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Interactive comment on "The observation of nitric acid-containing particles in the tropical lower stratosphere" *by* P. J. Popp et al.

P. J. Popp et al.

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We would like to thank the referee for a very detailed review of our manuscript. In the following text, the referee's comments are shown in italics followed by our response in plain type.

1)NAT formation Fig.7 shows that particles with diameters between 1.7 and 3 μm may have grown up to 14 days in a tropical layer that is supersaturated with respect to NAT. Assuming sedimentation times of Ÿ500 m/day for a 5 μm NAT particle and given that the NAT saturated region extends vertically 500 m above the measurement altitude, this implies that the larger observed particles (4.7 μm) must have grown in an area that is subsaturated with respect to NAT above 18.5 km. Fig.7 shows that particles with diameters larger than 3 - 3.5 μm fall from higher altitudes and have not formed in the

tropical NAT layer, hence cannot be explained with the present theory. Regarding the detection range of 1.7 to 4.7 micron and an equal distribution of particle sizes, particles with d >3-3.5 μm account for a significant fraction of the observed particles. How do the authors explain the formation of the larger NAT particles?

The particle sizes described in our paper are in units of microns in diameter. The largest observed particle diameter quoted in the paper is 4.7 microns. A particle of this size has a sedimentation rate of approximately 180 m/day at 80 hPa and 195 K, and will therefore reside in the NAT-stable region described in Fig. 4 for a longer period of time than it would if it had a sedimentation rate of 500 m/day. We note, however, that the back trajectories for the largest particles in Fig. 7 (that appear to fall from higher altitudes and never reach the background particle size in the top panel of Fig. 7) encountered NAT subsaturated air for an extended period, and the trajectory calculations cannot explain their existence. The text has been modified in section 5 (Particle growth and trajectory calculations) to more clearly state this position.

2) NAT particle composition Low concentrations of small ice particles (d<10μm) or cirrus clouds with an ice water content < 1 ppmv are not detectable with the presented instrumentation. Nitric acid measurements in cirrus clouds (Popp et al., JGR, 2004, Ziereis et al., GRL, 2004, Kondo et al., GRL, 2003) show nitric acid uptake of few pptv or more by cirrus clouds. These measurements suggest, that both ice particle growth as well as nitric acid uptake in ice crystals is a fast process given cirrus lifetimes of less than a day, compared to the calculated NAT particle growth times of several days. Hence, can the author unambiguously exclude having measured NAT or nitric acid in/on ice crystals? If so, then the authors then might change the title to: The observation of NAT particles in the tropical lowermost stratosphere. A discussion/exclusion of NAT/nitric acid in ice crystals could strengthen the conclusions of the paper.

In the second paragraph of section 3 (Particle Composition and Size), we thoroughly discuss the possiblity that we had observed ice particles containing nitric acid. As we state in the paper, given the current understanding of nitric acid uptake by ice particles

ACPD

5, S4838–S4843, 2005

described in the references listed above, small ice particles (diameters less than 10 microns and ice water contents less than 1 ppmv) are not likely to contain enough nitric acid to produce the instrument response shown in Figs. 2 and 3. Larger ice particles, if present, would have been observed by other instruments operating onboard aircraft during these flights.

Specific comments in order of appearance in the paper: Title: The particles have been observed in the lower stratosphere, the profiles of nearby sondes show a thermal tropopause extending over several km in altitude, hence the author might consider changing the title to: The observation of nitric acid containing particles in the tropical lowermost stratosphere or The observation of nitric acid containing particles near the tropical tropopause.

We prefer not to change the title of the manuscript because the description “tropical lower stratosphere” is correct.

Abstract: NAT particles with similar properties (n, d) have been observed in the Arctic stratosphere (Voigt et al., ACP, 2005). In the tropical troposphere, the particle growth times are enhanced compared to polar conditions, but the particle properties are similar. Are the observations a new category of NAT particles or are these NAT particles just measured at another location? It might be reasonable to not to circulate too many categories of NAT particles in literature, tropical NAT particles is just fine (see also introduction).

