Response to Reviewer 2

We thank the reviewer for his/her positive and constructive comments and for his/her thorough review. The reviewer comments are in plain font, the responses in *Italics.*

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.

In the introduction, we now write "…where production of new aerosol particles takes place in the UT from ***biogenic volatile organic material*** *brought up by deep convection…" to highlight the fact that our model is based on BVOC, whereas previous authors have mostly considered sulfur compounds or organics from pollution, including biomass burning. We have also added a paragraph to the introduction, making special reference to the work of Clarke and coworkers. See also our response to the first comment by Reviewer 3.*

In section 3.7, we refer extensively to previous work:

"The outflow regions in the UT present an ideal environment for particle nucleation, as had already been suggested in some earlier studies (Twohy et al., 2002; Lee et al., 2004; Kulmala et al., 2006; Weigelt et al., 2009)."

"Over marine regions and polluted continental regions, the particles observed in outflows and in the UT were mostly identified as sulfates (Clarke et al., 1999; Twohy et al., 2002; Kojima et al., 2004; Waddicor et al., 2012), and consequently H_2SO_4 has been proposed as the nucleating species."

We then go on to propose that, in contrast to these studies, organics may be the nucleating species, although a final proof still has to await our next campaign.

We also highlight the difference in the proposed mechanism of downward transport: "Large-scale entrainment of UT and MT air into the boundary layer has been suggested as the major source of new particles in marine regions (Raes, 1995; Katoshevski et al., 1999; Clarke et al., 2013). Over Amazonia with its high degree of convective activity, downdrafts are likely to play a more important role."

We never make the claim 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…". *In fact, the reviewer*

says so him/herself: "… as the authors discuss in Sections 1 and 3.7, this is not an entirely novel idea…".

Many more examples could be given. We feel that we have discussed previous work extensively in the introduction, in section 3.7, and in the conclusions. We find it difficult so see what more we could do to put our study in the context of previous work without repeating ourselves.

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.

In the community working on HOMs and ELVOCs there is currently no commonly accepted convention on terminology. Some authors suggest abandoning ELVOCs altogether and calling everything HOMs, while others are not using HOMs at all. In the Introduction we state "Extremely low volatility organic compounds (ELVOCs, which may be at least in part identical to HOMs)…". *In section 3.7 and the conclusions, we either use* "ELVOCs/HOMs*" or use the term that the authors of the papers use in the work that we are referencing.*

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

The values are meant to reflect the range of quartiles above 8 km. This has been clarified in the text.

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

There is an entire section devoted to this issue, section 3.4.1, which discusses the ratio between ultrafine and accumulation mode particles (expressed as ultrafine fraction, UFF). Averages for the particle concentrations in the different size classes are given in Table 2, to which we now refer to in the first paragraph of section 3.3 by "…, and average concentrations for the particle concentrations in the different size classes and altitude regions are given in Table 2*". We have also added the magnitude of the ratio in the text:* "On average, N_{CN} in the UT were almost five times as high as in the LT." *and* "On average, N_{acc} in the UT was only about half the concentration measured in the LT."

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).

We disagree. We started with log plots and switched to linear ones because they showed the differences much more clearly.

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?

The accumulation mode particles were measured by a UHSAS in a wing pod. There is no evidence for particle losses with this setup, which has been tested thoroughly, see also the paper by Walser et al. (2017) referenced in section 2.4.

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

The high values of the CCN fraction at this altitude are caused by the inclusion of a large number of measurements from flight AC20 on a horizontal leg at 11 km. This layer has only modest CN concentrations (around 1700 cm^{-3}), but elevated CCN, NO_y, CO, and aerosol nitrate and organics, with similar values to the biomass-burning-polluted boundary layer below. This flight was exceptional in that it was the only flight during the campaign on which we had evidence for transport of biomass smoke to the UT (see section 3.6). We included a short explanation and a forward reference to section 3.6 in the caption to Fig. 12a. We also added the following sentence in section 3.6: "Further evidence for the upward transport of biomass smoke was found in measurements on a horizontal leg at 11 km, which had only modest CN concentrations (around 1700 cm^{-3}), but elevated CCN, NO_y, CO, and aerosol nitrate and organics, with similar vales to the biomass-burning-polluted boundary layer below."

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

Corrected.

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

We corrected the label in Fig. 13.

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.

The concentrations in this region were not dramatically elevated, only up to about 1500 cm^{-3}. We changed the text to make this clearer. It would not have been appropriate to mention this region earlier, since it is specific to flight AC13, which is discussed in this paragraph as an illustrative example.

P24, L717: It is told here that the average rBC concentration below 5 km is 0.31±0.29 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?

Here, and everywhere else, we give averages and standard deviations, unless stated otherwise. To make this clear, we have added this definition of our notation in section 3.2.1, where it is used first.

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?

Actually, the fraction was 100%! The "almost" was left over from when we had not yet done the analysis for all cases, and has now been removed. We looked for such correlations, but could not find anything obvious. Unfortunately, since the mission objectives had been focused on aerosol/cloud-microphysics interactions, the flights were not designed to look into this issue. We plan to conduct a dedicated campaign in the future.

