Answer to referees

Two-year intercomparison of three methods for measuring black carbon concentration at a high-altitude research station in Europe

We thank the two reviewers for evaluating the manuscript and providing us constructive and useful comments.

Please find below reviewer comments in black and our responses in blue. The line numbers in the responses refer to the new version of the paper.

Anonymous Referee #1

Tinorua et al., 2024 "Two-year intercomparison of three methods for measuring black carbon concentration at a high-altitude research station in Europe" provides new findings on the uncertainties and specific artifacts encountered when using different techniques in determining the mass concentration of atmospheric black carbon. The manuscript is based on two years of atmospheric data and fits well with the scope of the journal. The text is well written and structured and derives rather consistent conclusions based on the analysis presented. This said, however, the analysis is rather superficial and presents mainly temporal variability of correlations and statistical uncertainty analysis. As such, the manuscript provides rather minor additions on top of the already published article by Tinorua et al., 2023 in ACP and does not evolve the analysis methodologies further towards the goals of this manuscript. The author should take advantage of the available size distribution data (existing based on Tinorua et al., 2023) and the measured aerosol optical properties (such as SSA and AAE) when evaluating the causes of the observed discrepancies in BC measurements. For example, when speculating on the ultrafine rBC particles from aviation (L342) or on the variability of the multiple scattering correction factor (P20-P21), these additional data could provide further insights for the underlying reasons behind the observations. Therefore, I would like to encourage the authors to incorporate into the analysis both the aerosol number size distribution and the aerosol single scattering albedo, as additional parameters to consider when different artifacts are evaluated.

REPLY:

Tinorua et al. (2024) is a manuscript on atmospheric processes occurring during 2019-2020 at the Pic du Midi. It describes the optical and microphysical properties of BC and shows the dependence of these properties with the boundary layer dynamics, the BC emission sources, transport pathways and chemical reactivity. The current paper is a technical manuscript that aims to highlight, quantify, and find the sources of the biases between the three most used methods to quantify BC mass concentrations. Thus, we do not believe that the current paper should be considered as an addition to Tinorua et al. (2024) but as a technical intercomparison study based on the same measurement campaign which could- but not only- be complementary to the ACP study.

We thank the reviewer for his relevant suggestion of exploring the aerosol particle size distribution to verify the presence of ultrafine particles from aviation, and thus explain the bias on M_{rBC} measured by the SP2. We plotted the aerosol size distribution as a function of the

M_{EC}/M_{rBC} ratio in Figure 7 (cf. Fig 7 below). This new Figure clearly shows a dominant presence of small particles below 20 nm when M_{EC}/M_{rBC} ratio reveals a significant positive bias. This mode is completely absent when the bias on M_{EC}/M_{rBC} ratio is negative or neutral.

A text on l. 355-359 has been added to describe Figure 7:

"To further investigate the role of small particles on the M_{rBC} bias, the aerosol number size distributions grouped by M_{EC}/M_{rBC} ratios has been plotted in Fig. S4 in the Supplement. The size distribution for which the highest ratios between M_{EC} and M_{rBC} has been observed clearly shows a contribution of small particles (<20 nm) up to 2 times higher than when the bias on the M_{EC}/M_{rBC} ratio is negative or neutral, supporting that the M_{rBC} underestimation compared to M_{EC} is probably due to undetected small rBC-containing particles."

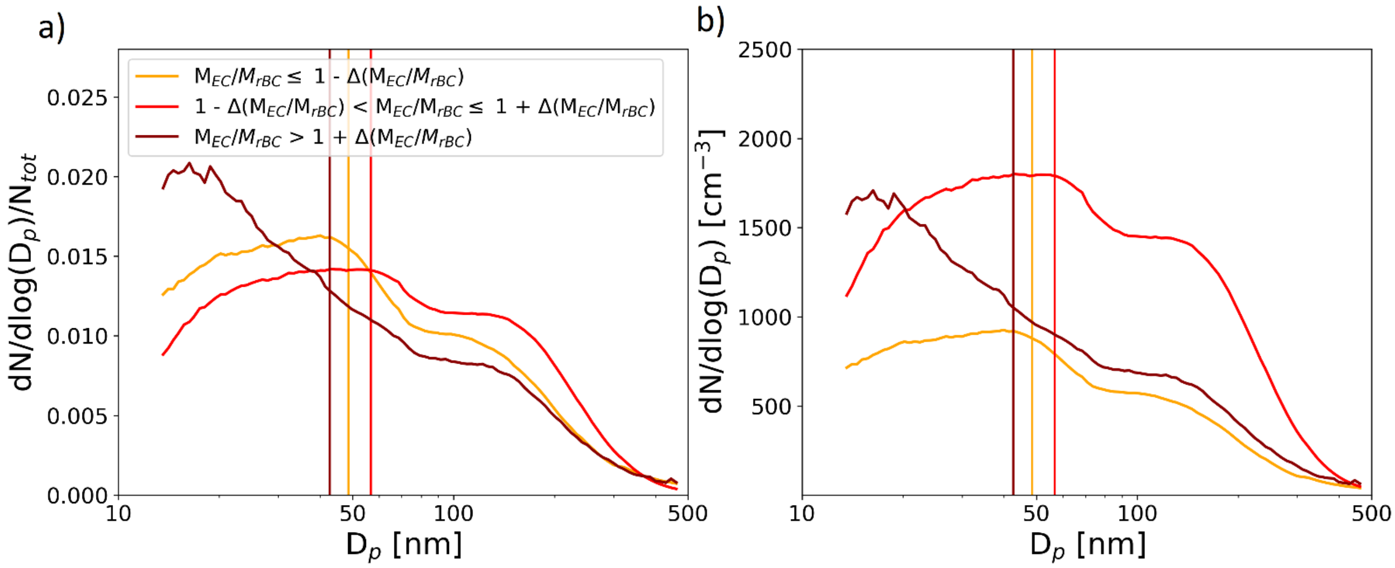

Figure 7: a) Number size distribution of aerosols measured by the SMPS, colored by M_{EC}/M_{rBC} ranges of values. b) is the same as a) but normalised by the total aerosol number concentration. Vertical lines highlight geometrical diameter corresponding to the color of the M_{EC}/M_{rBC} range.

