

Review of Aerosol characteristics and particle production in the upper troposphere over the Amazon Basin

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.

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.

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.
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?
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 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.

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

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”.

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 2000cm⁻³ are listed? 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.

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 gas-phase chemistry.

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?

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”.

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 SO₂ 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.

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. 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.

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

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.

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 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.

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

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.

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 pre-industrial atmosphere".

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

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?

Checklist

1. Does the paper address relevant scientific questions within the scope of ACP?

Yes

2. Does the paper present novel concepts, ideas, tools, or data?

Yes

3. Are substantial conclusions reached?

Yes

4. Are the scientific methods and assumptions valid and clearly outlined?

Yes

5. Are the results sufficient to support the interpretations and conclusions?

Yes, if a couple of sentences in the conclusions can be toned down slightly.

6. Is the description of experiments and calculations sufficiently complete and precise to allow their reproduction by fellow scientists (traceability of results)?

Yes

7. Do the authors give proper credit to related work and clearly indicate their own new/original contribution?

Yes

8. Does the title clearly reflect the contents of the paper?

Yes

9. Does the abstract provide a concise and complete summary?

Yes

10. Is the overall presentation well structured and clear?

Yes

11. Is the language fluent and precise?

Yes

12. Are mathematical formulae, symbols, abbreviations, and units correctly defined and used?

Yes

13. Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated?

No (or no substantial part of the text)

14. Are the number and quality of references appropriate?

Yes

15. Is the amount and quality of supplementary material appropriate?

Yes.