Brownness of Organics in Anthropogenic Biomass Burning Aerosols over South Asia

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Abstract

In South Asia, biomass is burned for energy and waste disposal, producing brown carbon (BrC) aerosols whose climatic impacts are highly uncertain. To assess these impacts, a real-world understanding of BrC's physio-optical properties is essential. For this region, the order-of-magnitude variability in BrC's spectral refractive index as a function of particle volatility distribution is poorly understood. This leads to oversimplified model parameterization and subsequent uncertainty in regional radiative forcing. Here we used the field-collected aerosol samples from major anthropogenic biomass activities to examine the methanol-soluble BrC optical properties. We show a strong relation between the absorption strength, wavelength dependence, and thermo-optical fractions of carbonaceous aerosols. Our observations show strongly absorbing BrC near the Himalayan foothills that may accelerate glacier melt, further highlighting the limitations of climate models where variable BrC properties are not considered. These findings provide crucial inputs for refining climate models and developing effective regional strategies to mitigate BrC.

Keywords: Brown carbon, Biomass fuels, Imaginary refractive index, Organic carbon

1. Introduction

Carbonaceous aerosols, such as black and organic carbon, make up most fine particulate matter ($PM_{2.5}$) emissions globally (Mcduffie et al., 2020; Roy et al., 2023; Kurokawa and Ohara, 2020; Crippa et al., 2018) and ~40% over South Asia (Tibrewal et al., 2024; Pandey et al., 2014; Sadavarte et al., 2019). Anthropogenic biomass usage for residential cooking and heating (Pandey et al., 2014; Habib et al., 2023; Navinya et al., 2023), residue burning for agricultural waste disposal (Kapoor et al., 2023b; Azhar et al., 2019) and biomass-fired brick kilns (Weyant et al., 2014; Tibrewal et al., 2023) are the common sources of these carbonaceous aerosols across South Asia (Tibrewal et al., 2024; Pandey et al., 2014; Sadavarte et al., 2019; Ohara et al., 2007) and many other developing countries (Bonjour et al., 2013; Yevich and Logan, 2003; Mcduffie et al., 2020). These aerosols perturb the Earth's energy balance, depending on their mixing state, size distribution, wavelength dependence of optical properties, and absorption

strength (Zhang et al., 2020; Neyestani and Saleh, 2022; Brown et al., 2018; Arola et al., 2015; Bond and Bergstrom, 2006). However, the extent of this perturbation remains uncertain (Szopa et al., 2021; Gliß et al., 2021). Over the last two decades, extensive research has focused on the climate impact of highly absorbing black carbon (BC) (Bond et al., 2013). In contrast, the climate implications of light-absorbing organic carbon (OC), termed brown carbon (BrC), have received relatively less attention and are thus less certain (Saleh et al., 2018; Brown et al., 2018; Saleh, 2020). The chemical composition of BrC varies significantly, and consequently its optical properties, as reported across previous studies, span orders of magnitude in the imaginary refractive index (k) values that determine its light absorbing strength (Chakrabarty et al., 2023; Choudhary et al., 2021, 2018, 2017; Dey et al., 2021; Kapoor et al., 2023a; Kirillova et al., 2016; Rana et al., 2020; Rathod et al., 2017; Saleh et al., 2018, 2014; Srinivas and Sarin, 2014). Previous studies often measured aged ambient BrC that are weakly absorbing ($k_{BrC,550}$ <0.01) due to photobleaching (Sumlin et al., 2017), hence some climate impact assessment studies have considered BrC as a weakly or non-absorbing particle (Lee et al., 2010; Sand et al., 2021; Zhang et al., 2020). However, this underestimates the impact of freshly emitted BrC that has high absorption strength ($k_{BrC,550}$ > 0.1) and resists photobleaching, resulting in extended atmospheric lifetime (Chakrabarty et al., 2023). Furthermore, the formation of light-absorbing secondary BrC and the enhancement of BC absorption due to OC coating (Rastogi et al., 2021; Bhowmik et al., 2024; Kapoor et al., 2022) add complexity to radiative transfer models.

BrC has a wide range of absorption strength; studies show $k_{BrC,550}$ varying from ~0.007 (Islam et al., 2022) to ~0.2 (Chakrabarty et al., 2023). In addition to the different methods used to derive BrC optical information, such variation is associated with the different combustion conditions (Saleh et al., 2018), ageing of BrC (Sumlin et al., 2017; Dasari et al., 2019; Romonosky et al., 2016; Chen et al., 2021), and secondary reactions (Wang et al., 2020; Kroll et al., 2007; Kroll and Seinfeld, 2008; Hecobian et al., 2010). An experimental study explained that the progressive transformation of BC precursors to BC results from different combustion conditions, which create the BrC-BC light-absorption continuum (Saleh et al., 2018). This continuum shows an increase in the absorption strength of carbonaceous aerosols that is associated with a decrease in wavelength dependence (w), solubility, and volatility (Saleh, 2020). Recent studies also observed such a relationship, but for a smaller range of k_{BrC} (<0.01) (Devaprasad et al., 2024; Luo et al., 2022). However, information about real-world source specific BrC absorption and its position in the BrC-BC continuum is lacking. Understanding this light absorption continuum alongside carbonaceous aerosol emissions aids BrC parameterization in climate models (Zhang et al., 2020; Saleh et al., 2014). Presently, because source specific BrC information is absent from emission inventories, many climate models inadequately account for BrC. Studies have used the BrC to BC ratio along with the k_{BrC} to understand its direct radiative effect (Park et al., 2010; Feng et al., 2013). Furthermore, other studies (Zhang et al., 2020; Neyestani and Saleh, 2022; Brown et al., 2018) have employed BrC parameterization schemes based on laboratory-generated data to address the climate impact of BrC, but this approach might not adequately represent real-world biomass burning conditions (Saleh et al., 2014; Lu et al., 2015). Hence, regions with high OC emissions and stronger BrC (S-BrC), also known as dark BrC ($k_{BrC,550}$ > 0.1), could have a high climate impact

due to persistent BrC, possibly underestimated in the absence of regional source specific BrC data.

