Aerosol observations and growth rates in the tropical tropopause layer

Reply to reviewer 2

1. Throughout the text particle number concentrations are sometimes reported for standard temperature and pressure, sometimes they are not. Please make a clear statement at the beginning how particle number concentrations are reported. Furthermore, I suggest to adjusting the figure axis titles where particle number concentrations are given in 1/cc instead of 1/cm3. Please use a consistent unit in the manuscript. You should also explain STP in the text.

Throughout this work we have always worked in Standard Temperature Pressure (STP). To clarify, we will add (STP) to the end of all our aerosol concentrations, in the text.

2. Number concentrations of Aitken mode particles are derived from the difference between two condensation particle counters with one being equipped with diffusion screens. However, diffusion screens never allow a 100% transmission efficiency as can be taken from Feldpausch et al. (2006). Please state in the text whether you considered the full transfer function for diffusion screens in your data analysis. This is a crucial point because it has a significant impact on the comparison of model data and observations. Actually the manuscript only mentions corrections for low pressure operation

We discussed the work of Feldpausch et al. (2006) in sections 2.2, and the end of section 5 we pointed out that 'the accumulation mode ..... counts arise from Aitken mode particles .... detected because of the broad efficiency cut-off imposed by the diffusion disks' . We do not believe that this transmission function is well enough known to allow a quantitative correction to the data. That is why we did not use this CPC quantitatively – however we do use the results qualitiatively to show that particles are growing over time to a size detectable through the diffusion screen.
The text in 2.4, regarding subtraction of one CPC from another, is in fact incorrect, and left over from an earlier draft of the paper before this problem was fully appreciated; we thank the referee for pointing this out. The paper (and figures) will be corrected accordingly.

3. Intercomparison of model results and observations is performed on the basis of integral number concentrations for the size range 10-100 nm and 100-1000nm, see Figure 10. The presented results are not discussed with sufficient detail because Aitken mode number concentrations vary over one order of magnitude for almost similar time out of cloud. On the other hand accumulation mode number concentrations also show two clusters of data, one with number concentrations close to 0 particles per cm3, and another cluster with values above approx. 10 particles per cm3. It is strongly recommended to discuss whether or not there is a link between high number concentrations in the Aitken mode and low number concentrations in the accumulation mode. A map of states of tropospheric aerosol was introduced by Schröder et al. (2002). Although this data were obtained in mid-latitudes the overall scheme is also valid for the TTL regions. I strongly recommend discussing Fig. 10 in relation to Schröder et al. (2002).
You may also try to plot data in a similar way as Schröder et al.
As discussed above the CPC with diffusion screen had a very broad efficiency curve that allowed particles as small as 30 nm to be measured with an efficiency of about 10% qualitatively (see page 2369, lines 1 -7). Therefore, the so called accumulation mode was only discussed qualitatively

because we could not discount the fact that significant numbers of sub-100nm particles might well have been detected by the CPC with diffusion screens. We were in fact able to replicate the 'accumulation mode' data curve (figure 10) by modelling particle growth from CPC1 to CPC2 (diffusion screens) with the broad efficiency curve of Feldpausch et al. (2006) [see page 2372, lines 10 – 18], but this is only a qualitative test, as argued above.

Schröder et al. (2002) made a comprehensive study of the free troposphere and tropopause region. We however limited our study to data gathered from a convective storm close to its outflow source. Therefore, we were looking at air that was in effect 'new' after its recent cloud processing. The Aitken and accumulation mode were suppressed by the cloud particles, shown by the very low number concentrations of both modes (see figure 7, after time 13.6). This effect was seen throughout the flight of 23 January and is indeed in agreement with Schröder et al. (2002): scavenging by large cloud particles reduces the aerosol number concentration in cloud. The two clusters of accumulation mode seen in figure 10 (right panel) show a change from 0 /cm3 up to ≈ 50/cm3; this we believe is due to growth of particles from the nucleation through to observable sizes, first detected by CPC1 (10 nm lower cut-off) and then, due to its wide efficiency, into the CPC2 (diffusion screens) detection range. As mentioned above the second CPC will start to observe particles as low as 30nm with approximately 10% efficiency. So it is the overlapping of the two CPCs in the lower/mid Aitken detection range (10-50 nm) which causes the sudden increase seen in 'accumulation mode' number concentrations (figure 10 – right-hand side). We cannot therefore compare our results with Schroder's.

4. The presentation of the material requires improvement. It is surprising that the results section is only half a page long. I suggest renaming section 4 as results section and move the current section 5 into section 4. I would also like to see results from the modelling study being presented in the results section because the model runs were performed for the specific observations conditions (I assume).

We prefer to keep section 4 and section 5 (results) separate because section 4 deals more with flight specifics and methodologies for detecting nucleation zones. The results section deals with the specific outcome of applying this methodology: the plot of aerosol growth against time out of convective anvil. We also prefer to keep the model as part of the discussions section because it was done as a test of our convective data and we believe it fits better with the general discussion of the in situ data and the validity of the nucleation hypothesis.

SPECIFIC COMMENTS

The specific comments have been amended in the paper.

Page 2371, line 22. What means a standard deviation of 1.237 nm for a log-normal size distribution?

Have placed a lognormal definition in the paper to clarify.

References: please check the references, currently they are not in line with ACP requirements. Each reference ends with a 4 digit number after the year of publication.

These extra four digits seem to have been added by the office and were not part of our latex file.

References

P. Feldpausch, M. Fiebig, L. Fritzsche, and A. Petzold. Measurement of ultra¯ne aerosol size distributions by a combination of diffusion screen separators and condensation particle counters. *Journal of Aerosol Science*, 37(5):577{597, 2006.

Schröder, F., et al.: Aerosol states in the free troposphere at northern midlatitudes, J. Geophys. Res., 107, 8126, 10.1029/2000jd000194, 2002.