Response to Reviewer 1 (H. Gordon)
The reviewer comments are in Arial, the responses in Times New Roman

Summary

This manuscript reports on new particle formation at high altitude in the Amazon region. I believe it is an important study and it will surely be highly cited. Addressing my "major" comments should not require substantial revisions to the manuscript.

We thank the reviewer for his/her positive statements and substantive comments and suggestions.

Major comments
Introduction

Given the relatively short length of the introduction the authors do an admirable job of reviewing the relevant literature. However, I think it is necessary to highlight a couple of key papers, which otherwise are a bit lost in the long lists of citations. I didn't read all the references, but from a random selection the Twohy (2002) and Weigel (2011) papers deserve a dedicated couple of summary sentences each somewhere in the introduction to compare them with the current work.

We have added the following sentences: "Twohy et al. (2002) observed particle concentrations up to 45,000 cm^{-3} over North America and suggested that they had been formed in situ from gas-phase precursors brought up by deep convection. Weigel et al. (2011) found similar concentrations in the UT over tropical America, Africa, and Australia, which they attributed to new particle formation from sulfuric acid and possibly organics." The Twohy et al. paper is cited three times in the introduction and four times in the discussion. The Weigel et al. paper is cited six times in the introduction and three times in the discussion. The results from both papers are compared to ours in the discussion.

Methods:

Section 2.10 outlines a sophisticated and valuable treatment of the back trajectories. Some minor clarifications on how the analysis was done, perhaps in the supplementary material, would be useful.

Specifics:

1. I think it may be helpful to show trajectories in longitude-altitude or (better) time-altitude space (e.g. for Figure S1). Would this shed any light on what the model is doing in areas of deep convection? The online HYSPLIT version gives these plots by default.

We have added a longitude-altitude plot to Figure S1. Like the vast majority of the UT trajectories, this one remains in the UT over the time frame considered. The trajectory model does not resolve individual convective elements, but only incorporates a general parameterization of vertical movement. See also the response to comment 3 below.

2. Please can the authors expand on the footnotes in Table 1? Are the maxima and minima that are given the maximum and minimum out of the five trajectories of the five

cluster centres they obtained from FLEXPART? Was the procedure explained in Figure S2 simply repeated for trajectories of each five possible cluster centres each time?

For simplicity, out of the five clusters, we consider only the center cluster given by FLEXPART. Therefore, the minima and maxima values of Table1 correspond only the values of center clusters trajectories within the flight leg time frame traced backwards up to 120 hours. This is now explained in the text. Doing the analysis for all five clusters would require an extraordinary amount of work and is not likely to give any other results, given the high abundance of deep convection in the basin. We have added the following sentence to the text: "For simplicity, out of the five clusters, we consider only the center cluster given by FLEXPART. Therefore, all trajectories mentioned hereafter refer to the center trajectory."

3. After the first contact with deep convection, (though not with the outflow of deep convection) presumably the five cluster centres diverge radically in horizontal and vertical positions as the air mass is vertically redistributed. Could the authors put the trajectories of the other four cluster centres on Figure S2 (or perhaps a copy of Figure S2, to help avoid confusion) as an example? Otherwise it is hard to see where the ranges in Table 1 for the time in gridboxes with deep convection are coming from. Ideally, it would be great to see how these clusters are transported in time-altitude space, as well. I note that Stohl et al (2002), where the clustering is introduced, does not report any validation of the algorithm in regions where deep convective clouds are present. Has this been done elsewhere? Are the five clusters really representative of the underlying distribution and does this affect the ranges for time spent in gridboxes with deep convection in Table 1? Given the huge vertical difference in winds (Figure 4 and line 449) one might speculate that the trajectories can be all over the place after contact with deep convection (though maybe not after contact only with an outflowing air mass).

The reviewer here points to a major problem with this and all other trajectory models. Fundamentally, they rely on the meteorological data from weather models which do not resolve individual convective elements. Convection is only represented in a parameterized way and therefore reflects the general vertical movement of an airmass, but not an individual parcel subject to a convective event. Thus, they cannot trace a parcel backwards through a convective event. The best they can do is show that a parcel came into the vicinity of a convective event, und thus was likely to be affected by the outflow. Coming close to a convective event does not make the parcels diverge, because the trajectory model actually does not see the event. Fundamentally, this is correct behavior, because the air in the outflow joins the general flow in the upper troposphere, and only those subparcels that actually came up through the cloud "should" have backtrajectories that go down through the cloud. Thus, if a back-tracked air parcel is not an outflow parcel, it should track backwards with the mean flow as represented by the model. It is thus legitimate to keep following it backward to perhaps encountering another region of convective outflow. The actual processes can only be resolved by a dedicated mission looking at the development of an individual outflow in a Lagrangian sense, which we hope to do in the future.

