

Review of “Aerosol characteristics and particle production in the upper troposphere over the Amazon basin”

General comments

In this study characteristics of aerosol particles over the Amazon basin are investigated using aircraft measurements. The study focuses on the layers of enhanced particle concentrations observed in the upper troposphere. The particles in these layers were found to differ from particles in the lower troposphere with respect of their concentration, size, and chemical composition. Authors show that in most cases air masses with high particle concentrations have previously been in contact with deep convective outflow. Therefore, they suggest that particles are formed in the upper troposphere from precursors vapors brought up by deep convection.

The study is of good scientific quality and certainly worth publishing in the ACP after some minor revisions. First of all, when reading the manuscript, one gets an impression that this is the first time when the conceptual model with the production of particles in the upper troposphere from material brought up by deep convection and the transport of particles back to the boundary layer is suggested (e.g. P2, L52–60). However, as the authors discuss in Sections 1 and 3.7, this is not an entirely novel idea. Therefore, the authors should make it clearer, what is new in their conceptual model, and what has been suggested before. More specific comments are presented below.

Specific comments

P4, L113–115: The use of terms is slightly unclear here. The current convention is to use HOMs to generally refer to highly oxygenated organic compounds, while ELVOCs are only those HOMs that have extremely low volatility. In some earlier articles all HOMs were called ELVOCs but this is not preferable.

P18, L523: Are these values means for different flights?

P18, L535: It would be good if authors presented typical ratios between concentrations in the upper troposphere and lower troposphere for different size ranges.

P19, L546: The enhancement of accumulation mode particle concentration as well as high total particle concentrations would be easier to see if particle concentrations were plotted using a logarithmic scale (this also applies to some other plots).

P21, L626: Could higher concentrations of CCN compared to accumulation mode particles be also caused by underestimation of accumulation mode particle concentration due to high losses?

P22, L646–647: Why there is a peak in CCN fraction at ~11 km?

P23, L664: Should UFF be low (instead of high) when discussing these more aged particles?

P23, L668: In Fig. 13 there seems to be AC10-F instead of AC07-F.

P23, L683: For me it is not obvious where this region with high CCN concentrations is in Fig. 11b. In any case, this region could be mentioned already when discussing the vertical distribution of CCN.

P24, L717: It is told here that the average rBC concentration below 5 km is $0.31 \pm 0.29 \text{ g m}^{-3}$. It would be good to clarify what 0.29 g m^{-3} means here (and elsewhere in this section); is it an uncertainty for the average?

P31, L915: Please report how large the fraction of the cases where these air masses had encountered deep convection is. Also, would it be possible to perform more statistical analysis of the connection between enhanced particle concentrations and deep convection, for example by studying correlation between time since contact and particle concentration?

P31, L926: Why the flight AC19 was different?

P34, L1009: Please report the correlation coefficient obtained for N_{CN} and O_3 .

P35, L1019: Please report the correlation coefficient. Also, adding a plot of NO_y vs N_{CN} could be useful.

P37, L1078: Check the terminology as VOCs (volatile organic compounds) cannot have low/very low volatilities by definition. Moreover, if low volatile vapors are removed in the cloud outflow, how can there be enough low-volatile vapors to form particles?

P37, L1100 & P38, L1128 & P40, L1194: Instead of “ELVOCs/HOMs” I would suggest using only “HOMs”. See also the comment above.

P38, L115: Stating that pure organic nucleation is “much more likely” than nucleation including both organic and sulfuric acid appears to be a too strong statement, especially when the authors do not have data on the vapor concentrations. In the summary section, the authors also write that “we propose that BVOCs in the cloud outflow are rapidly oxidized to HOMs/ELVOCs, which because of the low temperatures and low condensation sink can readily nucleate new particles and grow to sizes ≥ 20 nm within a few hours”. I would suggest modifying this to something like “... oxidized to HOMs, which because of the low temperature and low condensation sink can form new particles, possibly together with sulfuric acid, and condense on particles growing them to sizes > 20 nm”

P39, L1160: The “Summary and conclusions” section is very long and partly seems to repeat some things discussed in the previous section. Therefore, I would suggest making the summary section shorter, especially the end of the section (starting from the line 1205). If needed, some of the text could also be moved to the previous section.

Technical corrections

P1, L38: Change “September/October” to “September–October”

P2, L47: Change “depleted in aerosol particles” to “depleted of aerosol particles”

P2, L49: Please change hyphen in “5-72” to en dash (–). This should be changed everywhere in the manuscript where ranges of numbers are shown.

P2, L56: Change “biogenic volatile organic carbon” to “biogenic volatile organic compounds”.

P3, L74: Change “are” to “they are”

P3, L81: Rephrase this sentence so that it does not begin with “where”.

P3, L82: Check the use of verb tenses in the whole manuscript. For example here “was” should be changed to “has been”.

P4, L109: Please rephrase the sentence.

P7, L212: Modify the reference to follow the journal’s guidelines.

P8, L244: Change to “The DMPS data *were* then analyzed *by* taking into...”

P10, L278: Change “on the S” to “on S”.

P10, L284: Change “by M. Pöhlker et al.” to “by Pöhlker et al.”

P15, L422: Please check that the reference style follows the journal’s guidelines.

P20, L591–593: The description of ultrafine fraction should be presented in a clearer way.

P21, L610: Remove “M.” and add this also to the reference list.

P23, L662: Please change “at one extreme are” to “at one extreme there are”. Also, change “at the other extreme are” to “at the other extreme there are”.

P23, L689: The description of volatile fraction is not clear here; it is not explained what N_{nonvol} stands for.

P25, L722. Change “June/July” to “June–July”

P26, L751: Please use subscripts for chemical compositions (e.g. SO_4 , NH_4 ...)

P26, L752: When using abbreviation “BB” for the first time, please write the whole word.

P31, L903: Change “can this be reversed” to “this can be reversed”

P34, L1000: Change “close” to for example “strong”

P34, L1015: Change “2056” to “20:56” etc.

P36, L1064: “Fig. 20” should be “Fig. 24”

P36, L1064: I would suggest using some other term than “classical nucleation events”, as a reader may confuse it with the classical nucleation theory. The term is used also elsewhere in the manuscript.

P37, 1090–1091: Rephrase the sentence “the low particle surface area in the UT presents very little competition to nucleation from a condensation sink”, as it is slightly unclear.

P40, L1171: Please make it clear that “UT aerosol was fundamentally different from the aerosol in the LT” is the result of this study. Also, I would suggest combining this and the previous paragraph.

Table 2: Please state in the table caption what the numbers reported in the table are: means with their uncertainty ranges?

Figure 1: It is difficult to see the difference between normal and “heavier” lines, so I would recommend using some other way to distinguish them.

Figures 2–4: As the manuscript includes so many figures, I would consider moving these figures (or at least some of them) to the supplementary material.

Figure 7b: In many of the figures (especially the lower panels) font size and line thickness/dot size should be increased.

Figure 10b: The values in the figure seem to be fractions, not percentage values as indicated by the figure label.

Figure 19a: There seems to be something wrong with the y-axis label.