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Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #1

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General Comments

This paper presents some nice detailed results of aerosol composition in an undersampled region (the tropical upper troposphere). The paper is well organized and presented, including the figures that convey a large amount of information in useful format. It represents an important contribution in terms of results as well as discussion of likely aerosol sources and processes active in this region. My main substantive comments (detailed below) relate to assessing possible indirect effects of convection on the aerosol chemistry, such as increased actinic flux and/or lightning sources of nitrogen compounds. On the other hand, I suggest that a few sections could be shortened without losing context (while ACP does not have a length limit, the guidelines do request "concise" contributions).

Specific Comments

9402, line 24: Does PALMS really measure a “large fraction of aerosol number” even in the mid-latitudes and subtropics? Information on p. 9404 and 9412 indicates that most of the number was below the PALMS size limit for this data set, and I suspect this is also true for other UT regions. Based on the measurements discussed on p. 9403 (if for appropriate sizes) or theoretical arguments, it would be nice to discuss whether the composition of this smaller (presumably younger) aerosol is likely to be different from the larger particles that contribute most of the mass.

9406, Heading 3: Meteorological is misspelled.

9408, line 10: A little more information on the trajectory technique would be useful. It is “similar to Pfister et al. 2001”, but is it then the trajectory model of Schoeberl and Sparling (1995), coupled to the GEOS-1 analysis fields? Perhaps more succinctly, how is the current analysis different from the Pfister et al. one? Also, “convective influence parameter” could be more clearly defined on p. 9408–does a value of 1.0 just correspond to all trajectories in the cluster having intersected clouds at any point? I.e., trajectories that intersect a cloud at one point have the same weight as trajectories that intersect clouds many times, or is this history weighted somehow as well?

p. 9409-9410: A more detailed explanation of what markers are used to identify different particle types would be useful, for example in a table. Also, if particles are internally mixed, for example, EC or mineral dust with sulfate–how is it handled? It seems (p. 9403) that this paper uses a modification of the clustering technique used in Murphy et al. (2004)–please elaborate.

p. 9414: It should be noted that there are many aspects of the Ekman et al. model that probably do not apply to marine convection, namely the very strong updraft (max of 30 m s-1) and the extremely high initial Aitken concentrations in the boundary layer, which may (through latent heat release) affect the storm dynamics as well as precipitation rate. Finally, the Ekman et al. model does not seem to include resuspension of aerosol

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after entering cloud, which could be an important source of aerosol for regions where ice is growing at the expense of liquid water and in evaporating regions of the anvil itself.

Section 5: This is where I would suggest trying to be more concise. In most subsections, several different hypothesis are invoked to possibly explain the results, with substantial discussion of both those ideas that are discarded as well as those that are still possibilities. Spending less time on details of rejected hypotheses and cutting nonessential information as well as ideas already presented in earlier sections would help make a more focused, satisfying, paper. In particular, Sections 5.1.1, 5.2 and 5.3.1 could be condensed in this way.

Section 5.3.2: Could increased actinic flux due to convection play a role in increasing oxidation capacity in the TTL? While fewer trajectories may meet the convective influence criteria (physically passing through cloud) in the TTL, perhaps the enhanced actinic flux above anvils could still increase OH in this region.

Section 5.3.4: Since lightning produces much of the NOx and contributes to NOy in the tropical upper troposphere (i.e., Tie et al., JAC 2002), I wondered if this could be a source of nitrogen in the aerosol. If so, one might expect more nitrogen in the pre-AVE TTL, where the more intense continental convection had a strong influence. This is not discussed, but based on Fig. 11, pre-AVE and CR-AVE look similar. Still, if the lifetime of NOx in this region is several days as indicated by Tie et al., perhaps Asia could be a source for CR-AVE. Could the authors please comment on this?

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 9399, 2009.