The authors do acknowledge this briefly (line 929) and it may not be very important if one contact with outflow is usually enough to produce NPF. However, I think these uncertainties merit a bit more discussion in the text, some kind of demonstration in a supplementary figure as I suggest above, and a brief comment in the caption of Table 1.

We've attempted to clarify this situation as concisely as possible by modifying the text at line 929 (old) by writing:
"Because the model does not "see" the individual convective event that brings up an outflow, it cannot trace a parcel back into this outflow and back down to the boundary layer. On the other hand, an air parcel that passed through the vicinity of the outflow, but is not part of the actual outflow, will keep moving backward along the mean flow in the UT and may then encounter another outflow. Obviously, however, the uncertainty in the trajectory position increases with time going backwards, and is probably enhanced by passage near a region of active convection."
Given that our analysis shows that, in view of the frequency of convection over Amazonia and the generally long residence time of air parcels in the anticyclonic movement over the basin, almost all air parcels will pass near convection over a 72-hour time frame, it does not seem worthwhile to go much further in this analysis. See also our comment below in our response to remark 1 in the results section.

4. The 10-14km altitude range (e.g. line 463) seems quite high compared to many of the NPF bursts observed -one of the examples is at 7km. Some words on what happens at slightly lower altitudes would be useful, if this can be provided without huge extra effort.

Actually, the statement in line 463 was incorrect and, as can be seen in Table 1, the analysis was done for all enriched layers, including those at 7 km.

5. Is there a dependence of the NPF characteristics on trajectory type (A-E in Figure 1)?

We could not identify any obvious relationship.

Is it possible to draw general conclusions in addition to the discussion of specific flights and the statement that only a few daylight hours are needed for the NPF, in Section 3.5?

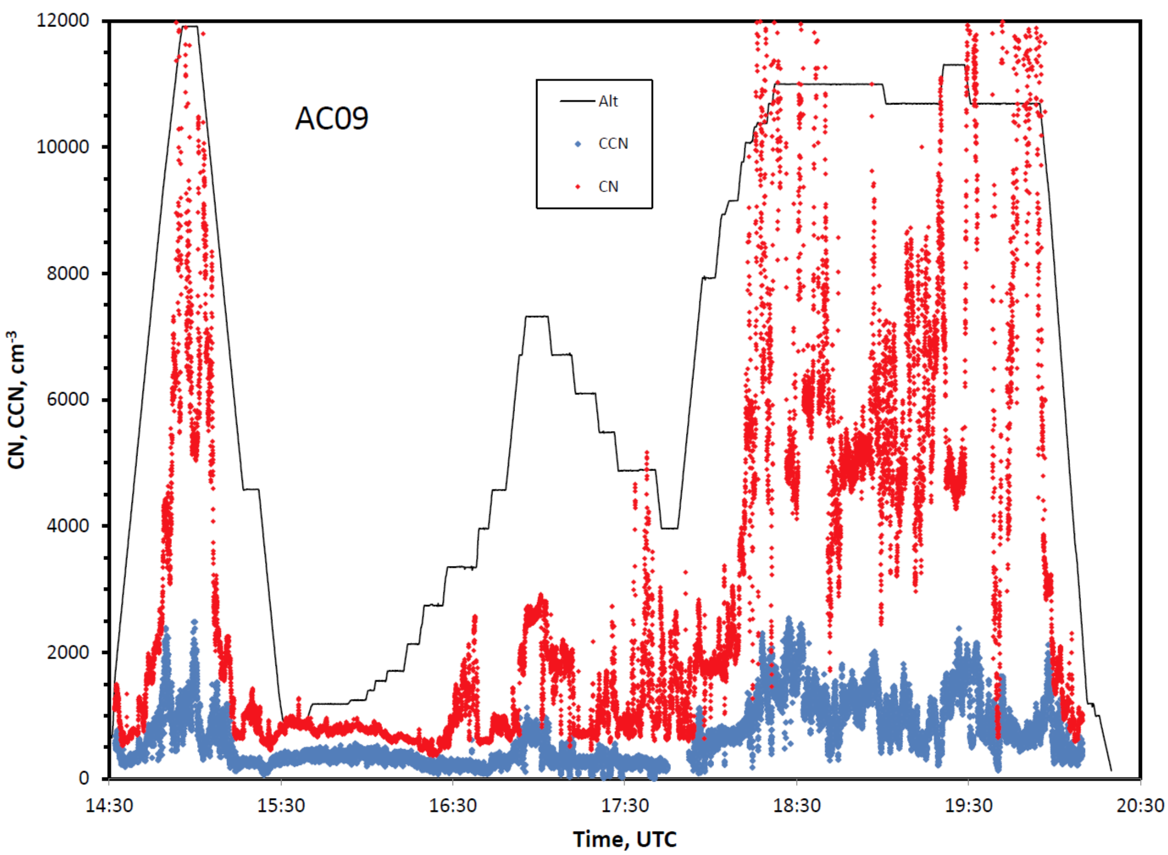
We don't feel that we can draw further generalizations based on the kind of data we have from this mission. To go further, different flight strategies and instrumentation would be required, which we plan to deploy on a future mission.

