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Comment

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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The two comments of Dr. Fan reveal finally the reason for failure of the ice scheme called “KC” scheme in Fan et al. paper (2009). The ice nucleation scheme by Khvorostyanov and Curry (KC) was developed for the interstitial deliquescent aerosol. This was emphasized in the title of the major papers (JAS, KC04, 05): “The theory of ice nucleation by heterogeneous freezing of deliquescent mixed CCN. Parts 1 and 2.”, and was clearly explained in the text of these and other papers. As follows from both Dr. Fan’s 2 comments, Dr. Fan does not believe that there was interstitial aerosol in

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MPACE cloud, as stated in the 2nd Fan's comment: "So, they are not cloud residuals or interstitial aerosols as the authors claimed." Therefore, it is clear that in the paper Fan et al. (JGR, 2009), the KC scheme was applied for evaluation of freezing of a substance that Dr Fan assumed did not exist. Thus, application of KC scheme in the paper by Fan et al. was internally contradictory and incorrect. The ice scheme called in Fan et al. "KC scheme" has nothing in common with the real KC scheme for deliquescent CCN and should not be called "KC". Our recommendation is not to use the KC scheme without proper understanding of the scheme. Unless this is done, further discussions do not make sense.

Replies to specific comments.

1) Fan: The authors may have some misunderstanding about the MPACE_B case. I just want to clarify a few things here. 1) The aerosol measurements (two modes of 72.2 and 1.8 cm-3) for MPACE_B were obtained by a CN counter from NOAA CMDL, which were surface measurements (see the documentation for the intercomparison of this case: <http://science.arm.gov/wg/cpm/scm/scmic5>). So, they are not cloud residuals or interstitial aerosols as the authors claimed."

Reply. Dr. Fan's knowledge of the MPACE data is substantially incomplete. The aerosol data in this case were compiled from 2 sources: a Hand-Held Particle Counter (HHPS-6) on unmanned aircraft (below cloud layer but at altitudes, which is missed by Dr. Fan) and the near surface CN counter from NOAA CMDL. The data and bimodal distribution were considered as representative not for the surface as Dr. Fan assumed, but for the subcloud layer (see Fridlind et al., 2007; Morrison et al., 2008, cited web site). Further, if Dr. Fan thinks that there was a sharp jump in the vertical aerosol distribution between cloud and subcloud layers, it must be proven, otherwise it is merely speculation. Here we argue that the aerosol was well mixed in the cloud topped boundary layer. The cloud-topped boundary layer (CTBL) in MPACE was well mixed, as seen in the vertical profiles in the paper by Verlinde et al. (2007). Further analysis can be done based on the general picture of Cold Air Outbreaks (CAO). Cloud streets seen in satellite

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images for this case are formed, as usually during CAO, due to superposition of the mean off-ice flow and cellular convection (e.g., Agee, 1980; Cotton and Anthes, 1989; Brummer, 1992; Chlond, 1992; etc.). That is, each air parcel with and without cloud particles in such CTBL moves up and down along the spiral trajectory and performs many loops in its motion. This vertical motion includes both cloud and subcloud layers (including aerosol, interstitial and in subcloud layer), ensures mixing between both layers, including aerosol, and forms this unified, well vertically mixed CTBL. None of the models participating in MPACE intercomparison reproduced such dynamics (including LES, Fan et al., and our ACPD paper); but we can only hope that a simplified single-column representation can approximate this picture. But a subsequent brilliant paper by Solomon, Morrison et al. in MWR (2009) described well this picture with cloud rolls, intermittent vertical velocities and mixing between cloud and subcloud layers. Each air parcel traveling ~ 200 km from the ice edge to Barrow could perform several vertical loops ensuring good mixing of all properties including aerosol. Therefore, we can conclude that aerosol measurements in subcloud layer can be representative also in the cloud layer.