The recent Carbonaceous Aerosol Emissions, Source Apportionment and Climate Impacts (COALESCE) field emission measurement campaigns and questionnaire surveys in India (Navinya et al., 2023; Kapoor et al., 2023b; Tibrewal et al., 2023; Habib et al., 2023) have prepared a comprehensive inventory encompassing both formal (transportation, industries and power generation) and informal (residential, agricultural residue burning, and brick production) emission sectors (Venkataraman et al., 2020; Tibrewal et al., 2024). It recognizes the substantial contribution of anthropogenic $PM_{2.5}$ in India arising from biomass fuel burning practices for residential cooking and agricultural residue burning (Kapoor et al., 2023b; Tibrewal et al., 2024; Habib et al., 2023). Recent studies have highlighted considerable biomass consumption for residential heating and brick production (Tibrewal et al., 2023; Navinya et al., 2023). Figure 1 shows 91% of the OC emissions (3 Tg y^{-1}) over India are from three sources: residential cooking (COOK), heating (HEAT), and agricultural residue burning (AGRI), with most emissions from the Indo-Gangetic plain (~50%) (Tibrewal et al., 2024). The unexplored climate impacts of OC emitted from these biomass-based sources make the Indian subcontinent particularly prone to environmental challenges.

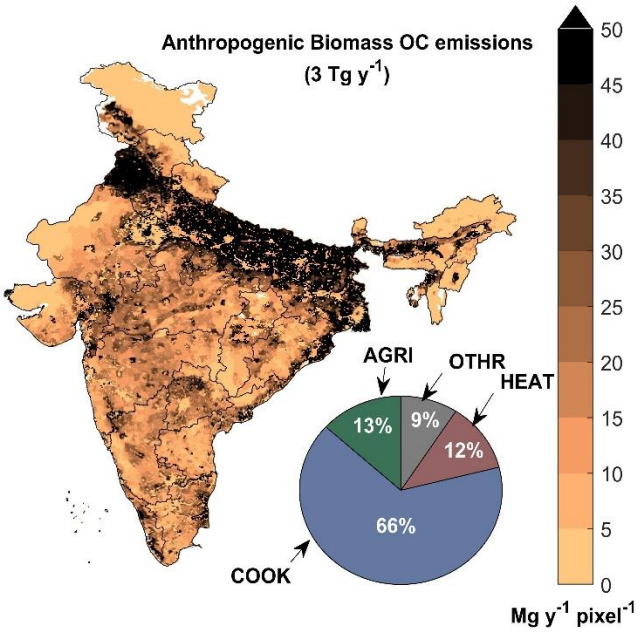

Figure 1. The spatial distribution of OC emissions (Mg y^{-1} $pixel^{-1}$) from three major sources—agricultural residue burning (AGRI), residential cooking (COOK) and residential heating (HEAT)—covers ~91% of the total OC emissions (3.3 Tg y^{-1}) over India. OC emissions are taken from the Carbonaceous Aerosol Emissions, Source Apportionment and Climate Impacts (COALESCE) Speciated Multipollutant Generator (SMoG)-India emission inventory for the year 2019 (Tibrewal et al., 2024). Here, the pixel size is 5×5 km. The pie chart represents the shares of anthropogenic biomass burning sources in the total OC emissions over India. Other sources (OTHR) of OC include brick production, transportation, industries, and power generation.

This study leverages samples of aerosol particle emissions collected on filter substrates during the COALESCE field campaign to evaluate BrC-BC light absorption continuum behavior in real-world biomass burning emissions. Using a UV-Vis spectrophotometer, it examines BrC derived from major biomass fuel sources such as cooking, heating, agricultural residue burning, and brick production. The study aims to connect BrC with the thermo-optically resolved carbon fractions to parameterize BrC absorption over South Asia. Further, it endeavours to couple source specific BrC properties with the BC-to-organic-aerosols (OA) ratio to explore the spatial variability of the absorption properties of BrC emitted across India.

2. Data and Methods

2.1. Data Collection

The field-based emissions measurement campaign (Figure S2) was conducted from Oct-2021 to Apr-2022 in rural parts of Gujarat and Maharashtra, two western India states. These locations were selected based on their representativeness for the fuels and devices commonly used in South Asia based on previous studies (Navinya et al., 2023; Kapoor et al., 2023b; Tibrewal et al., 2023; Habib et al., 2023). The primary aim of this campaign was to capture physical, chemical, and optical information about the emissions from biomass sources: agricultural residue burning, brick production from clamps, cooking, and heating. The versatile source sampling system, as described by Kumari et al. (2024) and Venkataraman et al. (2020), consists a multi-arm inlet design adapted from Roden et al. (2006) to function as an area plume sampler, positioned 1 to 1.5 meters above the emission source (Figure S2). The system comprises eight arms that aspirate aerosols, which are then combined in a mixing plenum to ensure representative sampling of the smoke plume. Aerosols drawn through the inlet pass through a 2.5 μm cut-off cyclone, subsequently being divided into two streams: for real-time and gravimetric measurements. Aerosols in the gravimetric stream were collected on quartz filter substrates for offline laboratory analysis over the entire duration of the experiment, encompassing ignition, flaming, and smoldering phases, in order to obtain a sample representative of the complete combustion cycle. The temperatures of the emitted plumes were diluted by the surrounding air, reaching levels close to that of ambient air before entering the multi-arm sampler. This ensured that the emissions had undergone gas-to-particle partitioning, corresponding to the properties of emissions used in climate models. In this study, we utilized aerosol-laden quartz filter substrates from 14 different fuel and source combinations (Table S1) to understand soluble BrC absorption ($Mm^{-1} = 10^6\ m^{-1}$) and total OC concentration ($\mu g\ m^{-3}$).