Results:
1. I'm reluctant to suggest additions to an already long and comprehensive study. However, I do feel information is lacking on the air masses in the UT in which particle concentrations were low (except for the immediate cloud outflow region, which is already described). Clearly from Fig. 7a quite a few segments with fewer than 2000 particles/cm3 were seen. At line 661 the authors could remind readers of this by changing "two distinct aerosol populations" as "two types of elevated aerosol population".

Done.

If studying the air masses with very low particle concentration shows significant differences in their interaction with deep convection compared to the air masses with high particle concentrations, the authors' conceptual model may become more powerful: it may be possible to suggest contact with deep convection is a necessary condition for particle production in these situations. If no significant differences are found, this would also be interesting, though it would certainly not invalidate the conceptual model, as there are many possible explanations for the absence of NPF. Thus, could the authors consider either adding another (shorter!) Table 1, where at least some of the flight legs where aerosol concentrations in the UT were below 2000 cm-3 are listed?

We felt this was a very valuable suggestion by the reviewer and examined our data for such legs. To our disappointment it was almost impossible to find such segments. Because of the high variability of the CN concentrations in the UT, the times when N_{CN} was below 2000 cm^{-3} were in almost all cases very short, and would not lend themselves to a meaningful analysis of airmass history. To illustrate this, we show a full time series plot of the measurements from Flight AC09 in the supplement:



The only exception to this were segments that were within a Cb outflow.

We were able to find only six segments, where N_{CN} was consistently below 3000 cm^{-3}, and which were not identifiably part of an outflow. These are listed in Table S1 in the supplement. The segments from flights AC16 and AC18 were well away from clouds, whereas those from AC19 and 20 were in the vicinity of Cbs, but not clearly in an outflow. The segment L from AC19 is low in CN, but actually has a relatively high $N_{CCN0.5}$, and may not really be significantly different from the aged enriched segment E2, which follows immediately after it. The airmass trajectory types in these segments do not contain type D, i.e., recirculation within the Amazon basin. Notably, the air in the segments from AC20, which had the lowest particle concentrations, had come in straight from the Pacific within the last 48 hours. We added the following text to section 3.5.2:

"To test whether there was a difference in the airmass histories between segments with high and low N_{CN}, we searched our data for suitable segments with low N_{CN}. However, because of the high variability of the CN concentrations in the UT, the times when N_{CN} was below 3000 cm^{-3} were in almost all cases very short, and would not lend themselves to a meaningful analysis of airmass history. To illustrate this, we show a full time series plot of the measurements from Flight AC09 in the supplement (Fig. S7).

We could find only six segments, where N_{CN} was consistently below 3000 cm^{-3}, and which were not identifiably part of an outflow. These are listed in Table S1 in the supplement. The segments from flights AC16 and AC18 were well away from clouds, whereas those from AC19 and 20 were in the vicinity of Cbs, but not clearly in an outflow. The segment L from AC19 is low in CN, but actually has a relatively high $N_{CCN0.5}$, and may not really be significantly different from the aged enriched segment E2, which follows immediately after it. Consequently, we don't have a data set that would allow a representative analysis of the history of airmasses with low particle concentrations. Notably, however, the airmass trajectory types in these segments do not contain type D, i.e., recirculation within the Amazon basin. The air in the segments from AC20, which had the lowest particle concentrations, had come in straight from the Pacific within the last 48 hours, but may also contain some outflow air."

Is there any systematic difference in the timings at which the air masses with few particles first made contact with deep convection, and at which the air masses with many particles made contact? I appreciate that the authors may prefer to leave this for further work if the analysis has not already been done.

Again, we feel that in view of the complexity of the airmass histories, dedicated campaigns are needed to resolve this question.

2. From Figure 5, the relative humidity at 7-10km altitude is very low – apparently unusually low (line 414 ish). It may be interesting to look for evidence of the RH enhancing or suppressing the particle number concentrations- if there is any effect of RH visible, this might suggest that the new particle formation is not at the kinetic limit for the vapours involved (or that water is important for the chemistry leading to the NPF). However, again I appreciate that this kind of investigation may be more appropriate for future studies with instrumentation better able to measure organic gasphase chemistry.

