Atmos. Chem. Phys. Discuss., 4, S2980–S2993, 2004
www.atmos-chem-phys.org/acpd/4/S2980/
European Geosciences Union

Atmospheric
Chemistry
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Discussions

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

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Received and published: 14 December 2004

Thank you to the referee, who in addition to insightful questions, provided very helpful suggestions to restructure some parts of the manuscript and thereby improve the presentation. We have incorporated all restructure suggestions in the revised manuscript.

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,

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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.

Suggested text included on P5809, L 6: "Other non-equilibrium models simulate, in lee wave clouds, growth and evaporation of STS particles (Meilinger et al., 1995), growth, evaporation and freezing of STS particles as well as formation and growth of solid particles (e.g., Carslaw et al., 1998; Luo et al., 2003: Larsen et al., 2004). The heating/cooling rates in these models are on the order of 5-80 K/hour while the heating rate in the heated inlet is $\sim$*1.5*$\cdot 10^7$ *K/hour. The basic equations for the growth/evaporation of the particles in these models are the same as in our model. However, we do not consider nucleation or freezing of particles since our model is an evaporation model at temperatures much higher than the formation temperature of NAT. While we follow a particle in the inlet, the other non-equilibrium models follow air parcel trajectories. The models described in Meilinger et al. (1995), Carslaw et al. (1998), Luo et al. (2003) and Larsen et al. (2004) are all Lagrangian or quasi-Lagrangian in radius space just as the model described here. However, direct comparison of these non-equilibrium models with the model described here can not be conducted since the lee-wave models concentrate on supersaturation cases, while our model is made for high subsaturation cases, and our heating rates far exceed anything found in the natural systems being modeled by the other non-equilibrium models."*

Note that we do not include Carslaw et al. (1999) for comparison since they do not say anything about the number density or size of NAT particles.

b) STS model, Fig.2 The authors show the weight fractions of HNO_3, H_2O and H_2SO_4 as function of the inlet distance. To better understand the model behavior 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 visualize the model behavior at the inlet tip. The initialization 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?

We have produced a new figure following suggestions from the referee. The air- and particle- temperatures increase quickly in the inlet. The difference in vapor pressure and partial pressure becomes quickly large. Therefore, any changes in partial pressure in the inlet will not affect particle evaporation in the inlet. We have therefore assumed that the partial pressure (mixing ratio) in the inlet is constant. The assumed ambient air mixing ratio is 5 ppmv for $\mathrm{H_2O}$ *and 10 ppbv for* $\mathrm{HNO_3}$.

According to the STS model, for a 1 $\mu\mathrm{m}$ *STS particle at 250 K, the evaporation time is about 0.04 s. At 290 K the modeled evaporation time is about 0.017 s. We have here defined evaporation time as the time it takes to evaporate a PSC particle to SSA.*

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.

We have included the time dependent vapor pressures and radius for NAT particles in the same figure as for STS particles (Fig. 2). However, the weight fractions of $\mathrm{HNO_3}$ *and* $\mathrm{H_2O}$ *are assumed to be constant for NAT evaporation and are not included in the figure.*

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 1 μm 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

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NAT evaporation time quantitatively to the evaporation time of STS (or ice)?

According to the model, a 1 μm NAT particle will evaporate back to SSA in about 0.1s if we use vapor pressures determined from latent heat of evaporation for pure $\mathrm{H_2O}$ and $\mathrm{HNO_3}$. The true evaporation time is probably somewhere between the estimated STS evaporation time (0.4s) and the transition time for the particles in the inlet (0.1s). We have not tried to calculate evaporation of NAT at 290 K since the model under predicts the observed NAT evaporation at 250K.

A very rough estimate of the evaporation time of a NAT particle at similar conditions at 250K (neglecting the Kelvin effect, the $\mathrm{HNO_3}$ 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 10^{-4} sec small enough to model such fast processes particularly at 290K?

No, the observed evaporation time at 250 K is not more than 6 orders of magnitude faster than the model. Modeled evaporation time for a 2 $\mu\mathrm{m}$ particle is probably 3-6 times slower than observations. Time steps $>$ and $<$ 10^{-4} s were tried. Shorter time steps did not change results whereas larger ones did. Thus 10^{-4} s is used.

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

Both modes are assumed to contain NAT particles. We chose these situations because this part of the cloud probably only consists of NAT particles. Note that the small mode is only slightly different from SSA.

d) Assuming a monomodal size distribution of the sulphate aerosol, where does the second mode in the STS layers come from (Figures 3 and 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?

We agree the second mode of the size distribution must result from a nucleation barrier, suggesting NAT mixed with the STS particles. The following text is added to comment on this: "The shapes of the PSC size distributions in Fig. 3 all show a weak second mode near 2 μm. This mode must result from a nucleation barrier, suggesting NAT mixed with the STS particles."

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?

This issue is not a problem for these measurements. The heated inlet orientations were horizontal in Dec. 2001 and vertical in Dec. 2002. In both cases the ambient inlet OPC used for comparison had its inlet with orientation the same as the heated inlet. Thus both measurements should suffer similar losses. For the vertical sampling configuration there is only a slight anisokineticity. For horizontally pointing inlets there is a large aspiration efficiency unless the balloon is floating; however, there is no way to correct the heated inlet measurements for aspiration since the size distribution for particles entering the inlet is unknown. Text was added to indicate that the comparisons were always between instruments with similarly oriented inlets.

