Responses to the comments of the Editor

We thank the Editor for the valuable comments, which have greatly helped us to further improve the manuscript. Please find below our point-by-point responses (in blue normal font) to your feedbacks (*in black italic*). The changes in the revised manuscript are in green.

Dear Authors,

I have reviewed the comments from three referees and your responses. While two of them recommended this work to be published in Atmospheric Chemistry and Physics, Referee #3 has pointed out the technical issues for quantifying BC absorption enhancement due to lensing effect using the filter-based measurement techniques that can affect the subsequent argument regarding suppression of lensing effect by BrC at the shorter wavelength. Overall, I find this manuscript presents valuable observations to advance our understanding on atmospheric BC and BrC absorptions and to address the potential impacts of BC-BrC interactions on total aerosol absorptions. Nevertheless, I would like to make two major comments on the revised version that are related to some of the comments from Referee #3 before considering this work to be published in Atmospheric Chemistry and Physics.

We thank the Editor for the evaluation of our revised manuscript and constructive comments on remaining issues. We are confident that we have fully addressed these issues and hope that the modified version of the manuscript can be accepted in ACP.

1) Section 3.5: The authors speculate that the observed BC absorption enhancement (Eabs) is primarily due to the lensing effect of BC coatings (line 365). However, it is possible that only a fraction of BC coating proxy (e.g., OOA+BBOA+NO3+SO4) are internally mixed with airborne BC. I suggest the authors to further comment whether the filter-based Eabs can be also caused by those chemical components that are externally mixed with BC and perhaps other factors such as calibration uncertainties and particle morphology. If so, the filter-based BC lensing effects (or Eabs) can be fundamentally different from the "true" lensing effects for airborne BC, and hence the two terms should not be interchangeable although the filter-based Eabs values are not significantly different from those reported in the literature based on in-situ measurements (lines 395-398). If this is the case, it is strongly recommended to use the operational-defined term "Filter-based Eabs" all the time (including the sub-heading) to avoid over-interpretation of data/misleading conclusion.

We appreciate the Editor's comment. Indeed, only a fraction of the proxy is expected to be internally mixed with BC, as not all particles contain a BC "core". This is already stated in lines 396-397 (in the newly revised manuscript), but does not affect our conclusions.

The Editor questions whether E_{abs} can be affected by uncertainties in the calibration of our filter-based measurements or be caused by the same non-refractory chemical components, but that are externally mixed with BC. We have now investigated the dependence of the calibration coefficients, C_{660nm}, on the proxy, for 18 PM_{10} samples measured with MWAA, and found no relationship: the Pearson's r is -0.08 and the slope is not significantly different from zero (p = 0.74). This suggests that uncertainties in the calibration might not affect the relationship between filter-based E_{abs} and the proxy. We cannot fully exclude the contribution of externally mixed non-refractory species deposited on the filters to the E_{abs}, even if the E_{abs} values we found are consistent with those determined based on in-situ measurements (lines 405-406 in the newly revised manuscript). Therefore, we have followed the Editor's suggestion and added "filter-based" next to lensing or E_{abs} throughout the manuscript text (including sub-headings), in order to provide a clear distinction between in-situ- and filter-based lensing and to avoid over-interpretation. The second paragraph of Sect. 3.5. has been modified as follows:

Figure 4c presents MAC_{BC} as a function of a proxy for the BC coating thickness, i.e., the ratio between the combined concentrations of major-SIA, OOA and BBOA and EC (NR-PM/EC; Table S4). While the variability in MAC_{BC} is not driven by the EC sources (Fig. 4b), MAC_{BC} increases linearly with NR-PM/EC < 33 consistently, unlike for other tested proxies (including the total OA mass, OA:EC, OOA:OA, OOA:EC or Sulfate:EC), indicating a filter-based BC lensing effect due to coating by multiple non-refractory components. The presence of coated BC particles is supported by the observation of compacted BC particles from SEM measurements (Fig. S13). We have examined the relationship between C_{660nm} and the proxy and found them to be independent, indicating that uncertainties in the Aethalometer calibration do not affect the resulting relationships between E_{abs} and the proxy. While we attribute the filter-based (apparent) BC absorption enhancement to lensing, future studies should evaluate its potential dependence on chemical components that are externally mixed with BC, including tar-balls absorbing at longer wavelengths (Sect. 3.4), as well as calibration uncertainties and/or the deposited particle morphology.

2) Section 3.6 and 3.7: While the suppression of filter-based BC lensing induced by BrC coatings is a novel observation that can provide important insight into our understanding of BC-BrC interaction and total aerosol absorption as well as uncertainties of filter-based measurements for quantifying BrC absorption, I agree with Referee #3 that the closure calculation involves many assumptions and simplified concepts that may lead to over-interpretation of results. Overall, I think it is still important to report the possibility of filter-based BC lensing suppression due to BrC coatings and such data analysis approach to the scientific communities for future research but the authors should tone down some of the related arguments/conclusion throughout the manuscript. Both quantifiable and non-quantifiable uncertainties described in Appendix C should be clearly presented in the main text to ensure the reader can easily recognize the major uncertainties and limitation of the closure calculation.

Alex

We agree with the Editor. as we had done in the revised abstract, we have now replaced the word “evidence” by “indication” throughout the main text when discussing the “inferred” filter-based lensing suppression at shorter wavelengths. While quantifiable uncertainties are already discussed in Sect. 3.7 together with Fig. 8 (lines 474-476 in the newly revised manuscript), we have now added in the end of Sect. 3.7 a sentence commenting also on potential unquantifiable uncertainties and referring to Appendix C for a more detailed discussion:

“[…] Finally, we note that our optical closure is limited in terms of interpretation of lensing effects, due to unquantifiable uncertainties potentially associated with filter sampling artifacts, possible chemical interactions between airborne BrC molecules or with BC, and the use of simplified Mie calculations to obtain the particulate BrC absorption (Appendix C).”

We have also added in Sect. 4 the following sentence, to provide a more balanced discussion on the implications of our findings as suggested by the Editor:

“While the optical absorption closure approach presented here involves multiple assumptions and simplified concepts, our results provide useful experimental insights into understanding BrC/BC interactions and total atmospheric aerosol absorption, as well as uncertainties of filter-based measurements for quantifying BrC absorption. If lensing suppression occurs due to internal mixing of BC and BrC as is apparently the case for many samples in our study […]”