# Response to Anonymous Referee

**Article: "Amazonian Aerosol Size Distributions in a Lognormal Phase Space: Characteristics and Trajectories", by Gabriela R. Unfer et al., Egusphere 2023-1361.**

Dear Editor,

The authors thank both reviewers for their helpful comments and suggestions. Our responses to each comment are developed hereafter, along with an indication of changes made in the revised version of the text. As a summary, the revisions to the manuscript include the following highlights:

- All plots of the Results section have a version in 2D as scatter plots of N versus $D_g$ with sigma in a color scale presented in the Supplement.
- A table containing the summary of all fit parameters is presented in the Supplement.
- A more detailed description of the code-fitting methodology and a discussion about the separated analysis of the modes sub-50 nm and 50-100 nm (Aitken).
- The text was improved in readability and the abstract was rewritten.
- New references were added to improve the discussions.

The individual reviewer comments and responses are included in the following document, where reviewer comments are presented in **bold** and the author comments in *italics*.

Sincerely,

Gabriela R. Unfer, on behalf of all co-authors

**Reviewer 2:**

**Main comments:**

**This paper presents analysis of long-term aerosol particle number size distribution (PNSD) data from the ATTO tower through characterising the PNSD in a three-dimensional phase space represented by 1) the geometric mean diameter; 2) the geometric standard deviation; 3) the number concentration of lognormal modes fitted to the PNSD. The manuscript deals with an important topic: finding new ways to analyse the emerging long-term data sets of aerosol particle characteristics, which are critical for enhancing our understanding on e.g. aerosol-cloud-climate interactions, and is therefore approapriate for the scope of ACP. I think the approach for investigating trends and behavior of the PNSD as a function of various environmental parameters is interesting and novel, although the exact value for potential applications is to be demonstrated in future studies. The manuscript therefore has scientific value that is of potential interest for the readership of ACP. There are, however, some aspects of the manuscript that should be improved before publication can be recommended - related to both the scientific approach and the presentation quality:**

*Dear Reviewer, we would like to thank you for your comments and suggestions; they were significant in improving and clarifying some essential aspects of the manuscript content. In the Editor's letter, we explain the main changes in the manuscript, and below, we listed these aspects related to your recommendations.*

**General:**

**- The reference list of the manuscript is rather limited with a high proportion of work from the authors themselves. I understand that when it comes to PNSDs in the tropical atmosphere this is hard to avoid, but perhaps there is previous work on PNSD characteristics from other parts of the world worth mentioning here? Please consider also adding some discussion and comparisons to past work to the Discussion and conclusions section.**

*As recommended, we increased the diversity of the cited works and included comparison discussions on the text. Please find below the new references added.*

Boucher, O: Atmospheric Aerosols: Properties and Climate Impacts, 1, Springer Dordrecht, https://doi.org/10.1007/978-94-017-9649-1, 2015.

Dada, L., Paasonen, P., Nieminen, T., Buenrostro Mazon, S., Kontkanen, J., Peräkylä, O., Lehtipalo, K., Hussein, T., Petäjä, T., Kerminen, V.-M., Bäck, J., and Kulmala, M.: Long-term analysis of clear-sky new particle formation events and nonevents in Hyytiälä, Atmos. Chem. Phys., 17, 6227–6241, https://doi.org/10.5194/acp-17-6227-2017, 2017.

Khadir, T., Riipinen, I., Talvinen, S., Heslin-Rees, D., Pöhlker, C., Rizzo, L., Machado, L. A. T., Franco, M. A., Kremper, L. A., Artaxo, P., Petäjä, T., Kulmala, M., Tunved, P., Ekman, A. M. L., Krejci, R., and Virtanen, A.: Sink, Source or Something In-Between? Net Effects of Precipitation on Aerosol Particle Populations, Geophys. Res. Lett., 50(19). https://doi.org/10.1029/2023GL104325, 2023.

