drop activation: when parameters are unknown, they are hypothesized and the sensitivity of calculations to the choice of the parameters is assessed.

DeMott, d. Page 2675, lines 16-19: "...In practice, observations (e.g., Marcolli et al. 2007) indicate that even similar sized and chemically-similar particles possess a spectrum of ice nucleation abilities."

Reply # 10. Yes, there can be spectra over many parameters. KC theory allows derivation of such spectra. It can be similar to the various spectra in drop activation derived by KC in several recent papers (JGR-2006, 2007; JAS-2008, 2009). These further developments are under way for ice, but this refinement does not preclude KC in its present form from being useful.

2. Thermodynamic constraints on heterogeneous ice nucleation schemes

DeMott, a. As noted above, this section could be fine as a hypothesis for the existence of thermodynamically-restricted regimes, but this should serve as the basis/motivation for experimental evaluation and refinement of theory.

Reply # 11. See our reply # 4. Again, only general thermodynamics was used in derivation of this criterion. We totally agree that this theory should serve as the basis/motivation for experimental evaluation and we await new measurements from DeMott.

3. Evaluation of phase state simulations.

DeMott, The following comments lead me to suggest that this entire section is invalid and needs to be removed.

Reply. We do not see how the authors comments below (individually or collectively) in any way invalidates the entire section. Besides providing an additional data set against which to compare the KC parameterization, evaluating an ice nucleation parameterization for application in climate models against such a climatological data set would seem to be absolutely essential.

DeMott, a. Page 2679, lines 18-26: It is not clear why it is important to note the similarity between the DW04 and KC schemes, a point made by EDK09 to show that both have some undesirable features due to idealization of the ice nucleation behavior of a population of particles.

Reply # 12. The similarity between empirically derived DW04 scheme and theoretically-derived KC schemes is noted because it shows that two schemes, developed from completely independent methods, yield similar results, which are substantially different from PDA08 scheme.

DeMott, b. Page 2680, discussion of parcel model simulations: This section repeats what we already know from EDK09, although the authors misinterpret the meaning.

The simulation performed by EDK09 using the PDA08 ice nucleation scheme was an idealized parcel simulation simply intended to cover a wide temperature range. There was no intent to mimic a specific cloud case, just specification of a steady updraft for an unrealistically long period. The simplest comparison...

Reply # 13. The only difference of our simulations relative to EDK09 is a) using the exact original data of EDK09, but represented in a different form (mixed phase fractions), CK10 showed really unrealistic performance of PDA08 parameterization that yields liquid cloud at –35 C and lower with almost 100 % probability; b) they do show better performance of KC scheme (and indirectly DW04) for the same case.

As to "idealized parcel simulation simply intended to cover a wide temperature range", this approach in EDK09 and in PDA08 was wrong in principle. As we explained in CK10, the experimental data given in EDK09 were obtained in "a wide temperature range", but in many experiments under variable initial conditions. The same with all the other data, Fletcher (1962), Cooper (1986), etc. The same with KC05 parcel simulations, there were done for many various initial data. Every time, nucleation occurs in rather narrow T-range, but due to averaging over various cases, we obtain wide and smooth T-curves. When EDK09 tried to reproduce it in one run, this is merely misinterpretation of the real data. Just one even cannot represent several tens and hundreds of events.

DeMott, c. Page 2681, more discussion of the parcel model simulations... " the "constraints" imposed in the PDA08 scheme lead to a substantial underestimation of heterogeneous ice nucleation." What is the basis for this conclusion?

Reply # 14. See Reply # 13. EDK failed to reproduce cloud phase state, which is something that EDK failed to notice. Our examination of phase state makes this immediately clear. The "constraints" are from the CFDC observations, which is the basis for PDA08: the residence time in CFDC is 7-15 s (PDA08), which should cover nucleation time, but this nucleation time in EDK simulations is several hours, in sharp

conflict with PDA08 and indicates that the time in CFDC is insufficient as well as crystal concentrations. Another reason was indicated in MPACE: missing particles greater 1.5 mcm, while measured coarse mode was 1.3 mcm. As to 1000 L-1, this was choice of EDK09, not ours, They could choose one-two orders of magnitude smaller number in the 2nd mode, this would be closer to CFDC measurements and could yield more realistic crystal concentrations. So, referee should address this question to EDK, not to us.

DeMott, d. If the authors believe that present measurements greatly underestimate ice nuclei number concentrations active by the mechanisms they purport to describe theoretically, they need to provide hard evidence as to why measurements are in error. Further, they might like to explain: ....