2.2. Estimation of BrC Properties

We used 4.5 ml of methanol solvent and dissolved two 0.25-inch diameter punches of quartz filters. After 1 hour of sonication, the extracted solvent was passed through a 0.22 μm polytetrafluoroethylene membrane syringe filter (Fisherbrand™) to remove insoluble debris. The absorption of this methanol-soluble OC (considered as BrC absorption) was estimated by using a UV-visible spectrophotometer (LAMBDA 35, PerkinElmer) with a working range of 300 nm to 900 nm and a spectral resolution of 1 nm. The equation shown below was used to estimate the absorption coefficient at any given wavelength (Chakrabarty et al., 2023; Sarkar

et al., 2019; Satish and Rastogi, 2019; Srinivas and Sarin, 2013, 2014; Bikkina et al., 2020; Boreddy et al., 2021; Choudhary et al., 2017, 2018, 2021, 2022; Dasari et al., 2019; Dey et al., 2021; Kirillova et al., 2016; Mukherjee et al., 2020; Rajeev et al., 2022; Rastogi et al., 2021; Rathod et al., 2017; Rana et al., 2020; Shamjad et al., 2016b, 2018).

$$b_{abs,BrC,\lambda} = \frac{(A_\lambda - A_{700}) \times V_{Extract} \times \ln(10)}{V_{Sampled} \times L \times f_{filter\ area}} \quad (1)$$

In equation 1, A_λ is absorbance at wavelength λ, $V_{Extract}$ is the volume of solvent extract used (4.5 ml in this study), $V_{Sampled}$ is the volume of air sampled, $f_{filter\ area}$ is the fraction of filter area used for the analysis, and L is the optical path length (0.01 m). Given that soluble BrC does not absorb at 700 nm and longer wavelengths or, at best, absorbs very little, the absorption at 700 nm (A_{700}) was used to normalize absorbance to account for signal drift within the instrument, which is a limitation of this method. In this study, the estimated BrC only includes the methanol-soluble component and may not fully represent total BrC, including its insoluble components. The estimated BrC absorption could be underestimated due to excluded insoluble BrC and tarball structures, which possess high absorption strength (Corbin et al., 2019; Chakrabarty et al., 2023, 2010). The underestimation may be more pronounced as particle light absorption strength increases, i.e., closer to the dark-BrC region, since particle solubility is inversely proportional to light absorption strength (Saleh, 2020). In brief, Saleh (2020, and references therein) reviewed and categorized different BrC classes based on their volatility, using UV-vis spectrometry, optical closure (Aethalometer, Cavity Ring-Down Spectroscopy, and photoacoustic), and electron energy loss spectroscopy techniques. While UV-vis spectrometry misses out insoluble particles, optical closure techniques consider absorption by particles regardless of their solubility. However, they have uncertainties associated with separating BrC light absorption from the total aerosol light absorption. In this study only two data points, observed marginally in the dark-BrC region, might be affected.

Quartz filters were examined using a Magee Scientific DRI multi-wavelength thermo-optical carbon analyser with the IMPROVE-A protocol to estimate the elemental (EC) and organic carbon (OC) concentrations (Chow et al., 2007). Thermo-optically resolved carbon fractions (OC1, OC2, OC3, OC4, EC1, EC2, and EC3) were used after pyrolytic correction to reconstruct the total organic carbon and total elemental carbon fractions (Chow et al., 2007). For the purpose of representation in Figure 3, pyrolytic carbon was assigned to OC4. These fractions are associated with the volatility of the OC (Kapoor et al., 2023a; Shetty et al., 2023; Tohidi et al., 2022; Vodička et al., 2015; Soleimanian et al., 2019; Ma et al., 2016), as these OC fractions are measured under increasing temperature peaks (140, 280, 480, & 580°C) during thermo-optical analysis. Hence, OC1 exhibits relatively higher volatility compared to OC2, while OC2 is more volatile than OC3, and so forth. In this study, pyrolysis corrected EC was treated as a proxy of BC to facilitate the comparison with other studies. The uncertainty associated with OC and EC measurements are 5 and 10% respectively (Cheng et al., 2021; DRI Manual, 2015). Cheng et al. (2021) reported an overall uncertainty of approximately 10% for methanol-soluble k_{BrC} determined through UV-vis spectrophotometry. When accounting for the

5% manufacturer-reported uncertainty in OC concentration, the corresponding uncertainty in the absorption coefficient is estimated to be around 10%.

