Responses to Reviewer #1 of the manuscript "Vertically resolved aerosol variability at the Amazon Tall Tower Observatory under wet season conditions" by Franco & Valiati et al., submitted for publication in Atmospheric Chemistry and Physics

Dear Editor, we would like to thank reviewer #1 for the valuable comments and useful suggestions to improve our manuscript. Below, you can find answers and actions for each individual comment. To make it easier to identify the individual answers and actions, we used the following color code strategy:

- **In black are the reviewer's comments.**
- **In blue are the author's responses.**
- ***In blue and italics are the text modifications we made in the manuscript.***

General comments:

This article provides and analyzes very valuable information of the sub-micron aerosol in a remote Amazon site. It informs not only number concentration and size distribution (10 nm to 400 nm) but also extensive and intensive optical properties of the aerosols, at two different heights, thanks to the well known ATTO tower. This kind of data is very scarce in the region, since most of the (few) air quality monitoring stations in South America are placed in urban regions.

As the authors claim, comparing continuum measurements at different heights is very important, since it provides information regarding sources (new particle formation, biogenic emissions, long range transport) and helps validate and improve aerosol models and remote sensing retrievals. The location of the site allows to study a pristine Amazonian atmosphere, with impact of Africa long range biomass burning, and in addition provides insight regarding the effect of downdraft events.

For instance, they observe the higher relevance of BC at 325 m due to the effect of the African plume (and optical aging of BrC); they show new particle formation near the canopy (sub-50 nm) and growth during the day through aging (accumulation mode); they observe the rate of new particle formation after wet deposition; etc.

The article provides very relevant figures (both main article and supplement) that are adequate to derive the conclusions in the main text. The article is well written and supported with previous results from literature, adequately discussed.

I recommend publication after minor revision.

We thank Referee #1 for the very constructive comments and useful suggestions. They helped us to clarify important aspects of our discussions and, thus, to improve the manuscript overall.

Specific comments:

Section 2.3: if aerosol volume is estimated from SMPSs (as I imagine), it would be relevant to introduce in this section a comment on that, and maybe the equations that connect the measured size distributions with the aerosol volume (here or in the Supplement).

Many thanks for the suggestion. Indeed, we used the SMPS data to retrieve the aerosol volume. Section 2.2 was updated, and we added the following text to detail the process:

The fine mode number concentration and the aerosol volume were obtained by integrating the PNSD. For the particle volume, it was considered that all particles are spherical, following single-particle characterizations developed with Amazonian aerosols (Wu Li et al., 2019).

Section 2.4: I believe it would be important to make clear which cut size (if any) have each optical instrument. For instance, usually the aethalometers use PM2.5 size cut, but not always. From the discussion it seems that nephelometer and MAAP let coarse aerosols in (effect of large biogenic aerosols is discussed), but it is not clear what sizes are allowed.

Thanks for pointing that out. All the optical instruments measured with PM2.5 size cut. We added the following line to the manuscript to highlight it:

All optical instruments operated with a size cut of 2.5 μm.

Section 2.5: The adjustment of m and k with Mie's calculations is interesting. Do you think is reasonable to assume sphericity for these particle sizes? Do you have microscopy information for similar samples?

Yes, sphericity is a reasonable assumption for submicrometer aerosols during the wet season at the ATTO forest site. Single-particle characterization of aerosols using ED-EPMA (energy-dispersive electron probe X-ray microanalysis) has shown that SOA (secondary organic aerosols) and ammonium sulfate comprised 73 %–100 % of submicrometer aerosols (Wu et al., 2019). The authors report that most submicron SOAs were internally mixed with ammonium sulfate, having a circular shape in the images. The overall circular morphology of SOA-ammonium sulfate mixed particles indicate that they are mostly in aqueous droplets at the time of sample collection rather than in crystalline form. Therefore, the spherical assumption is reasonable.

Section 3.2: "Although differences were observed for the SAE and the vertical profile, indicating different aerosol coarse mode populations, the ω0,637nm indicates

that the fine mode aerosol population in both highs is very efficient in scattering radiation." I would like the authors to comment a little further on this phrase (and other connected phrases below in the article). In the one side, I understand that you did not measure the coarse mode aerosols in this work, but that is plausible that there are more coarse biogenic particles at 60 m, which should be expressed as a lower a SAE. But here you see small differences in SAE in the vertical profile, and you assign it to the fact that "the fine mode aerosol population in both highs is very efficient in scattering radiation". Am I correct? Later in this section, you also suggest that the differences in the accumulation mode median diameter could influence the aerosol optical properties. Please comment a little bit more on your view regarding the impact of coarse and fine mode aerosols in scattering coefficient and SAE. Maybe comparing the effects that increase or decrease the scattering coefficient and SAE and the suggested balance among these factors/causes at the end of this section? It can also be connected with complementary results and discussion in Sections 3.5 and 3.6.

