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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Formation of solid particles in synoptic-scale Arctic PSCs in early winter 2002/2003" *by* N. Larsen et al.

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In their paper Larsen et al. present measurements of polar stratospheric clouds (PSCs) from several balloon flights in December 2002 in the Arctic winter stratosphere. The measurements are of excellent quality and provide new insight into the formation processes of HNO3 hydrate particles. I can only congratulate the authors to this success. The paper is generally well written and should in my opinion eventually be published as a paper in ACP. However, concerning the evaluation of the data and the modelling I have a number of points which should be addressed before publication.

The paper presents three central findings:

(1) that the observations were "unique in the sense that the PSC particles seem to have formed in the early winter under synoptic temperature conditions and not being

influenced by mountain lee waves";

(2) that "the applied surface nucleation scheme results in reasonable agreement between observations and microphysical simulations," but that "reducing the calculated freezing rates by a factor 10-20" compared to a previous formulation of this process had been necessary in order to obtain this agreement;

(3) that the ensemble of liquid and solid particles measured in situ was "in good agreement with SAGE-III measurements from the same period", thus illustrating this instrument's ability to measure and distinguish different type PSC particles.

I go through my major points in the order of these topics.

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Topic (1): Unique early winter observation not influenced by mountain lee waves

There have been papers in the past claiming that many, if not all, of the observations of HNO3 hydrate particles in the Arctic stratosphere so far might have been related to ice clouds upstream, where mesoscale mountain waves had possibly forced the temperatures of passing air parcels temporarily far below synoptic scale temperatures, giving rise to ice formation. The relatively good understanding of the formation of ice in the first place and subsequently of HNO3 hydrates on this ice would then lead to a straight forward string of arguments and to an almost quantitative understanding of the entire hydrate formation process. This may then serve as input to a vortex-wide simulation of denitrification.

It has been a worry to me that recently a number of papers stated that a freezing process occurring at temperatures above the ice frost point was necessary to explain both the occurrence of solid phase PSCs early in the winter and of denitrification. I was not fully convinced by these papers because they were either referring to winters which had produced lots of ice (e.g. documented by mother-of-pearl cloud observations) or had not given sufficient attention to possible limitations of the underlying synoptic

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scale model. The inconclusiveness of previous findings was further enhanced by work suggesting that mesoscale ice clouds, despite their patchiness, could without much problem lead to widespread and non-patchy HNO3 hydrate clouds downstream.

In this sense I find the present paper much more convincing. The arguments made concerning the temperatures upstream of the measurements are lucid. Larsen et al. therefore quite rightly call their observations "unique" and that "the solid particles nucleated above the ice frost point". On the other hand they resort to previous much less compelling studies in arguing that "it has become clear that a freezing process above T(ice) is required". If this had indeed already been clear, the presented observations would not be so unique at all. By not explaining this issue in some detail and by avoiding an assessment of previous studies this paper loses momentum in an unnecessary manner. For example, they could argue that recent large scale studies have been successful only after assuming the existence of such a process, whereas here the existence is proven (as much as it can be proven). This is an important point. It is in the end up to the authors if want to express this more clearly in a revised version of the paper, but I would strongly like to stimulate them to do so.

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Topic (2): Applying a surface nucleation scheme to reach agreement between observations and microphysical simulations

Under topic (1) convincing arguments have been presented that there must be a nucleation mechanism unrelated to ice formation. The stage is set for a new and scientifically relevant approach to model these observations, simply based on ECMWF-derived trajectories without having to worry about unresolved wave activity. The authors use a comprehensive microphysical model to simulate the observations and to draw conclusions based on these calculations. The situation appears to be clear, namely that a hydrate particle production rate coefficient of 7e-9 - 1e-8 cm-3(air) s-1 is required to lead to satisfying model results, as the authors show in the middle of their "Discussion"

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section. Now one task of the model approach could be to test the constraints on this value of the rate coefficient (and potentially to some degree on its possible temperature dependence). This could be explored with the model by fine-tuning to the wealth of microphysical and optical data available from these balloon flights. In addition, sensitivity calculations with respect to other hydrate particle production rate coefficients, e.g. as applied by Carslaw and co-workers in their vortex-wide calculations, could be performed.

Instead, the authors take a detour and prefer to mix the modeling of their new finding with an interesting but unproven formulation of a "surface nucleation scheme". Surface-mediated nucleation is an interesting idea, but is at the present time disputed (see Tabazadeh, "Commentary on 'Homogeneous nucleation of NAD and NAT in liquid stratospheric aerosols: insufficient to explain denitrification' by Knopf et al.", Atmos. Chem. Phys. Discuss., 3, 827-833, 2003, and the roughly 10 comments following this publication). While I think that testing Tabazadeh's formulation of the surface nucleation rate is one thing amongst many that one can try out on the new measurements, I do not think that it is a good idea to thoroughly bind these two new and still uncertain issues together. I would argue that this connection does not really help either issue. What does it mean that "reducing the calculated freezing rates by a factor 10-20, the model is able to simulate the observed particle size distributions"? Is this supporting or weakening the surface nucleation hypothesis? The statement "the applied surface nucleation scheme results in reasonable agreement between observations and microphysical simulations" is sufficiently tuned down when the authors write that "the observations and microphysical simulations presented here may not clarify whether surface effects ... or heterogeneous nucleation (Drdla et al., 2002a) is responsible for the higher production rates". But then I wonder why this approach is picked in the first place, when a simple volume-proportional rate would have done the same job so much simpler and causing less suspicion. In summary, I would suggest the authors consider performing model runs based on more straight-forward assumptions.