Furthermore, OC concentration and $b_{abs,BrC,\lambda}$ were used to calculate the mass absorption coefficient ($MAC_{BrC,\lambda}$). The imaginary refractive index of BrC ($k_{BrC,\lambda}$) was estimated by considering the density (ρ) of freshly emitted OC to be 1500 kg m^{-3} (Liu et al., 2013; Shamjad et al., 2016), using the following relation (Jennings et al., 1979):

$$k_\lambda = \frac{\rho \times \lambda \times MAC_\lambda}{4\pi} \quad (2)$$

The same equation was used in many previous studies, some of which cover the same geographic region (Shamjad et al., 2018; Bikkina and Sarin, 2019; Shamjad et al., 2016b; Rana et al., 2020; Liu et al., 2013; Zhang et al., 2020). In addition, an absorption Angstrom exponent (AAE) between 365 and 550 nm ($AAE_{365/550} = -\ln[b_{abs,BrC,365}/b_{abs,BrC,550}]/\ln[365/550]$) was also estimated to understand the spectral dependence of the BrC absorption coefficient. Similarly, w (AAE-1) indicates the spectral dependence of the imaginary refractive index between 365 nm (a commonly used wavelength for studying BrC absorption) and 550 nm (the peak of solar radiation intensity). In this study, we have used w and k for the ease of comparison with previous studies (Saleh et al., 2014; Lu et al., 2015; Luo et al., 2022; Saleh et al., 2018). However, AAE and MAC can also be used interchangeably.

2.3. Spatial variation of BrC absorption

The relationship between fuel and source averaged $k_{BrC,550}$ and the BC to OA ratio ($k_{BrC,550}$ = 0.0365(±0.006)×(BC/OA) + 0.0047(±0.0037), R^2 = 0.93) was established using field-collected fuel samples. Similarly, w was also calculated as a function of the BC to OA ratio (w=5.355(±0.50)×exp(-0.428(±0.25)×(BC/OA), R^2=0.60). Here, OA was derived by multiplying OC by a factor of 1.8, a methodology consistent with previous studies (Turpin et al., 2001; Chow et al., 2015; Navinya et al., 2020; Provençal et al., 2017; Kumar et al., 2023), and aligned with the considered OA density (Kuwata et al., 2012). Although this factor does not impact the R-square (R^2) of the relationship, it facilitates comparisons with other studies that have utilized the BC to OA ratio to derive $k_{BrC,550}$. The spatial distribution of BC and OC emissions from the SMoG-India emission inventory (Tibrewal et al., 2024) was integrated into the equation, after converting OC into OA using the same factor, to calculate the nationwide $k_{BrC,550}$ and w for the major (~90%) OC emitting sources: AGRI, COOK, and HEAT. Additionally, we derived overall k_{BrC} and w values through a weighted averaging approach, incorporating OC emissions (Figure S5) as weights, along with source-specific information (Figure S3). BRICK (brick production) was omitted because field-based samples were limited to clamp kilns, but not available for other major brick production technologies, including Bull's trench kilns and vertical shaft brick kilns (Weyant et al., 2014; Tibrewal et al., 2024, 2023).

3. Results and Discussion

3.1. BrC-BC Absorption Continuum

The measured $k_{BrC,550}$ values varied from 0.0007 to 0.1199, while w ranged from 7.52 to 1.00, highlighting inverse dependence of k_{BrC} on w (Figure 2). A previous study using synthetic fuels under different combustion conditions reported a similar observation, based on experimental measurements (Saleh et al., 2018). In this study, different field-collected sources and fuels reflected real-world variations in burning practices. An equation fitted to the data ($w = 0.1917/(k_{BrC,550} + 0.02886)$) has an R^2 value of 0.58, and an extension of this curve with 95% prediction bounds overlaps the BC absorption region (k_{550} = 0.6-0.8 and w = ~0-0.2) (Bond and Bergstrom, 2006; Saleh et al., 2018; Liu et al., 2018; Gyawali et al., 2013). The range of $k_{BrC,550}$ and w observed in this study spans across three broad classes of BrC (weak, moderate and strong) suggested by Saleh (2020) for different combustion conditions. They suggest that while combustion processes emit particles containing a mix of different BrC classes, smoldering biomass emissions are skewed more toward weakly absorbing BrC (W-BrC), while high-temperature biomass combustion emissions are skewed more toward moderately and strongly absorbing BrC (M-BrC and S-BrC). In the present work, some data points, mainly from cooking and heating, exhibit greater spectral variation (larger w) than that suggested for M-BrC, while falling within its $k_{BrC,550}$ range. Changing combustion conditions were observed during several experiments, where both flaming and smoldering combustion phases occurred, while particles were collected as a time averaged filter sample. Here, the greater spectral dependence in M-BrC measurements, implies that these samples would exert stronger light absorption in the near-UV range, than typical M-BrC. The thermo-optically resolved carbon fractions show a decline in the total OC fraction, mainly in OC1 and OC2 (relatively high volatile fractions), with increasing BrC absorption strength from weak to moderate (Figure 3a). A simultaneous increase of EC highlights the dominance of BC absorption as the strength of BrC absorption increases, as also reported previously (Saleh et al., 2014; Chakrabarty et al., 2023). Relationships between BC, OC and BrC properties, reported by Saleh et al. (2014), are useful in parameterizing BrC absorption in radiative and climate models (Brown et al., 2018; Neyestani and Saleh, 2022; Wang et al., 2018).

