Response to reviews:

We would like to express our sincere gratitude to the reviewers for their insightful and constructive feedback on our manuscript titled *"Strong Influence of Black Carbon on Aerosol Optical Properties in Central Amazonia during the Fire Season."* We deeply appreciate the time and effort invested in reviewing our work, and the comments provided have significantly improved the clarity, depth, and quality of the manuscript.

In this response, we outline the changes made to address the reviewers' major and minor comments. Each point has been carefully considered, and we have revised the manuscript accordingly to ensure that the scientific rationale is more clearly presented, the results and discussions are better structured, and the potential sources of uncertainty are adequately addressed. Additionally, we have improved the manuscript's readability and the presentation of key findings to enhance its overall impact.

We hope that the revisions meet the reviewers' expectations and that the revised manuscript is now suitable for publication in *Atmospheric Chemistry and Physics*. We believe that the changes made have significantly strengthened the manuscript and provide a clearer narrative of the importance of our study in the context of aerosol radiative forcing and its implications for climate modeling.

We are grateful for the thoughtful feedback provided, and we look forward to any further suggestions that may arise during the next stages of the review process.

Sincerely,

Rafael Stern, on behalf of all co-authors

Response to Reviewer 1:

General Comments

This manuscript presents a comprehensive study of submicrometer aerosols during the dry season in central Amazonia, focusing on their chemical composition and optical properties. The study employs state-of-the-art analytical techniques, including Positive Matrix Factorization (PMF) and Multi-linear Regression (MLR), to provide valuable insights into the contribution of different aerosol components to radiative forcing. The findings, particularly the high scattering efficiency of equivalent black carbon (eBC), are novel and carry significant implications for climate modeling. However, there are several areas where the manuscript can be improved, both in terms of scientific content and presentation. Below are specific comments.

Response: We are very grateful to Reviewer 1 for the thorough evaluation of our manuscript and for recognizing the novelty and importance of our findings regarding the high scattering efficiency of eBC in central Amazonia. We also appreciate the acknowledgment of our methodological approach using PMF and MLR to disentangle the chemical and optical properties of submicrometer aerosols.

We have carefully considered all the reviewer's constructive suggestions and implemented substantial improvements to both the scientific content and the overall presentation of the manuscript. These include:

- A restructured and more focused Introduction, with a clearer articulation of the scientific motivation, research gap, and relevance of the study to broader atmospheric and climate science.

- Expanded discussion on the mechanisms behind the high MSE of eBC, including a comprehensive review of recent literature on the effects of coatings and atmospheric aging, and the inclusion of supporting references.

- Enhanced comparisons with previous studies, emphasizing how our results align with or differ from those observed in other regions or under different pollution regimes.

- A detailed treatment of uncertainties and methodological assumptions, now more prominently addressed in the main text and supported by new supplementary materials.

- Numerous editorial improvements, including revised figure formatting, improved table descriptions, consistent terminology (e.g., use of "Amazonia" throughout), and streamlined language to improve clarity and readability.

We hope these revisions address the concerns raised and substantially enhance the manuscript's clarity, scientific rigor, and impact. We thank the reviewer once again for the insightful feedback, which has helped us strengthen the manuscript significantly.

Major Comments:

1. *Review: Introduction: Lack of Logical Flow and Clear Scientific Questions:*

    The introduction provides a rich background on Amazonian aerosols, their sources, and their climatic impacts, but it lacks a clear logical progression that leads to the importance of this study. Specific issues include:

    While the first four paragraphs review broad background knowledge, they do not adequately justify the importance of studying eBC and organic aerosols in the Amazon. The scientific problem and research gap are not clearly articulated.

    The statement "the intrinsic optical properties of each aerosol species are still rare" is insufficient as a scientific rationale. The authors need to more explicitly identify why studying these properties is critical and how the study addresses unresolved questions in the field.

    Logical inconsistencies in content, such as between lines 71–74, need to be addressed. For example, the discussion of secondary oxidized aerosols (lines 71–73) and scattering efficiencies (line 74) appears disconnected and lacks a clear link to the study's objectives. The author should ensure logical transitions between sentences and paragraphs and ensure all background information directly supports the scientific rationale of the study.

Response: We thank the reviewer for highlighting the need for improved clarity. We have revised the introduction to better define the scientific problem and clearly articulate the gap in knowledge. Specifically, we now emphasize the importance of studying the optical properties of eBC and organic aerosols in the Amazon, and we have explicitly connected these studies to broader climate modeling implications.

