The authors would like to thank the anonymous referee # 2 for taking the time to review the manuscript and for considering our study suitable for publication in ACP. We thank them for the suggested bibliography that allowed us to improve the discussion section of the manuscript. We hope that they'll find our responses to their comments adequate.

Below you will find the list of the referees' observations (bold), followed right after by the author's responses (normal font) and the respective changes made to the manuscript (italic), highlighting the sections that were modified.

1. Some studies have shown that the Single Particle Soot Photometer, when relying on scattering measurements to determine optical particle size and mixing state, can be influenced by microphysical characteristics. Please refer to relevant explanations for further clarification.

References

Wu, Y., Cheng, T., Zheng, L., Zhang, Y., and Zhang, L.: Particle size amplification of black carbon by scattering measurement due to morphology diversity, Environmental Research Letters, 18, 024 011, https://doi.org/10.1088/1748-9326/acaede, 2023.

Luo, J., Hu, M., Qiu, J., Li, K., He, H., Sun, Y., and Geng, X.: Technical note: Numerical quantification of the mixing states of partially-coated black carbon based on the single-particle soot photometer: Implication for global radiative forcing, EGUsphere [preprint], https://doi.org/10.5194/egusphere-2024-1155, 2024.

We thank the reviewer for pointing out the limitations of the diameters of BC-containing particles derived from the SP2 measurements. They indeed need to be considered when discussing the size distributions inferred from the scattering signals measured by the SP2. However, the analysis of the size distributions of coated particles and their scattering signal was reserved for a separate article focused on the microphysical properties of BC, which is currently in preparation. In the present study, we limited to analyzing the mass concentrations obtained from the SP2 (that rely in the incandescence signal emitted by the cores of BC) and their comparison to the absorption coefficients associated to them. The classification of mass concentrations based on the mean mass equivalent core diameters in Figure 6 is based on the peaking of the mass size distribution, which is estimated from the incandescence signal.

2. Some studies have also found that the Aethalometer model, when used for tracing the sources of black carbon, can be influenced by the microphysical properties of black carbon. Please cite relevant literature to support this statement.

References

Virkkula, A. (2021). Modeled source apportionment of black carbon particles coated with a light-scattering shell. Atmospheric Measurement Techniques, 14(5), 3707–3719.

Luo, J., Li, Z., Qiu, J., Zhang, Y., Fan, C., Li, L., Wu, H., Zhou, P., Li, K., and Zhang, Q.: The Simulated Source Apportionment of Light Absorbing Aerosols: Effects of Microphysical Properties of Partially-Coated Black Carbon, J. Geophys. Res.-Atmos., 128, e2022JD037291, https://doi.org/10.1029/2022JD037291, 2023.

Luo, J., Li, D., Wang, Y., Sun, D., Hou, W., Ren, J., Wu, H., Zhou, P., and Qiu, J.: Quantifying the effects of the microphysical properties of black carbon on the determination of brown carbon using measurements at multiple wavelengths, Atmos. Chem. Phys., 24, 427–448, https://doi.org/10.5194/acp-24-427-2024, 2024.

We thank the reviewer for enriching the discussion on the AAE with recent literature support. The following paragraph was included in the discussion of section 3.3.2.