Reply # 15. First of all, it is not the task of the theory and theoreticians – to prove that CFDC measure all IN. This should be proven by experimentalists who operate CFDC. To our knowledge, this has not been done. We indicated in CK10 the two reasons, see our previous reply #14; here are some additional points. Numerous experiments have found measured crystal concentrations much lower than IN concentrations measured by chambers or other devices when known ice multiplication mechanisms could not work (e.g., Hobbs and Rangno, 1990; Rangno and Hobbs, 1990, etc). If CFDC or other chambers measured all IN, this problem would be solved to high extent, and many problems with "ice multiplication" would not occur. Substantial undercounting of IN by CFDC was remarked by most modelers in MPACE. The absence of measurable IN during 90 % of the time or insensitivity of CFDC to large IN stimulated several hypotheses of ice nucleation, some like evaporation-freezing (we don't deny them) may look exotic as compared to the much simpler KC mechanism. Simulations in EDK09, and many others cited in CK10 have shown that nucleation time is much greater than allowed in CFDC, 7-15 s; in EDK09, it can reach several hours. This list could be continued, but it is not our job, we would expect from DeMott et al. to prove that all IN are measured by the CFDC. A curve with dependence of Nc on the residence time or chamber length L with saturation and plateau at some L, could constitute such a proof. Until this is done, we regard all CFDC data to be inadequate to falsify our theories of heterogeneous ice nucleation.

The following discussion suggested by referee can be very long and certainly should not be the subject of our reply. We can only briefly try to outline some possible reasons relative to the items in referee's list.

DeMott, d. 1) Low ice crystal number concentrations measured in laminar flow orographic wave cloud scenarios.

Reply # 16. E.g., short residence time of an air parcel in an orographic wave, and low values of supersaturation.

DeMott, d. 2) The general agreement of average ice nuclei number concentrations as measured (EDK09) with ice crystal concentrations found in clouds when ice initiation is presumably isolated (Cooper, 1986), a reference the authors oddly twist to corroborate their model in Section 4.

Reply # 17. This is the typical example of referee's logic: when EDK09 is close to Cooper, this is correct agreement, but if KC is close to Cooper as shown in CK10, this is "oddly" incorrect agreement. Simulations with the KC scheme shown in CK10 are also for the cases when ice nucleation is isolated, just ceased, not for the whole cloud life cycle; that is, can be compared with Cooper (1986).

DeMott, d. 3) The presence of liquid water to low temperatures in local regions of cumulus clouds (e.g., Rosenfeld and Woodley, 2000; Fridlind et al. 2004).

Reply # 18. Yes, there are observations of liquid water occurring at temperatures of –35 C locally in cumulus clouds and also in arctic stratus clouds, but climatologically of the existence of liquid water at such low temperatures is insignificant. The PDA08 parameterization with EDK09 tests predicts liquid almost always at –35 C, contrary to the extensive climatology of cloud phase observations.

DeMott, "d. 4) Ice nucleation results from laboratory studies, such as shown in Figures 1 and 2"...., and further almost 4 pages of the text of review plus 2 figures with description of experiments in AIDA chamber with Sahara dust.

Reply # 19. The AIDA results provide interesting material for our further meditations, but not for this paper. Recall that the KC scheme considers deliquescence-freezing, i.e., the presence of soluble fraction is required as in many mixed aerosols. The dust particles considered by the referee may have zero or very small soluble fraction and nucleation on them cannot be calculated using the KC scheme. In this AIDA case, nucleation could proceed via deposition mode. This is confirmed by referee's remark: "Note that the peak humidity at the cloud formation point is not resolved and may easily have exceeded a few % for the CCN concentrations and expansion rates used." Such high water supersaturation is a typical picture for the insoluble or almost insoluble particles. Another confirmation from referee's description: "Similar to the second AIDA expansion result, water saturation is required in the CFDC prior to the onset of significant ice nucleation. " This is a typical picture of ice nucleation on insoluble particles (Young, 1993, see there 2 figures from DeMott's dissertation (1990) on this; Pruppacher and Klett, 1997). Thus, the ice nucleation could proceed via deposition mode and should be different from any predictions of KC theory for freezing. Then the referee's comparison of this experiment with KC04 is irrelevant. We could try to simulate this dust experiment, but not in this paper.

DeMott, p. C271. "e. Page 2681, paragraph starting line 18 and continuing through the next page: The comparison of idealized parcel model ice mass fraction versus climatological values measured in clouds or prescribed in global models has potentially nothing at all to do with nucleation on measurable heterogeneous ice nuclei."

Reply # 20. The reviewer fails to understand that an ice nucleation parameterization developed for climate models must provide a climatologically realistic distribution of ice clouds, which is the point of this section.

DeMott, p. C272. “f. Page 2682, lines 21-23: I believe that the prediction of homogeneous freezing influence beginning at -34.5 C in EDK09 is fully expected for the onset of action of homogeneous freezing in up to 40 micron drops”.

Reply # 21. Yes, we agree, this is a reasonable explanation. Note that if it was a more realistic parcel model or an Eulerian model with smaller drops, not 40 mcm, freezing in EDK09 with PDA08 occurred only at about –40 C, which also indicates inefficiency of PDA08 scheme.

DeMott, p. 272. “g. Page 2682, line 25, on through to the end of section 3: This is another instance of turning an EDK09 argument around inappropriately. The high values of the DW parameterization are no more realistic than the high predicted values of KC04.”