2. **Mechanisms of eBC Scattering Efficiency:**

Review: *The study identifies a surprisingly high mass scattering efficiency (MSE) for eBC, suggesting a potential role of coating in enhancing light scattering. However, the manuscript does not provide sufficient experimental or theoretical discussion to explain this phenomenon. It is recommended to include additional analyses or citations to studies focusing on eBC coatings and their optical effects. If possible, provide size distribution or coating thickness data to support the hypothesis of enhanced scattering due to coating effects.*

Response: We appreciate the reviewer's suggestion to discuss eBC scattering efficiency in more detail. Firstly, we have strengthened literature review on the effect of eBC coating by adding the following references: Schwarz et al. (2006); Cheng et al. (2008); Titos et al., 2012; Metcalf et al. (2013); Wang et al. (2016); Tasoglou et al. (2017); Saturno et al. (2018); Tao et al, 2019; Kleinman et al., 2020; Zhang et al., 2020; Romshoo et al. (2021); Li et al. (2024). The main text has been modified as described below:

In the Introduction section, where it read:

> "While Organic Aerosol (OA) originates from both primary emissions, as well as secondary formation from gaseous precursors (Martin et al., 2010), black carbon is mostly primarily emitted from incomplete combustion, and in remote areas of the Amazon it is associated with Amazonian or transatlantic forest fires (Artaxo et al., 2013; Holanda et al., 2020). The sign and magnitude of the ARI forcing are dependent on several parameters such as particle origin, size distribution, mixture and age, notably affecting the light-absorbing component of OA, termed brown carbon (Laskin et al., 2015; Saturno et al., 2018; Li et al., 2024)."

Now it reads:

"Aerosol particles known for efficiently absorbing radiation - such as BC - often also exhibit significant scattering efficiencies, which are strongly influenced by their size, chemical composition, and the extent and nature of their atmospheric aging and coatings (Bond and Bergstrom, 2006; Schwarz et al., 2006; Yu et al., 2010). Although chemical aging has shown to enhance light absorption due to the coating of the BC core by condensing semi- and intermediate volatility organic compounds or coagulation with other particles (Darbyshire et al., 2019; Metcalf et al., 2013; Saturno et al., 2018b; Tasoglou et al., 2017; Wang et al., 2016), primary biomass burning aerosols have also been associated with high scattering efficiencies (Hand and Malm, 2007; Malm et al., 2005). Coating by non-absorbing material, such as Organics (Romshoo et al., 2021), has been shown to increase BC scattering by a factor of 3-24 depending on the size, morphology, aging stage, coating thickness and composition of the BC particles (He et al., 2015). Conversely, sulfate and water coating have also shown to increase elemental carbon particle diameter, playing a stronger role on its scattering efficiency, more than absorption (Cheng et al., 2008; Yu et al., 2010). Precisely quantifying distinct ARI for each chemical species, and especially decreasing uncertainties on the ones with high potential to both absorb and scatter radiation such as BC is critical to improve our understanding and prediction of the atmospheric system and improve climate models."

In the Results and Discussion section, where it read:

"Our result of a pronounced MSE of the eBC represents an opposite trend than the observed in the transition from the dry to the wet season at a site more impacted by the fires, where the MSE of BBOA was higher than eBC's (Ponczek et al., 2021)."

Now it reads:

“The pronounced MSE of the eBC (7.62-13.58 $m^2 g^{-1}$, Table 3) is strongly corroborated by other studies which found remarkably high scattering efficiency related to BC, especially when the particles undergo atmospheric processing and aging, such as in the case of our study (Bond and Bergstrom, 2006; He et al., 2015; Malm et al., 2005; Pitchford et al., 2007; Romshoo et al., 2021; Schwarz et al., 2006). It has been demonstrated that while aerosol scattering efficiency increases with increasing size, age and distance from the source, the absorption efficiency remains nearly constant (Kleinman et al., 2020; Zhang et al., 2020). MSE of elemental carbon in a rural area ranged from 5.4-66.2 $m^2 g^{-1}$, and the high increase was found to be related to sulfate addition during cloud processing (Yu et al., 2010). Recently, on a comparable method, MSE for eBC has been estimated at 6 $m^2 g^{-1}$ in a site located in Western Amazonia. Located within the deforestation arc, the site is strongly impacted by fresh, sometimes local emissions, in contrast to regional or long-range transport of fires impacting Central Amazonia (Ponczek et al., 2021). In regions impacted by urban pollution MSE of eBC was 2.6 $m^2 g^{-1}$ (Tao et al., 2019), and found not to influence MSE for coarse mode particles (Titos et al., 2012).”

In the Conclusions section, we added these sentences:

“Aerosol observations at the heart of Amazonia are extremely challenging. Although our analysis during the dry season yielded robust results, limitations remain—particularly concerning the size distribution of BC and its absorption across multiple wavelengths. To advance our understanding, future studies should prioritize extensive field and laboratory observations aimed at better constraining aerosol coating formation mechanisms and their impact on the radiative scattering properties of BC particles in Amazonia. Increasing the precision of the quantification of the eBC contribution to light scattering has the potential to improve models and decrease uncertainties in global radiative forcing estimations.”