The discussion in line 414ff (old) refers to the column moisture content and precipitable water, not to the relative humidity in the upper troposphere. However, to follow up on the reviewer's suggestion, we examined several flights (AC07, AC09, AC13, and AC18) for relationships between RH and N_{CN}. We found a tendency for the layers with high N_{CN} to be associated with moister layers (RH>50%), but also many exceptions. This relationship may simply have to do with the fact that moisture was brought up with the convective clouds, or there may be a relationship with the actual particle formation process, but at this point we have no way to answer these questions. We added a couple of sentences on this in section 3.5.3. We are planning a future campaign dedicated to process-level studies of NPF in the UT.

3. Related to comment #1, can the authors suggest some possible explanations for why the areas of extremely high particle concentration (suggestive of very recent new particle formation) are usually organised in thin layers?

The outflow from convective clouds tends to become stretched into relatively thin layers due to velocity shear and subsidence, especially when transported over considerable distances (for a discussion, see Eastham and Jacob, 2017, and references therein).

Conceptual model:
In general, I find the arguments in this section compelling and I have only minor comments, see below.
Conclusions:
At lines 1230-1238, the authors point out that in pre-industrial times, the mechanism they propose would operate unchanged, while sources of low-altitude particles would be diminished, meaning that upper-troposphere new particle formation may in some cases become the dominant source of CCN in the boundary layer. They further propose that the aerosol profile in polluted continental regions may be flipped in the pre-industrial compared to the present day.
The authors do make it clear that these statements are speculative, and I appreciate the need to be concise. However, at lines 1223-1224 I think they should additionally point out that the pre-industrial atmosphere may not have been particularly pristine in many places, with large marine, volcanic and fire emissions leading to uncertain but possibly high concentrations of boundary layer particles. It would be enough to modify “strongly affected by anthropogenic aerosols” to “strongly affected by anthropogenic or natural primary aerosols”.

Done.

Furthermore, to justify the arguments in the paragraph “The conceptual model proposed here implies…” the authors need to show evidence that in present-day *polluted* areas, concentrations of particles greater than say 3nm in diameter are usually lower at high altitude than they are at low altitude. A very brief look at flight data from INTEX over the eastern USA suggested to me that there is still plenty of particle production in the upper troposphere in polluted regions (in these areas, of course there are more particles in the BL, but also more SO_2 making particles in the UT). There is a modification to the gradient of the aerosol profile over the industrial period (modelling studies suggest this is true even as a global average, see for example Fig. 1a of http://onlinelibrary.wiley.com/doi/10.1002/2017JD026844/abstract) but to say “turned upside down” seems a bit strong.

A climatology of aerosol concentrations in the UT is available from the CARIBIC project. This shows median particle concentrations (> 12 nm) in the region 200-300 hPa to be ~3500 cm^{-3} over North America, ~2500 cm^{-3} over Europe, and ~3000 cm^{-3} over India (Ekman et al., 2012). Of course, there are elevated values at particular place and times, such as those the reviewer refers to, but they appear to be more the exception than the rule. In contrast, the averages measured at ground level at polluted continental sites worldwide range between 3400 and 19,000 cm^{-3} in the compilation by Andreae (2009). This is quite close to being the exact opposite of the distribution measured during ACRIDICON-CHUVA, where the averages (±std.dev.) were 7700±7970 cm^{-3} in the UT and 1650±980 cm^{-3} in the LT. This information has been added into the Conclusions text. But, so as not to over-generalize, we have modified the statement to “… has been turned upside down, at least in many polluted regions”.

Minor comments

The text is well written and logically structured, but as it is long, the introduction of more cross-referencing between sections to relate different parts of the text together would be very helpful. For example, it would be helpful to reference Figures 4 and 6 at the appropriate places in the paragraph starting on line 471.

Done.

Also at line 662 it would be helpful to remind the reader that the two aerosol populations were already introduced at line 547, to confirm the distinctions drawn are the same in the two cases.

Done.

Structurally, the one concern I have is that Section 3.4 and Section 3.5 start with essentially the same question, then Section 3.4 deals with one part of it and then 3.5 introduces another possible source (immediate outflows) and most of the section is then spent dealing with this new issue that was not previously introduced. Can the authors think about whether it is possible to organise these sections more rigidly and flag up the most important messages more strongly?

We have added some introductory sentences at the beginning of section 3.4 that inform the reader what to expect in sections 3.4 and 3.5.

