

Interactive comment on “Spectroscopic evidence for large aspherical β -NAT particles involved in denitrification in the December 2011 Arctic stratosphere” by W. Woiwode et al.

Anonymous Referee #2

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The authors provide a nice analysis of Fourier transform IR spectroscopic measurements in an Arctic PSC in December 2011. The measurements are analyzed under a variety of radiative transfer situations and with a variety of possible PSC size distributions. The authors convincingly demonstrate the signatures of β -NAT in the data and provide good justification for including the presence of a lower cloud layer and a size distribution with a median radius near $5 \mu\text{m}$ in the second mode. The paper is well written and should be published. I have three somewhat major comments.

- 1) For those not up on the latest PSC literature. How does β -NAT differ from other NAT? What are the other NAT forms? I assume there is an α -NAT. Is there a γ -NAT?
- 2) What is the temperature history of the β -NAT particles sampled? Since the mea-

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surement is in December do the presence of these particles provide any information on the possible formation of NAT through heterogeneous nucleation? Did the temperature fall below T_{ice} in the previous days so the nucleation pathways would come through ice? I realize the authors may not wish to pursue this in an already long paper, but it is a natural question which people will be curious about and it may not take much work.

3) The discussion of the differences between the condensed hno_3 mixing ratio from the various possible size distributions from the different instruments is confusing. Specifically: In section 4, is there an explanation for why the *in situ* particle size measurements overestimate the condensed phase nitric acid by about a factor of 2 compared to the MIPAS measurements and compared to what may be realistically expected at these altitudes? Even the MIPAS measurements of ~ 9 ppbv are not consistent with the HNO_3 gas phase deficit shown in Fig. 18, which appears to be about 2-3 ppbv.

15.8-9 “The bimodal size distribution A corresponds with 18.2 ppbv gas-phase equivalent HNO_3 . . . from the FSSP-100 observations, which corresponds to 18.5 ppbv of gas-phase equivalent HNO_3 ”. By gas phase equivalent, I assume the authors mean the gas phase mixing ratio should all the condensed phase hno_3 be converted to gas phase? If this is the case please state it more explicitly.

This discussion of condensed hno_3 is the most confusing of the paper. What is excess hno_3 , and condensed excess hno_3 ? Excess to what? I assume excess to some predetermined gas phase mixing ratio determined without particles involved. It is not clear how the gray and black hno_3 profiles differ in Fig. 18. From the profiles I guess that the black profile is without particles, but this is not stated.

The authors conclude that the hno_3 in the MIPAS determined size distributions, $\sim 8\text{-}9$ ppbv, is consistent with the gas phase hno_3 , but would this not mean that all the hno_3 is condensed? If I understand Fig. 18 correctly there should be about 2 ppbv hno_3 in the particle phase, implying that this is most consistent with CLAMS. The authors should rewrite this section to clarify this discussion.

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17.5-10. Here is a plausible explanation for the discrepancy between the in situ and MIPAS estimates of condensed HNO_3 , but this possibility should be described in section 4 where this discrepancy is discussed.

Minor comments:

Figure 2. Why are there no MIPAS circles above the flight altitude of the Geophysica? 7.16, “The close agreement of the temperatures measured by MIPAS-STR, the UCSE data and the ECMWF data with calculated TNAT around flight altitude suggests that the observed PSC was composed of β -NAT.” Does this correspondence really specifically indicate β -NAT, or just NAT? I have never seen such temperatures specifically target a particular type of NAT. Also what is β -NAT and how does it differ from ??? What is/are the other option(s)? This should be discussed in the introduction.

8.11 “In Figure 9a, the particle size distributions derived by Molleker et al. (2014) from the FSSP-100 and CDP observations (black and magenta, respectively) during the PSC encounter are shown.” This figure shows observations I think, not a derived size distribution. Please clarify.

9.27 “into the simulated spectra by to the simulated” Fix.

10.10-32 and Fig. 10. Do we really need to put the reader through all these Mie calculations since the temperatures are clearly too warm for STS and ice? It seems a waste of space to include Figs 10 e)-h) and the discussion of ice and STS, for all the reasons listed at the bottom of page 10. Just state these reasons up front as to why STS and ice are not considered.

Comment related to most of the Figs 10 -16. A little more effort on the legends in the figure captions would be appreciated. I recognize the space limitations, but the readers do not know what NAT_{coa} means. Why the coa? Nor do the readers know what SP means. I can guess SP10 may mean an aspect ratio of 10 and SP01 an aspect ratio of 0.1, but no directions are provided. Why SP? Use the space to indicate the type of

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calculation, the type of particle, the aspect ratio, and if possible the median diameter of the second mode.

13.13-19. Does the fact that the particles may not be homogeneous, throughout the layer sampled, play a role in diluting/spreading the signal? Are there some minimum number of NAT particles required for the signature?

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