3. **Comparisons with Other Studies:**

Review: While the manuscript cites relevant literature, a more detailed comparison with similar studies would enhance its impact:

Compare the reported MSE, MEE, and SSA values with those from other regions and seasons, particularly regions strongly impacted by urban or biomass burning pollution.

Discuss how the findings align or contrast with previous studies on eBC optical properties, especially in terms of scattering contributions.

Response: In response to the reviewer’s request for further comparisons with other studies, we have expanded the section discussing the reported MSE, MEE, and SSA values. The whole section 3.3 was re-written. We were previously citing these studies (Hand and Malm, 2007; Yu et al., 2010; Wang et al., 2015; Luo et al., 2020; Ponczek et al., 2021; Velazquez-Garcia et al., 2023). We have included studies from regions impacted by urban or biomass burning pollution to show how our findings compare and contrast, such as Levin et al., 2010; Cheng et al., 2015; Wu et al., 2019; Tian et al., 2022; Saide et al., 2022; Pani et al., 2023; besides what was added regarding comment 2.

4. **Uncertainty in Optical Properties:**

Review: The derived optical properties (e.g., MSE, MEE) rely on specific assumptions, such as absorption cross-section values for eBC. The manuscript should:

Clearly state the sensitivity of the results to these assumptions.

Discuss potential sources of uncertainty in the measurements and their implications for the reported efficiencies.

Response: We thank the reviewer for the opportunity to improve clarity of the text. We moved the section where the sensitivity and potential sources of uncertainty in the measurements are discussed to the beginning of section 3.3. We also improved clarity of some sentences, figures and tables legends and added Table S6 and Figure S6.2 to the supplement.

Where it read:

"A previous study found that eBC absorption cross-section for the Amazon was 12.3 m^2 g^{-1} (Saturno et al., 2018b), and we tested our dataset applying this value. The result is that eBC mass concentrations would become half of what they are reported, with no change in σ_a, SSA, but MSE would double, while eBC contribution to MEE (Figure 8) would remain unchanged. Due to some methodology differences between our study and (Saturno et al., 2018a) (they measured refractory Black Carbon using a single-particle soot photometer SP2, with a higher cut-off, possibly leading to a sub-estimation of the mass), and the fact that applying the absorption cross-section value they found would make MSE of eBC be an order of magnitude higher than the others (Supplement Figure S6), we opted to remain with the more stablished value of 6.6 m^2 g^{-1}."

"Concerning the contribution of AN to the PM1 mass concentration, we tested the MLR removing AN, and the results were comparable, especially for eBC (Supplement Table S5.1). We also tested the robustness of the method by running 100 times MLR on random 50% of the data, yielding similar results (Supplement Table S5.2)."

Now it reads:

"A previous study found an eBC absorption cross-section in Amazonia of 12.3 m^2 g^{-1} (Saturno et al., 2018b), and we tested our dataset applying this value (Supplement, Figure S6.1). The result is that eBC mass concentrations would decrease by half, with no change in σ_a and SSA, but MSE would double, while eBC contribution to MEE (Figure 8, Figure S6.2) would remain unchanged. Due to some methodology differences between our study and (Saturno et al., 2018b) (they measured refractory Black Carbon using a single-particle soot photometer SP2, with a higher cut-off, possibly leading to a sub-estimation of the mass), and the fact that applying the absorption cross-section value they found would make MSE of eBC be an order of magnitude higher than the others (Supplement Figure S6.1), we opted to remain with the more established value of 6.6 m^2 g^{-1}."

"We applied the multiple linear regression (Section 2.4.2) to our dataset, and the resulting coefficients successfully predicted the observed scattering (R^2 = 0.86, Figure 6), confirming the validation of this methodology to estimate the specific contribution of each chemical group to the optical properties. We tested the MLR removing AN (due to its low contribution to the PM1 mass concentration, close to the ACSM detection limit, and therefore, possible artifacts), and the results were comparable, especially for eBC (Supplement Table S5.1). We also tested the robustness of the method by running 100 times MLR on randomly selected 50% of the data, yielding similar results (Supplement Table S5.2). All standard errors were small (Table 3), and the Variance Inflation Factor was around 3 for IEPOX-SOA, BBOA, AS and AN; 5.20 for OOA, and 6.19 for eBC. The abovementioned tests suggest that typical MLR caveats such as collinearity had minimal effect on the observed final results."

Minor Comments:

5. Abstract:

<u>Review:</u> The abstract effectively summarizes the main findings but could provide more emphasis on the broader implications of eBC's scattering role for climate modeling.