The discussion of the trajectory results (3.5.2,3.5.3) probably merits a new section 3.6.

We prefer to retain the current structure, as we think it is appropriate to the discussion.

Line 93: the authors might cite here only the papers which really focus on UT NPF: the Carslaw (2017) citation seems out of place in this paragraph.

The reference has been deleted.

Line 197 or 218: please state approximate distance between inlet and instrument, to put these flow rates and efficiencies in context. Also for the UHSAS and CCNC.

The length of the line to the CPC was about 2 m, to the CCN about 1.8 m. The flow in the inlets was increased by using a variable flow bypass to reduce particle losses. The UHSAS is mounted in a wing-pod and has no inlet line.

The authors convincingly demonstrate NPF is the only possible source of the particles. However, they should emphasise the sentence at line 843-845 more, where the key reason for why the particles cannot come from long range transport is explained (even though it is fairly obvious). This could be done by forward referencing Section 3.5 from line 553, or restructuring slightly as suggested above.

Done, by the new introductory sentences at the beginning of section 3.4.

Line 806: please label the citation to Schulz as ‘submitted’, or ‘in preparation’, here. I couldn’t find the paper.

Done.

Line 1087 The authors should specify that the CERN CLOUD chamber studies so far published only provide the temperature dependence of inorganic NPF. NPF involving organic molecules may behave quite differently, though NPF is still obviously expected to increase at lower temperatures (all other things being equal). Similarly, the Yu (2017) study does not fully account for the gas-phase chemistry (as this chemistry is not fully characterised the authors had little choice), so it treats NPF of organics rather similarly to that for H2SO4.

Cautionary sentence added: “Note, however, that these temperature dependencies are based on measurements for inorganic NPF, and that while the trends for organics are expected to be similar, the magnitude of the increase in nucleation rates for organics may be quite different.”

Line 1123 The Gordon (2016) modelling study didn’t quite suggest “dominant mode of new particle formation in the pre-industrial atmosphere”, perhaps replace by “in large parts of the preindustrial atmosphere”.

Done.

On page 68, the footnote labels to Table 1 all read “a”.

Corrected.

Fig S1 caption: aren’t the parcels zoomed in approximately a 6x6 degree box, not 3x3? Despite the valuable efforts of the authors to make things clear with the colour scale of the trajectories and marking the GOES time on the figure, I found the way this was phrased in the caption a little confusing. If I understand, the snapshots are zoomed in a box centred at the parcel location at the time shown on the top ***of*** the snapshots, ***in parentheses backwards from the parcel start***. Perhaps the authors could add something like the italicised words/phrases to the caption?

The reviewer must be referring to Figure S2 (not S1). Yes, the boxes are 6x6 degrees and we corrected that in the caption. We added the wording on the number of hours in parentheses.

Andreae, M. O., Correlation between cloud condensation nuclei concentration and aerosol optical thickness in remote and polluted regions: Atmos. Chem. Phys., 9, 543–556, 2009.

Eastham, S. D., and Jacob, D. J., Limits on the ability of global Eulerian models to resolve intercontinental transport of chemical plumes: Atmos. Chem. Phys., 17, 2543-2553, doi:10.5194/acp-17-2543-2017, 2017.

Ekman, A. M. L., Hermann, M., Gross, P., Heintzenberg, J., Kim, D., and Wang, C., Sub-micrometer aerosol particles in the upper troposphere/lowermost stratosphere as measured by CARIBIC and modeled using the MIT-CAM3 global climate model: J. Geophys. Res., 117, D11202, doi:10.1029/2011jd016777, 2012.

Twohy, C. H., Clement, C. F., Gandrud, B. W., Weinheimer, A. J., Campos, T. L., Baumgardner, D., Brune, W. H., Faloona, I., Sachse, G. W., Vay, S. A., and Tan, D., Deep convection as a source of new particles in the midlatitude upper troposphere: J. Geophys. Res., 107, 4560, doi:10.1029/2001JD000323, 2002.

Weigel, R., Borrmann, S., Kazil, J., Minikin, A., Stohl, A., Wilson, J. C., Reeves, J. M., Kunkel, D., de Reus, M., Frey, W., Lovejoy, E. R., Volk, C. M., Viciani, S., D'Amato, F., Schiller, C., Peter, T., Schlager, H., Cairo, F., Law, K. S., Shur, G. N., Belyaev, G. V., and Curtius, J., In situ observations of new particle formation in the tropical upper troposphere: the role of clouds and the nucleation mechanism: Atmos. Chem. Phys., 11, 9983-10,010, doi:10.5194/acp-11-9983-2011, 2011.