"As demonstrated by Virkkula et al. (2021), Luo et al. (2023) and Luo et al. (2024), an added uncertainty needs to be considered when interpreting the results of the Aethalometer model, since AAE depends not only on the nature of the absorbing particles but also on several of its time-evolving microphysical parameters such as the mean core diameter, morphology, mixing state, coating thickness, coating material, among others. Since the aethalometer method heavily relies on the wavelength dependency of absorption, the variations of AAE due to the mentioned factors has shown to result in artificial positive and negative contributions of biomass burning (BB, also known as solid fuel combustion), even when no brown carbon (BrC) is present. In the present study, the AAE_{TR} *and* AAE_{BB} *values selected to apply based on the AAE frequency distribution giving values of 0.85 and 1.57, respectively. This meant using an* AAE_{TR} *lower than what is typically used in the literature. As modelled by Virkkula et al. (2021) and Luo et al. (2023), AAE<1 are typically associated to compact BC particles that changed their original aggregate morphology due to the coating of other materials onto their surface, which might not be representative of the main population of BC in the urban sites. Using these values of AAE resulted in higher contributions of BB to absorption than those obtained from the second method of absorption source apportionment performed in the present study. These contributions remained >0 even outside the biomass burning season and during the wet season, when traffic emissions are expected to be the main source of absorbing aerosol. Similarly, an incorrect attribution to BB was observed by Virkkula et al. (2021) and Luo et al. (2023), which was slightly higher when using the pair of lower AAE suggested by Zotter et al. (2017), 0.9 and 1.68, in the presence of non-absorbing material coating entirely (Virkkula, et al., 2021) or a fraction (Luo et al., 2023) of the BC cores. In the present study, when the average AAE of all the non-BB sources was used to represent traffic (*AAE_{TR}*=1), which also is closer to the mean of the frequency distribution of AAE, the agreement of both source apportionment methods improved. However, this change in* AAE_{TR} *resulted in negative contributions of BB to absorption of the order of ~5% during the wet season, period in which the removal efficiency of wet deposition is highest, and the presence of other combustion sources is minimum. This incorrect negative contribution of BB to absorption is in the range of the uncertainties associated to the application of the aethalometer method using an* AAE_{TR}*=1 for freshly emitted BC found by Luo et al. (2024)."*

3. Many researchers have utilized various instruments to measure and study black carbon in different regions. To further elaborate on the novelty of this paper, please discuss the unique characteristics of black carbon in the metropolitan area of La Paz and El Alto, Bolivia, compared to previous studies in other regions and similar settings.

We consider that the study site itself represents a novelty because little is reported on BC in most Latin American cities, and even less in high-altitude conditions. Moreover, the observed influence of BB in the absorption measured in these two Andean sites, despite being hundreds of kilometers away from the source and having a mountain chain in between, makes the sampling site even more unique. Despite the presence of two major sources of (potentially) absorbing aerosols, dust and BB, the frequency distributions of AAE were narrow and dominated by traffic-like combustion sources. As described in the manuscript, the exclusion of a large source of "potentially absorbing" particles, as the Altiplano dust, from being one of the contributors to absorption was not evident a priori, and to the best of our knowledge had not been evidenced before. Likewise, the MAC values estimated in the urban sites, validated through two methods of measuring the mass concentrations of BC, are within the lower range of MAC values found in the literature for urban background sites. These values quickly increase in their ~20 km journey to CHC-GAW towards the highest values of MAC reported in the literature. The estimated MACs and the size distributions of BC peaking below 80 nm raise up the

question if these characteristics are the result of combustion processes in extreme atmospheric conditions. Lastly, the influence of sources outside BB and traffic are rarely characterized in other sampling sites. Based on pre-existing studies of source apportionment performed in the same site, two main objectives were fulfilled. Firstly, the validation of the estimations of BB contribution to absorption obtained from the aethalometer method and the evaluation of different selection methods of representative AAE_{TR} and AAE_{BB}, obtaining similar uncertainties than those predicted by modeling studies. Secondly, the characteristics of the site allowed the implementation of an alternative way of physically characterizing multiple sources of absorbing particles through their contribution to absorption, and their respective AAE and absorption efficiency. Once more, to the best of our knowledge very few studies have been able to do so in ambient conditions.

4. What are the specific implications of black carbon measurements in this region for understanding regional and global climate, as well as air quality changes? Additionally, what recommendations would you make for future black carbon measurements?

In terms of regional and global climate, the observed rapid increase by a factor ~3 in the estimated MAC values needs to be further studied. The reasons for this observed increase require better understanding to be able to quantify by how much the enhanced absorption due to coating is underestimated. If common patterns are found amongst other regional sites, this absorption magnification would need to be considered in climate modeling. In terms of air quality, the expected reduced combustion efficiency could have a role in the observed concentrations of absorbing aerosols. Since high-altitude extreme conditions of hypoxia (leading to increased ventilation) increase the risks associated to air pollution exposure, understanding better the combustion processes and their main sources could help improve and or adjust the locally existing air quality policies.

Finally, absorption measurements from filter-based instruments are extremely susceptible to instrumental artifacts. For this, I would recommend measuring absorption using an independent method to validate the measurements in CHC-GAW to better understand the rapid increase of MAC coefficients, as well as ideally the onsite comparison of the absorption measured after denuding the absorbing particles. As suggested by Virkkula et al. (2021), absorption measurements are incomplete without a continuous monitoring of the size distribution of BC and their coating. I consider that based on the experience gained in the present study, prolonged measurements of rBC on the three sites would largely contribute to untangling the unanswered questions.