Reply # 22. The high values in both KC and DW were caused in EDK to high extent by the choice of high concentrations 500-1000 L-1 in the coarse fraction. In any case, KC and DW both predict a more realistic phase state than EDK.

4. Assessment of parameterized ice particle concentrations

This is a too lengthy referee's remark to cite it all. Briefly, here the reviewer attacks the feedbacks and vertical velocities in our suggested parameterization, Eq. (13) in CK10. An example: “Equation 13 encapsulates the effects of nucleation and negative feedbacks and so is not recommended for application in a global model until the nucleation scheme is modified properly.”

Reply # 23. This remark shows the referee's fundamental lack of understanding of the current parameterizations of ice nucleation in GCMs. The KC parameterization, in particular, eq. (13) in CK10 stands in one line with the other theoretical parameterizations of ice nucleation developed over the last decade, e.g., Kärcher and Lohmann (2002, 2003), Gierens (2003), Liu and Penner (2005), Liu et al. (2007), Chen et al. (2008), Barahona and Nenes (2008, 2009) and others. These parameterizations all include

negative feedbacks accounted for by the supersaturation equation, which limits crystal concentrations. They all include in some form dynamical factors, in particular, vertical velocity. The various ways in which these subgrid velocities are specified in GCMs are discussed in applications (Ghan et al., 1997; Morrison and Gettelman, 2008). Thus, all of the referee's criticism against the KC scheme applies also to all the other cited theoretical parameterizations that include a dependence on supersaturation and/or vertical velocity. So DeMott's remarks and review essentially reject all modern theoretical parameterizations of heterogeneous ice nucleation. The vertical velocity describes the dependence of crystal concentration Nc on supersaturation arising from adiabatic cooling of the cloud, Nc does depend on w, as many parcel simulations show, including EDK09. Contrary to all these theoretical parameterizations that include dependence on dynamical factors or vertical velocity, PDA08 does not include any dynamical factors; that is, Nc with PDA08 should be the same in vigorous convection, in small synoptic uplift and in slow advective cooling, contrary to observations.

5. Simulations of Mixed-Phase Arctic Cloud Experiment (MPACE)

DeMott. I consider the exercise done here to be premature and, intending no disrespect, to be yet another instance of publishing cloud model simulations that give the false notion that the authors have solved the topic of ice formation in the atmosphere.

Reply # 24. The authors (CK) did not write that they “have solved the topic of ice formation in the atmosphere”, but simply showed that application of KC scheme in a simplest way in this particular case with measured aerosol properties allows to obtain measured crystal concentrations. DeMott seems to suggest that no cloud models or climate models with ice clouds be conducted at this time, given disagreements about the ice nucleation parameterization. In the absence of an adequate set of observations, theoreticians and modelers have made substantial progress on this topic.

DeMott. “, without regard to the numbers of ice nuclei available via the mechanism prescribed (it is absolutely not the total CCN population),” Reply # 25. Yes, we agree, the text in all these places should be corrected and clarified as we already explained in replies to Fan's comments. Of course, not all aerosol serves as IN and it is only a small fraction. But even concentrations 0.01-0.02 cm-3 (10-20 L-1) in the coarse mode (missed by CFDC in MPACE due to size or time limitations) are sufficient to produce Nc $\sim$ 10-30 L-1 observed in MPACE. We remind DeMott here that EDK09 bravely and without explanations used IN concentrations in the coarse mode of 1 cm-3 (1000 L-1 !!!), "without regard to the numbers of ice nuclei available via the mechanism prescribed", and which is about 2 orders of magnitude greater than typical CFDC measurements. Why is EDK allowed to make such an assumption, but not CK?

DeMott, p. C274. a. ..."The first point, one also made by Santachiara et al. (2009), is a valid concern if all of the 10 L-1 are active as ice nuclei. This seems unlikely in this case based upon the best measurements presently published regarding the similarly limited fractions of large aerosols active as ice nuclei (Berezinsky and Stepanov, 1986)."

Reply # 26. Yes, this would be unlikely if all coarse mode was 10 L-1. However, the coarse mode in MPACE was 1.8 cm-3 = 1800 L-1 (Morrison et al., 2008). If the fraction that can serve as IN is 10-20 L-1, this is $\sim$ (10-20)/1800 $\sim$ 0.005 - 0.01, a very small fraction of all coarse mode. This fraction of all aerosol that can serve as ice nuclei needs to be measured by experimentalists. If it was not measured, this is not the fault of the theoreticians.

DeMott, p. C274. b. ..." I will state this once again: based on the preponderance of evidence, the potential number that may become ice crystals at any temperature is not the total aerosol number."

Reply # 27. We absolutely agree with this, as explained just above, and all confusing formulations will be corrected in the paper. We also state once again, if only a tiny fraction of all aerosol, with concentrations as estimated before, 0.005-0.1, serves as ice nuclei, this is sufficient for good glaciation. And once again, experimentalists should produce measurements that give us this fraction. Until then, we can only vary in sim-

ulations this fraction and aerosol properties, and solve the "inverse" problem: what properties aerosol should have to give reasonable crystals.

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