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Interactive comment on "Supersaturation, dehydration, and denitrification in Arctic cirrus" *by* B. Kärcher

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I am glad that Dr. Nelson published his thoughts about the use of the deposition coefficient α in cirrus cloud modeling and the physics behind it. I generally agree with most of his arguments, and do think they apply to many other modeling studies as well.

In particular, it became clear that the few facts known about α and related ice crystal growth models hold for relatively warm temperatures (above 250 K) and do not directly apply to the cold upper troposphere / lower stratosphere region.

One little detail is that one might argue about the term "near-equilibrium conditions" of ice in the atmosphere. I would argue that a growing body of field observations, the present and other model studies show that atmospheric ice crystals frequently experience many percent or even tens of percent of supersaturation (and subsaturation), and are hardly found closer to equilibrium. Nice examples of how RH varies in observed cirrus are given by Popp et al. (2004, their Figures 2–5) and Ovarlez et al. (2002, their Figure 1). If growth is shut off only for much smaller supersaturations (fractions of a percent), for which Dr. Nelson provided evidence in his comment, then this growth inhibition should hardly occur in the cold upper troposphere. One known exception may be tied to surface impurites hindering water vapor deposition (Gao et al., 2004). Another implication is that the experiments which support $\alpha = 0.5$ at low temperatures (Haag et al., 2003) include ice growth at high supersaturation conditions and thus may well be applicable to the present Arctic case study.

With all that being said, I will certainly be happy to add a note of caution in Section 2.3 of the final manuscript and mention that the use of a fixed value for α is a simplification and refer to this interactive discussion.

As for the question of whether the results significantly change when α is reduced from 0.5 to 0.05, I tend to agree with Dr. Nelson that one cannot seriously fix the problem by just repeating the calculation with a low α. Nevertheless, I have checked this point (see below).

In the framework of the standard growth model I have used, variations of α within a factor of 2 or so around the baseline value of 0.5 do not significantly change the results because the growth of the typically supermicron-sized ice particles then tends to be diffusion-limited rather than kinetically controlled.

A separate calculation performed for this reply shows that a value of 0.05 increases the number of ice crystals locally by a factor of 2, because high supersaturations last longer and allow more ice particles to be formed. More importantly, the cloud development changes because the crystals stay smaller and sediment more slowly. The calculated vertical extension of the lower nucleation zone increases and the duration of the nucleation event in it increases significantly. This causes the column crystal density, ice

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water path, and visible optical depth after 4 h of simulation to be a factor of roughly 10, 3, and 3 higher than in the baseline run with $\alpha = 0.5$, which is probably unrealistic. The lidar backscatter data do not provide sufficient information to directly constrain number and size of the ice crystals in the cirrostratus cloud. Note that the neglect of small-scale variability in vertical winds and heterogeneous ice nucleation effects may also modify the predicted cloud properties.

Also other studies underline that the standard growth model should be used along with $\alpha > 0.2$ or so. First, we have analyzed in great detail homogeneous freezing processes and resulting ice particle properties measured in a coolable aerosol chamber under controlled, close-to-atmospheric conditions. The result was that "We have run the baseline case with $\alpha = 0.05$ and found no combination of T and total H_2O within their uncertainty limits that predicted the number of ice particles consistent with the measurements." (Section 6.3 and Figure 14 in Haag et al., 2003). Second, we have analyzed field measurements of cirrus cloud properties. The result was that with $\alpha = 0.05$, "Values $n_i = 100\,\mathrm{cm}^{-3}$ are [now] predicted with high probabilities [of 0.03], but no ice crystals with concentrations above 20 cm^{-3} have been observed." (Section 3.4 and Figure 6 in Kärcher and Ström, 2003). Finally, the failure of using small α to explain atmospheric observations of tropopause cirrus has also been reported by Jensen et al. (2005, paragraph [34] in their Section 4.3).

Of course, this evidence is no proof that real deposition coefficients to be used in more accurate ice crystal growth models could indeed be very different from unity, and could exhibit a supersensitive dependence on supersaturation as outlined by Dr. Nelson. It merely supports the fact that standard growth models used together with α close to unity are capable of explaining atmospheric observations in many cases.

Gao, R.S., et al., Evidence that ambient nitric acid increases relative humidity in low-temperature cirrus clouds. Science 303, 516–520, 2004.

Haag, W., et al., Numerical simulations of homogeneous freezing processes in the

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aerosol chamber AIDA. Atmos. Chem. Phys. 3, 195–210, 2003.

Jensen, E., et al., Formation of a tropopause cirrus layer observed over Florida during CRYSTAL-FACE. J. Geophys. Res. 110, D03208, doi:10.1029/2004JD004671, 2005.

Kärcher, B. and J. Ström, The roles of dynamical variability and aerosols in cirrus cloud formation. Atmos. Chem. Phys. 3, 823–838, 2003.

Ovarlez, J., et al., Water vapour measurements inside cirrus clouds in Northern and Southern hemispheres during INCA. Geophys. Res. Lett. 29, doi:10.1029/2001GL014440, 2002.

Popp, P.J., et al., Nitric acid uptake on subtropical cirrus cloud particles. J. Geophys. Res. 109, D06302, doi:10.1029/2003JD004255, 2004.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 1829, 2005.