Based on Tinorua et al., 2023, the measured aerosol SSA was rather high (>0.9). How does the CxMAC value depend on the aerosol optical properties and the aerosol particle size distribution? Do the mass correlations present additional dependence on them?

REPLY: Since the SSA is calculated with σ_{abs}, which is retrieved from σ_{ATN} using the C value, we believe that investigating the MAC*C dependency on the SSA is not relevant. We can notice that Yus-Díez et al. (2021) were able to show an increasing C value with the SSA because they had two different measurements of $\sigma_{abs,}$ which is not the case in our study.

In addition, I have some minor comments for the authors to consider, presented below.

Specific comments:

- Please, be specific when defining "MAC" – do you mean MAC of the aerosol or of the material black carbon? Especially in the introduction (lines 49-52) it is slightly confusing what is meant by MAC.

REPLY: The sentence in l. 48-50 has been modified as follows:

"Quantifying M_{eBC} acquired by optical methods is challenging because it requires the assumption of a BC mass absorption cross section (MAC) value translating the absorption coefficient (σ_{abs})."

- L54 Note that "optical method" could include also other than filter-based absorption measurements.

REPLY: This has been replaced in l. 54 by the term "filter-based optical methods".

- L 358-360 Consider simplifying and sharpening the key point of the sentence. For example, it seems rather intuitive that possibly "the SP2 missed the detection of a mode that is centered at lower diameter than the lower limit of detection of the SP2".

REPLY: The sentence in l. 368-369 has been modified as follows:

"(1) the extrapolation of the first mode peaking at ~ 100 nm is inaccurate for masses lower than 90 nm, which is the lower size detection limit of the SP2".

- L 367-368 Referring to a study by Wei et al., 2020, the MAC values provided here are now a bit different than in introduction, also specifying that the MAC is for BC (material?). Please double check this reference and the correct values.

REPLY: This mistake has been corrected in l. 377-378 as follows:

"Nonetheless a wide range of MAC of BC from 3.8 to 58 m^2 g^{-1} at 880 nm has been reported from field and laboratory measurements (Wei et al., 2020)."

- L388 Please provide a bit more information on the model and how it was applied, e.g. for which altitude and what meteorological data were utilized.

REPLY: Some details about the Hysplit model has been added in Section 2.3, l. 205-209 as follows:

"The Hysplit model (Hybrid Single-Particle Lagrangian Integrated Trajectory, Stein et al., 2015) has been used to retrieve the precipitation event along the 72-h trajectory of air masses arriving at the measurement site. The model was initialised to the PDM altitude, using 3-hourly atmospheric data of 1-degree spatial resolution from the Global Data Assimilation System (GDAS) of the National Centers for Environmental Prediction (NCEP).”

- L394 I would not recommend calling this observed behavior a “trend”. L394 Explain what “measurement artifact” is suspected to explain the difference.

REPLY: The sentence of l. 404-406 has been modified as follows:

“The absence of correlation between C×MAC and $\Delta M_{rBC}/\Delta CO$ in spring may be due to a measurement artifact during this season, such as the dominant presence of dusts particles which can affect the C correction of the AE33 (cf. Fig. 10a. and associated text).”

References :

Stein, A. F., Draxler, R. R., Rolph, G. D., Stunder, B. J. B., Cohen, M. D., & Ngan, F. (2015). NOAA’s HYSPLIT Atmospheric Transport and Dispersion Modeling System. *Bulletin of the American Meteorological Society*, *96*(12), 2059–2077. https://doi.org/10.1175/BAMS-D-14-00110.1

Tinorua, S., Denjean, C., Nabat, P., Bourrianne, T., Pont, V., Gheusi, F., & Leclerc, E. (2024). Higher absorption enhancement of black carbon in summer shown by 2-year measurements at the high-altitude mountain site of Pic du Midi Observatory in the French Pyrenees. *Atmospheric Chemistry and Physics*, *24*(3), 1801–1824. https://doi.org/10.5194/acp-24-1801-2024

Wei, X., Zhu, Y., Hu, J., Liu, C., Ge, X., Guo, S., Liu, D., Liao, H., & Wang, H. (2020). Recent Progress in Impacts of Mixing State on Optical Properties of Black Carbon Aerosol. *Current Pollution Reports*, *6*(4), 380–398. https://doi.org/10.1007/s40726-020-00158-0

Yus-Díez, J., Bernardoni, V., Močnik, G., Alastuey, A., Ciniglia, D., Ivančič, M., Querol, X., Perez, N., Reche, C., Rigler, M., Vecchi, R., Valentini, S., & Pandolfi, M. (2021). Determination of the multiple-scattering correction factor and its cross-sensitivity to scattering and wavelength dependence for different AE33 Aethalometer filter tapes: A multi-instrumental approach. *Atmospheric Measurement Techniques*, *14*(10), 6335–6355. https://doi.org/10.5194/amt-14-6335-2021