Dear editor:

We sincerely thank the editor for the careful evaluation and valuable suggestions.

Regarding your concern about "(1) the extrapolation between wavelengths using AAE", we have strengthened the discussion of the associated uncertainty and the limitations of our framework in the revised manuscript and the Supplement, as detailed below.

Regarding your concern about "(2) the temporal mismatch between long-time-average filters and high PAX measurements", we have clarified that the PAX data were averaged over the same 23.5 h window represented by each filter sample. We have also added an explicit discussion in Sect. 2.4.1 of the manuscript and Sect. S5 of the Supplement acknowledging that unresolved intraday covariance between AAE_{coated} and $B_{\mathrm{abs\_coated}}(870)$ may introduce bias when a single daily AAE_{coated} is applied to the corresponding daily-averaged PAX value. Accordingly, our interpretation is restricted to daily and longer time scales.

Detailed point-by-point responses to reviewer comments are provided below (in blue). We believe that the revised manuscript has been substantially strengthened and now meets the high standards of *Atmospheric Chemistry and Physics*. We sincerely appreciate your valuable guidance and careful consideration throughout the review process.

Yours sincerely,

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Response to Reviewer #1

General comment:

This work has high potential merit for deconvolving the affect of mixing state and various species to the optical properties of urban aerosol. It represents a creative approach to bridging the gap between techniques that are sensitive to different morphologies and wavelengths. However, in its current state, this manuscript remains incomplete.

We sincerely appreciate the reviewer's positive assessment of the creativity and potential value of this work. We also appreciate the reviewer for identifying the remaining limitations. In the revised manuscript and Supplement, we have strengthened the discussion of the methodological uncertainties, the implications of daily averaging, and the interpretation of the short-wavelength BrC blocking signal. Detailed point-by-point responses are provided below (in blue).

Minor Comment:

1. The thermal/optical analysis has a significant wavelength and loading dependent adjustment factor which introduces uncertainty in to the AAEcoated calculation.

We thank the reviewer for raising this important concern. Multiple scattering by the filter matrix and particle-loading effects can bias filter-based attenuation, and the magnitude of these effects is wavelength- and loading-dependent (Arnott et al., 2005). To reduce variability associated with the filter substrate, filters from the same batch were used for both the thermal/optical analysis and the integrating-sphere measurements. We emphasize, however, that using the same filter batch does not eliminate filter artefacts; it only reduces an additional source of substrate-to-substrate variability.

In addition, we applied the wavelength- and EC-loading-dependent attenuation correction factors of Chow et al. (2021) to reduce loading artefacts. These empirical factors were developed from the CSN dataset (n = 23,904), spanning a broad range of EC loadings, and were designed to restore the attenuation–EC relationship toward the approximately linear behaviour expected after correction. In our data, the corrected seven-wavelength absorption spectrum is well represented by the fitted power-law relationship (Figure S3), providing an internal consistency check for the derived AAE_{coated}. Nevertheless, this correction cannot remove all uncertainties inherent to filter-based optical measurements. We have now made this implication explicit in Sect. 3.4 of the manuscript and Sect. S7 of the Supplement: residual filter-related systematic effects in AAE_{coated} can propagate into the extrapolated $B_{abs\_coated}(\lambda)$, $B_{abs,lensing}(\lambda)$, and $E_{abs}(\lambda)$, with greater sensitivity at wavelengths farther from the directly measured 870 nm PAX channel. Conclusion point (5) was revised accordingly.

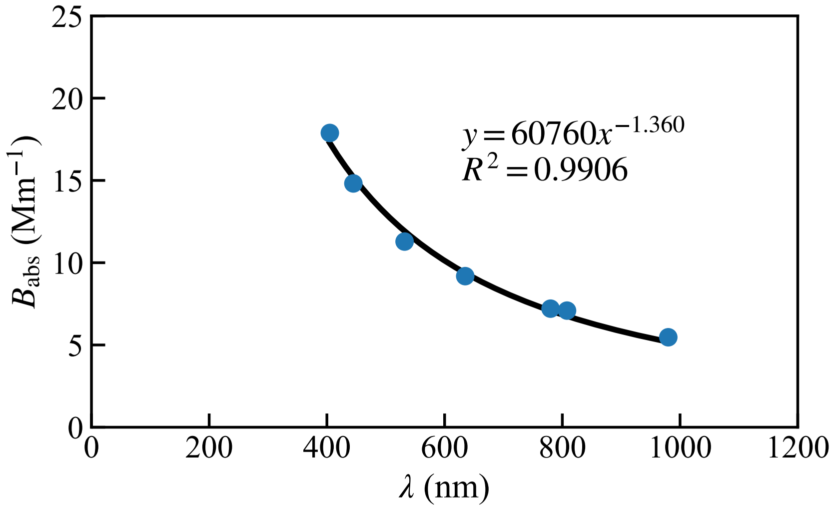

Figure S3 Absorption coefficients and the fitted curve measured by the DRI Model 2015 Multi-Wavelength Thermal/Optical Carbon Analyser on 19 February 2023.

2. There should be a discussion of the affect of using other BrC surrogates besides humic acid would have in the DWIC determination. E.g. suwanee river fulvic acid or nitroaromatic compounds.

We appreciate this valuable comment. We agree that the choice of BrC surrogate can affect the quantitative DWIC partitioning because atmospheric BrC is chemically and optically diverse, and no single standard can reproduce the full range of its spectral properties. Kwon et al. (2018) compared the optical properties of humic-material standards, including HASS and SRFA, while Lin et al. (2016) showed that nitroaromatic compounds can be important BrC chromophores, particularly at near-UV and visible wavelengths. These studies support the reviewer's point that alternative BrC standards may yield different spectral calibration behaviour.

