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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.

N. Larsen et al.

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Reply to Thomas Peter.

We thank Thomas Peter for the extensive and very constructive review of our paper, pointing to aspects which can be improved to bring the scientific message forward in a more clear way.

Topic no. 1, namely the uniqueness of the observations. As noted by Thomas Peter we use the word 'unique' (only once in the abstract) about the meteorological conditions that prevailed during the period of observations, allowing us to obtain measurements of PSCs in the early winter that were not influenced by mountain lee waves. However, during Thomas Peter's discussion the 'uniqueness' is turned towards the issue that the solid particles nucleated above the ice frost point, which is strictly speaking not what we meant by 'unique'. Thomas Peter's arguments are well taken, and if our observations,

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better than what has been the case in previous studies, almost ‘prove’ of the existence of a freezing process above the ice frost point, we take Thomas Peter’s arguments for this as a recognition.

Topic no. 2, applying a surface nucleation scheme to reach agreement between observations and microphysics. (This reply also addresses the specific comment no. 1 of referee #2). The requirement of a hydrate particle production rate of 7x10-9 - 10-8 cm-3(air) s-1 is derived from our observations, namely that around 10-3 hydrate particles per cc are produced in less than 1.75 days. We don’t see how we should ‘test the constraints of this value’ since this is a required value that the observations put on the applied nucleation mechanism. However, we have shown in Figure 6 and discussed in the text how an increased hydrate particle production, using no adjustment factor, affects the size distribution and uptake of HNO3 in the particles.

We have applied the surface nucleation mechanism of Tabazadeh et al. (2002) because this, to our knowledge, is the latest published available parameterisation of hydrate nucleation out of STS working at temperature above the ice frost point, in addition to the volume nucleation parameterisation suggested by Tabazadeh et al. in Science 291, 2591-2594, 2001. Although this surface nucleation parameterisation is based on laboratory experiments, we are aware of the caveats of extrapolating the laboratory results to stratospheric conditions. This is mentioned in the text and reference is given to the scientific debate on this issue. Actually, our results clearly show that the surface nucleation parameterisation results in too high hydrate particle production which may reflect an inaccuracy of the extrapolation of the lab data to stratospheric conditions.

Thomas Peter suggests also performing volume-proportional nucleation simulations, but what parameterisation should be applied? Tabazadeh et al. in their Science paper suggested a volume-proportional parameterisation, which was much debated, and actually Tabazadeh et al. (2002) later suggested the surface-proportional parameterisation since this gives a more consistent agreement with the lab results. Anyway, we have now also applied the volume-proportional nucleation of Tabazadeh et al. (2001), and again we can reproduce the observation reasonably well by applying a nucleation adjustment factor; in this case by increasing the volume-nucleation rates by a factor 10. This is now mentioned in the text. In both cases we don't think it would be appropriate to translate the nucleation adjustment factors into adjustments in the applied extrapolations behind the parameterisations since we don't believe that our measurements will justify this. In this respect more laboratory investigations on particle nucleation are needed. Likewise for heterogeneous nucleation, there are many free parameters that could be adjusted to fit our observations (fraction of liquid particles holding a solid nucleus, size distributions of the solid nuclei, freezing temperature etc.). However, it is questionable if much would be learned about heterogeneous nucleation from such fitting in microphysical simulations.

This leads us to believe that our observational data set cannot be used to settle the issue whether surface or volume nucleation or heterogeneous nucleation are responsible for the formation of the observed hydrate particles. We have stated this clearly. We don't take the surface nucleation as a 'standard for producing solid PSC particles' - we have only applied this mechanism because of the available published parameterisation and the arguments presented by Tabazadeh et al (2002) in favour of the surface contra the volume nucleation. We have now emphased this more clearly by expanding, as follows, the sentence (page 14, line 6) to meet the referees' comments:

"We have applied the surface nucleation mechanism of Tabazadeh et al. (2002) because this, to our knowledge, is the latest published available parameterisation of hydrate nucleation out of STS working at temperature above the ice frost point. Secondly, the surface nucleation seems more consistent with laboratory experiments (Tabazadeh et al., 2002), compared to a corresponding volume nucleation parameterisation (Tabazadeh et al., 2001; Salcedo et al., 2001). However, we have also made simulations with the latter volume-proportional nucleation and could obtain equally good agreement with the observations if these nucleation rates were increased by a factor 10. Hence, the observations and microphysical simulations presented here may not clarify whether surface or volume nucleation or heterogeneous nucleation (Drdla et al., 2002a) is responsible for the observed high hydrate production rate - an issue which would require further particle nucleation experiments in the laboratory."