The extended discussion of the uniqueness of the LP-EA and CHC-GAW sampling sites, and the implications of our results in regional in global climate previously provided responding to questions 3 and 4 of reviewer #2 were summarized and included in the conclusions section of the manuscript as described below. We thank the reviewer for giving us the opportunity of expanding and improving this section through their questions.

"Long-term records of eBC concentrations and properties in the high-altitude cities of La Paz (LP) and El Alto (EA) were documented for first time. The average background concentrations of eBC in La Paz (LP) and El Alto (EA) are comparable amongst the sites and lower than the concentrations reported for other high-altitude Latin-American megacities. The different local meteorological conditions make the concentrations of eBC in EA much higher during the evening compared to LP. A factor of two of difference in magnitude can be observed between working and non-working days at both sites, as well as between the dry and the wet season, indicating the important role that weekly anthropogenic activities and meteorology play in modulating the eBC mass concentrations. The influence of the conurbation can also be observed at the global station CHC-GAW, located ~20 km away, showing concentrations that are roughly 35% of what is measured simultaneously at the urban area (within the

hours CHC-GAW is influenced by the urban mixing layer, ~10-16h). This illustrates the importance of the export of BC emissions from the LP-EA conurbation at the regional scale.

Despite the specific conditions of the urban sites, the intrinsic optical properties of BC in LP-EA are not fundamentally different than at other urban sites. The MAC values and the AAE estimated for both cities show that at an urban background level, eBC mass concentrations are dominated by relatively fresh vehicular emissions that do not undergo drastic ageing processes. Nevertheless, the peaking at low diameters of the BC mass size distributions in the city of La Paz is a phenomenon that is not expected at an urban background site and remains to be further investigated. Despite the presence of two major sources of (potentially) absorbing aerosols, dust and BB, the frequency distributions of AAE were narrow and dominated by traffic-like combustion sources. The relatively low estimated MACs and the size distributions of BC peaking below 80 nm raise up the question if these characteristics are the result of combustion processes in extreme atmospheric conditions and remains to be investigated. Understanding better the combustion processes in high-altitude and their main sources could help improve and or adjust the locally existing air quality policies.
MAC values quickly increase in their ~20 km journey to CHC-GAW towards the highest values of MAC reported in the literature. This observed rapid increase by a factor ~3 in the estimated MAC values needs to be further studied. If common patterns are found amongst other regional sites, this absorption magnification would need to be considered in climate modelling.

The main sources of absorbing particulate matter in LP-EA are rather local. Vehicular emissions are the first target to tackle from an air quality perspective as well as to reduce the impact these emissions can have on climate. Other sources contributing to emitting BC are open waste burning (particularly in EA) and regional agricultural biomass burning happening in the lowlands across the Andes. Identifying the influence of open waste burning in absorption is noteworthy since it occurs in many cities in developing countries, in Bolivia and elsewhere. No significant contribution to absorption from dust was observed in the visible-IR range, despite being reported by previous studies as one of the mains sources of PM_{10}, in the region.

It was observed that the Aethalometer method can overestimate the contributions of biomass burning, as predicted by previous studies, and more so in the presence of a third source of absorbing aerosol particles, such as open waste burning. The multilinear regression allowed to evaluate and constraint the results of the aethalometer method. In addition, it made possible the estimation of the source-specific MAC values, the source contribution to total absorption and the source-specific absorption Ångström exponent (AAE) of the sources directly or indirectly associated to vehicular emissions, biomass burning and open waste burning. We consider that this represents a relatively simple methodology for evaluating the results of the aethalometer method in measuring sites where a PMF analysis has been performed, and where concurrent influence of multiple sources can be expected.

Rigorous policies controlling the open waste burning and the size/state of the circulating vehicle fleet are therefore imperative to reduce the impact of BC on climate and on health of the inhabitants of the conurbation. Furthermore, the detection of ultrafine BC (Black Carbon) particles with exceptionally small diameters at an urban background site is a phenomenon that requires further investigation since it represents a potential higher risk of exposure to ultrafine particles of the local population."