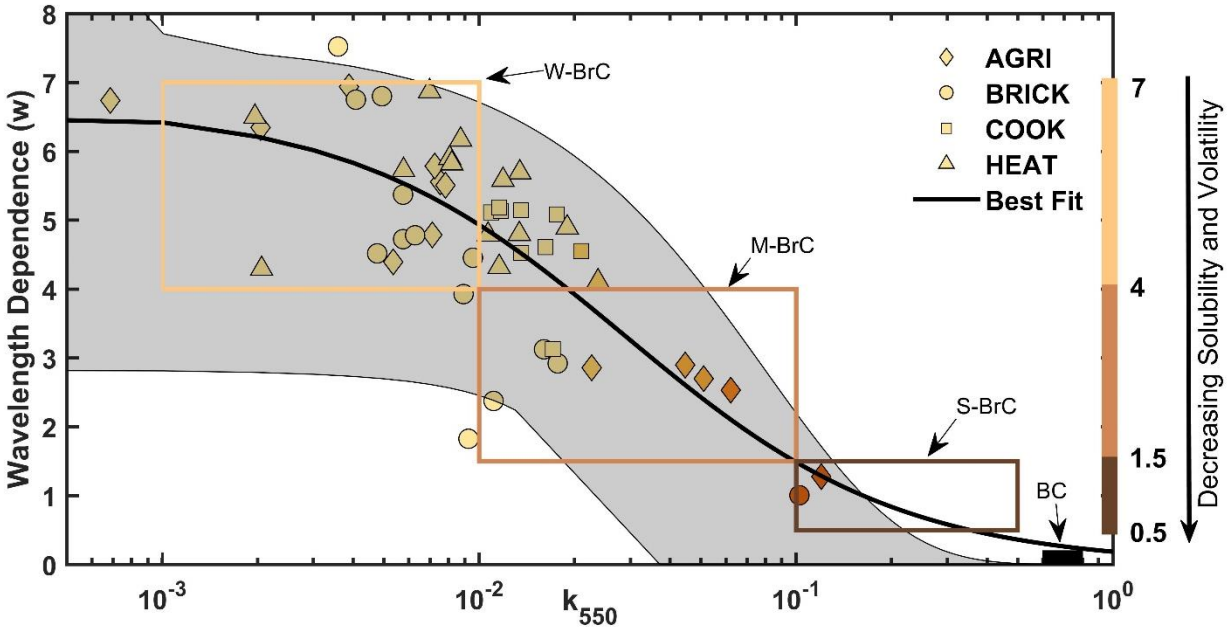

Figure 2. BrC-BC light absorption continuum in semi-log scale, showing the wavelength dependence (w = AAE-1) of the imaginary part of the refractive index (k_{550}) versus the imaginary part of the refractive index at 550 nm (k_{550}), w = 0.1917(±0.074)/($k_{BrC,550}$ + 0.02886(±0.014)). Here, the BC region lies between k_{550} = 0.6 - 0.8 and w = 0 - 0.2. The BrC classes are defined per Saleh, (2020): strongly absorbing BrC (S-BrC), moderately absorbing BrC (M-BrC), and weakly absorbing BrC (W-BrC). Arrow in the right indicates reduction in the solubility and volatility with increase in k_{550} (Saleh, 2020). The shaded grey area represents the continuum reported by previous studies (Saleh et al., 2014; Lu et al., 2015; Luo et al., 2022; Saleh et al., 2018), and the equations for the shaded area are given in the supplementary information, S1 and figure S1. The right axis displays the range of wavelength dependence for the three BrC classes.

3.2. Source Specific BrC

We observed that the variability of source specific BrC properties is larger within a source category than among different source categories. Figure 3b shows no significant changes in $k_{BrC,550}$ among different source categories. However, there are much larger differences among individual data points in a source category, because of varying fuels, meteorology, and burning practices. The $k_{BrC,550}$ means from agricultural residue burning, brick production, cooking and heating are respectively 0.026 (±0.035), 0.015 (±0.026), 0.015 (±0.003), and 0.010 (±0.006) (Figure 3b). A large variation in $k_{BrC,550}$ was observed during agricultural residue burning, with banana, which has a high moisture content (Tock et al., 2010), showing a $k_{BrC,550}$ of 0.008, and pigeon pea (an oil seed legume) having a $k_{BrC,550}$ of 0.082. In comparison, $k_{BrC,550}$ varies from 0.006 (final-stage) to 0.022 (initial stage) during brick kiln operation and from 0.002 (crop residue) to 0.013 (firewood) during residential heating. This contrasts with cooking, where deliberate efforts are made to ensure efficient burning of fuel for meal preparation. Hence, BrC properties in cooking emissions do not vary much ($k_{BrC,550}$= 0.015±0.001). Our study observed $k_{BrC,365}$ of ~0.1 (±0.01) for cooking, which is higher than lab-measured values (0.014-0.054)

for the same fuels at 350 nm (Rathod et al., 2017). We observed that the $MAC_{BrC,365}$ stayed between 1.5-2.5 $m^2\ g^{-1}$ for all the source-fuel combinations, except for pigeon pea residue burning ($MAC_{BrC,365}$=4.01 $m^2\ g^{-1}$). The current findings are comparable with the $MAC_{BrC,365}$ value of 2 (±0.5) $m^2\ g^{-1}$ from Indian airmasses influenced by agricultural residue burning (Satish et al., 2020). The values reported in our study are in the upper range of ambient $MAC_{BrC,365}$ (0.62-2.3 $m^2\ g^{-1}$) reported previously over India (Sarkar et al., 2019; Shamjad et al., 2018; Satish et al., 2020; Rastogi et al., 2021; Rana et al., 2020; Kirillova et al., 2016; Dey et al., 2021), which could be due to photobleaching of ambient BrC that decreases MAC. However, our estimation of $MAC_{BrC,365}$ aligns well with the previously reported source-specific values (1.09-2.53) (Pandey et al., 2020; Debbarma et al., 2024; Rathod et al., 2017). The observed AAE_{BrC} (~5.23 ± 1.51, range 2-8.5, see Table S1) is comparable with previous observations (~5.31 ± 1.67, range 2.3-6.8) for biomass burning over India (Islam et al., 2022; Pandey et al., 2020; Rathod et al., 2017; Satish et al., 2020). In agricultural residue burning, banana residue shows the lowest $k_{BrC,550}$ (0.008), and BC to OA ratio (0.030) (Table S2). In contrast, pigeon pea residue burning has the highest $k_{BrC,550}$ (0.082) and BC to OA ratio (2.054). A similar relationship between $k_{BrC,550}$ and BC to OA ratio has also been observed in other source-fuel combinations and used to parameterize $k_{BrC,550}$ and w (Figure 4).

