

***Interactive comment on* “Thermal stability analysis of particles incorporated in cirrus crystals and of non-activated particles in between the cirrus crystals: Comparing clean and polluted air masses” by M. Seifert et al.**

Anonymous Referee #2

Received and published: 8 August 2003

General Comments The paper describes measurements of the volatility of dry residuals of ice crystals and interstitial particles in cirrus clouds sampled through a counterflow virtual impactor (CVI). The measurements were made during the INCA project and cover two field experiments in Punta Arenas, Chile and Prestwick Scotland, UK. The campaigns both took place in the austral summer/autumn of 2000. Very little is known about ice nuclei in cirrus and yet the microphysical properties of cirrus have a control the radiative properties of these clouds, which are very important climatically. An understanding of the ice nuclei that control ice particle nucleation in cirrus is extremely important and such measurements as these serve to increase understanding

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in this subject area. I believe these results are worthy of publication in ACP after taking into account the comments below, however, the authors should address the issues of artefacts in the inlet raised by Referee #1, a subject that I not feel qualified to comment on. The paper is well written and free from typographical errors.

Main comments: I would like to see a discussion on the overall numbers of aerosol observed in comparison with other studies.

The introduction starts with an overview of ice nucleation (lines 10-18 page 3661). However no recent literature is discussed. There are a number of papers by US and European groups discussing the main aspects here and these should be cited in the discussion.

Pg 3664 line 4-5. The cabin temperature is around 50-70 K warmer than the outside air. This means the aerosol have been heated significantly. BaltenspergerŠs group have shown significant changes in size distribution occur by sampling with a DMA inside a laboratory at a mountain top side as compared to direct measurements at ambient temperature. Others such as Ziemann observe thermal volatilisation of particulate organic at very low temperatures.

Pg 3665 line 1-5: If the particles are composed of volatile material with a small involatile core, then the CPC/volatility arrangement will detect them as involatile particles, yet the particle properties may be very different from either a particle composed entirely of involatile material. The particle may well have homogeneously nucleated. A discussion of the way the volatility instrument deals with internally mixed particles such as this is vital to the interpretation of the results and may caveat some of your conclusions. You really must introduce these limitations to the technique for the reader to be able to interpret the result for themselves.

Page 3666 lines 15-25 and Figure 1 and 2: The authors need to point out how the changes in lifecycle of IN in different clouds affects the superposition of point in the diagrams. In IN rich air many crystals grow quickly suppressing the supersaturation

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with respect to ice and presumably restricting the points to just over 100% RH_i. With fewer ice nuclei for a cloud with the same dynamics and temperature the maximum RH_i would increase. A discussion of the variability of this diagram resulting from change in either IN, temperature or dynamics would be useful to guide the reader through the plot. It should be emphasised that RH_i is the RH above ice, I believe referee 1 missed this point. It would also be useful to show the variability of N_{int} on the same style of plot so the reader can assess the ratio of N_{cv} to N_{int}. Pg 3671 lines 5-9 Your arguments here do not take into account a significant fraction of your involatile material being internally mixed with sulfate/water solution drops. If, the particles are well aged this might be expected to be the case. These particles may well be homogeneously nucleated, you evaporate the volatile material but count the particle as it leaves an, albeit small, involatile core.

Pg 3672 lines 5-10 The description of the effect of the volatility technique appears here, surely this should come much earlier. You also need to make the comment that as you don't measure the size you don't know the relative amounts of volatile and involatile material in the particles. If there some involatile material remains it is counted just the same if it were a 200 nm solution droplet with a 20 nm involatile core as if it is a 200 nm involatile particle.

Pg 3673 line 4-5 I agree with referee 1 I don't think there are sufficient numbers of sea salt particles at these sizes to account for the very high numbers you observe.

Pg 3673 Your conclusions need to be amended given the above comments and certainly caveated with points raised about the technique not being sensitive to particles which are volatile but have small involatile cores. Your parting comments are very valuable, a tandem DMA would help greatly to understand the measurements you have made.

Minor comments: Pg 3663 line 5 the instruments have already been described elsewhere. Where? Give references

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Pg 3663 line 15-19 You need to state and reference the method you used for calculating the enrichment factor and correcting it.

Pg 3664 line 14: The interpolation between the CPCs with 5 nm and 14 nm to 10 nm only works if there is no recent new particle formation or pollution in the sampled region. If the dN is small relative to N then the correction may be valid but not if dN/N is larger than say 10-20% How large was this difference?

Pg 3664 were the counting efficiencies of the CPCs (say at 30 nm) the same for all the counters? An offline DMPS test would be sufficient here.

Pg 3667 lines 9-11. I don't agree with the statement that for volatile particles the gradient from warm to cold colors occurs with increasing N_{cvi} and also slightly with RH_i . Rather there is a band of warm colors with an increased fraction of the involatile particles occurring closest to the regions of cloud formation and evaporation (are these the first to nucleate, largest and therefore last to evaporate?) and also occurring at the maximum ice number concentration. Is this not surprising? I don't believe you have discussed this adequately.

Pg 3669 line 13 in the ambient air should read in the out of cloud total aerosol population.

Pg 3669 lines 20-23 A discussion of the meaning of the figures 1 and 2 (see above) should be sufficient to rule out any formation/growth and evaporation of the crystals in the region 95-105% you are selecting.

Pg 3672 line 17 not necessarily more aged but greater heterogeneity of sources. What evidence do you have for the increased age of SH aerosol? It needs to be presented in the paper, as far as I know this is not common knowledge.

Typographical errors Pg 3663 line 13 should read: downstream of the inlet

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 3659, 2003.

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