

## ***Interactive comment on “Nitric acid trihydrate (NAT) formation at low NAT supersaturations” by C. Voigt et al.***

**Anonymous Referee #1**

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Understanding the details of how HNO<sub>3</sub>-containing PSCs form and subsequently denitrify the winter polar stratospheres remains elusive. The recent observations of dilute populations of large PSC particles and the associated modeling of their role in denitrification represent important steps forward. The present manuscript presents more observations of such large particles, thereby expanding an otherwise meager dataset in the community. The authors note that the special formation conditions inferred for the observed particles have not been inferred before and use these conditions to constrain the formation process. They further speculate that impurities, namely meteoritic material, might be the underlying cause for the nucleation of the dilute population. Overall, this is an interesting paper with unique data and should be published. How-

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ever, there are some important points that the authors need to consider in making a revised manuscript.

Important points:

Abstract. The sentence: 'These particles have the potential to grow further and to remove HNO<sub>3</sub> from the stratosphere, thereby enhancing polar ozone loss.' needs to be qualified. Are the authors suggesting that this applies to the particles they observed or to HNO<sub>3</sub> particles in general? If the former, some justification should be provided. If the latter, it should be made clear that it does not necessarily apply to particles observed. The point is that all PSC particles do not have the same potential to denitrify.

Same comment applies to p8581 In 7-9. Is this a general or specific comment?

P8583 In 6 State the volume sampling rate.

P8583 In 7 The particles can only be resolved individually if they are all equally spaced in the sample volume, which of course will never happen. Suggest clarifying this point.

P8583 Add comment about the water instrument and measurement accuracy and uncertainty.

P8585 In10 In4 Change to 'These particles have an average number' and add volume sample rate here or above and add the distance for which the average is calculated.

P8585 In6-7 Define technically what is meant by 'enhanced fluctuations' and why that is evidence of particles.

P8585 In21-29 This paragraph combined with Fig 3 is confusing. The text and Fig 3 should be clear about where in the size distribution NAT and STS particles are assumed to reside, ie label STS and NAT. Are the only NAT particles above 2  $\mu\text{m}$ ? How much of the channel difference does the STS distribution containing 0.03 ppbv account for? What is known separately from the payload measurements about the background sulfate distribution and the amount of total sulfate present? Most importantly, what is

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the sensitivity of histogram fit to the size distributions of NAT and STS particles above and below 2 $\mu$ m? It is clear that the 4-5  $\mu$ m particles are sized with low uncertainty. But given the stochastic nature of evaporating large numbers of smaller particles containing HNO<sub>3</sub>, the size distribution is likely very uncertain (ie. not unique in forming the observation histogram), particularly when one allows that selective nucleation and growth of the background aerosol can be occurring. What is the role of the background signal outside of the PSC in this analysis and how does it contribute to the uncertainties?

P8586 In14 I think that 'agreement' is stretching the concept when one is using the detection of 3 particles. Maybe 'basic consistency' is a better description.

P8587 In2-10 How is the uptake of HNO<sub>3</sub> on ice taken into account here?

P8588 In16 The units are not correct here. Should be '0.3 ppbv-s' or something similar. This is also the case for the x axis of Fig 3A.

P8589 In5-7 What is the importance of seeing these small particles for the first time?

P8592 In17 Explain/define 'obviously potent' and explain the basis for 'sufficiently low surface area'

P8594 In1-2 How does the Biermann result hold here, ie what is the difference between the freezing of STS and the formation of NAT from STS? Are they the same process? Also what is the mass difference between the proposed meteoritic mass and the STS mass, ie is the former immersed in the latter or is the meteoritic material dominate the particle composition?

Smaller points:

Title would be better if it included 'particle' as 'Nitric acid trihydrate (NAT) particle formation at low NAT supersaturation' and perhaps 'in the polar stratosphere'

Abstract and throughout. The notation ' $T_{nat} < T_{nat} - 3.5K$ ' and similar designations would be clearer if they were changed to ' $T_{nat} < (T_{nat} - 3.5K)$ '.

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P8580 In19 Change to 'visible as a colourful'

P8581 Ins1-5 'Dentrified' is not defined. Change to: 'Those large NAT particles can denitrify through sedimentation, which irreversibly transports HNO<sub>3</sub> to lower altitudes ().'

P8581 In 10 Change 'confined' to 'constrained'

P8581 In 14-15 Change to 'we investigate the potential role of meteoritic particles in NAT nucleation.'

P8581 reference footnote. The Schlager unpublished reference might be augmented or replaced by the Grooss et al APCD 2004 paper.

P8582 In 5 Change to 'project characterized'

P8582 In 17 Change to 'of the Geophysica'

P8582 In 25 Change to 'stripped from the'

P8583 In 6 Is 'normal' accepted usage? I would prefer 'standard'.

P8583 In 7 Delete 'of the instrument' and replace '-<' with 'less than'

P8583 In13-17 Improve structure by changing to 'The forward scattering spectrometer probe () measures the radiation scattered by particles that isokinetically pass through the HeNe laser. The probe size range is 0.4 to 23  $\mu\text{m}$  in diameter, divided into 30 channels. Due to ambiguities in the Mie scattering coefficient, the particle sizes between 0.7 and 1.9  $\mu\text{m}$  cannot be resolved and hence are detected as a single size.'

P8583 In21 Add 'of particles starting from x km above the aircraft.'

P8583 In22 Change to 'detection limit of the instrument is 3-4% volume depolarization at 532 nm.'

P8583 In24 Change to 'The temperature was'

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P8583 In26 Define what a PT100 sensor is.

P8583 In28 Change to 'the Rosemount data'

P8584 In10 Change to 'On the inbound flight leg'

P8584 In10 State why STS cannot produce isolated peaks.

P8585 In23 Change to '(thick horizontal black lines'

P8585 In16 Change to 'the instrument background noise outside of a PSC'

P8585 In20 and 22 Define the lower limits of the size ranges.

P8591 In6 Change to 'trajectory'

P8591 In14 Change 'during the' to 'for'

P8591 In22 Change to 'sediment less than 500m. Therefore, the likelihood that sampled particles sedimented from'

P8592 In2 Change 'summarized' to 'systematic'

P8595 In17 It is incorrect to state 'increase the time scales' I believe the intent is to state 'increase the formation rate' or something comparable.

P8596 In3 Change to 'particles represents a plausible pathway'

Figure 4 caption Change to 'The slowing of NAT growth at very'

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Interactive comment on Atmos. Chem. Phys. Discuss., 4, 8579, 2004.

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