Kulmala, M., Petäjä, T., Nieminen, T., Sipilä, M, Manninen, H. E., Lehtipalo, K., Dal Maso, M., Aalto, P. P., Junninen, H., Paasonen, P., Riipinen, I., Lehtinen, K., Laasonen, A., and Kerminen, V-M.: Measurement of the nucleation of atmospheric aerosol particles. Nature Protocols, 7, 1651–1667, https://doi.org/10.1038/nprot.2012.091, 2012.

Mäkelä, J. M., Koponen, I. K., Aalto, P., Kulmala. M.: One-year data of submicron size modes of tropospheric background aerosol in Southern Finland, Journal of Aerosol Science, 31, 595-611, https://doi.org/10.1016/S0021-8502(99)00545-5, 2000.

Sogacheva, L., Saukkonen, L., Nilsson, E. D., Dal Maso, M., Schultz, D. M., De Leeuw, G., and Kulmala, M.: New aerosol particle formation in different synoptic situations at Hyytiälä, Southern Finland, Tellus B: Chemical and Physical Meteorology, 60:4, 485-494, https://doi.org/10.1111/j.1600-0889.2008.00364.x, 2008.

Tunved, P., Hansson, H.-C., Kulmala, M., Aalto, P., Viisanen, Y., Karlsson, H., Kristensson, A., Swietlicki, E., Dal Maso, M., Ström, J., and Komppula, M.: One year boundary layer aerosol size distribution data from five nordic background stations, Atmos. Chem. Phys., 3, 2183–2205, https://doi.org/10.5194/acp-3-2183-2003, 2003.

Wang, J., Krejci, R., Giangrande, S., Kuang, C., Barbosa, H. M. J., Brito, J., Carbone, S., Chi, X., Comstock, J., Ditas, F., Lavric, J., Manninen, H. E., Mei, F., Moran-Zuloaga, D., Pöhlker, C., Pöhlker, M. L., Saturno, J., Schmid, B., Souza, R. A. F., Springston, S. R., Tomlinson, J. M., Toto, T., Walter, D., Wimmer, D., Smith, J. N., Kulmala, M., Machado, L. A. T., Artaxo, P., Andreae, M. O., Petäjä, T., and Martin, S. T.: Amazon boundary layer aerosol concentration sustained by vertical transport during rainfall, Nature, 539(7629), 416–419. https://doi.org/10.1038/nature19819, 2016.

**- The manuscript needs to be improved for readability and precise use of terms and language. Some examples are provided in the specific comments below, but generally e.g. references to analysis or figures that are only to be presented later in the manuscript should be kept to minimum, and concepts and terms should be referred to as precisely as possible.**

*Based on your suggestions, the full text was improved for readability, and the mention of later analysis/figures was minimized.*

**- It would be interesting if the authors could, for example at the end of the introduction section, reflect on the potential research questions that could be answered using the**

**approach presented here - beyond following temporal patterns and responses to precipitation events. What larger-scale implications might the results presented here have?**

*We envision that the analysis of aerosol population in the lognormal phase space can be helpful, for example, in understanding the distribution of particles under different synoptic systems or even interannual variabilities, like in El Nino/La Nina. It is also possible to compare different global warming scenarios regarding aerosol distributions. One could analyze the phase space for different geographic regions by plotting the different aerosol populations and analyzing how they cluster in the phase space. Another possibility is plotting particle growth for different new particle formation events and possibly extracting the parameterizations.*

*This discussion has been added to the introduction section*

**- My main potentially scientifically major comment has to do with the fact that it appears from Fig. 2 and the text that the division between the "sub-50 nm" and "Aitken" modes was somewhat arbitrary - if I understand correctly, a predefined size-cut at 50 nm was simply used instead of letting the fitting algorithm find the best number of modes and their parameters. This constraint makes it difficult to use the fitted "sub-50" and "Aitken" mode bevior for analysis of the underlying microphysics and chemistry - because it is not clear whether one can justify the choice of the modes in terms of them representing clearly different aerosol populations. Could the authors please clarify their methodological choices in this regard and reflect on the potential implications for the interpretation and usefulness of the presented results?**

