

Interactive comment on “Detection of reactive nitrogen containing particles in the tropopause region – evidence for a tropical nitric acid trihydrate (NAT) belt” by C. Voigt et al.

Anonymous Referee #1

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General

Voigt et al. present observations during one flight in the tropical tropopause layer (TTL) over Africa on August 8 2006. Analysing in-situ measurements of total reactive nitrogen (NO_y), total and gas phase water vapour, and FSSP observations of particles, they find evidence for nitric acid containing particles with very low particle number densities (less than 10E-4 cm⁻³), with sizes of up to about 6 microns (diameter). These observations strongly resemble those presented by Fahey et al. [2002] of observations made in the Arctic polar night vortex. The manuscript argues that the particles must be NAT, but I have not seen hard evidence that the stoichiometric ratio really corre-

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sponds to NAT. The analysis of the measurements is complemented with an analysis of the possible origin of these particles using trajectory calculations, which could also give much-needed clues about the NAT nucleation process. The manuscript is well written, but could be more precise in some cases (detailed below). The topic falls certainly in the scope of ACP, and I recommend publication after considering the following suggestions.

The manuscript does not make a strong case that the possible existence of NAT particles as observed in the TTL is of particular importance to either chemistry or denitrification - the particles occur in very low number densities, with correspondingly low surface area, and sedimentary mass flux. As far as I can see it, the real importance is that apparently particles are observed similar to the 'NAT-rocks' in the polar vortex, where they play an important role; and where - disturbingly enough - after 20 years of intense research it is still not clear how exactly NAT forms in the atmosphere. Hence the detection of NAT particles in the TTL may help to resolve these problems. The paper presents attempts to analyse possible formation mechanisms, but remains - unfortunately - somewhat inconclusive. In particular the trajectory study would deserve a more elaborate treatment. It would be helpful if the authors could calculate the trajectories with a range for poorly constrained parameters such as temperature, H₂O and HNO₃ mixing ratios, as well as imposed upwelling speed. The one sensitivity calculation mentioned (with temperatures lowered by 2K) clearly shows that the results are highly sensitive, and that we need more calculations 'sweeping' the parameter space in order to obtain a feeling for the likelihood that all particles nucleated in the TTL, or that some apparently 'fall out of the sky' and hence must have formed, e.g. on larger (ice) particles. Also, it would be probably fair to mention the work of Fueglistaler et al. [2002] where they showed that the concept of calculating particle trajectories indeed can give important clues and constraints on NAT nucleation, and subsequent 'cloud' evolution.

Detailed suggestions and corrections

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Title: Please remove 'evidence' - the measurements are really only over Africa, and in the abstract you only mention that there is a 'potential' for such a belt. Also, the 'belt' as shown in Figure 7 is really more a set of patches than a continuous belt.

P14148/L15: How large could the contribution be from PAN?

P14151/L5: What H₂O and HNO₃ partial pressures did you use to calculate Tice and TNAT?

P14152/L8: It may not have been observed because there was no attempt to do so? Please clarify.

P14152/L26: It may be helpful to give here a number for 'high'. (I see that you give some numbers on the next page, but the information should be available also here.)

P14153/L15: Typo, 'the the' should be 'to the'.

Section 6: The description of trajectory calculations should be more comprehensive. Which temporal resolution did you use for the temperature/wind fields? Were the trajectory calculated using data on the model levels or (standard) pressure levels? From the fact that you impose a mean upwelling (which is not a good approximation in the TTL, where conditions are far more variable than in the polar vortex where this strategy is often employed) I deduce that you discard the vertical wind field, and calculate the trajectories isentropically (with the superposed vertical velocity)? Also, please note that the ascent is not radiatively 'driven', but balanced, and I assume that the 0.3mm/sec should correspond to clear sky conditions; but note that at 15km this velocity is zero. Could this be a clue to the behaviour of the trajectories that virtually 'explode' in time reverse mode?

P14156/L14: I suggest to replace 'routinely' with 'frequently'.

P14157/L7: At an earlier stage you quote Luo et al. [2003] which show very high NAT supersaturation could occur in the polar vortices. The maximum of SNAT = 6 quoted here is much lower than the values found by Luo et al, a fact that should be shortly

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mentioned.

Figure 6: Caption: For at least 1.5 days prior ... Please rephrase.

Fig 7/CCM Model E39C-A: Results of this model are relevant only if (a) it gets temperatures at the tropopause correct (this information is missing in the paper), and (b) it gets H₂O and HNO₃ concentrations correct. Please give some more details about concentrations here. Also, an annual mean supersaturation may not be the best way to show the information.

Additional references

Fueglistaler, S., et al., 2002. Large NAT particle formation by mother clouds: Analysis of SOLVE/THESEO-2000 observations, *Geophys. Res. Letts.*, 29 (12), doi:10.1029/2001GL014548.

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