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Topic (3): Good agreement with SAGE-III measurements from the same period

The Section 4 on Optical Modeling is not very well linked to the rest of the paper. The statements made here on the in-situ/remote sensing agreement and the satellite instrument's capability to distinguish type-1 PSC particles with different sizes is well taken, but the material presented would really need much more background information to be convincing. Questions such as "what indices of refraction were assumed in the T-matrix calculation?", "what was the aspect ratio of the particles?", "is the result in Fig.5f obtained by merely investigating one shape?" would need to answered first. However, I am not convinced that even after having answered these questions, that this material really belongs into this paper or would be necessary. Just as I am not sure whether all people who have helped or even guided the major effort within the EU-projects CIPA and MAPSCORE, which is behind these results, has been appropriately acknowledged. In conclusion I would suggest to simply eliminate this part from the revised version, and to concentrate on the topics (1) and (2) above. If not, the deficiencies mentioned here need to be addressed.

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Minor points:

Abstract:

1. Correct spelling of "Vintersol".

2. "Calculated extinction(indices) are in good agreement" - what does indices here mean?

3. "It appears that all PSC observations show the presence of a background population of solid particles, occasionally mixed in with more dominating liquid particles." What does Şmore dominatingŤ mean? In an optical sense?

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Introduction:

1. "... homogeneous freezing of ice out of STS requires temperatures 3-4 K below the ice frost point temperature which, in mountain leewave conditions, may lead to solid particle formation". This sentence says that ice formation leads to formation of solid particles, e.g. ice, which is probably not what is meant.

2. I would argue that a missing dehydration signal is a poor indicator for the requirement of a "freezing process above T(NAT)".

Meteorological Conditions:

1. "The PSC particles discussed in this paper can be assumed to have formed in synoptic scale cooling events ...". The word "events" sounds strange, as it was more an extended synoptic scale cooling phase.

Measurements:

1. I find the discussion on the vertical and horizontal OPC inlets very confusing: "One OPC used a vertical inlet and the second a horizontal inlet, to, perhaps, reduce bias for measurements on descent. In practice this was not the case. More large particles were observed with the vertical inlet during both ascent and slow descent, and measurements from the vertical inlet are presented here." What bias? What was not "the case"? Is the vertical inlet pointing up or down? If there were more particles in one than the other, there is after all a bias, no? Does the inlet produce an enhancement factor for condensed matter? Therefore, are number densities too high and need to be corrected downward (by the enhancement factor)?

2. "Similar interpretations have been reached from lidar measurements of PSCs, e.g. Shibata et al. (1997, 1999), Shibata (1999), Stein et al. (1999), Biele et al. (2001), and Toon et al. (2000)." This sounds as if the data presented here would by themselves also lead to the interpretation that there are solid particles externally mixed with droplets at the core of the liquid PSC. However, strictly speaking, although the size measurement

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is suggestive such a conclusion cannot be reached based on what is shown here. This is basically, because S(perp) is not shown. Instead of making an unnecessary claim the authors should either show S(perp) (which is not trivial) or go straight to the other papers, e.g. to Biele et al. who devoted most of their entire analysis exactly to this point.

Optical Modeling:

1. "vertical plots" are profiles?

Discussion:

1. "If hydrate particles form by homogeneous nucleation, these production rates are orders of magnitude higher than derived from laboratory experiments by Knopf et al. (2002)." This statement is hard to understand. May be what is meant is: "If the observed hydrate particles had formed by homogeneous nucleation, the required production rates would be orders of magnitude higher than those derived from laboratory experiments by Knopf et al. (2002)." If this is what was meant, why not be more clear and simply say: "The observed hydrate particles may not have formed by homogeneous nucleation, as the required production rates would then clearly contradict rates derived from laboratory experiments by Knopf et al. (2002)."

2. "Synoptic scale solid PSCs may of course also appear upwind of the cold region if the particles have formed in a previous cooling cycle and survived at temperature below TNAT, but this may not be as prominent as the appearance on the downwind side." This is confusing and possibly not true.

Figures:

1. Generally many labels and legends are by far too small to reproduce well.

2. Figure 2: at 606 K the first mode radius of 34 nm is unusually small. This mode would correspond to only about 38 pptv of H2SO4? Normally I would expect r ˜ 70 nm and 100-200 pptv H2SO4? Also, in the microphysical modeling section the authors

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mention a uni-modal lognormal size distribution with number density 9.3/cc, median radius r = 70 nm, and geometric standard deviation sigma = 1.4. This corresponds to ~ 150 pptv H2SO4, which sounds reasonable.

3. Figure 2: at 527 K the fit to the size distribution (right lower panel) seems awful for large particles (r ~ 3 micron). Why?

4. Figure 4: I am not sure why ES 3 Dec (550 K) and ES 7 Dec (550 K) have been omitted from the plot. This makes the plot hard to read.

5. Figure 5: It is confusing to have t = 0 at a point 24 hours before the measurement. t = 0 at the time of the measurement would help.

6. In Figure 5, the CN density appears to be constant although the air is cooling adiabatically (from 192 K to 183 K), and hence air density is decreasing by 13 %. CN density must therefore drop from 10/cc to below 9/cc. Why does this not occur?

I thank the authors for a paper showing exciting data and I apologize for submitting this comment only 2 days before the end of the discussion period.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 2485, 2004.