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

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Specific comments

1. Measurement constraints

I agree that the discussion paper provides not enough information on which observations the model case study is based and how the model has been initialized. I will expand Section 3.1 in this regard.

As mentioned in Section 3.1, I am referring to the measurements taken with the GKSS Raman lidar near Kiruna, Sweden, published by Reichhardt et al. (2002), in particular their Figure 1b. As the observation is described there, and no further in situ information (e.g., from aircraft measurements) is available, I find it not necessary to repeat the lidar image in the discussion paper, but will describe the image in more detail in the revised manuscript. The observations suggest an almost continuously acting uplift, little wind shear, and little change of horizontal wind direction during cloud development. Multiple fall streaks were present, and cloud cells at the top boundary near the tropopause, probably caused by mesoscale dynamical variability.

The initial vertical profiles of $\Theta, T,$ and S_i have been taken from the model study of Lin et al. (2005), who independently study formation pathways (homogeneous versus heterogeneous nucleation, among other topics) of this cloud. The profiles have been taken from a nearby radiosonde. The linear sections in the Θ and T profiles could be smoothed out, but this wouldn't affect the results. The radiosonde ice saturation profile has been used as a first guess by Lin et al. (2005); toghether with the imposed vertical ascent, $S_i(z)$ has been tuned such that the overall characteristics of the cloud could be reproduced.

The use of these initial conditions enables a direct comparison to the Lin et al. (2005) simulations with regard to cirrus properties, which was intended. In the revision, I will add more detail about the agreement between these and the APSC*m*-1D model results (ice number densities and ice water content). Given the complexity of the models, this good agreement is encouraging.

In sum, the simulation is certainly idealized, but it is partly based on information from atmospheric soundings and its salient geometrical characteristics are roughly consistent with lidar measurements. I do not make an attempt to model the small-scale cloud structure explicitly (as it cannot be constrained by the lidar observation), rather I place emphasis on the dehydration and denitrification potential of this cloud type (see also item 3 below).

2. Supersaturation relaxation time

I agree that a plot of the e-folding time τ for the reduction of the water vapor content above ice saturation will support the discussion of elevated in-cloud supersaturations

(s). I refer to my reply to reviewer 1 for a description of how τ will be calculated (Kärcher, 2005, p.S543f).

The new figure shows that τ generally decreases inversely proportional to nr in the relaxation phase and ranges between 4–150 min. These rather long time scales render the appearence of quite persistent in-cloud supersaturations of 0.2 or so (as shown in Figure 3) plausible. The highest supersaturations (0.4–0.5) only occur prior to nucleation and are comparatively rare events.

Jensen et al. (2005) present a graph showing τ versus the cloud specific surface area density A_i, showing very similar values of τ. I will also add a figure showing the 1D time history $A_i(z,t)$. The highest values appear in the early fall streak region (2000–4000 μm^2/cm^3), below the fall streak values 500–1000 μm^2/cm^3 prevail, and the thin cloud top region is characterized by values $<$500 μm^2/cm^3.

I will mention in the revision that the findings concerning τ are in general agreement with Jensen et al. (2005). In particcluar, high ice supersaturations are only found in homogeneous freezing regions or when ice particles sediment into supersaturated air.

Concerning the latter point, I found it surprising that even a modest updraft of 5 cm/s can lead to persistent supersaturation of the order 0.2 in regions below cloud top (for which reason this is highlighted in the conclusions). I guess this is also the case in the subtropical tropopause cirrus analyzed by Jensen et al. (2005), although they do not explicitly note the magnitude of the large-scale updraft present in their underlying NCEP analysis fields.

3. Profiles representing irreversible dehydration and denitrification

It is true that a final precise quantification of these processes should include sublimation of the entire cloud, but I would like to keep Figure 6 as is to demonstrate the potential for dehydration and denitrification to occur. In the end, the conclusions wouldn't change.

Changes in the vertical wind speed w still consistent with the lidar data (in particular the evolution of the cloud top height and the lower cloud boundary) would lie within ∼3–7 cm/s. Corrections to the assumed initial $S_i(z)$ profile may need to be introduced if w is changed to maintain a good overall agreement. The conclusions regarding dehydration and denitrification would not change in this case.