<u>Response:</u> We thank the reviewer for the opportunity to improve clarity of the abstract.

Where it read:

"The dominance of eBC over light scattering, in addition to absorption, depicts a surprisingly high role of this important climate agent, indicating the need to further investigate the chemical processing and interaction between natural and anthropogenic aerosol sources over remote tropical forested areas."

Now it reads:

"The dominance of eBC over light scattering, in addition to absorption, depicts a remarkably high role of this important climate agent, with potentially broad implications for more precise radiative forcing quantification, increasing climate modelling precision, representing deep contributions to Earth's climate system comprehension."

6. Figures:

<u>Review:</u> Improve the clarity and readability of some figures. For example, in Figures 2 and 4, use consistent scales and labels to make comparisons across factors easier.

<u>Response:</u> We thank the reviewer for this opportunity to increase clarity and readability of the figures. Now Figure 2b has the same timestamp as Figure 2a (1 hour instead of the previous 12 hours, one unique legend with a better color code, year information on X axis). Figure 4 was also improved (X axis labels).

7. Table 3:

<u>Review:</u> clearly indicate which values are derived from measurements vs. calculations.

It will be better to add a schematic figure summarizing the contributions of different aerosol components to light scattering and absorption.

<u>Response:</u> We thank the reviewer for pointing this out. Now the table contains only MSE values, and we added the text "resulting from the MLR" to the legend to improve clarity. The contribution of each aerosol component to light scattering is shown in Figure 7b. Our measurements technique - Multi Angle Absorption Photometer (MAAP, model 5012, Thermo Electron Group, Waltham, USA) (Müller et al., 2011) - minimizes the interference from non-absorbing aerosols, and therefore is considered as a reliable measurement of absorption, dominated by eBC at wavelength 637 nm.

8. Lines 289-291:

<u>Review:</u> The inference oversimplifies the source attribution of black carbon (eBC) by directly linking it to biomass burning. While biomass burning is indeed a significant source in the Amazon during the dry season, local and regional sources, such as emissions from Manaus (e.g., diesel generators and vehicular emissions), must also be considered. A more nuanced approach would be to discuss the dominant sources in the region, then use the correlation between eBC and biomass burning markers (if available) to strengthen the attribution to biomass burning.

<u>Response:</u> We thank the reviewer for the opportunity to improve clarity and scientific precision of the text. It is important to note that, as described in Section 2.2, potential influence of diesel

generator and Manaus plume were removed from the dataset, allowing to focus on aerosol population representative of Central Amazonia basin.

Where it read:

"Since OOA corresponds to more than half of PM1 (Table 1), this high correlation indicates that most of the submicrometer aerosols measured during the dry season in Central Amazonia are, in general, influenced by biomass burning emissions, since eBC is an important combustion tracer."

Now it reads:
"This suggests that a significant fraction of the aged submicrometer aerosols measured during the dry season in Central Amazonia is largely influenced by biomass burning emissions, in combination with other combustion sources such as sporadic urban plumes transported from Manaus. In addition, co-variability between aerosol species is expected due to strong washout events that, although less frequent, can still occur during the dry season and impact multiple aerosol components simultaneously."

9. Language:

Review: Some sentences, particularly in the Introduction and Discussion, are overly long and could be simplified for better readability.

Response: We thank the reviewers for the opportunity to improve clarity of our article. All the sentences have been extensively revised and restructured to be more concise and improve readability.

10. Code and Data Availability:

Review: The manuscript mentions that data will be made available upon acceptance. It would be more transparent to provide a DOI or link to a repository containing at least some preliminary data.

Response: We updated the data availability. It now reads:

"Codes and data are currently available in this link:

https://zenodo.org/records/15345166; DOI: 10.5281/zenodo.15345166."

11. *Review: The discussion of diurnal variations could be expanded to include a more detailed analysis of potential drivers (e.g., boundary layer dynamics, local emissions).*

Response:

Where it read:

