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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

VTJP Phillips

vaughanp@hawaii.edu

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I agree with the reviewer, Paul DeMott, that the present paper cannot be published. I agree with all of Paul DeMott's comments. Here are some of my own.

General Comments:

It is commendable that the authors of the present paper for several years have been developing classical physics theory to attempt to explain ice nucleation. It seems

promising that in future, progress in observing the probability distribution of contact angles and nucleating efficiencies among insoluble aerosol particles, or their cumulative effects, may eventually allow their theory to make accurate predictions. However, there seems little point in construing the current version of the KC scheme as realistic, as the present paper attempts to. In nature, if freezing of insoluble aerosol particles were almost a "Heaviside" function, with all of them freezing somewhere near -10 or -20 degC in a water-saturated cloud, as predicted by the current KC scheme, then this would have been discovered by the community many decades ago. Although the present paper criticizes the CFDC probe, this is only one of many experimental tools (e.g. the AIDA chamber) for observing heterogeneous ice nucleation, and all of them show only a gradual increase of freezing fraction during prolonged supercooling.

Detailed Comments:

On 16th April 2007, Vitaly Khvorostyanov emailed me the complete codes for his heterogeneous ice nucleation scheme, which included an input for the relative humidity (S_w). I did not change these codes in any way when producing the intercomparison of the KC scheme with other schemes in my 2008 paper (named 'PDA08' in the present paper by Vitaly Khvorostyanov and Judith Curry). 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 maximum water supersaturation". I could not follow this advice as I was comparing various schemes partly with laboratory data obtained by fixing the humidity at water saturation, for my 2008 paper.

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.

It is false to suggest that the response of supersaturation to the appearance of heterogeneous ice is always inseparable from this heterogeneous ice nucleation: in fact, it can be perfectly separable from it, as proven by laboratory experiments to study ice nucleation in which the humidity is externally controlled, or in natural clouds when the humidity is maintained close to water saturation by liquid. It is the diffusional growth of cloud-ice and -droplets as well as the ascent rate, and not the event of ice nucleation itself, that immediately control the supersaturation. The fact that the supersaturation and heterogeneous ice nucleation can sometimes be tightly coupled and inter-dependent (e.g. when supercooled cloud-liquid evaporates away completely) in nature does not mean that they both always form just one single process. The supersaturation is coupled to many other processes too in a natural cloud. Either cloud-droplets, heterogeneous ice, or ice from some other source, may control the supersaturation in any natural cloud.

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 Khvorostyanov without making any changes to it, in an adiabatic parcel for my 2008 paper (Phillips et al. 2008). This parcel necessarily had no microphysics, except for heterogeneous ice nucleation, in view of my direct comparison with laboratory data only concerning this same ice nucleation. 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 to the vapour growth of ice. Despite this difference, both implementations by myself and Eidhammer et al. produced a similarly unrealistic behavior of the KC scheme in terms of its freezing fraction attaining unity at warm temperatures (-10 to -20 degC), whenever water saturation (e.g. due to supercooled cloud-liquid) persisted during appearance of heterogeneous ice. So, provided there is sufficient ascent for water saturation to persist, it is not very important to the KC scheme's behavior whether

or not the supersaturation is predicted or prescribed: vast numbers of ice particles are produced either way, with all insoluble aerosols becoming frozen near about -10 to -20 degC. This is unrealistic.

The claim on page 2685 of the present paper, that my 2008 paper implements the KC scheme "without the S_w-dependence" is misleading. In fact, both Phillips et al. (2008) and Eidhammer et al. (2009) implemented the KC scheme by including the S_w input to it. The KC scheme's predicted ice nucleation depends on S_w in all our implementations. Phillips et al. (2008) merely prescribed a fixed value for it.

This requirement that the KC scheme can only be applied in a cloud model makes it difficult to be validated with high acuity against real laboratory experiments, which tend to involve the supersaturation being externally imposed. It also means other processes may be unwittingly "tuned" in the cloud model when any ice concentration observed is being predicted. This tuning compensates for the fact that, as noted by Phillips et al. (2008) and Eidhammer et al. (2009), the KC scheme over-predicts number concentrations of heterogeneous ice by orders of magnitude when water saturation persists. A plot shown by Eidhammer et al. (2009, Figure 5 therein) is quite lucid about how this tuning effectively would work: as vertical velocity is increased from a few cm/sec up to about 0.5 m/sec, the supercooled cloud-liquid evaporates later and later during appearance of heterogeneous ice from the KC scheme. Humidity collapses later and at cooler temperatures, raising the eventual ice concentration. But beyond about 1 m/sec, no such tuning is possible because all insoluble aerosols are frozen. The eventual ice concentration is determined by ascent rate and temperature, as the KC scheme is over-active.