Specific comments: P = page, L = line
Abstract: Generally, the abstract is written a bit circumstantially. The authors might go

through it again.

The abstract has been rewritten to make it less tentative.

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.

We will include the transition period.

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.

References to Dye et al., 1992 and Fahey et al., 2001 were added to the introduction, as well as three references for ice PSCs (Goodman et al., 1989; 1997; Deshler et al., 2003b).

h) A comparison with existing models is required.

See answer under section a).

2 Model description

i) P 5809 L 12

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

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We do not know the liquid phase diffusion coefficients (D_l) for $\mathrm{HNO_3}$ and $\mathrm{H_2O}$ in a STS particle, however, if we assume that D_l is on the order of 10^{-5} cm^2s^{-1} then the characteristic liquid phase diffusion time is on the order of 10^{-5} s. The characteristic time for gas diffusion from the particle is on the order of 10^{-2} s for $\mathrm{H_2O}$ and 1 s for $\mathrm{HNO_3}$ and is far greater than the liquid phase diffusion. We also assume that liquid phase diffusion is not a problem since the weight fraction of $\mathrm{H_2O}$ and $\mathrm{HNO_3}$ are both around 0.5.

j) P 5810 L 7
Is this assumption valid?

Pruppacher and Klett (1997) shows accommodation coefficients for water molecules on an ice surface with particle surface temperature below -40°C and air temperatures near 0°C. The accommodation coefficients are close to 1. We know of no other information for a liquid particle at these temperatures. Thus we can not say if this assumption is valid, and we have no information on accommodation coefficients for $\mathrm{HNO_3}$. Other investigators (Larsen, 2000) also assumes the accommodation coefficients to be unity.

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.

We have assumed constant mixing ratio in the inlet (5 ppmv H_2O and 10 ppbv HNO_3). The partial pressures for one STS and one NAT case will be shown in the revised figure 2. The details for the vapor pressures over NAT and STS, the flow and the temperature distribution in the inlet are moved 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

We agree with this comment and have followed the suggestions to reorganize the sections here and below.

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

Ok

n)P5813 L 23

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

The temperature precision is $\leq \pm$ *0.5 K. For the flight on 6 December 2002 there were 5 ambient temperature measurements. Wyoming was responsible for four of these, consisting of two Vaisala sensors and two thermistors. In addition Joelle Ovarlez, CNRS-Laboratoire de Meteorologie Dynamique (LMD), had one high precision thermistor measurement of air temperature for her frost point hygrometer measurements. On initial ascent to 20 km, 4 of the 5 temperature sensors agreed to within less than 0.5 K. These were the two Vaisala sensors, one Wyoming thermistor and the LMD sensor. One Wyoming sensor was warmer. Thus through initial ascent to 20 km we are confidant that temperatures were 1.0 K above* T_{STS} *until 19.5 km. Text was added to the manuscript to clarify that temperatures do not support the interpretation that STS particles appear between 16 and 20 km.*

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

Ok

p) Is the time step appropriate?

Yes. See point c).

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

Ok

r) P 5815 L22

The authors compare different things. Is that helpful?

Point well taken. We will remove this comparison.

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 meant 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

By incomplete evaporation we mean that the observed size distribution with the heated OPC show that particles have not evaporated to SSA (assuming SSA does not *have a large second mode). We have changed the section title to: “4.1 STS particle evaporation at 250 K”*

t) P 5816 L14 - L21

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

Ok

The authors should mention the transition time in the inlet.

Transition time will be mentioned in the introduction (see point f) and in the model section.

u) 3.1.2 STS, complete evaporation (2002) see point s) What is meant by complete evaporation? change to 4.2 (Complete) evaporation of STS particles at 290K

By complete evaporation we mean that the PSC particles evaporated to SSA. We have changed the section title to: “4.2 STS particle evaporation at 290 K”

v) P 5817 L11

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

Although the focus of Larsen et al. (2004) was on NAT, because it is now the most interesting PSC particle, the PSCs in Dec. 2002 were dominated by STS. Thus the statement in the manuscript is correct and supported by the Larsen et al. reference. No change is made to the manuscript.

w) Could the authors verify that the second mode in the particle size distribution in

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Fig.4B is NAT, e.g. by calculating the evaporation of NAT particles at 290K? Could this also be other nuclei?

We don't see it possible to verify, by additional calculations at 290 K, that the second mode in Fig. 4b is NAT. Already the model is unable to reproduce the observed evaporation at 250 K, and the primary problem in this calculation is how to handle the $\mathrm{HNO_3}$ *saturation vapor pressure, particle phase, and molar ratio. Calculations at 290 K would only exacerbate this problem and not provide a convincing conclusion.*

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

Ok

y) P 5819 L7 -L 25

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

We changed the title to: NAT particle evaporation at 250 K

z) 4 Conclusions is now 5 Conclusions

Ok

Fig.1

Flight date is not easily read

Point well taken. We will change the date to 6. Dec 2002.

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.

New figure is produced.

Fig.3

Labels and flight date (9 Dec 2001) are not easily read also in other figures. Incomplete axis labeling 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.

Ok

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$>$1 μm higher compared to the measurements?

Thank you for this comment. It lead to the discovery of a small bug in the fitting algorithm that, on rare occasions, caused the program to adopt the fit to the previous layer. This problem has been fixed and the correct Figure 4 appears in the revised manuscript, with thick lines used for the model results.

Fig.5

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

Ok

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