"The BBOA diurnal profile is different from that of the other PMF factors (Figure 4). While the OOA and IEPOX-SOA mass concentrations increase during the daytime due to photochemical oxidation processes, the BBOA mass concentration is fairly constant (Figure 4). Since the BBOA is a biomass-burning indicator, it is composed of mostly primary particles, so its concentration does not depend on photochemical activity. This pattern is different than the stark decrease in mass concentrations of fresh biomass burning particles during daytime, observed in southwestern Amazonia (Brito et al., 2014), where there were constant local sources of fires and the diurnal cycle was mostly determined by the boundary layer increasing and diluting the particles in a bigger area during daytime (Andreae et al., 2015). The lack of a clear diurnal pattern in our study for BBOA seems to confirm the regional origin of the aerosol particles, likely transported from distant biomass burning sources in the eastern parts of the basin, and long-range transport with complex interactions between residual and nocturnal layers (Darbyshire et al., 2019). An additional confirmation of the long-range transport is the relatively flat pattern of the eBC diel cycle, although there is a small but noticeable increase in the eBC diurnal mass concentration during daytime (Figure 4), which may indicate some lensing effect due to the increase in the particle coating (Denjean et al., 2020). While the diel cycle of the NH_4 and NO_3 show practically no variation, the SO_4 indicates the influence of photochemical processes (Figure 4) or atmospheric transport. The higher boundary layer in the afternoon (Fisch et al., 2004) may favor the downward transport of particles originally emitted in distant regions (Darbyshire et al., 2019). An additional possible explanation for this observed increase in SO_4 during the afternoon is biogenic sources of SO_4 precursors."

Now it reads:

"The daily profile of BBOA differs significantly from other factors (Figure 4). While OOA and IEPOX-SOA mass loadings increase during the day, likely due to photochemically driven oxidation processes, BBOA remains relatively constant throughout the day, despite the daytime dilution effect of a rising boundary layer (Andreae et al., 2015). Interestingly, this pattern contrasts with the pronounced daytime decrease in fresh biomass-burning aerosol concentrations reported in southwestern Amazonia (Brito et al., 2014), where local fire emissions were more prevalent. The absence of a clear diurnal cycle for BBOA in our study corroborates a regional, rather than local, origin—likely from biomass-burning sources located in the eastern Amazon. The flat variability of this primary factor reflects transport over long distances and the influence of complex vertical mixing, including interactions between residual and nocturnal layers (Darbyshire et al., 2019).

Further supporting this hypothesis is the relatively flat daily cycle of eBC, although a slight daytime increase is observed (Figure 4), possibly due to lensing effects as particles acquire coatings during transport (Denjean et al., 2020). Unlike eBC, NH_4 and NO_3 show minimal diurnal variation, while SO_4 exhibits a daytime increase, consistent with secondary production via photochemical reactions from biogenic sources, or atmospheric transport processes. The rise in the boundary layer during the afternoon (Fisch et al., 2004) may facilitate the entrainment of particles from above the boundary layer (Darbyshire et al., 2019)."

12. Review: *The manuscript needs to provide further explanation for why the MSE (mass scattering efficiency) calculated by PMF decreases with increasing wavelength. Is this related to the optical properties of the aerosols, changes in particle size, or their mixing state? Providing some theoretical background or relevant literature would help clarify this observation.*

Response: In our analysis, we observe that all components except AS exhibit a decreasing trend of MSE with increasing wavelength, which is a typical behavior of submicrometric aerosol particles. This wavelength dependence is consistent with Mie theory, which predicts stronger light scattering at shorter wavelengths for particles smaller than or comparable to the wavelength of light. The variability in the slopes of MSE across the different PMF factors is likely due to a combination of factors, including: Differences in mixing state (internal vs. external); variations in intrinsic refractive indices among the aerosol components; temporal dynamics of the aerosol populations, as reflected by the factor loadings throughout the diurnal cycle, which influence the particle size distribution associated with each source.

We have now revised the manuscript to better contextualize this behavior.

The following text has been added on L. 406: "As shown in Figure 7a, the MSE of all components except AS decreases with increasing wavelength, which is consistent with the typical behavior of submicrometric aerosols. This spectral dependence can be attributed to Mie scattering theory, where smaller particles scatter shorter wavelengths more efficiently (Hand and Malm, 2007; Malm et al., 2005). Nonetheless, the variability in the MSE slopes among the different components reflects a complex interplay between aerosol mixing state, refractive index, and size distribution dynamics—particularly the diurnal evolution of each factor's contribution to the total aerosol population (Figure 4). It is particularly interesting that AS exhibits a distinct spectral behavior, typically associated with coarse-mode aerosols, denoting stark differences in its sources and atmospheric processing compared to the other components. Sulfate in Amazonia has been associated with secondary production from biogenic emissions and mixing with primary biogenic organic aerosols (PBOA) (Martin et al., 2010b; Pöhlker et al., 2012), as well as with coarse-mode particles such as dust and sea salt transported over long distances (Brito et al., 2014; Wu et al., 2019). It is remarkable that the MLR analysis captured this behavior, considering that the ACSM is limited to non-refractory species in the submicron range and is not particularly efficient at detecting the sources likely involved. This highlights the sensitivity of the MLR approach to broader aerosol population dynamics, which were captured by the optical instruments operating with a PM_{10} inlet, suggesting the influence of coarse-mode aerosol sources."

