Atmos. Chem. Phys. Discuss., 4, S2652–S2660, 2004
www.atmos-chem-phys.org/acpd/4/S2652/
European Geosciences Union

Atmospheric
Chemistry
and Physics
Discussions

Interactive comment on "Technical Note: Evaporation of polar stratospheric cloud particles, in situ, in a heated inlet" *by* T. Eidhammer and T. Deshler

Anonymous Referee #2

Received and published: 22 November 2004

Review of Technical note: Evaporation of polar stratospheric cloud particles, in situ, in a heated inlet submitted to ACPD by Eidhammer and Deshler

In their paper, Eidhammer and Deshler develop a model, which calculates the size of polar stratospheric cloud (PSC) particles, that have evaporated in the heated inlet of their optical particle counter (OPC). From their calculations it turns out, that this instrument can be used - in combination with an ambient OPC with a non-heated inlet - to identify the phase of polar stratospheric cloud particles. Thus the authors get substantial new information on the physical state of PSCs, whether those clouds are composed of purely liquid ternary solution particles (STS) or whether a small fraction of NAT particles is mixed in a predominantly STS cloud. Therefore, this paper presents an interesting novel concept, which might also be considered for furture measurements in other types of clouds.

The paper shows excellent in-situ measurements of the particle size distribution and concentration of polar stratospheric clouds (PSC) and stratospheric sulphate particles in two years (2001 and 2002) with different OPCs. One measures the ambient particles, the other has a 80cm long heated inlet, to evaporate the condensates and measure the residues of PSCs, mainly stratospheric sulphate particles. The effect of two different inlet temperatures (˜250K and ˜290K) and a bend in the inlet on the size of PSC particles is measured and compared to measurements of the ambient PSC size distribution.

The authors develop a model, which calculates the evaporation of STS and NAT particles in the heated inlet at temperatures ˜250K and ˜290K during a particle transition time of ˜0.1sec. The evaporation process strongly depends on the difference between the partial and the vapour pressure of the different species. It is controlled by the diffusion in the gas phase as diffusion in the liquid is by orders of magnitude faster under stratospheric conditions. The model takes into account discontinuities of the Diffusion coefficient at the particle surface, the effects of latent heat due to evaporation, the Kelvin effect, the particles losses in the bend and the flow and temperature distribution in the inlet. In time steps of 10ˆ-4 sec, the model calculates the change in particle radius, vapour pressure, density, latent heat, modified diffusion coefficient, modified thermal conductivity and particle temperature.

Specific assumptions are made for the calculation of the vapour pressure of NAT because there are no measurements at temperatures above 200K and stratospheric partial pressures. Therefore the author use an approach based on Toon et al., (1986) to extrapolate the vapour pressures of H2O and HNO3 over binary solutions changed below the melting point in proportion to the latent heat of the trihydrate or by using the latent heat of the pure substances. This introduces a large uncertainty in the model.

The authors compare their model results to particle size distribution measurements with the heated inlet OPC. At 250K the evaporation of STS particles turns out to be incomplete and there is a good agreement between the model results and the observations. At 290K the STS particles evaporate completely into sulphate aerosol. In 20% of the observations despite complete evaporation of the STS particles, there remains a second mode in the size distribution, suggesting the presence of low number densities of NAT particles mixed in the STS cloud. Concerning the NAT PSCs at 250K, results of calculations using the latent heat of the pure materials agree better with the measurements than the calculations using the latent heat of the trihydrate, but the correct evaporation may lie in-between the two extreme assumptions.

The paper is generally fairly well written and should eventually be published in ACP. I detail below a number of points, which should be addressed by the authors before publication.