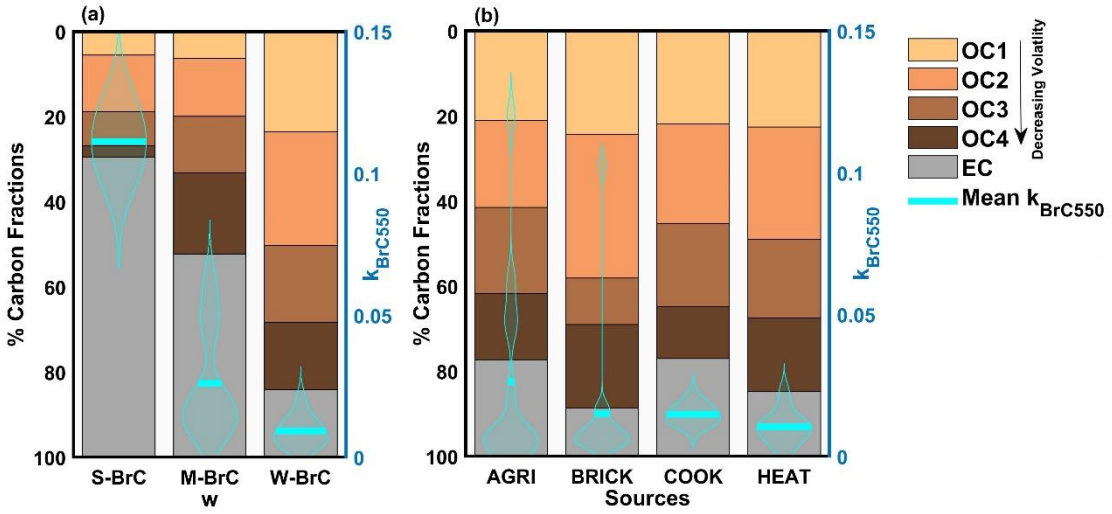

Figure 3. (a) $k_{BrC,50}$ distribution (right) and thermo-optically resolved carbon fractions (left) with varying BrC classes based on wavelength dependence. Here, the strongly absorbing BrC (S-BrC), moderately absorbing BrC (M-BrC) and weakly absorbing BrC (W-BrC) ranges are respectively <1.5, 1.5 - 4, and 4< in wavelength dependence (Saleh, 2020). **(b)** source specific $k_{BrC,550}$ distribution (right) and thermo-optically resolved carbon fractions (left). The distribution of violin plot shows the Kernel density. Arrow near legends indicates reduction in the relative volatility from OC1 to OC4 (Ma et al., 2016). For the purpose of representation, pyrolytic carbon was assigned to OC4.

3.3. Parameterization of k_{BrC} and w

We leveraged the significant correlation (p-value < 0.01) between the BC to OA ratio and the BrC properties ($k_{BrC,550}$, R^2=0.93; w, R^2 = 0.60) to build a relationship between these quantities.

Despite the variety of fuel burning technologies used, such as traditional stoves, open residue burning, and brick clamps, $k_{BrC,550}$ variability is explained (R^2 = 0.93) by the BC to OA ratio. We observed that $k_{BrC,550}$ varies linearly from 0.006 to 0.74 for BC to OA ratios of 0 to 20 (Figure 4a). Similarly, we explain w by using the BC to OA ratio to provide an approximation of the BrC absorption over the different wavelengths. We observed an exponential relation between w and the BC to OA ratio with an R^2 of 0.60 (w varies from 5 to ~0 for BC to OA ratios of 0 to 20, respectively) (Figure 4b). Relative to the present studies, the relationship used in climate modelling studies (Zhang et al., 2020; Neyestani and Saleh, 2022; Brown et al., 2018) given by Saleh et al. (2014) would overestimate the $k_{BrC,550}$ over South Asia (Figure S7). In contrast, previous studies (Saleh et al., 2014; Lu et al., 2015; Luo et al., 2022) underestimate the range of w observed in this study, which may result in an underestimation of $k_{BrC,365}$ (Figure S7). Such an underestimation would propagate uncertainties to radiative forcing calculations, especially over South Asia.