Clarification. Production of realistic crystal concentrations in KC scheme does not require IN concentrations as high as CCN of 50-100-200 cm^{-3} . When we wrote “comparable”, this meant that if IN is comparable with CCN, it still can produce realistic crystal concentrations, not too high. However, interstitial aerosol may freeze as IN and give reasonable crystal concentrations in KC scheme even if its concentration is as low as 1 cm^{-3} (1000 L⁻¹) or even smaller, down to 0.01 – 0.03 cm^{-3} (10-30 L⁻¹), but not as small as Fan et al. chosen, 0.2 L⁻¹. In Eidhammer et al, (2009) and our ACPD paper, it was found that this 1 cm^{-3} (perhaps even smaller) in the coarse mode can produce crystal concentrations from a few to a few hundreds or thousands per liter. Such concentrations of interstitial aerosol, ~ 1 cm^{-3} , especially with lower than average soluble fraction, not activated into drops, are certainly available in clouds. Thus, Dr. Fan’s premise of the complete absence of interstitial aerosol is incorrect. Even if Fan et al. desired to use IN concentrations NIN measured by CFDC, they did it in

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an incorrect way: this 0.2 L-1 was an artifact of temporal averaging and did not exist in reality. It is not a problem of Fan's work, but a more general consequence of the strategy adopted for MPACE model intercomparison project. Although NIN in MPACE B-period was below the CFDC threshold (0.15-0.2 L-1) during the 90 % of the time, there were significant local concentrations higher than 10 L-1 and noticeable NIN were measured during ~ 10 % of the time (see, e.g., Fig. 6 in Morrison et al., and other MPACE materials). Besides, as discussed in Morrison et al. and Fridlind et al., CFDC may significantly undercount IN concentrations all the time, especially for sizes larger than its threshold of 2 mm. For some reason, the general assumption was to average over time, which yielded NIN ~ 0.16 -0.2 L-1. This averaging was obviously inadequate in application to studies of cloud glaciation and resembles "average patient's temperature over the hospital". If in simulations as input for KC or any other scheme not these "average 0.2 L-1", but more realistic periodically (locally) high NIN ~ 5 -15 L-1 were used in the same temporal proportions as measured by CFDC (e.g., each tenth time step, i.e., 10 % of the time), this would yield a periodical "cloud seeding". The crystals formed after "seeding" will be stored in a cloud for a sufficiently long time, will be accumulated, could maintain mixed cloud state and produce crystal concentrations closer to the observed. Similar seeding effects and their effects on radiation were simulated by M. Ovtchinnikov 20 years ago at CAO using a 2D model with spectral bin microphysics (see Kondratyev et al., 1990a, b, c). The agreement for MPACE case could be better with corrected (increased) NIN data of CFDC. If such strategy was chosen by Fan et al., the results would be different. Another option: if Fan et al. tested IN concentration increased from CFDC data by 1-2 orders of magnitude as Fridlind et al. and Morrison et al. did, simulated crystal concentrations would be much closer to observed.

2) Fan: The composition was recommended to use ammonium sulfate.

Reply. For Dr. Fan's information: the composition was unknown (see cited papers and web), it was only hypothesized for simplicity and "recommended" for model intercomparison. As indicated in Fridlind et al. (2007), composition was assumed to be

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ammonium bisulfate (not sulfate as Fan assumes) for purposes of drop activation, not for crystal nucleation. The actual composition was certainly much more complicated, see e.g., JGR, v. 106 (2001) with SHEBA issue, edited by Curry, and similar sources – Big and Leck (JGR, 2001); Fridlind et al. for references and discussions.

3) Fan: If there is any soluble fraction, which is the premise of the KC scheme, in the aerosols of the coarse mode with mean radius 1.3 microns, they will immediately become droplets in cloud at RH of 100%.

Reply. Not at all. If soluble fraction of the coarse mode was smaller than in the fine mode, as is typical, then the mode 1.3 microns would remain unactivated as CCN and could serve as IN in the KC scheme. By the way. In our simulations we did not analyze in detail which mode gave the major contribution for ice nucleation. However, nucleation rate is proportional to the particle surface, and larger particles can give greater contributions, which has been shown in simulations by Eidhammer et al. (2009, EDK09). EDK included as IN only large mode aerosol with radius about 1 micron and concentrations less than 1 cm^{-3} . This mode was not activated into drops (contrary to Dr. Fan's assumption), remained as interstitial aerosol, and parcel model simulations in EDK09 produced then high crystal concentrations with KC scheme and Diehl and Wurzler's (2004) scheme. In this respect, Fan's et al. work is in sharp conflict with the EDK09 simulations: EDK09 state that KC strongly overestimates crystal concentrations, and Fan et al. say that KC underestimate them. Our analogous parcel simulations in ACPD with only coarse mode as IN and IN concentration $\sim 1 \text{ cm}^{-3}$ produced similar to EDK09 results, which give realistic cloud phase state and concentrations between Fan et al. and EDK09. Thus, only coarse mode, mostly not detected by CFDC, with IN $\sim 1 \text{ cm}^{-3}$ and smaller could ensure crystal nucleation in MPACE.

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