Experimental evidence of the lensing effect suppression for atmospheric black carbon containing brown coatings

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Accounting for the wavelength- and source-dependent optical absorption properties of the abundant light-absorbing organic (brown) carbon (BrC) and the mixing state of atmospheric black carbon (BC) are essential to reduce the large uncertainty in aerosol radiative forcing. Estimation of BrC absorption online by subtraction is highly uncertain and may be biased if not decoupled from the potential BC absorption enhancement (lensing) due to non-refractory (organic and inorganic) coating acquisition.

Here, the reported total particulate absorption is based on long-term, filter-based seven-wavelength Aethalometer (AE33 model) data, corrected for multiple scattering effects with Multi-Wavelength Absorbance Analyzer (5λ MWAA) measurements. Using ultraviolet-visible spectroscopy absorbance measurements along with particle size distributions obtained by a scanning mobility particle sizer, we have conducted Mie calculations to assess the importance of source-specific extractable particulate BrC (Moschos et al., 2018) versus BC absorption.

For the species-specific optical closure, the wavelength dependence of bare BC absorption is estimated using MWAA measurements upon successive filter extractions to remove the influence of BrC/coatings. The lensing contribution, supported by observations from field-emission scanning electron microscopy coupled with energy-dispersive X-ray spectroscopy, is estimated at longer wavelengths using a refined proxy for the BC coating thickness. The approach is validated independently by applying a novel positive matrix factorization-based approach on the calibrated total AE33 absorption data.

Based on the observational constraints established in this study, we demonstrate for various distinct case studies that the interplay between lensing and BrC absorption results in lower than expected BC absorption at shorter wavelengths. This indicates that the volume additivity assumption is not valid for particulate absorption by internally mixed heterogeneous atmospheric aerosol populations. These comprehensive experimental analyses verify the BC lensing
suppression predicted for simplified core-shell structures containing moderately absorbing BrC (Lack & Cappa, 2010). The implications discussed in this work are relevant for co-emitted species from biomass burning or aged plumes with high BrC to BC mass/absorption ratio.

References