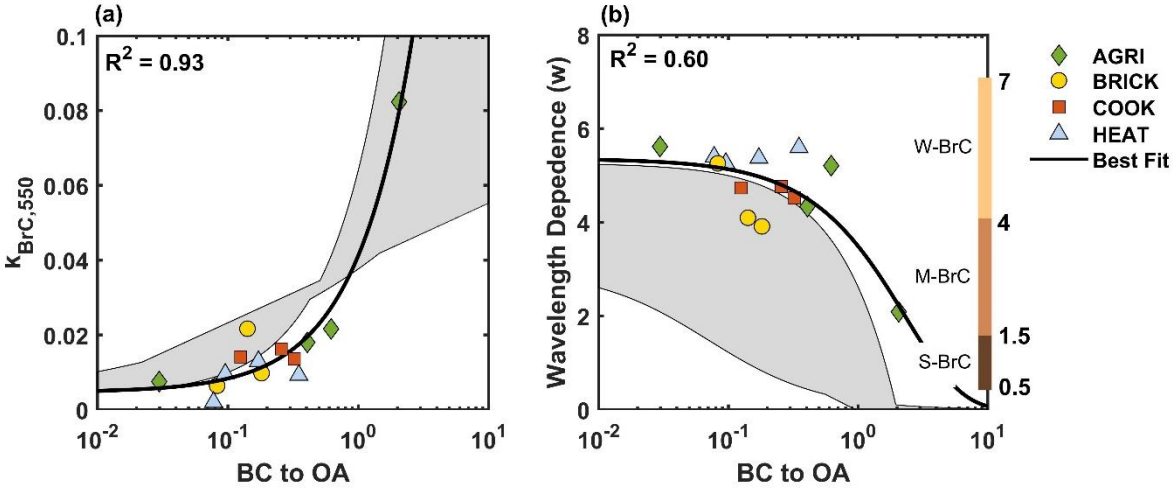

Figure 4. (a) Mean $K_{BrC,550}$ versus BC to OA ratio ($k_{BrC,550}$= 0.0365(±0.006)×(BC/OA)+0.0047(±0.0037), R^2=0.93) and **(b)** w versus the BC to OA ratio (w=5.355(±0.50)×exp(-0.428(±0.25)×(BC/OA)), R^2=0.60). Here, OA = OC x 1.8: the factor 1.8 is widely used to convert OC into OA (Turpin et al., 2001; Chow et al., 2015; Navinya et al., 2020; Provençal et al., 2017; Kumar et al., 2023). The grey shaded area represents the relationship reported by previous studies (Saleh et al., 2014; Lu et al., 2015; Luo et al., 2022), and the equations for the shaded area are given in the supplementary information (S2, S3 and figure S1). The right axis in Figure 4b displays the ranges of wavelength dependence for the three BrC classes.

3.4. Spatial Differences in $k_{BrC,365}$ and w

Several studies have reported ambient BrC absorption in the South Asian region (Dey et al., 2023; Srinivas and Sarin, 2013, 2014; Bikkina et al., 2020; Boreddy et al., 2021; Choudhary et al., 2017, 2018, 2022; Dasari et al., 2019; Dey et al., 2021; Kirillova et al., 2016; Mukherjee et al., 2020; Rajeev et al., 2022; Rastogi et al., 2021; Rana et al., 2020; Shamjad et al., 2016b, 2018), while most climate models continue to consider weakly absorbing BrC absorption (Sand

et al., 2021; Feng et al., 2013), invariant of sources and combustion conditions. Feng et al. (2013) simulated global BrC absorption by using 2- to 5-fold weaker k_{BrC} values than those observed in our study, and they noted underestimation over South Asia owing to presence of strongly absorbing BrC. Other studies (Brown et al., 2018; Zhang et al., 2020) have used 2- to 3-fold higher $k_{BrC,550}$ values (Saleh et al., 2014; Mcmeeking, 2008) than observed in this study to simulate the global radiative impact of BrC. Hence, neglecting the spatial variability of k_{BrC} could lead to bias in understanding its radiative impact. Thus, we calculated emissions-weighted BrC optical properties across the Indian region to demonstrate their spatial heterogeneity in the region. The relationships shown in Figure 4 were used to make a spatial map of $k_{BrC,550}$, $k_{BrC,365}$, and w, with emission strength from the COALESCE SMoG-India emission inventory (Tibrewal et al., 2024). SMoG-India is a multi-sectoral, multi-pollutant data set available at 5 km grid resolution, developed under the COALESCE network (Venkataraman et al., 2020), which also facilitated the collection of samples used in the present study.

Figure 1 shows the large OC emissions over the Indo-Gangetic Plain, with annual emissions ranging from 50-70 Mg y^{-1} $pixel^{-1}$ (pixel size is 5×5 km), while other regions emit ~10-20 Mg y^{-1} $pixel^{-1}$. Emission weighted spatial information about w (range: 4.3 - 5.3) and $k_{BrC,550}$ (0.006 – 0.023) aids the estimation of $k_{BrC,365}$. Figure 5a shows $k_{BrC,365}$ ranges from 0.05 to 0.14, indicating strong absorption in the UV-visible wavelengths. The Himalayan foothills show large k_{BrC} compared to other parts of India, mainly due to high BC to OA emissions from the predominant heating activity. A recent study highlighted the low photobleaching rate of BrC near Himalayan regions due to the low ambient temperatures (Choudhary et al., 2022). The coincidence of dark BrC particle emissions in this study, along with their reported extended lifetimes, could result in snow darkening on deposition, along with accelerated snow and glacier melting. The northwestern region of India exhibits the highest OC emissions from agricultural residue burning (Figure S5), primarily from straw residue burning (Kapoor et al., 2023b), which has a relatively low BC to OA ratio. Consequently, the k_{BrC} remains lower compared to other regions, such as Maharashtra and Andhra Pradesh, where oilseed crop burning is prevalent (Kapoor et al., 2023b), resulting in higher BC to OA ratio and k_{BrC} values. Heating activities are particularly intense in the colder areas, especially in the Himalayan foothills, with a higher use of firewood in the eastern India (Navinya et al., 2023), leading to significantly higher BC to OA ratios, and elevated k_{BrC} in the northern and eastern regions (Figure S3). In the Central Indo-Gangetic Plain, particularly in Uttar Pradesh and Bihar, dung cake is more commonly used for heating (Navinya et al., 2023), which contributes to very low k_{BrC}. The variation in the BC to OA ratio across India due to cooking activities is minimal (0.075-0.125) compared to that from agricultural residue burning (0.025-0.2) and heating (0.025-0.25), resulting in substantially low spatial variation of $k_{BrC,365}$ (0.06-0.08) from cooking (Figure S3). The $k_{BrC,550}$ values of combustion aerosol emissions from India vary from 0.006 to 0.023 (Figure S6), with some hotspots scattered across the country. These numbers highlight the order of magnitude increase in $k_{BrC,365}$ compared to $k_{BrC,550}$, with higher values over Eastern and Northern India. An earlier investigation also noted elevated modelled BrC absorption in the Eastern regions of India (Zhu et al., 2021). The substantial emissions of BrC across the country, coupled with the high k_{BrC} values observed in certain other regions, suggest that BrC particles may have significant radiative impacts over the region.