Topic no. 3, optical modelling. We agree that the justification for including the optical modelling and comparison with SAGE-III could be better explained, and this has been done (p. 8, line 22):

"The good agreement with satellite measurements shows that our measurements could be representative for PSC properties in a larger geographical region in the Arctic during the period of investigation."

We have given the applied refractive indices and aspect ratios, noting that the calculated extinction only depends weakly on refractive index and particle shape. We have also acknowledged Beiping Luo and Christiane Voigt (ETHZ) for their contributions, together with members of the author team in the MAPSCORE project, to set up a database for fast T-matrix calculations (we are sorry this was not included in the submitted version).

Minor points:

Abstract: 1. Corrected.

2. We have omitted the word indices in the abstract. Extinction index is explained in the text.

3. Yes, dominating in an optical sense - corrected.

Introduction: 1. We think the sentence is correct: temperature 3-4 K below the ice frost point may lead to solid particle formation.

2. We think this is a minor point. One could argue that ice formation gives rise to a dehydration signal (e.g. Vömel et al., GRL 24, 795-798, 1997). If hydrate particle formation, responsible for observed denitrification, would have required a preceding

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ice particle formation, a dehydration signal should accompany the denitrification.

Measurements: 1. The previous text was confusing to everyone but the authors. The text has been modified and is clearer now we think.

2. We agree that we should have made one more argument for saying that the large mode in the size distributions are not due to liquid particles. The following sentence will be added at page 7, line 12:

"If the large particle mode in our profiles of PSC size distributions was due to liquid particles, this would also be reflected as bimodal size distributions of the background sulphate aerosol, which is not the case as seen in the corresponding OPC measurements with the heated inlet."

Optical modelling: 1. Corrected.

Discussion: 1. We think we have made it quite clear that our measurements cannot settle the issue of by which process the hydrate particles formed, cf. the above reply to topic 2.

2. We shall remove this sentence.

Figures. 1. Will be changed if needed in the production.

2. Comments are well taken. See reply #2 to the anonymous referee for an explanation of this difference. Text is added to the manuscript to clarify these differences in the first mode of the size distributions from the unheated and heated inlet OPCs.

3. Thank you for this comment. The data symbols plotted were, due to an error in the graph on Terry's part, not the right data file. When the correct data are included the fit is indeed much better as seen in the new 527 K panel in Fig. 2.

4. ES 3 and ES 7 are not shown in the left-hand panels (temperature history for liquid particles at 550 K) because liquid particles were not observed at this altitude in these flights. Will be explained more clearly in the caption.

5. Time is UT hours on the day of observation. The observations were obtained shortly before midnight between 6 and 7 December.

6. In the time interval between t=-24 h and t=24 h the total particle number density drops from 10.364 cm-3 to 9.7950 cm-3. This change in CN concentration is hard to discern on the log-scale plot in Figure 1c. In the same time the temperature drops from 191.3 K to 187.0 K, and the pressure drops from 17.67 hPa to 16.32 hPa during the adiabatic expansion. This leads to a drop in air density from 3.21862x10-2 kg m-3 to 3.0407x10-2 kg m-3, (i.e. a decrease of 5.5%) meaning that the total number of particles per kg of air is conserved.

Reply to Referee # 2. We thank the referee for taking the time to review our paper.

1. Please see above under topic 2.

2. The difference between these two size distributions at this altitude is caused by the fact that the heated inlet size distributions are well constrained by measurements at three sizes, CN, 0.15 and 0.25 um. With three measurements a single mode log-normal distribution will fit the measurements nearly perfectly. For the unheated inlet size distributions often, as apparent in Figure 2, 606 K, particles $>$ 0.15 um are experiencing some growth into PSC particles, thus their concentration increases over the background SSA aerosol. Since the fitting algorithm requires three measurements for the first mode size distribution it uses the concentrations at the same three sizes as the heated inlet, but because of the growth of some particles $>$ 0.15 um, the first mode distributions are generally broader and this leads to a smaller first mode radii. Thus it is not strictly correct to equate the first mode of the size distribution at 606 K with SSA. We believe the more correct representation of the SSA unimodal lognormal size distribution is given by measurements from the heated inlet and these were what were used to initialize the PSC growth model. Text is added to explain this difference when the microphysical modelling is introduced.

3. We shall mention in the caption to Figure 1 from where the 12 ppbv HNO3 originates:

... consistent with a mean-vortex value, measured by the Envisat-MIPAS instrument late November 2002.

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

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