The KC scheme, as shown by Eidhammer et al. (2009), has an extremely high bias. The present paper shows how the KC scheme only predicts ice concentrations that seem plausible ($<$ 10-100 per litre) when one of these 'lucky' situations occurs:- (1) very few IN are in the environment, as in the Arctic; or (2) sufficiently weak ascent for all supercooled cloud-liquid to evaporate away before all the insoluble aerosol particles are activated as ice, such that collapse of humidity to ice saturation then shuts down the over-active KC scheme (via its S_w input), as seems to occur in Fig. 6.

Neither of these two lucky situations apply to Figure 2 of the present paper, and so the lack of realism of the KC scheme is exposed. The same aerosol mixture used by Eidhammer et al. (2009) is applied to compare two versions of the KC scheme with the PDA08 parameterization, in a parcel model. This aerosol mixture consists of about 1000 per litre of insoluble aerosols (soot and dust), according to Eidhammer et al., who sent the authors of the present paper the data-files for the mixture. Both of the authors' own versions of the KC scheme shown in Fig. 2 predict about 1000 per litre of ice crystals at about -15 degC. The corresponding predicted freezing fraction of insoluble aerosol must be of the order of unity.

Any freezing fraction approaching unity is very unrealistic, especially for such warm temperatures. First, laboratory data (AIDA) for dust at about -20 degC and water saturation from Field et al. (2006, ACP), plotted in my 2008 paper, show freezing fractions of the order of about 5%. Such laboratory observations do not rely on the CFDC probe, and give an independent measurement. If anything, the observations of dust from the ground done by Field et al. must over-estimate ice-nucleating ability of natural atmospheric dust, as the more active dust particles at a given size will be removed by ice nucleation during transport. Second, soot has even lower active fractions of the order of 0.1% or less, near about -20 degC(DeMott 1990).

The authors' own Figure 2 graphically demonstrates the inaccuracy of the KC scheme. This is just as shown in my 2008 paper and by Eidhammer et al., who actually included the response of supersaturation. So, why does the KC scheme seem realistic in Fig. 6 of the present paper when compared with observational data ? Inclusion of the response of supersaturation to the appearance of the ice allows the humidity to evolve and to become too low during a parcel simulation, because the vertical velocity is so weak (< 5 cm/sec) that the liquid evaporates away when heterogeneous ice from the KC scheme appears. (The liquid content from the runs plotted in Fig. 6 is not

shown.) Thus, unintentionally perhaps, a compensating balance of errors may have arisen between supersaturation with respect to water and too much ice produced by the KC scheme. One can see how this evaporation happens in the parcel simulations with the KC scheme by Eidhammer et al. (2009, Figure 5 therein), as noted above. It is one of the 'lucky situations' noted above. Why is the run with vertical velocity = 0.5 m/sec omitted from this Fig. 6 of the present paper ? Going back to Fig. 5 of the present paper one sees that if it were included it would probably predict at least an order of magnitude more ice crystals, taking it away from the plotted observational data. The ascent rates shown on the plot produce the requisite amount of evaporation of liquid to compensate for the KC scheme's over-active ice nucleation, causing a semblance of agreement with observations in Fig. 6. In other words, a physical process (cooling during ascent) in the cloud model controlling the scheme's (S_w) input seems to have been tuned to produce the correct ice concentration, even if unwittingly.

I think papers presenting heterogeneous ice nucleation schemes, such as the KC scheme, ought only to be published if they provide a direct comparison at high acuity (same temperature and humidity as that imposed in the experiment) with observations of this same ice nucleation. In laboratory experiments, humidity is artificially controlled and since the KC scheme solves the fundamental physical equations governing ice nucleation, it ought to be able to predict the ice nucleation in any situation, including one with the humidity fixed to water saturation. Supersaturation change is not the same process as heterogeneous ice nucleation and is linked to many different microphysical processes in mixed-phase clouds.

If the KC scheme predicts apparently reasonable ice concentrations for Arctic clouds, this may be because there are other processes occurring in them in nature that the cloud model in which the scheme is implemented is not representing. Many processes of ice initiation exist, including ice multiplication, which are poorly understood (Cantrell and Heymsfield 2005). Consequently, Figures 7 and 8 of the present paper do not provide validation of the KC scheme of heterogeneous ice nucleation.

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