General comments:

a) There are several publications of non-equilibrium models of supercooled ternary solution PSCs (e.g. Meilinger et al., grl,1995, Carslaw et al., jgr,1998, Larsen et al., jgr, 2004) or NAT particles (e.g. Carslaw et al., jgr, 1999, Fahey et al., sci, 2001, Luo et al., jgr, 2003) at lower temperatures compared to the present case. The paper would benefit from a comparison with existing non-equilibrium models.

b) STS model, Fig.2 The authors show the weight fractions of HNO3, H2O and H2SO4 as function of the inlet distance. To better understand the model behaviour and to allow a comparison with other models, I suggest to additionally show the temperature, (partial/vapour) pressures and radii versus time. The authors should start the time axis shortly before the particles enter the inlet to visualise the model behaviour at the inlet tip. The initialisation of the model is not detailed, e.g. the (partial) pressures in the inlet are missing, therefore it is hard to verify their model. Is it possible to quantify an evaporation time of STS at ˜250 and ˜290K?

c) NAT model It would be interesting to see time dependent model results of the evaporation of NAT particles at 250K. A figure similar to fig.2 could be shown for the NAT model.

For comparison, 1 μm NAT particle evaporates in few minutes at 200K at 50mbar, assuming 5 ppmv water and 10ppbv nitric acid neglecting effects of latent heat. At 250K a 1μm ice particle evaporates in less than 0.1 sec at similar (partial) pressures. In the model, NAT evaporation seems to be slightly slower (fig.5). Is it possible to quantify an evaporation time of NAT at ˜250 or ˜290K? Could the authors relate the NAT evaporation time quantitatively to the evaporation time of STS (or ice)?

A very rough estimate of the evaporation time of a NAT particle at similar conditions at 250K (neglecting the Kelvin effect, the HNO3 partial pressure and latent heat) is by more than 6 orders of magnitude faster compared to results in fig.5. Is that right? Therefore, is the time step of 10ˆ-4 sec small enough to model such fast processes particularly at 290K?

How has the NAT model been initialised? Do the authors use the measured bimodal PSC size distribution with a small mode of STS particles and a large mode of NAT particles?

d) Assuming a monomodal size distribution of the sulphate aerosol, where does the second mode in the STS layers come from (Figures 3 & 4)? As there is no nucleation barrier for the STS particles, the complete size distribution should shift. In my view, a second mode can only be present with a selective (heterogeneous) nucleation process. Could the authors comment on that?

e) The following question may be beyond the scope of this paper. There is an enhancement/reduction in the measured particle number density if the particle velocity in the inlet is different from the ambient particle velocity (anisokinetic sampling). The particle enhancement is size dependent. Has this effect been corrected for the PSC measurements?

Specific comments: P = page, L = line

Abstract: Generally, the abstract is written a bit circumstantially. The authors might go through it again.

1 Introduction

f) The authors could state in the introduction/abstract that they calculate the evaporation of STS and NAT particles in a time period of ˜0.1 sec at temperatures of ˜250 and ˜290 K at ambient partial pressures.

g) There are other in-situ measurements of PSC particle size distribution (e.g. Dye et al., jgr, 1992 or Fahey et al., sci, 2001) in literature, which could be cited.

h) A comparison with existing models is required.

2 Model description

i) P 5809 L 12

Would it be possible to estimate time scales for liquid phase diffusion under inlet conditions?

j) P 5810 L 7

Is this assumption valid?

k) What are the (partial) pressures in the inlet? The details for the vapour pressures over NAT and STS, the flow and the temperature distribution in the inlet also belong to the model section.

3 Results and discussion

l) This section is difficult to follow and the title is misleading. There are some paragraphs, that belong in the model section. Therefore I suggest to split this section: e.g.