13. _Review:_ *eBC is known to be a light-absorbing aerosol, but why does it have the highest MSE in this study, and why is it higher than in other studies? The authors should provide possible explanations, such as the influence of coating, particle size changes, or other mixing effects (Li et al., npj, 2024). This is crucial for understanding eBC's scattering properties.*

Response: We fully addressed this topic while addressing comment 2 from reviewer 1.

14. _Review:_ *The manuscript states that eBC does not show wavelength dependence, yet Figure 7a shows large fluctuations in MSE at shorter wavelengths. Can the authors explain these fluctuations? Additionally, while Figure 7a shows higher MSE for eBC at shorter wavelengths, its contribution to scattering is lower compared to other wavelengths. This seems contradictory. Please explain the relationship between these two observations.*

Response: We appreciate the reviewer's careful reading and comments regarding Figure 7a.

We would like to clarify that our statement in the manuscript refers to the MSE of **AS, not eBC**, as showing no clear wavelength dependence. We apologize if this was unclear and have revised the text to improve clarity.

Regarding the fluctuations in MSE at shorter wavelengths, if the reviewer is referring to the error bars shown in Figure 7a, this is indeed observed across several components. While the relative error does not vary dramatically across wavelengths, it tends to increase at shorter wavelengths. This is likely due to greater sensitivity of scattering to changes in aerosol size distribution at shorter wavelengths, as captured by the SAE. As discussed in our response to comment 12 above, the variability in SAE across the aerosol components is complex and reflects differences in mixing state, refractive index, and temporal dynamics, all of which influence the size distribution and its evolution over time.

As for the second point, we added this to section 3.3: "While the MSE of eBC does decrease with increasing wavelength, its slope (or more precisely, its SAE) is lower than that of other aerosol components. As a result, eBC retains a relatively higher fractional contribution to total scattering at longer wavelengths compared to components with steeper MSE declines. The absolute contribution to scattering is determined by both MSE and mass concentration, and although eBC mass concentrations are generally lower, its weaker wavelength dependence allows it to contribute proportionally more at longer wavelengths. Interestingly, AS exhibits a distinct behavior, likely associated with different sources and atmospheric processes than those influencing the other components. While the ACSM is limited to submicrometer aerosol, and does not capture contributions from larger particles, the variability of AS has been consistently observed in the nephelometer and MAAP measurements operated with a PM_{10} inlet, suggesting the influence of coarse-mode aerosol sources."

15. _Review:_ *The SSA value for eBC in Figure 8 is 0.57, indicating that scattering dominates its extinction. However, theoretically, black carbon, being a strong absorber, typically has a low SSA, often below 0.3 for pure black carbon (Wang et al., 2021). How do the authors reconcile this higher SSA for eBC in the study? Can this be explained by coatings or other factors? Further clarification on this point is needed.*

Response: We fully addressed this topic while addressing comment 2 from reviewer 1.

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Response to Reviewer 2:

General Comments:

1. Review: *The manuscript presents an analysis of aerosol chemical and optical properties recorded in Central Amazonia during the dry season 2013 from August to December. The recorded properties cover a comprehensive set of chemical parameters, mainly from an ACSM instrument combined with Positive Matrix Factorization, and numerous optical parameters which allow the determination of all relevant extensive and intensive optical properties.*

The study is carefully designed and well conducted. As one major finding, the authors identify the strong contribution of eBC to the aerosol scattering properties during the dry season. Besides this finding, the study contains numerous aerosol properties for remote Amazonian aerosol as an important region of the world. From this perspective, the manuscript fits very well into the scope of the journal and deserves publication.

Before being acceptable for publication, the presentation of the important results needs to be improved. In its current state, there is no clear logical flow of the presentation of the results and the manuscript is more of a measurement report style. The following revisions or adaptions are requested:

The Introduction section requires a rearrangement concerning the pre-existing knowledge about the properties of the Amazonian aerosol and a better reference to what is published in literature. Actually, there is no clear storyline visible.

Response: We fully addressed this topic while addressing comment 1 from reviewer 1.

2. Review: *Overall, a more detailed discussion of the reported results in their relationship to the pre-existing knowledge is required. In its current version, the manuscript appears more like a measurement report which is focusing on the reporting of observations. For a research paper, it must become clearer what are the novel findings related to what we already know.*

Response: We fully addressed this topic while addressing comment 2 from reviewer 1.

3. Review: *The sub-sections of the results chapter lack a clear structure of presentation. It is difficult to identify the key findings among all the details listed here. A careful restructuring of the presentation is highly recommended.*

Response: We thank the reviewer for the opportunity to increase clarity in our results presentation and discussion. The whole section 3 was substantially restructured to be less descriptive, being able to better highlight the key findings of our study.

4. *Review: To allow the critical evaluation of the reported results, information about the uncertainties of the reported optical parameters is necessary. Respective values are reported for the chemical composition.*

Response: We fully addressed this topic while addressing comment 11 from reviewer 1.

