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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Nitric acid partitioning in cirrus clouds: a synopsis based on field, laboratory and model studies" *by* M. Krämer et al.

M. Krämer et al.

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Reply to Referee 1:

General comment:

At first, I want to thank both referees for their helpful suggestions. Following their recommendations, I have vastly revised the manuscript.
The major changes are:

- The part of the manuscript dealing with the coverages of ice surfaces with nitric acid is removed from the manuscript and maybe submitted as an extra publication.
- The part on the partitioning of nitric acid between the gas phase, interstitial and

ice particles is expanded. Ammonia containing particles are considered for the field experiments; additional model sensitivity studies on the dependence of the partitioning on the initial nitric acid content and the in-cloud RH_{ice} are included.

- Because of the large changes of the manuscript, I also have changed the title to:

  'Nitric acid partitioning in cirrus clouds and the role of interstitial aerosol'

Reply to the specific comments:

- **Point 1:** This important point is more clearly explained now throughout the manuscript, for example read new section 3.1, page 5, left column:

  'Ice particle (HNO_3^{ice}): For both particles types, only very little HNO_3 was taken up by the ice particles (see also *Meilinger et al., 1999*) under cold cirrus conditions, while in warm cirrus the main fraction of HNO_3 is scavenged by ice particles . This increase is related to the ice surface area (A = $f(H_2O)$), which in turn increases with the temperature (see also Figure 3).

  In summary we find that both HNO_3^{ptcl} and HNO_3^{ice} are controlled by the temperature of the cirrus:

  $$HNO_3^{ptcl} = f(1/T)$$
  $$HNO_3^{ice} = f(A(H_2O(T)))$$

  for both types of interstitial particles, ternary and quaternary solutions. In cold cirrus clouds with low water content and a small ice surface area the partitioning is in favour of the interstitial particles and in warmer cirrus clouds with a greater water content and a large ice surface area the uptake on ice preponderates.'

- **Point 2:** Is concerned with the cancelled part of the manuscript.

- **Points 3 and 4:** Read new section 2.1.1, page 2, right column.

  'A forward-backward NO_y detector *Feigl et al., 1999*; *Ziereis et al., 2000* is used to measure HNO_3^{ice}. Both inlet tubes are heated to 30°C. In the forward pointing inlet ice particles are evaporated and NO_y contained in/on ice particles is released in

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addition to the gas phase NO_y. Any particle surviving the inlet tube is evaporated downstream in a gold converter which is heated to 300°C. In the converters gas-phase NO_y is reduced to NO. Downstream the converters NO is detected by chemiluminescence technique *Feigl et al., 1999*. The difference between two NO_y signals measured with the forward and the backward pointing inlet of the detector is interpreted as the NO_y on particles, which corresponds approximately to HNO_3. The accuracy of the measurement is estimated to $\pm$50% for particles larger than 4μm.'

and

New scetion 2.1.4, page 3, right column:

'As shown by *Schlager et al., 2000*, the integrated ice surfaces are underestimated by a factor of 2–3 when comparing with the ice water amount measured with the FISH instrument. Therefore, *Schlager et al., 2000* corrected the ice surface on the basis of the H_2O measurements ...'

Comment: 80% uncertainty indeed means that the value ranges from 20 - 180%, a factor of plus or minus 2 means a range of 50 - 200%.

- **Point 5:** Read new section 2.2, page 3, right column:

'2.2 Calculation of nitric acid in interstitial particles
The composition of the particles plays a major role for the uptake of HNO_3 in the interstitial aerosol, whereby the occurrence of H_2O, H_2SO_4 and NH_3 mainly influences the amount of particulate HNO_3 *Kärcher and Solomon et al., 1999*. The chemistry of the particle population in the upper troposphere is not well known, but sulfur and possibly ammonium compounds are expected to exist in this altitude. In cirrus clouds the water saturation with respect to ice is large enough so that non-activated but hygroscopic particles grow by water uptake to interstitial liquid particles, the 'haze mode' (see section 1 and Figure 1, bottom panels). These interstitial liquid particles consist of the original particle substances and water, whereby organic or insoluble material are assumed not to influence the HNO_3 uptake.

For the estimation of HNO_3^{ptcl} we assume two different types of interstitial parti-

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cles, namely ternary (HNO_3 – H_2SO_4 – H_2O) and quaternary (HNO_3 – H_2SO_4 – H_2O – NH_3) solutions. ’

- **Point 6:** Is concerned with the cancelled part of the manuscript.
- **Point 7:** Read footnote 1 on new page 4, left column:

  ’We assume that in the field experiments the interstitial and ice particles are equilibrated with the gaseous HNO_3. This assumption is based on box model calculations on uptake of water and HNO_3 by interstitial and ice particles for the conditions of POLSTAR 1997 performed by *Meilinger et al., 1999*. *Meilinger et al., 1999* show that at the point of observation the ice particles were exposed to gaseous HNO_3 for 25 hours which is long enough to equilibrate with the gas phase.’
- **Figure 1:** Vertical labels are added.
- **Figure 2:** Is replaced by a new Figure.
- **Figure 3:** Legend is added.
- **Figure 4:** Is cancelled.

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Interactive Comment