3 PSC and background aerosol observations from P 5812 L 5 to P5813 L 26

m) P5813 L 1-9

I suggest to put the temperature (as well as the flow) description at the end of the model section: e.g. 2.3 Temperature and flow in the inlet

n)P5813 L 23

Could the layer between 16 and 20 km be STS, what are the temperature uncertainties?

o) Section 3.1 STS

P5814 L 1 - P 5815 L15 belongs to the model description e.g. 2.1 Details of the STS model

p) Is the time step appropriate?

q) P 5815 L16 - P 5816 L8 show the model results and therefore could be put in new section: 4 Model results and discussion

r) P 5815 L22

The authors compare different things. Is that helpful?

s) 5816 L8 3.1.1 STS, incomplete evaporation (2001)

The title does not get the main point. The date of the observation is not important for the reader. What is meat by incomplete evaporation? That the particles are not in equilibrium with the gas phase? I suggest to change to 4.1 (Incomplete) evaporation of STS particles at 250K

t) P 5816 L14 - L21

Flow description belongs to the model section 2.3 Temperature and flow in the inlet

Full Screen / Esc

Print Version

Interactive Discussion

Discussion Paper

The authors should mention the transition time in the inlet.

u) 3.1.2 STS, complete evaporation (2002)

see point s) What is meat by complete evaporation? change to 4.2 (Complete) evaporation of STS particles at 290K

v) P 5817 L11

Larsen et al (2004) concentrate on the NAT PSCs in December 2002?

w) Could the authors verify that the second mode in the particle size distribution in Fig.4B is NAT, e.g. by calculating the evaporation of NAT particles at 290K? Could this also be other nuclei?

3.2 NAT

x) P 5817 L 23 - P 5819 L7 belongs to section 2 Model description e.g. 2.2 Details of the NAT model

y) P 5819 L7 -L 25

I suggest to change to 4.3 Evaporation of NAT particles at 250K

z) 4 Conclusions is now 5 Conclusions

Fig.1

Flight date is not easily read

Fig.2

Include a time axis in Fig.2, fraction of inlet is not very helpful for the reader. I suggest to also plot in different panels the temperature, pressure, radius and partial/ vapour pressures as function of time. The time axis could start shortly before the particle enters the inlet. Change figure caption and description in the text accordingly.

Fig.3

Labels and flight date (9 Dec 2001) are not easily read also in other figures. Incomplete axis labelling and figure caption. The altitudes correspond to which pressures? The authors could explain a cumulative size distribution. The authors could use a thicker line for the model results.

Fig.4

The authors could use a thicker line for the model results. Why is the fitted size distribution in Fig 4b ambient STS at r$>$1μ higher compared to the measurements?

Fig.5

The authors could use a thicker line for the model results.

References:

Carslaw, K.S., M. Wirth, A. Tsias, B. P. Luo, A. Dörnbrack, M. Leutbecher, H. Volkert, W. Renger, J. T. Bacmeister, Th. Peter, Particle Microphysics and Chemistry in Remotely Observed Mountain Polar Stratospheric Clouds, J. Geophys. Res., 103, 5785-5796, 1998.

Carslaw, K.S., T. Peter, J. T. Bacmeister, S. D. Eckermann, Widespread solid particle formation by mountain waves in the Arctic stratosphere, J. Geophys. Res., 104(D1), 1827-1836, 10.1029/1998JD100033, 1999.

Dye, J. et al., Particle size distributions in Arctic polar stratospheric clouds, J. Geophys. Res., 97, 8015-8034, 1992.

Fahey, D.W., R.S. Gao, K.S. Carslaw, J. Kettleborough, et al.: The detection of large HNO3-containing particles in the winter arctic stratosphere, Science, 291, 1026-1031, 2001.

Larsen, N. et al., Formation of solid particles in synoptic-scale Arctic PSCs in early winter 2002/2003, Atmospheric Chemistry and Physics, 4, 2001-2013, 2004.

Full Screen / Esc

Print Version

Interactive Discussion

Discussion Paper

Luo, B., C. Voigt, S. Füglistaler, Th. Peter: Extreme NAT supersaturations in mountain wave ice PSCs: a clue to NAT formation, J. Geophys. Res., 108 (D15), 4441, doi: 10.1029/2002JD003104, 2003.

Meilinger, S. et al., Size dependent stratospheric droplet composition in lee wave temperature fluctuations and their potential role in PSC freezing, Geophys. Res. Lett., 22, 3031-3034, 1995.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 5807, 2004.