Specific Comments:

Figure 6

5. *Review: In the scatter plot of modelled versus measured scattering coefficients there are several groups of data visible which follow different regression lines. Obviously, there is an impact of the particle diameter, but that effect is not discussed.*

Response: We thank the reviewer for pointing this out. We improved the discussion and included the observed effect.

Where it read:

“However, no clear particle size dependency was observed for the radiation scattering in our study (Figure 6).”

Not it reads:

“ No clear particle size dependency was observed for the radiation scattering in most of the cases (regression fitting under typical conditions, of aerosol sizes in the range of 100-150 nm), except at events dominated by ultra fine particles, at around 50 nm (Figure 6). This is notably an underestimation of observed scattering at lower particle diameters.”

Figure 7

6. *Review: The MSE of eBC particles is much higher than the MAE value. A more in-depth discussion is requested what would be the physical properties (size distribution, complex refractive index) which would lead to such a surprising result.*

Response: We fully addressed this topic while addressing comment 2 from reviewer 1. We additionally want to highlight that this result is remarkable, rather than surprising, as it reads now (after revisions) in section 3.3 of the results, in lines 408-411:

“The pronounced MSE of the eBC (7.62-13.58 $m^2\ g^{-1}$, Table 3) is strongly corroborated by other studies which found remarkably high scattering efficiency related to BC, especially when the particles undergo atmospheric processing and aging, such as in the case of our study (Bond and Bergstrom, 2006; He et al., 2015; Malm et al., 2005; Pitchford et al., 2007; Romshoo et al., 2021; Schwarz et al., 2006).”

Minor Issues:

7. *Review: The authors use the terms “Amazon” and “Amazonia” as synonyms. For the readability of the manuscript, it is recommended to use the term “Amazonia” which*

describes the region, while “Amazon” refers more to an online ordering service. Overall, a careful language check is recommended.

Response: We thank the reviewer for the suggestion, and carefully updated the manuscript to improve language quality.

8. Review: *Line 31: The author’s name is „Pöhlker”, please correct.*

Response: We thank the reviewer for pointing out this mistake. It has been corrected.

9. Review: *Line 44: Should this be” biosphere-atmosphere coupling” ?*

Response: We thank the reviewer for pointing out this mistake. It has been corrected.

10. Review: *Line 51: The term “climatic scenario” is unusual. How about “future climate scenarios” ?*

Response: We thank the reviewer for the suggestion. It indeed reads much better. The text was updated accordingly.

11. Review: *Line 55: the authors state that aerosol radiative forcing depends among other parameters also on particle origin. While all other mentioned properties are physically based, particle origin is not a criterion of this kind. The underlying effect arises from particle chemical composition and its effect on the complex refractive index. Please rephrase accordingly.*

Response: We thank the reviewer for the suggestions to improve clarity and exactitude of the sentence.

Where it read:

“The sign and magnitude of the ARI forcing are dependent on several parameters such as particle origin, size distribution, mixture and age, notably affecting the light-absorbing component of OA, termed brown carbon (Laskin et al., 2015; Saturno et al., 2018b).”

Now it reads:

“The sign and magnitude of the ARI forcing are dependent on several parameters such as particles size distribution, mixture, aging processes and meteorological conditions, as well as the particle chemical composition and its effect on the complex refractive index, based, among other factors, on the origin of the particles (Laskin et al., 2015; Li et al., 2024; Saturno et al., 2018a).”

12. Review: *Line 63 – 65: the sentence seems to be incomplete, please check.*

Response: We thank the reviewer for pointing out this mistake.

Where it read:

“One of the dominating isoprene SOA pathways in the Amazon is through the OH attack, leading to hydroperoxy radicals and subsequently via the HO_2 pathway (Shrivastava et al., 2019; Wennberg et al., 2018).”

Now it reads:

“One of the dominating isoprene SOA pathways in Amazonia is through the OH attack, leading to hydroperoxy radicals (Shrivastava et al., 2019; Wennberg et al., 2018).”

13. <u>Review:</u> *Line 78 - 81: The last paragraph of the introduction requires revision. First, when referring to intrinsic optical properties, do the authors mean intensive optical properties? This sentence seems to be incomplete and requires revision.*

<u>Response:</u> We thank the reviewer for pointing this out. The text was improved.

Where it read:

"However, the intrinsic optical properties of each aerosol species are still rare (Velazquez-Garcia et al., 2023), notably associated with OA origins (Ponczek et al., 2021)."

Now it reads:

"However, intensive optical properties of each aerosol species are still rare (Velazquez-Garcia et al., 2023), notably associated with OA origins (Ponczek et al., 2021) and with BC behaviour."

