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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Numerical simulations of homogeneous freezing processes in the aerosol chamber AIDA" *by* W. Haag et al.

W. Haag et al.

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Reply to Paul DeMott, Referee 1

General Comments

Based on our analysis, we think indeed that the actvity-based parameterization of homogeneous freezing rates can be used to reasonably simulate cirrus formation by this process. We further believe that it can be applied for any updraft speed w (or cooling rate), and irrespective of how many aerosol particles from the population freeze.

We support this view by two arguments: the number of ice particles formed is mainly dictated by the competition between generating supersaturation by cooling (a process $\propto w$) and reducing supersaturation by water vapor deposition on nascent crystals. The latter argument is backed up by a theoretical description of the freezing process (Kärcher and Lohmann, 2002). The nucleation rate coefficient does not depend on w;

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We do not consider the very recent arguments about the possibility of a surface freezing mechanism in liquid particles. The papers appeared after submission of our analysis, and Tabazadeh et al. (2002) center their arguments around PSC formation. I would expect some discussion taking place about this work, too, as not all aspects of previous PSC freezing measurements have been covered. While the work of Djikaev et al. (2002) has some appealing theoretical aspects, its applicability with respect to freezing of multi-component aerosols, in particular in cirrus conditions, remains to be studied.

We think that the reviewer puts very much emphasis on the freezing rates; we argue that the pure magnitude of the rate is not the most influential factor; it can vary by orders of magnitude without changing the freezing threshold relative humidities (for particles of a given size at a fixed cooling rate) by more than a few percent (certainly outside the experimental uncertainties in measurements of RHI in the AIDA and in situ in the atmosphere). This argument is supported by theory (Kärcher and Lohmann, 2002), and by our Fig.11 (see dashed and dotted lines, along with the discussion in Sect.5.1, first paragraph).

We generally agree with the reviewer' second comment about the proposed use of the simulation methodology.

Specific Comments

1) "freezing thresholds" changed to "freezing threshold relative humidities"

2) The Koop et al. study discusses measurements in supermicron-sized droplets. The derived rate parameterization describes the rate of nucleus formation per unit volume of the drops, independent of drop size. (A possible Kelvin effect, that becomes important for very small particles building up a non-negligible internal pressure, is ignored.)

Using this parameterized rate, Koop et al. derive freezing threshold relative humidites for various droplet sizes, extending down to typical sizes of freezing aerosol particles, assuming a certain time span for the freezing event. As all measurements (within their

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intrinsic uncertainties) can be "fitted" by the same activity-based rate, the derived freezing threshold relative humidites are applicable to liquid particles of different chemical composition.

Therefore, we see no reason to actually modify our sentence l.16–19, although the actual measurements have been performed with rather large drops.

3) From Tab.1, we infer a range of 0.5–2.5 K/min; assuming adiabaticity, this corresponds to w in the range of roughly 100–500 cm/s. Indeed, this would indicate strong wave or convective activity. We add a note here that cooling rates lie in that regime.

However, we recall that we have no doubt about the applicability of the present methodology to weaker updrafts, for the reasons explained above under General Comments.

4) We add on l.21: "... is measured in an ex-situ mode with the ..." and refer to the companion paper after that sentence on l.22.

5) The average air temperature is not 1 K warmer than the average wall temperature, as the reviewer states, see Fig.2. On p.1471, we add in l.1: "... offset of ~ 0.13 K is observed ...."; add "mean" to gas and wall temperature; and refer to Möhler et al. (2002).

The sentence on p.1476, l.5-7, requires modification, because the small difference alone cannot explain the initial undersaturation. Instead, in addition to the experimental uncertainties of T and total [H_2O], inhomogenities of the wall temperature likely determine this offset.

The mean gas temperature as used here is representative for the AIDA volume due to rapid mixing of the air in the chamber. In contrast, the wall temperature data, measured at 4 locationsmay not cover the lowest wall temperature (that controls the ice coverage). Strictly speaking, we should compare the lowest wall temperature with the mean gas temperature.