The particles reported in our study are similar to NAT particles known to exist in polar regions. We contend that our observations represent a new category of particle because they were observed in the tropics, and agree that “tropical NAT particles’ is an appropriate term to describe the particles.

How do the authors determine the thickness of the observed particle layer? Have there been particle measurements above 18.1 km altitude and how long was the measurements time below 18 km? Is the altitude range of the particle observations mainly

ACPD

5, S4838–S4843, 2005

determined by the flight path and measurement times at different altitudes? Is that a particle layer?

We did not make measurements at altitudes higher than 18 km, and we cannot be certain the particle layer does not extend to higher altitudes. The text has been modified in section 2 (Particle detection) to more clearly state this position. Measurements were made at altitudes lower than 18 km and particles were never observed in these regions, leading us to conclude that the particles exist in a layer that starts at 18 km.

P10101 L13 The sampling volume has to be corrected for the particle enhancement factor. Therefore, I suggest and the known particle sampling volume

Agreed.

P10101 L25 change: for a short period

Agreed.

P10101 L27 see abstract

Agreed.

P10103 L1 Could those particles represent the large tail of a background ternary aerosol distribution?

As explained in the paper, these particles cannot represent the large tail of a background ternary aerosol distribution. Under the ambient conditions at which the particles were observed, the uptake of HNO_3 by the background aerosol population is not thermodynamically favorable. Furthermore, since there is no thermodynamic nucleation barrier to the uptake of HNO_3 by the background aerosol, all available aerosols would grow and gas-phase HNO_3 would be significantly depleted. We note in the paper that there is no significant depletion of gas-phase HNO_3 in the presence of the observed particles.

P10103 See major comment

ACPD

5, S4838–S4843, 2005

Agreed.

P10103 L23 slightly larger than

Agreed.

P10104 L20 Does the larger scatter in the downward facing channel data during particle observation periods (see Fig.3) results from the detection of smaller particles?

No. The particle observation period in Fig. 3 occurred soon after takeoff during the flight on 27 January. The signal-to-noise ratio of the CIMS instrument is slightly higher immediately after instrument startup, resulting in the apparent larger scatter during this period.

P10107 L8 It should be stated, that the NAT saturated area extends 500 m above the altitude of the measurements.

Agreed.

P10107 L19 What effect has the upwelling velocity on the particle trajectories? What is the quantitative difference to calculations without upwelling velocity?

The upwelling velocity serves to slow the sedimentation rate of the particles during the growth period. The text has been modified to more clearly state this position.

P10107 L21 Fig.7 shows that only the smaller particles (d<3 to 3.5μm) have nucleated in the NAT supersaturated area. Is that right? See also major comment 1. Please correct the text accordingly.

As stated above, the back trajectory calculations fail to explain the existence of these larger particles. They do not nucleate in a region above the NAT supersaturated region. The text has been modified accordingly to more clearly state this position.

P10107 L21 change the trajectories to the particle trajectories

Agreed.

ACPD

5, S4838–S4843, 2005

P10107 L24 A major fraction of the particle trajectories ends in a localized region between 55 to 65 E and 22 to 26 S.

Thirteen of the 27 trajectories shown in Fig. 6 end in the localized region between 55 to 65 degrees E and 22 to 26 degrees S.

P10108 L11 The authors might derive a nucleation rate for their particle observations.

Reporting a nucleation rate for the observed particles is beyond the scope of our manuscript, and might be of limited value given the low number of particles observed.

P10110 L6 10 Considering also smaller NAT particles, which should exist but cannot be detected with the set of instruments results in an increase of that fraction.

Agreed.

P10110 L22 change af. to af

Agreed.

P10111 L19 What is meant by the total temperature?

Total temperature is the temperature observed at the aircraft probe, which represents the ambient temperature plus a component contributed by ram heating at the surface of the probe.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 10097, 2005.