

Reply to comment by J. Fan: Interactive comment on “Assessment of parameterizations of heterogeneous ice nucleation in cloud and climate models” by J. A. Curry and V. I. Khvorostyanov, Atmos. Chem. Phys. Discuss., 10, C97–C98, 2010.

J. A. Curry and V. I. Khvorostyanov

For convenience of discussion, we cite below remarks from Dr. Fan’s comment, and our reply follows.

Fan: I am not convinced by using the background aerosol concentration as an input to the KC scheme in the mixed-phase clouds. I think it is more understandable to use aerosol concentrations in cloud. In the case of MPACE, most of background aerosols would serve as CCN and in-cloud aerosol concentration would be very low.

CK: Reply. First, Dr. Fan here disputes not with our simulation of MPACE but with Dr. Fan’s own invention that Dr. Fan says is somehow based on our work, but that is irrelevant to our work. We used *only* the aerosol that was measured in MPACE near Barrow under cloudy conditions as all other modelers of this project did. As indicated in Morrison et al. (2008) and other works, there was aerosol bimodal size spectrum with concentrations of 72.2 cm^{-3} in the fine mode and 1.8 cm^{-3} in the coarse mode. This was the residual aerosol that left after cloud formation, coexisted with cloud, and could serve as CCN in case of further drop formation, or as ice nuclei, since it was mixed aerosol and insoluble fraction could serve as catalyzing agent in ice nucleation.

The statement of Dr. Fan “*In the case of MPACE, most of background aerosols would serve as CCN and in-cloud aerosol concentration would be very low*”, is in conflict with general conceptions of cloud formation commonly adopted in present-day cloud physics and in conflict with measurements in MPACE. It is well known that the fraction of background aerosol (CN) that serve as CCN in cloud $\text{CCN}/\text{CN} = 0.2$ to 0.6 , with a median value of 0.5 (see e.g. the book by Pruppacher and Klett (1997, chapter 9, pages 288-290). Thus, only about 50 % of “background aerosol” become CCN and activate, and the rest exist in cloud as the interstitial aerosol, deliquescent but not activated into drops. The boundary between activated CCN (drops) and interstitial aerosol was calculated e.g. in Sedunov (1973), Ghan et al. (Atmos. Res., 1993; 1995), Khvorostyanov and Curry (JGR, 1999) and many others; it determines the concentration of interstitial aerosol and indicates that concentrations of activated and non-activated (interstitial) aerosol are comparable. Indeed, it was confirmed in MPACE. The drop concentrations in period B varied in the range about $40\text{-}90 \text{ cm}^{-3}$, and concentrations of residual aerosol after activation, were 72 and 1.8 cm^{-3} in the 2 modes. The existence of this interstitial aerosol was found experimentally in many thousands of cloud measurements over more than six decades but Dr. Fan denies this fact for some unclear reasons. If “*most of background aerosols would serve as CCN and in-cloud aerosol concentration would be very low*” as Dr. Fan assumes, then evaluation of Köhler’s critical radius and supersaturation would not be necessary, and calculation of drop activation would be very simple: just put $N_{\text{drop}} = N_{\text{CCN}}$. We would be astonished if this is the way that drop activation is treated in that spectral model by Dr. Fan.

It is this interstitial aerosol that may serve as IN in the KC scheme and does produce reasonable crystal concentrations, as this our paper in ACPD shows (Figs. 7, and 8). One may ask, if so, why these IN were not measured by the CFDC? Fridlind et al. (2007) and others noted that CFDC undercounts IN concentrations, and we tackled already in the ACPD paper this question. We see at least 2 possible reasons: a) insufficient time of nucleation (with 7-15 seconds available in CFD chamber vs required for ice nucleation 5-10–60 min or more, as numerous cloud

models show), and b) size limitation of CFDC with maximum detectable IN radius of $0.75 \mu\text{m}$ (Fridlind et al., 2007). Fig. 2 in Morrison et al. (2008) shows measured aerosol spectra, and it is seen that 2 right data points in the coarse-mode spectrum lie at $r > 0.8 \mu\text{m}$, i.e., beyond the CFDC limit. The concentration in this mode is $N_a = 1.8 \text{ cm}^{-3} = 1800 \text{ L}^{-1}$. If only 1 % of this coarse-mode spectrum at $r > 0.8 \mu\text{m}$ produced crystals, there would be $\sim 20 \text{ L}^{-1}$, but these IN would not be detected by CFDC because are beyond its limit. And it is this aerosol that is allowed to serve as IN in KC scheme, which does produce reasonable crystal concentrations.

Fan: In the cirrus cloud case, one can use background aerosol concentrations because aerosols do not serve as CCN to form cloud liquid. Maybe KC scheme can predict reasonable ice formation for the pure ice cloud case but not for the mixed-phase cloud case, since CCN takes the vast majority of the aerosols.

CK: Reply. See reply provided above.

Fan: Some other mechanisms such as droplet evaporation freezing may contribute to ice formation since IN is very low in the mixed-phase clouds. That is the whole point of Fridlind et al 2007 and Fan et al 2009 - trying to find other ice formation mechanisms possibly contributing to ice formation in the mixed-phase clouds where IN is low (because most of aerosols serve as CCN first).

CK: Reply. First, the statement “since IN is very low in the mixed-phase clouds” is also wrong in many cases, since in many mixed-phase clouds the concentration of IN can be sufficient and high, see Pruppacher and Klett (1997), and especially numerous works by Hobbs, Rangno and team, where abundant IN were measured. Second, we do not deny that other mechanisms that may act in addition to the old “classical” 4 modes of ice nucleation. As we write in ACPD, we agree with previous works as Fridlind et al. (2007), in particular, the lower maximum in crystal concentration can be caused by evaporation-freezing or other mechanism. However, before searching for various additional or exotic schemes, it is always worthy to test the traditional mechanisms, and we showed that they do work in MPACE.

Fan: It is not well justified why the background aerosol concentration should be used in the mixed-phase clouds since most of aerosols are CCN instead of IN. Therefore, it is still too early to say for sure that it is wrong in Fan et al 2009 by using the very low IN concentration - that could be the case in the mixed-phase clouds.

CK: Reply. Once again, we used not “background”, but real aerosol measured under cloud conditions that could serve as IN but was not detected by CFDC.

Of course, everybody is free to use whatever input concentrations they want, but since Fan et al. (2009) used the concrete scheme that they called “KC” scheme, there are 2 limitations or requirements.

1) If the KC scheme is used, it should be used with proper understanding as developed by the authors. Fan et al. used the KC equations but with incorrect inputs. Hence their conclusions about “KC scheme cannot produce measured crystal concentrations”, are incorrect since they were based upon using incorrect input values. Our ACPD paper shows that with proper application the KC works and gives quite reasonable results for MPACE.

2) If such strong modifications are applied to KC scheme as was done in Fan et al. (input concentration was decreased by 3-6 orders of magnitude), which make the whole scheme senseless, it must not be called “KC scheme”. The scheme that was applied in Fan et al. can be called, e.g., “Fan’s et al. ice scheme with the T - S_w dependencies as in KC but with zero input

aerosol concentration”. The zero input had to produce zero output as it happened, but the sense of such experiment is unclear, and it should not be ascribed to KC scheme.

References

Curry, J. A., and V. I. Khvorostyanov: Assessment of parameterizations of heterogeneous ice nucleation in cloud and climate models”, *Atmos. Chem. Phys. Discuss.*, 10, 2669-2710, 2010.

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