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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Composition analysis of liquid particles in the Arctic stratosphere" *by* C. Weisser et al.

Anonymous Referee #2

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General comments

This paper describes new, comprehensive measurements of synoptic-scale PSCs, including the first composition measurements of synoptic-scale PSCs. The data is particularly interesting because of differences in PSC composition compared to previous case studies, all of which were under leewave conditions. Also, the first quantitative measurements of HCl uptake by PSCs are presented. The subject matter, novelty and quality of the data, and the overall quality of the paper all make it appropriate for publication in ACP. However, there are some aspects of the paper that could be strengthened in order to maximize its relevance.

One key finding of this paper is that NAT particles coexist with STS particles. However, this critical feature of the data is at times ignored in the data analysis. For example,

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the analysis of ACMS compositions only compares the measurements to modelled *liquid* compositions. Even if ACMS does only measure liquid particles, the presence of NAT will affect the liquid compositions. However, it also seems likely that ACMS is sampling some NAT particles, especially if the NAT mode radius is near 1 μm, as suggested by the Larsen et al. [2004] companion paper. Details on the ACMS relative sampling efficiency of NAT and STS particles must be included. The contribution of NAT to the measured compositions needs be quantified, or at the least a NAT upper limit estimated. Also, the paper would be significantly enhanced with some indication of the relative volumes of NAT and liquid particles in the OPC measurements – even just adding another figure similar to Figure 3c, but with integrated volume as the x-axis.

The inconsistency between modelled and measured H_2O/HNO_3 mole ratios is intriguing and warrants more than one short paragraph of discussion. A brief mention is made that these measurements were under synoptic scale conditions whereas previous measurements, which agreed with models, were made under leewave conditions. However, this is counterintuitive: synoptic scale conditions should be closer to equilibrium. Are there other differences between the measurement conditions? Conversely can specific cases be identified where the conditions appear nearly the same in the two datasets, allowing an accurate assessment of similarities and differences? As mentioned previously, implications of liquid/NAT coexistence need to be discussed. Uncertainties in the model values should be assessed, for example due to uncertainty in the total HNO_3. Finally, how is it possible for the modelled volume to compare well with measurements if the model composition is in error? Although an explanation of the discrepancies may remain elusive, providing a couple additional paragraphs will assist readers in better understanding the nature of the discrepancies.

Similarly, the discussion of the HCl weight percent could also be expanded to address a few of the uncertainties in the analysis. How does a mix of NAT and STS particles affect the analysis? What might the model HCl weight percent be for liquid solutions with the measured H_2O/HNO_3 mole ratios? How large a temperature error would be

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necessary to explain the discrepancy?

Specific comments

title: is this truly an analysis of liquid particles? Should synoptic scale be mentioned in the title, since this is the most important difference relative to previous publications?

abstract: the conclusion that liquid particles can mask the solid particles is not included here.

p. 2515, l. 29 to p. 2516, l. 7: Cl has previously been measured in PSCs, and this early work should be referenced [Pueschel et al., JGR 94, 11271, 1989; Gandrud et al., JGR 94, 11285, 1989].

p. 2518, l. 17-19: the absence of ice particles is not proof that temperatures remained above the ice frost point, given that $\sim$3 K supercooling is necessary for ice to freeze [Koop et al., 2000]

p. 2521, l. 8-10: presumably the depolarization values are only low between 500 and 580 K?

p. 2521, l. 19-20: The final phrase needs revision, since the overall PSC development *is* ultimately the factor determining the sandwich structure. Perhaps "rather than some features related to the development of the *solid-phase particles*" is more appropriate.

p. 2521, l. 24: not all measured ratios exceed the model, in particular for times greater than 76000 UT.

p. 2522, l. 4: provide more details on the "small corrections for the transmission efficiency" – isn't this correction dependent upon an assumed size distribution?

p. 2522, l. 17-18: it is not clear that this demonstration is so "impressive" given the large scatter (on a log scale!) in the data, the wide range of assumed HNO_3 values, and the many points that lie outside the model envelope even below T_{STS}.

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p. 2522, l. 25-27: Equilibrium is a critical assumption in the model calculations and this must be clearly stated.

p. 2522, l. 28: Modify to "There are also data that do not fit the STS or NAT *equilibrium* model predictions" – equilibrium is the flaw in the model calculations, not the assumed composition.

p. 2523, l. 6: the high concentration of the liquid droplets (which is presumably the same concentration as below 500 K) is not the factor responsible for masking the NAT particles, but rather the surface area or volume.

p. 2524, l. 12: again, it is not the high number density that is responsible.

p. 2524, l. 20-24: This conclusion is much too strong, especially given the discrepancy in HNO_3/H_2O mole ratios, but also given the wide scatter in aerosol volumes shown in figure 3.

Figure 3: limiting the range of HNO_3 to a maximum value of 11 ppbv is probably too restrictive (The Larsen companion paper assumes 12 ppbv; in SOLVE, values of nearly 15 ppbv were measured in early December). Using a larger value of HNO_3 would clarify whether the high measured volumes are inconsistent with the model.

Technical corrections

p. 2518, l. 11: it would be very useful to insert a paragraph here that briefly summarizes all the other measurements available on the balloon payload; currently this information is scattered across several pages. Is the payload identical to what was previously flown? This would also be a convenient place to put the discussion of HNO_3 uncertainties from p. 2522, so that it is introduced before all of the calculations that depend upon an assumed value.

p. 2520, l. 5: what does "low mass 18 resolution" mean?

Zhang et al., 1993: is the reference for this paper correct? Zhang et al., JPC 97,

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8541-8548, 1993 seems more appropriate.

Figure 1, panel c is difficult to read. Fewer size bins would probably help, as would a better choice of colours. Also, a coarser vertical resolution or some other modification is recommended to avoid all the data dropouts.

Figure 1 caption: "assuming 5 and 11 ppbv nitric acid" Is 5 ppbv shown?

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 2513, 2004.