*In Central Amazon, where our measurements were taken, the maximum number of modes is three, as presented and discussed in Franco et al. (2022). In our study, three modes were not necessarily always fitted; the code was free to decide between one and three. Regarding the diameter ranges, the study of Franco et al. (2022) and other studies like Machado et al. (2021) have already shown that in the ATTO region, the size ranges of 10 to 50 nm, 50 to 100 nm, and 100 to 400 nm are representatives of the aerosol modes and that they present different behaviors.*

*Specifically, the separation of the literature Aitken mode into two (sub-50 nm and 50 to 100 nm) did not affect the behavior of our Aitken mode itself since the concentration of the sub-50 nm mode is low compared to the other modes. Furthermore, it was beneficial since we could see clearly from the analyses that the sub-50 nm mode presented distinct results from the Aitken mode. Machado et al. (2021) already had shown that this mode presents an increase in concentration after rainfall events, in contrast with the Aitken (50-100 nm) and accumulation modes, which present a decrease in concentration. The segregation of the Aitken mode into two brings rich information about the formation of new particles in the Amazon.*

*We improved the discussion of the modes fitting and modes selections in the methodology section to clarify the lognormal fit. In addition, the typical dry and wet season distributions are in the Supplement (Fig. S1), which corroborates our methodology.*

**Specific comments:**

**- Abstract, p. 1, e.g. lines 23-24 and 25-26: The abstract should be stand-alone and understandable without having to read the manuscript in detail. It is very difficult to understand e.g. what it means that "the sub-50 nm mode appears as a curved cone, the Aitken mode as a semi-sphere, and the accumulation mode as a cylinder" without looking at the plots in the manuscript. Also what does a "positive linear slope" mean in "The diurnal cycle of sub-50 nm particles in the dry season shows a positive linear slope as a function of all three fit parameters." - i.e. which variable has a positive linear slope as a function of the three fit parameters? Do you perhaps mean that all three fit parameters have a positive linear slope as a function of time? Please revise the abstract for readability through e.g. defining which modes where fitted to the data, accurate definition of parameters and clearly highlighting the key conclusions that can be summarized without having to read the entire mansucript.**

*The abstract was rewritten and improved. Regarding the positive linear slope, we meant the change of every parameter with respect to time. But for clarity, we changed it to just "a linear cycle", since it depends on the starting point of the time to determine whether it is positive or negative, but the trajectory is linear overall.*

**- p. 2, lines 46-48: PLease revise the sentence starting with "Improving aerosols parameterizations..." for clarity, English language and readability.**

*The sentence was rephrased.*

**- p. 2, line 64 and p. 3, line 66: What do you refer to with the term "comparatively" - as compared with what? Please specify if possible.**

*The term refers to the comparison between the concentration in each of the seasons to the typical concentration in the Amazon and also worldwide. This clarified sentence was added to the text.*

**- p 3, lines 89-91: Perhaps it is appropriate also to refer to the study by Hussein et al. from 2005 (Hussein, T., Dal Maso, M., Petäjä, T., Koponen, I. K., Paatero, P., Aalto, P. P., Hämeri, K., & Kulmala, M. (2005). Evaluation of an automatic algorithm for fitting the particle number size distributions. Boreal Environment Research, 10(5), 337-355. http://www.borenv.net/BER/pdfs/ber10/ber10-337.pdf) here? Also, what do you mean by "isolating the variability of isolated modes" - please consider revising for readability.**

*The reference was included and the referred sentence was improved. We meant that when working with the lognormal fit, one can show the variability of every mode separately.*

**- p. 4, lines 130-131: Was it always appropriate to fit three modes or were there instances when a different number of modes would have represented the size distribution better? If yes, what kind of error might the choice of three modes introduce to the results presented? Furthermore, did you fix the size ranges assumed for the three modes or let the code decide this. What might this imply for the results? Please add a brief elaboration on these questions and a justification of the chosen approach (in terms of numbers of modes and size ranges assumed).**