Adding to item 1 from above, more important is the issue of small-scale temperature fluctuations. The addition of such fluctuations unresolved in the present study will lead to more numerous but smaller ice crystals. This may reduce the potential of the cloud to transport water and nitric acid vertically due to smaller ice crystal sedimentation speeds. However, to include a realistic mesoscale forcing pattern would deserve a study on its own. At any rate, I will address this sensitivity in the revised manuscript.

4. Potential for important dehydration effects

I fully agree here and will spell out explicitly that large numbers of this type of simulations are required to arrive at robust conclusions.

5. Denitrification

Here I refer to my reply to reviewer 1 (Kärcher, 2005, p.S5).

In brief, there is laboratory evidence for the fact that growing ice surfaces lead to enhanced uptake of HCl and HNO_3 (cited in Kärcher and Basko, 2005). It is very difficult to infer details of this process from existing field measurements, as trapping is a process that integrates uptake over the entire life cycle of individual crystals, and cannot be simply correlated to local variables. Concerning the validation of trapping with field data, this is a major challenge.

I believe trapping cannot be neglected in a real cloud as the ice particles hardly stay at saturated conditions. Details depend on a variety of factors, such a the likelihood of adsorbed molecules to escape from the ice surface before new ice layers are added, the ice growth rate, the trace gas partial pressure, among others (Kärcher and Basko, 2004). It seems, however, that if trapping is inefficient, then surface adsorption is even

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less efficient. Trapping becomes inefficient when adsorbed molecules desorb at a faster pace than new ice layers (leading to burial of the adsorbed HNO_3 molecules) are added. The time scale for ice growth is quite rapid ($\sim \mu$m/min or less), so for inefficient trapping, desorption must be even faster. In this case, a pure adsorption/desoprtion equilibrium would also predict little uptake.

The present cloud simulations suggest that trapping is important for HNO_3. To emphasize this point, I will add to Figure 7 a conventional calculation where the amount of HNO_3 that would be adsorbed in equilibrium (dissociative Langmuir isotherm according to Popp et al., 2004) is computed without keeping this amount in the ice phase. It is considerably smaller than the amount predicted to be trapped, so is the denitrification.

I calculate surface coverages $\theta \sim 0.01$ or lower with the conventional Langmuir model using an enthalpy of adsorption Q of 10.5 kcal/mol in the baseline case. The amount ϕ of HNO_3 associated with cirrus particles reaches only 0.4% after 7 hours in this scenario, compared to 20% in the trapping scenario with the same Q.

6. Sensitivity of the trapping process for HNO_3

Species that do not easily desorb from the ice surface can be trapped very efficiently as their residence time at the surface is typically longer than the time needed to grow several monolayers of ice. In contrast, highly volatile species may escape the ice surface faster than they become buried, rendering trapping inefficient.

Kärcher and Basko (2004) have shown that the cross-over occurs at Q values comparable to the latent heat of sublimation ($\sim$12 kcal/mol). In the simulations, we use an estimate of 10.5 kcal/mol derived through a chromatographic technique at low partial pressures. This is why the present results show a strong sensitivity on the assumed desorption rate.

As indicated, the trapping model requires independent validation (perhaps best from laboratory measurements) to draw final conclusions. This and the above arguments

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will be clearly spelled out in the revised paper.

7. FSSP measurements of small ice crystals

I fully agree, this point is well taken. I will remark that it is extremely difficult to interpret FSSP data below about 10 μm due to ambiguities associated with Mie oscillations and often poorly known deviations from sphericity.

Bearing this in mind, I believe it is still worth pointing out that the left tails of the red size distributions in Figure 4 closely resemble such FSSP data taken in young cirrus (Schröder et al., 2000, their Ci data in Figures 1,2,4,5,8), even though this similarity is hard to justify.

Editorial Comments

1. Reference to Kelly et al. (1991) will be added.

2. This will be clarified during the final production process.

References

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