14. <u>Review:</u> *Line 90: Should it read "seasonal floodings" ?*

<u>Response:</u> We thank the reviewer for pointing out this mistake. It has been corrected.

15. <u>Review:</u> *Line 92: Please check the logic of this sentence.*

<u>Response:</u> We thank the reviewer for pointing this out. The text was improved.

Where it read:

"The seasonality at the region of this site in central Amazonia has been previously defined as the wet season from 1 December – 14 June, and the dry season from 15 June – 30 November (Andreae et al., 2015)."

Now it reads:

"The wet season in this region is typically from 1 December – 14 June, and the dry season from 15 June – 30 November (Andreae et al., 2015)."

<u>Review:</u> *Line 124: It should read "0.28 µg m^{-3}.*

<u>Response:</u> We thank the reviewer for pointing out this mistake. It has been corrected.

16. <u>Review:</u> *Line 165 – 166: This sentence is difficult to understand and requires a logical rearrangement.*

<u>Response:</u> We thank the reviewer for pointing this out. The text was improved.

Where it read:

"After rain events and other moments when the atmosphere is very clean, both α_s for σ_e values are very low, and therefore the ratio between them (SSA, Equation 4) becomes unrealistically high. We therefore calculated SSA for α_s for σ_e > 1 Mm^{-1}."

Now it reads:

"After rain events and other moments when the atmosphere is very clean, all the optical parameters are close to zero, and therefore the ratio between them becomes unrealistically high. We therefore calculated SSA only when σ_s and σ_e > 1 Mm^{-1}."

17. *<u>Review:</u> Section 2.4.2 needs re-arrangement since here, in contrast to the rest of the manuscript, numerous words are added in parenthesis. Please adjust to the style of the manuscript.*

<u>Response:</u> We thank the reviewer for pointing this out. The text was updated accordingly.

18. *<u>Review:</u> 13: Line 210: This sentence needs logical rearrangement.*

<u>Response:</u> We thank the reviewer for pointing this out. The text was improved.

Where it read:

"This is very similar to the wet season and wet to dry season transition in the same site (> 80% (Artaxo et al., 2013; Chen et al., 2015; Whitehead et al., 2016)), but high compared to other continental urban areas, such as across Europe (~30-50%, (Chen et al., 2022)) and lower than the strongly impacted by biomass burning region in Southwestern Amazonia (90%, (Brito et al., 2014)."

Now it reads:

"The PM1 aerosol composition was dominated by the organic fraction (77±5%, Table 1, Figure 2b), similar to what was found in the same site in wetter conditions (Artaxo et al., 2013; Chen et al., 2015; Whitehead et al., 2016), but lower than what was found in a region highly impacted by biomass burning in southwestern Amazonia (Brito et al., 2014). In continental urban areas, such as across Europe, the organic particles represented a much lower fraction of the total particles mass (Chen et al., 2022)."

19. *<u>Review:</u> Line 247: This sentence needs logical rearrangement.*

<u>Response:</u> We thank the reviewer for pointing this out. The text was improved.

Where it read:

"It presented the largest m/z 44 fraction (Figure 3), and therefore, this is the factor with the highest estimated O:C ratio (Chen et al., 2015)."

Now it reads:

"This factor has the highest estimated O:C ratio, which is evident in the observed m/z 44 fraction (Figure 3, note the different scales)."

20. *Review: Figure 3 requires the explanation of the relevance of m/z = 44.*

Response: We thank the reviewer for pointing out this need for clarification. We have now expanded the explanation in the manuscript as follows:

Where it read:

"The m/z 44 is formed mainly by the fragment CO_2^+, typical of the thermal decarboxylation of the organic acids groups (Alfarra et al., 2004)."

Now it reads:

"The m/z 44 signal predominantly arises from the CO_2^+ ion fragment, which is typically generated by thermal decarboxylation of carboxylic acid functional groups in organic aerosols (Alfarra et al., 2004). Therefore, m/z 44 serves as a valuable marker for the extent of aerosol oxidation and the presence of oxygenated organic compounds, providing insight into aerosol aging and secondary organic aerosol formation processes."

21. *Review: Line 264: What is meant by the IEPOX-SOA molecule?*

Response: We thank the reviewer for pointing this out. The text was improved.

Where it read:

"IEPOX-SOA molecule"

Now it reads:

"IEPOX-SOA particles"

22. *Review: 17: Line 316: Please rephrase "long-distance particles".*

Response: We thank the reviewer for pointing this out. The text was improved.

Where it read:

"long-distance particles"

Now it reads:
"particles originally emitted in distant regions"

23. *Review: Line 354: In Table 2, there are no numbers in parentheses, please correct.*

Response: We thank the reviewer for pointing out this mistake. It has been corrected.