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Optical and chemical properties of wintertime light-absorbing aerosols in the eastern Indo-Gangetic Plain, India

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Light-absorbing carbonaceous aerosols such as black and brown carbon (BC and BrC) and humic-like substances (HULIS) have pronounced effects on the earth's radiative balance and tropospheric photochemistry. In India, large heterogeneities exist for BC and organic carbon (OC) emission inventories, which necessitates regionally-representative ground-based measurements. Such measurements are spatially scattered for BC, rare for BrC and non-existent for HULIS. This severely limits a robust understanding of the optical and chemical properties of these aerosols, and consequently, their climate effects. To address this issue, the present study reports optical and chemical properties of wintertime (December 2018-February 2019) BC, BrC and HULIS at a rural receptor site in the highly polluted eastern Indo-Gangetic Plain (IGP), India. A 7 wavelength aethalometer was deployed to measure time-resolved BC mass concentration, and absorption coefficients (b_{abs}) and Angstrom exponent (AE) of BrC. Separation of aqueous and organic BrC (BrC_{aq} and BrC_{org}) and HULIS fractions via a multi-step chemical extraction procedure followed by optical measurements (UV-Vis, fluorescence and FT-IR), and supplementary measurements of OC, water-soluble organic carbon (WSOC) and ionic species led to better insights into the potential chromophore composition and their relative importance in constraining aerosol optical properties.

The daily averaged BC mass concentration was 15.4±9.5 μg m^{-3} during winter, where the biomass burning (BB) contribution was 25±5%. The diurnal profile of BC_{BB} and BrC light absorption coefficient (b_{abs_BrC}) showed a prominent morning peak (0700-0800 H) characterized by mixed fossil fuel and biofuel emission and a gradual increase towards night due to enhanced primary BB emission from cooking activities and lowering of the mixing depth. The regionally transported BB plume from northwestern IGP contributed substantial BC and BrC to this receptor location in the eastern end of the corridor, which was supported by concentration-weighted air mass trajectories (CWTs).

The BrC_{org} light absorption at 365 nm ($b_{abs\_BrC\_org}$) was almost 2 times compared to that of BrC_{aq} ($b_{abs\_BrC\_aq}$) (36±7.1 vs 18.3±4.3 Mm^{-1}), which suggested a dominance of non-polar polyconjugated BrC chromophores. This was also supported by the increasing trend of water-insoluble BrC from 49±10% at 365 nm to 64±21% at 550 nm, with averaged contributions of 49±8% at 300-400 nm and 67±9% at 400-550 nm, respectively. A strong correlation between WSOC and NO_3^- (r=0.78, p<0.01)

and WSOC and NH_4^+ ($r=0.63$, $p<0.01$) indicated the possibility of nighttime secondary organic aerosol formation. A prominent fluorescence peak at ~409 nm for BrC_{aq} confirmed the presence of HULIS, and $b_{abs\_BrC\_aq}$ was dominated by the low-polarity HULIS-n fraction. AE of individual HULIS fractions increased by 7-36% towards the more polar HULIS-a and highly-polar water-soluble organic matter (HPWSOM) compared to the less polar HULIS-n for the 300-700 nm range. Distinct FTIR peaks at 3400 cm^{-1}, 1710 cm^{-1} and 1643 cm^{-1} suggested abundance of C-H, C=O and C=C functional groups, respectively, in the BrC chromophores. Overall, it appeared that the regionally transported BB plume significantly enriches BrC and HULIS in the eastern part of the IGP corridor.