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Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #2

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The paper describes outstanding measurements of nitric acid containing particles in the tropical lower stratosphere, defined as new category of NAT particles in that region. The particles with low number densities ($<$10ˆ-4 cmˆ-3) and diameters up to 5 μm were observed near 18 km altitude over vast areas. The authors suggest, that these NAT particles had grown up to 14 days near the tropical tropopause, hence might be common in the tropics. The paper concludes by a discussion of the particle nucleation process and the atmospheric implications of NAT particles in the tropics.

The paper is very well written, clear, concise and well structured. The presented data are of excellent quality. The authors give a thorough description of the particle measurements and the particle formation conditions. The paper presents a valuable contri-

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bution to science and definitely merits publication after the major comments have been addressed.

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 ˜500 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 sub-saturated 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?

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 conclu-

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sions of the paper.

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.

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).

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

P10101 L13

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

and the known particle sampling volume

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P10101 L25

change: for a short period

P10101 L27

see abstract

P10103 L1

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

P10103

See major comment

P10103 L23

slightly larger than

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?

P10107 L8

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

P10107 L19

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

P10107 L21

Fig.7 shows that only the smaller particles (d$<$3 to 3.5μm) have nucleated in the NAT

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supersaturated area. Is that right? See also major comment 1. Please correct the text accordingly.

P10107 L21

change the trajectories to the particle trajectories

P10107 L24

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

P10108 L11

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

P10110 L6

10 % of the air masses could contain NAT particles. add: with the observed properties.

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

P10110 L22

change af. to af

P10111 L19

What is meant by the total temperature?

Summarized, the authors present an excellent paper and I can only congratulate the authors to their success.

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