We modify the above sentence accordingly.

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6) We add on l.9: "We drive the simulations by prescribing the initial aerosol size distribution and the temporal evolution of measured temperature, pressure, and total water mixing ratio, similar ..."

7) l.20, we replace "removing" by "reducing"

On l.24-26, we write: "At each time step, we decrease the modeled ice water content by subtracting the value $q(t) - q(t + \Delta t) > 0$, starting from the ice mass present in the largest bin of the ice crystal size distribution until the modeled total water content becomes equal to value indicated by the FISH instrument."

We hope this clarifies that we do not reduce ice particles in each size, but that the losses primarily occur due to removal of large crystals.

8) It is possible to truncate the distribution, but we believe that the associated uncertainties are covered by the sensitivity study summarized in Tab.3.

9) We will add two points in Fig.4, showing the onset of ice formation as predicted by the model (the time and RHI where n_i increases above 0.001 cm^{-3}) and as predicted by the AIDA measurements (the time and RHI where the depolarisation signal starts rising). These numbers are implicit in Fig.7, but showing them in Fig.4 supports the discussion of freezing thresholds in this paragraph. Actually, both points are very close together near 162 %.

10) Studies are underway to investigate this point with support from FTIR measurements in the future.

11) We refer to the companion paper, where this issue will be addressed in the description of the experimental set-up.

12) We add on p.1475, l.16: "The deposition coefficient α is equal to 0.5."

We are not sure if we understand your remark. Nucleation and ice growth are convoluted in the sense that a retardation of ice growth at low values of α causes more

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particles to freeze as the RHI is kept high for a longer period of time. This explains the sensitivity analysis discussed in Sect.6.3. The freezing nucleation rate of a liquid, however, does not depend on the deposition coefficent of gas molecules at the liquid surface.

13) As the onset of freezing is controlled by water activity, the actual onset RHIs are not sensitive to changes of either total H_2O mixing ratios of temperature, as shown in Figs.12+13 (top panels). Within the variations of [H_2O] and T shown there, the onset RHIs given in Fig.11 vary by only 0.3 %. We note that in the discussion in Sect.5.1.

The changes of the freezing onset times are more sensitive to variations of [H_2O] and T. This can be inferred from Figs.12+13 (bottom panels) and is discussedin Sect.5.2.

14) The freezing onset RHI increases when the freezing particle size decreases. When going from high to low T, we have a growing tendency for the largest droplets to have a non-equilibrium (water poor) composition, because of reduced water condensation rates. (Given a certain cooling rate; the non-equilibrium effect increases, i.e., affects more and more aerosol particles with smaller sizes, when w increases.) This explains why most ice initially comes from particles $< 1\,\mu$m, and, consistent with this fact, why the freezing onset RHIs increase.

Possibly the reviewer is confused because we have not mentioned the above anti-correlation between T and large-size portion of the freezing particle spectrum. We will add a note in the para on p.1483.

15) see 13)

16) We do not (fully) agree. We believe that the processes studied are not fundamentally different when other cooling rates are used, for the reasons considered above (General Comments). We cannot see why a faster rise of RHI (this is the only effect higher w have) should alter basic features of the homogeneous freezing process in a chemically uniform aerosol, besides the fact that the portion of aerosol particles that

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freeze increases. Truely, this study provides no sense for that, so we state this more explicitly in the last paragraph of the conclusions.

Concerning unresolved factors like the effect of organic aerosol components, we simply rely on the range of apllicability of Koop et al.'s measurements, who investigated a large number of very different liquid systems. We'd like to see experimental studies that convincingly demonstrate that some liquid systems exhibit freezing thresholds markedly different from those predicted by the activity-based results. As this study deals exclusively with liquid H_2SO_4, however, we add in l.10 "...freezing rates in H_2SO_4/H_2O aerosols provided by ...".

Interactive comment on Atmos. Chem. Phys. Discuss., 2, 1467, 2002.