*The code did not necessarily fit three modes. It was free to fit between one and three. In addition, the decision of three modes and the range in diameter was based on a statistical analysis of 6 years of data (Figure 2 in Franco et al., 2022), where the maximum of modes and the ranges is clear.*

*The mode fitting code started with a fixed guess based on the statistical diameter position of each one of the three modes. Later, the code did two optimizations to correct the guess: one was done based on the other modes' positions, and the other was an optimization of the three modes together. After these three steps, it is expected to obtain more precise fits. We studied the modes separately in our analyses, considering all the fitted distributions in 1 year. So, in the end, we had enough data points for every mode, but not necessarily they were always fitted together in the time resolution of 5 minutes. However, since we present the means in our analyses, we have the statistical representation of every mode for each studied case.*

**- p. 7, line 72: Please revise the expression commenting that "the SMPS used was limited to 10 nm" for accuracy. I guess you want to say that the lower detection limit of the SMPS was 10 nm.**

*The referred expression was improved as recommended.*

**- p. 7, line 76: Instead of "constant dispersion" do you mean "constant standard deviation"? Can this depend on the the size ranges that you have constrained (if that is the case)?**

*Yes, by constant dispersion we mean constant standard deviation, as seen by the projection (shadow) on the sigma axis. Regarding the size ranges, the code first considered the same range of sigma for all modes, varying from 1.1 to 1.55 nm, and then in the optimization step, the code allowed a new range of the maximum of 1.2 times the first fit.*

*Although the ranges in the axis are different, in Fig. 2, it is noticeable that the variations in the sigma values of the sub-50 nm and the accumulation are more spread out vertically than the ones in the Aitken mode, which is nearly around 1.2 and 1.3 nm. You can check this on the new 2D plot in the Supplement. In Fig. S2b the colors are mainly green, exactly between 1.2 and 1.3 nm, while the others (Figures S2a and S2c) have a greater variation.*

**- p. 8, lines 215-216: Please revise the sentence starting "the accumulation mode dominates...". I believe you want to say that "accumulation mode dominates over Aitken mode" and at the end "prevalent" instead of "equivalent".**

*You are correct when we tried to say that the accumulation mode dominates over the Aitken mode. For the second part, the word "equivalent" fits better since we meant that both modes have the same overall concentration.*

**- p. 9, lines 242-243, the sentence saying "reaching a maximum during the night probably due to late afternoon rainfalls": What about the importance of boundary layer dynamics?**

*We included in the text the effects of the nocturnal boundary layer in the sub-50nm concentration dynamics. Rainfall increases the ultrafine particle number, and the nocturnal boundary layer keeps the concentration nearly constant during the night.*

**- p. 9, line 243: with "initiates" do you perhaps mean "begins"?**

*Exactly.*

**- p. 12, lines 305-306: Please revise the the sentence "Since lightning and precipitation peak simultaneously (Mattos et al., 2017) the following results (Figure 5) are intercomparable, promoting a complete characterization of the aerosol-precipitation interaction." for clarity and readability.**

*Thank you for the comment. The text has been improved and we hope it is clear now.*

**- p. 13, line 314: Instead of "sensible" do you perhaps mean "pronounced" or something similar?**

*Exactly. The word has been changed as recommended.*

**- p. 13, line 317: Please revise the sentence starting as "In addition, it was shown..." for readability and English language.**

*The sentence has been rephrased and we hope it is now improved.*

**- Same as above, the sentence starting "In fact, it can be seen...": Where exactly can this be seen? Please specify.**

*The whole paragraph has been rewritten and we hope it is improved now.*

**- p. 14, line 327: Please revise the sentence for readability, clarity and precise use of English. Specifically, what do you mean by "background trajectories" - please specify.**

*The text has been improved and we hope it is clear now. The new sentences are: "The following analysis explored the background aerosol concentration in the morning considering afternoons with and without precipitation. It was considered time trajectories from 6 to 12 LST."*