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Interactive comment on "Aerosol composition of the tropical upper troposphere" by K. D. Froyd et al.

Anonymous Referee #2

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The paper presents an interesting and important data set of particle composition measurements in the tropical tropopause layer. Measurements were made using the PALMS single particle mass spectrometer and a variety of results is presented concerning the vertical profiles for different types of aerosol, aerosol sources, transport processes into the TTL, transport across the tropopause, etc. The paper is well-written and results are presented in a concise and structured way. The detailed analysis of the data reveals a lot of new findings. Aerosol composition data from the TTL region is extremely scarce and therefore this data is very valuable to the atmospheric aerosol community. I recommend publication in ACP after consideration of the minor points listed below.

Minor points:

C1801

- Section 3.2. The determination of "convective influence" should be discussed in more detail, especially as it is used quite a bit for the data interpretation later on. Some questions come to mind when reading the description given here, eg: are trajectory calculations aborted once a region is reached with significant convective influence? Or can the same air parcel have flown through various convectively influenced regions? It should be mentioned how optically thick convective clouds are distinguished from other clouds. Can it happen that trajectories pass below an anvil but have not been affected by convection? How has the method from Pfister et al., 2001, been used/validated/modified since 2001? Also, Figures 2 and 3 are very small and it is hard to discern the details. It would make sense to make these figures 2 pages each (one for a) and one for b)), for example. Some of the data in Fig 3 is difficult to interpret, e.g. there are lots of dark blue dots in Fig 3b (left panel) indicating often convective influence up to 18-20 km altitude but the right panel indicates only a very small convective influence for these altitudes and this is also stated in the text. Nevertheless, the dark blue dots indicate to me that there is apparently some marine convection also reaching the upper TTL and even the stratosphere. Please comment.
- For the various markers used to identify certain aerosol types please list the actual mass/charge signals used for analysis (e.g. a list of m/z values used for determination of the particle types listed in line 23/24, p. 9404).
- To assess the significance of the various statements it is important to get an impression of the number of analysed particles as a function of altitude, e.g. for the altitude bins of Fig 5 and Fig 8. Some of the data points presented here are probably based on a much larger number of analysed particles (14-18 km altitude) than others. This information could be given as a table. It is also somewhat misleading to just connect the data points if there is insufficient data for the altitude range in between. (e.g. Fig 5b between 4 and 10 km altitude).
- For the data presented in Fig 6 it is surprising to see that there is almost always a minimum at exactly 10 nm between the nucleation mode and the larger particle mode.

Is there any explanation for this? I would expect that there should be more often particles growing over into the larger mode (like CR-AVE at 13 and 18 km) and/or the gap in between the modes should not always be located at 10 nm. Could there be a problem with the algorithm to calculate the size distribution?

- Section 5.5. Can processes of secondary ice formation be excluded to explain the differences between the ice crystal number concentration (\sim 20-100 L-1) and mineral dust particle concentration (\sim 5 L-1)?
- Did you observe in the TTL any mercury containing particles as in Murphy et al., Science, 1998, or lead containing particles as in Murphy et al., 2007 or Cziczo et al., Nature Geoscience, 2009? Please comment.

Technical corrections:

- p. 9402, I. 29, correct "a small numbers"
- p. 9426, l. 6, correct "altitudes profiles"

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