The first highlighted sentence was divided and more clearly phrased. The scattering efficiency and albedo are not necessarily related to the observed difference in the SAE at both heights.

Furthermore, the balance between the fine and coarse modes between the two heights has contrasting effects on the scattering coefficient and the SAE. For the scattering coefficient, the fine mode is likely more important than the coarse mode, due to the larger fraction of aerosol surface area being linked to fine mode particles. In this scenario, the larger fine mode volume observed at 325 m suggests that the particle cross-section is larger as well, which explains the higher scattering coefficient. On the other hand, the more prevalent coarse mode at 60 m affects the SAE greatly. Schuster et al. (2006) found that the effective diameter of the fine mode particles is less important than the fine/coarse mode ratio for the SAE, which is likely the reason that this intensive property is so different between the two height levels.

In order to improve this discussion, we included the following text at the end of Section 3.2:

As most of the particles found above the canopy are within the fine mode range, the scattering coefficient is largely dependent on the surface area of fine mode aerosols, and less on the coarse mode mass concentration. This is likely the reason for the higher scattering coefficient at 325 m, as the larger particles have higher scattering cross-sections. Nevertheless, Schuster et al. (2006) found that the SAE is less dependent on the effective diameter of the fine mode particles, and more influenced by the balance between fine and coarse mode aerosols. A strong vertical gradient of coarse mode particles, more prevalent at 60~m, is likely the reason behind the large difference in the SAE.

Section 3.5, p.18: "The results suggest that aerosols at 325 m, as observed in Figure S6, are likely to be more processed and are less efficient in scattering

radiation. In contrast, smaller aerosols in direct contact with fresh VOC emissions from vegetation are more likely to scatter radiation more efficiently. Another possibility that might explain the results is that the higher apparent MSE at 60 m is likely due to the presence of a coarse mode, which is not detected by the SMPS. In fact, Prass et al. (2021) observed that bioaerosols account for about 70% of the aerosol coarse mode at ATTO, with higher concentrations at 60 m, which decreases with height.". Are there previous results that suggest that "smaller aerosols in direct contact with fresh VOC emissions from vegetation are more likely to scatter radiation more efficiently"? Or do you believe that the second hypothesis is more likely?

In fact, both hypotheses are likely, and further analyses are required to discriminate which one is the more important. From the perspective of the fresher and smaller particles, they are mainly constituted by secondary organic aerosols (SOA) derived from the oxidized volatile organic compounds (VOCs). Holanda et al., 2023, have shown that, at 325 m high at the ATTO site, pristine aerosols, which are mainly formed by oxidized SOA (80% of the mass fraction), have a significantly higher single scattering albedo (SSA) than long-distance aged aerosols, which are larger, with a relevant coating size and are more absorbing, even during the wet season. The SSA, just like the mass scattering efficiency (MSE), is an intensive aerosol property, and both are directly correlated; the bigger the SSA, the bigger the MSE. The lensing effect may prevail for the coarse mode particles, turning the biological particles, which are the main aerosols in this particular size range, the main source of radiation scattering.

To improve the quality of this discussion, we added the following text to the manuscript:

Holanda et al., 2023, have shown that, at 325 m high at the ATTO site, pristine aerosols, which are mainly formed by oxidized SOA (80% of the mass fraction), have a significantly higher single scattering albedo (SSA) than long-distance aged aerosols, which are larger, with a relevant coating size and are more absorbing, even during the wet season. The SSA, just like the mass scattering efficiency (MSE), is an intensive aerosol property, and both are directly correlated; the bigger the SSA, the bigger the MSE.

Another possibility that might explain the results is that the higher apparent MSE at 60 m is likely due to the presence of a coarse mode, which is not detected by the SMPS. In fact, Prass et al., 2021 observed that bioaerosols account for about 70% of the aerosol coarse mode at ATTO, with higher concentrations at 60 m, which decreases with height. Both hypotheses are likely, and further analyses are required to determine which is more relevant.

Technical corrections:

P.18, "Shorter wavelengths have higher scattering efficiencies:"
It seems to me that the correct expression would be "Scattering efficiencies are higher for shorter wavelenghts"
(to my mind, aerosols HAVE higher or lower scattering efficiencies, not wavelenghts).

Thanks. We corrected the sentences in the manuscript.

References:

Schuster, G. L., Dubovik, O., and Holben, B. N.: Angstrom exponent and bimodal aerosol size distributions, Journal of Geophysical Research: Atmospheres, 11.

Wu, Li, et al. "Single-particle characterization of aerosols collected at a remote site in the Amazonian rainforest and an urban site in Manaus, Brazil." Atmospheric Chemistry and Physics 19.2 (2019): 1221-1240.

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