Responses to the review of the manuscript "Occurrence and growth of sub-50 nm aerosol particles in the Amazonian boundary layer", by Franco et al., submitted for publication in ACP - manuscript ACPD 2021-765

Dear editor, we would like to thank you and both reviewers for their valuable comments and useful suggestions to improve our manuscript. Below you can find answers and actions for each individual comment from the reviewers. In order to make it easier to identify the individual answers and actions, we used the following color code strategy:

- **In black, the referee's comments.**
- **In blue, the author's responses.**
- ***In blue and italic, the text modifications we made in the manuscript.***

Responses to Anonymous Referee #2

Article "Occurrence and growth of sub-50 nm aerosol particles in the Amazonian boundary layer", by Franco et al., ACPD 2021-765

Referee comment on "Occurrence and growth of sub-50 nm aerosol particles in the Amazonian boundary layer" by Marco A. Franco et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-765-RC2, 2021

This manuscript investigates the origin and growth of sub-50 nm particles over Amazonia. The presented analysis relies on several years of measurements in the boundary layer, extending earlier more campaign-based results on this topic. The paper is scientifically sound and, in most parts, very well written. I have a few, mostly minor issues to be considered further. After addressing these issues, the paper is, in my opinion, acceptable for publication in ACP.

We would like to thank referee #2 for the constructive comments and useful suggestions. They were helpful for clarifying important aspects of our analysis and discussion on the results.

The authors should provide some reasoning for selecting the periods G1 to G2 when looking at different times of the day. At first sight, it seems like this choice only covers 4 roughly equally long periods covering both nighttime (G1 and G4) and daytime (G2, G3). Furthermore, it seems evident that many of the differences observed between these 4 groups are simply the results of typical diurnal development of the boundary layer and its interactions with the rest of the troposphere. This feature should be explicitly brought up when discussing the results.

The choice of periods G1 to G4 was made due to particularities in the frequency of occurrence of growth events, as seen in Figure 7e. We have divided daytime events into two parts: those that occur in the morning, between 6:00 and 12:00, and those that occur in the afternoon. Morning events are those that are influenced by the evolution of the mixing layer, are mainly driven by photochemistry, and are the ones that occur in greater intensity. Afternoon events are mainly dominated by convective events. The nocturnal events were divided according to the evolution stage of the PBL. Events between 18:00 and 00:59 would be those that are still influenced by some kind of turbulence reminiscent of the mixing layer, but which may also still have had some influence from convective events. The growth events between 01:00 and 05:59 would be the most curious and they are not directly driven by photochemistry or variations in the PBL, since at this time the nocturnal PBL would already be well established. These events, in particular, occur under clear sky conditions and are the ones that raise the most questions about their origins.

In order to make the choice of periods explicit in the results sections, we have modified the text, which now reads as:

To further investigate typical conditions or processes related to the observed growth events, we here discriminate between different groups based on their daily frequency distribution. The growth events (see Figure 7f) were divided into four groups (G1 to G4), where G1 and G4 represent nighttime and G2 and G3 represent daytime events. The daytime events were divided considering the occurrence of two frequency peaks: the first peak, representing 53 % of the growth events, is included in G2, covering the time from 06:00 to 11:59 LT. The afternoon increase, representing 21 % of the growth events, is included in G3, covering the time from 12:00 to 17:59 LT. The nocturnal events were divided according to the evolution stage of the PBL. Events between 18:00 and 00:59 (G4), with 10 % of the events are still influenced by some PBL turbulence, but may also have had some influence from convective events in the late afternoon. The growth events between 01:00 and 05:59 (G1), with 16 % of the total growth events are the most enigmatic ones. They are not directly driven by photochemistry or variations in the PBL, since at this time the nocturnal PBL is already well established. Different mechanisms such as air mass entrainment into the PBL by, e.g., intermittent turbulence (Dias-Júnior et al., 2017), nighttime rainfall events, or even an unknown biogenic source could play a role in the aerosol particle dynamics of in this time period.

As a final note, was there some reason for having one hour (00:00-01:00) that was not included in any of the groups G1-G4?

The missing one hour was a typo, which we corrected in Table 2 of the revised manuscript. The correct label is 18:00 - 00:59.

Minor comments

While NPF may occur in both local and regional scales, occurrence of NPF leading to observable growth requires usually NPF taking place over relatively large spatial scales. Therefore, it somewhat misleading to claim that occurrence of NPF depends strongly on local conditions of individual sites (page 3, lines 3-4). Therefore, rather than emphasizing just local conditions, it should be noted that conditions (both emissions and meteorological conditions) over regional/synoptic scales are important in this respect.

Thanks for pointing that out. Indeed, NPF with the observable growth occurs at relatively large spatial scales and it should be more clear in the manuscript. Thus, we improved the text on P3, l 3-4 of the revised manuscript, which now reads as:

The occurrence of NPF is dependent on the local conditions at individual sites, including meteorology, biogenic emissions, and air pollution levels, but regional and synoptic scales are also very important for this process. Particle growth events lasting on the order of hours are particularly influenced by larger geographic scales.

Page 26, line 9: "early afternoon" does not match with the time period 06:00-11:00.

Thanks. We improved the text, which now reads as:

During daytime, 53% of the growth events start in the morning between 06:00 and 11:00 00 LT, with a pronounced peak at 07:00 LT, showing a relation to the photochemistry and links with the evolution of the PBL.

Page 26, lines 11-12: The author mention coagulation as an explanation for the observed decrease in N<50 and state that coagulation results in the growth of these particles. It is a relatively simple procedure to estimate the approximate growth rate of a mode of particles due to self-coagulation when knowing the particle number concentration in this mode. I recommend the authors to make this exercise. To me, it seems that for the typical values of N>50, coagulation can explain only a minor fraction of the observed growth of sub-50 nm particles, suggesting that this growth is mainly

due to condensation or other gas-to-particle conversion processes.

We agree with the reviewer, indeed the condensation plays a major role in the process, followed by a minor contribution of the coagulation process. Thus, we clarified the text, which now reads as follows:

The subsequent decrease of $N_{<50}$ is likely due to condensation of semi- and less-volatile organic species on the sub-50 nm particles, resulting in growth.

References:

Dias-Júnior, C. Q., Sá, L. D., Marques Filho, E. P., Santana, R. A., Mauder, M., and Manzi, A. O.: Turbulence regimes in the stable boundary layer above and within the Amazon forest, Agricultural and Forest Meteorology, 233, 122–132, https://doi.org/https://doi.org/10.1016/j.agrformet.2016.11.001, https://www.sciencedirect.com/science/article/pii/S0168192316304257, 2017.