In principle, another BrC surrogate, such as SRFA, could be used, but its wavelength-specific calibration relationships at 420 and 650 nm would need to be established under the same ISS conditions and be sufficiently distinct from those of

CarB for stable dual-wavelength separation. Thus, a higher AAE than CarB alone is not sufficient to guarantee quantitatively identical DWIC results. Nitroaromatic compounds further illustrate the diversity of atmospheric BrC and would likewise require appropriate wavelength-specific calibration if used as standards. We have revised Sect. S4 of the Supplement accordingly and added Lin et al. (2016) to the Supplement references.

Because the surrogate choice can propagate into the derived BC/BrC partitioning, we now treat it explicitly as a source of methodological uncertainty. Consistently, Conclusion point (5) has been revised to state: "In addition, further work is required to better constrain the influence of BrC chemical composition and filter-based measurement uncertainties, including potential positive and negative artefacts and changes in particle morphology during sampling." (Lines 526–529).

3. There is still no discussion on how the daily filters may misrepresent optical properties that change throughout the day.

We appreciate the reviewer's attention to this critical issue, and we agree that our previous wording did not fully explain the consequence of intraday variability. The online PAX observations and offline ISS measurements are placed on the same 23.5 h daily basis, so our analysis is designed to compare daily-integrated/daily-averaged aerosol optical properties, not to reproduce their diurnal evolution. Nevertheless, a limitation remains: AAE_{coated} is derived from an integrated daily filter, whereas PAX measures $B_{\mathrm{abs\_coated}}(870)$ at high temporal resolution before it is averaged over the corresponding filter-sampling period. If AAE_{coated} and $B_{\mathrm{abs\_coated}}(870)$ covary substantially within a day, applying one daily AAE_{coated} to the daily-averaged PAX value can smooth that covariance and may introduce bias into the reconstructed daily spectrum. We have now added this limitation explicitly in Sect. 2.4.1 of the manuscript and Sect. S5 of the Supplement. Accordingly, we restrict our interpretation to daily and longer (seasonal and annual) time scales and do not make claims about diurnal changes in aerosol optical properties.

4. Section 4 conclusion #3 is overstated. The uncertainty analysis, which in my opinion is incomplete, shows that this technique has an uncertainty of ~12%, which means an Eabs value of <1 is only probable at the shortest wavelengths, which is extrapolated beyond the wavelengths used to calculated AAEcoated (405 - 980 nm.)

We thank the reviewer for highlighting this important issue. We agree that Conclusion point (3) was overstated. In particular, E_{abs} values below 1 occur only at the shortest wavelengths, where $B_{abs\_coated}(\lambda)$ is extrapolated using AAE_{coated} derived from the 405–980 nm filter measurements, and the uncertainty analysis gives an uncertainty of approximately 12.4% at 420 nm. We have therefore revised the manuscript at three levels. In the Abstract, the spectral contraction is now described as "a pattern suggestive of a BrC 'blocking effect'" and the field measurements as providing "observational indications consistent with this blocking effect." In Sect. 3.3.1, the short-wavelength decline in E_{abs} is described as being "likely associated with" BrC blocking, and the field result as "suggestive field-based evidence consistent with this phenomenon rather than definitive proof of the underlying mechanism." In Conclusion point (3), the heading is now "Observational Indication of the BrC Blocking Effect," and we explicitly state that further investigations are needed to directly verify the controlling processes. We have also added cautionary discussion in Sect. 3.4 and Sect. S7 of the Supplement regarding residual filter-related uncertainties. Together, these changes make clear that the short-wavelength results are suggestive rather than definitive evidence for the blocking mechanism.

5. Finally, the foundational value of this work, babs_coated, is based on applying an AAE from filter-based absorption measurements to an in situ measurement. Filter based measurements can have both positive and negative artifacts and may change the morphology of the particles during sampling. While this shortcoming is acknowledged in the final section of the paper, there is no discussion of the impacts on the conclusions.

We appreciate this important comment. We agree that this limitation has a direct bearing on the wavelength-resolved conclusions. The PAX measurement of

$B_{abs\_coated}(870)$ itself is in situ, but $B_{abs\_coated}(\lambda)$ at other wavelengths depends on the filter-derived AAE_{coated}. Consequently, residual filter artefacts in AAE_{coated}, including multiple-scattering/loading effects not fully removed by the empirical correction and possible changes in particle morphology during collection, can propagate into the reconstructed $B_{abs\_coated}(\lambda)$, and hence into $B_{abs,lensing}(\lambda)$ and $E_{abs}(\lambda)$. The conclusions that depend most strongly on the extrapolated spectral shape, especially those at the shortest wavelengths, are therefore more sensitive to these uncertainties than the directly measured 870 nm absorption. The use of the same filter batch and the Chow et al. (2021) loading correction reduces, but does not eliminate, these effects.

To make this impact explicit, we have added corresponding discussion to Sect. 3.4 of the manuscript and Sect. S7 of the Supplement. We also revised Conclusion point (5) to state: "In addition, further work is required to better constrain the influence of BrC chemical composition and filter-based measurement uncertainties, including potential positive and negative artefacts and changes in particle morphology during sampling." In light of these limitations, we interpret the exact magnitude and spectral dependence of the derived enhancement cautiously, particularly at wavelengths farthest from the directly measured PAX wavelength.

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