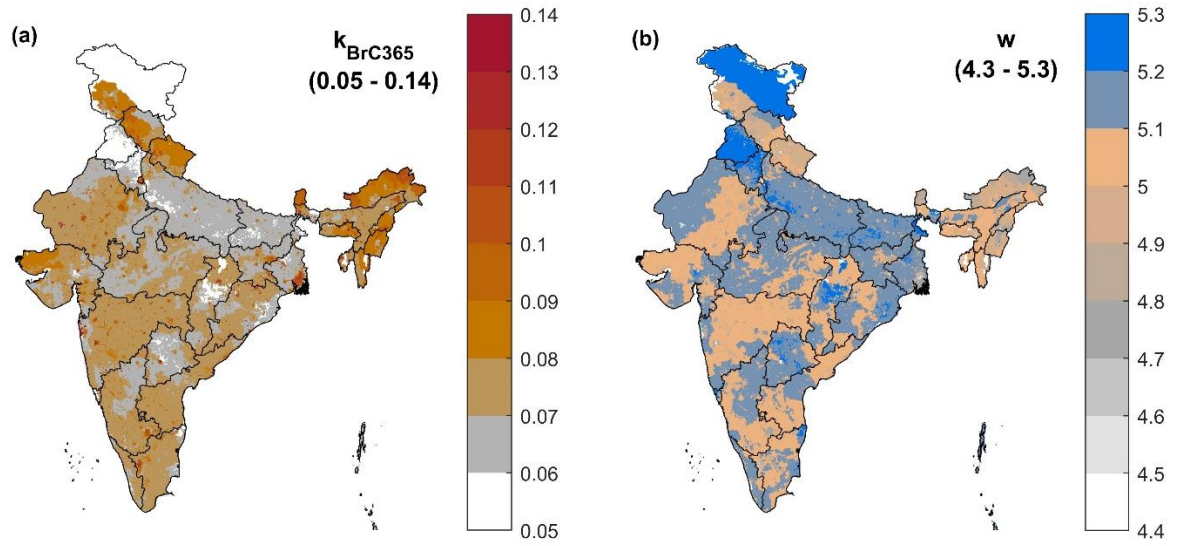

Figure 5. The spatial distribution of **(a)** $k_{BrC,365}$ and **(b)** wavelength dependence (w). The BC to OA ratio is taken for the year 2019 from the COALESCE SMoG-India emission inventory (Tibrewal et al., 2024).

4. Implications

The variability in $k_{BrC,near\text{-}UV}$ across modelling studies, ranging from 0.045 (Zhang et al., 2020) to 0.168 (Lin et al., 2014), arises from methodological, fuel, and burning condition disparities in the studies reporting BrC absorption properties from lab-based biomass combustion (Kirchstetter et al., 2004; Chen and Bond, 2010; Lack et al., 2012). However, our study, by using field measurements of a variety of sources, introduces source- and fuel-specific k_{BrC} values, enhancing modelling capabilities for a more nuanced understanding of the radiative and climate impacts of BrC. Additionally, the observed varying wavelength dependence (w), linked with the BC to OA ratio in this research, amplifies uncertainty when it is assumed to be constant in models (Zhang et al., 2020). Compared to the findings of this study, typical BrC parameterization schemes (Saleh et al., 2014; Lu et al., 2015; Luo et al., 2022) in climate models tend to overestimate $k_{BrC,550}$ while substantially underestimating wavelength dependence, which may misrepresent near-UV BrC absorption in world regions with biomass combustion emissions resembling those in South Asia. Additionally, this study's findings aid in pinpointing biomass fuels and activities, including burning of some agricultural residues and residential space heating, that are both prone to emitting stronger absorbing BrC ($k_{BrC,550} > 0.1$) and prevalent across developing nations. These variations in k_{BrC} with sources and fuels lead to spatial variations in emitted BrC properties. In the Himalayan foothills, residential space heating produces stronger absorbing (and more persistent) BrC emissions, and the deposition of these emissions increases the potential risks of increased snow darkening and accelerated glacier melting. Leveraging this information with emission inventories enables the identification and potential interventional targeting these biomass fuels and activities, towards reducing both their local health and global climate impacts.

Acknowledgements

The authors thank the Fulbright Program {Fulbright-Kalam Climate Fellowship Award No. 2913/FNDR/2023-2024 to CN} and the IIT Bombay – WashU program for financial support to conduct a part of this work at Washington University in St. Louis. This work was supported by the Indian Ministry of Environment Forest and Climate Change under the NCAP-COALESCE project {Grant No.14/10/2014-CC(Vol.II)}. The views expressed in this document are solely those of authors and do not necessarily reflect those of the Ministry. The Ministry does not endorse any products or commercial services mentioned in this publication.

Data Availability

The data used in this study are provided in the supplementary information of the article.

Conflict of Interest

The authors declare that they have no conflict of interest.

Author contributions

Conceptualization: CV, RKC, CN, TSK; **Methodology, Formal Analysis:** RKC, CN, TSK; **Software, Visualization:** CN; **Data Curation:** CN, TSK, GA; **Writing - Original Draft:** CN; **Writing - Review & Editing:** TSK, RKC, CV, HCP, GA; **Supervision:** RKC, CV, HCP.

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