P31, L926: Why the flight AC19 was different?

Most of this flight took place outside of the Amazon basin, off the east coast of South America over the Atlantic.

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

Because of the great variability in the O_3 concentrations in the UT, there is no general correlation for the entire mission (r^2=0.02). For individual flights, modest but significant correlations emerge, which are still affected by the high variability of both variables. We added the following text:
"Because of the great variability in the O_3 concentrations in the UT, there is no general correlation between N_{CN} and O_3 for the entire mission (r^2=0.02). For individual flights, modest, but statistically significant, negative correlations can be found, e.g., an r^2 value of 0.13 (N=8509) in the UT on flight AC09. The scatter plot in Fig. S08 shows that high O_3 concentrations were always associated with low N_{CN}, but that there were low-O_3 regions in the UT both with and without enhanced particle concentrations."

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

Again, there is no significant overall correlation. As pointed out in the text, the relationships are very complex because the transformation of NO and the formation of particles both occur on short timescales that cannot be resolved by a general correlation analysis. In the paper, we provide some examples of these interactions, but a full analysis if the nitrogen oxide chemistry and its role in aerosol formation in the UT must await a dedicated mission.

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?

We replaced "VOCs" by "organic compounds". However, we remind the reviewer that the terms LVOCs and ELVOCs are very commonly used in the literature. The low-volatile vapors that form the new particles are produced by the oxidation of volatile vapors by photochemistry in the UT, as discussed in the subsequent paragraphs. For a better flow of the discussion we have moved the paragraph with this discussion up, to follow directly after the statement referred to by the reviewer.

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

We responded to this suggestion already 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”

We changed “much more likely” to “likely” and changed the sentence in the summary to include the possible role of H_2SO_4, as suggested by the reviewer.

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.

We disagree. This paper describes a very complex data set with a large range of information from many different instruments, from atmospheric transport models, and remote sensing. In the Summary and Conclusions we have tried to pull this information together in a concise way. The summary part must necessarily repeat, to a certain extent, things that have been said before. The end of the section is very important, since it puts the results into a “big picture” perspective.

Technical corrections

P1, L38: Change “September/October” to “September–October”
Done
P2, L47: Change “depleted in aerosol particles” to “depleted of aerosol particles”
Done
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.
Done
P2, L56: Change “biogenic volatile organic carbon” to “biogenic volatile organic compounds”.
Done
P3, L74: Change “are” to “they are”
Done
P3, L81: Rephrase this sentence so that it does not begin with “where”.
Done
P3, L82: Check the use of verb tenses in the whole manuscript. For example, here “was” should be changed to “has been”.

"was" is correct here.
P4, L109: Please rephrase the sentence.
Done
P7, L212: Modify the reference to follow the journal's guidelines.
This reference is a place holder and will be updated when the final files are prepared.
P8, L244: Change to "The DMPS data *were* then analyzed *by* taking into..."
Done
P10, L278: Change "on the S" to "on S".
Done
P10, L284: Change "by M. Pöhlker et al." to "by Pöhlker et al."
Since there are references to two different Pöhlkers as first authors, we use the initial to differentiate them. If the journal does not like this, the copyeditor is free to change it.
P15, L422: Please check that the reference style follows the journal's guidelines.
This can be checked by the copyeditor.
P20, L591–593: The description of ultrafine fraction should be presented in a clearer way.
We can't think of a clearer way. The definition equation is clear and unambiguous. If the editor disagrees, we would appreciate a suggestion for a better expression.
P21, L610: Remove "M." and add this also to the reference list.
Reference deleted.
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".
Done
P23, L689: The description of volatile fraction is not clear here; it is not explained what Nnonvol stands for.
Definition added.
P25, L722. Change "June/July" to "June–July"
Done
P26, L751: Please use subscripts for chemical compositions (e.g. SO4, NH4…)
These are in fact not chemical formulae, in which case they would have to be written also with the ionic charges, but abbreviations that are commonly used in the AMS literature.
P26, L752: When using abbreviation "BB" for the first time, please write the whole word.
Done
P31, L903: Change "can this be reversed" to "this can be reversed"
Retained as is. Can be changed by copyeditor if necessary.
P34, L1000: Change "close" to for example "strong"
"close correlation" is very common usage. We don't think "strong" would be better.
P34, L1015: Change "2056" to "20:56" etc.
This notation is very common in the meteorological literature.
P36, L1064: "Fig. 20" should be "Fig. 24"
Corrected
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.
We put "classical" in quotes to distinguish it from other usages.
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.

Done

P40, L1171: Please make it clear that “UT aerosol was fundamentally different from the aerosol in the LT” is the result of this study.

Done

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

Done

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.

We increased the thickness contrast between the lines.

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.

We disagree. This meteorological information is essential to understand the context of the mission.

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

The figures were somewhat preliminary. They will be updated for the final submitted files.

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

Percentages are one way to express a fraction.

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

Fixed.