

Atmospheric Brown Carbon: Sources, Optical Properties, and Chromophore Composition

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ABSTRACT

Brown carbon (BrC) constitutes a significant portion of organic carbon (OC) and exerts a substantial influence on air quality, atmospheric chemical processes, and the impact on climate dynamics. The intricate chemistry of BrC arises from multiple mechanisms and source types, yielding a wide spectrum of spectral properties. Consequently, identifying representative chromophore species to develop reference material is crucial to capture the full diversity of BrC found in the atmosphere and to ensure its precise determination. This also emphasizes the need for a more comprehensive measurement method to link BrC's optical traits with its chemical composition. One of the objectives of this review is to evaluate the existing level of understanding concerning the molecular compositions of BrC compounds by analyzing recent field and laboratory data. This paper revises and expands a part of the prior database containing critical assessments of the primary and secondary sources, photochemistry of multi-phase chemical reactions involving BrC, potential candidate BrC chromophores, and recent studies on optical properties of BrC in the cryosphere, to draw lessons from recent advances that lead to improved BrC representation in the atmosphere.

Keywords: Brown carbon (BrC), Secondary formation processes, Optical properties, Sources, Reference material

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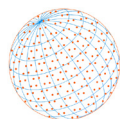
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1 INTRODUCTION

A majority of organic aerosols (OAs) are known to effectively scatter visible light. Nonetheless, a notable and widely fluctuating fraction of carbonaceous organic aerosols absorbs radiation in the near-UV (300–400 nm) and visible ranges. This type of aerosol, termed as brown carbon (BrC), is a subset of OA that predominantly absorbs light in the short-visible and ultraviolet (UV) spectral range (Chakrabarty *et al.*, 2010). These compounds typically display light brown to yellowish coloration. This is true particularly during the initial stage of combustion, when there is substantial emission of yellow and brown particles (Andreae and Gelencsér, 2006).

Typically, the light-absorbing attributes of BrC are measured through the wavelength-dependent imaginary component of the refractive index ($k_{\text{BrC-365nm}}$) or the mass absorption cross-section (MAC). The imaginary component of the BrC refractive index, which reflects the absorption of light by particles, covers a broad spectrum of values, ranging from 10^{-4} to 10^{-2} at ultraviolet and lower visible wavelengths (Chakrabarty *et al.*, 2010; Alexander *et al.*, 2008; Sumlin *et al.*, 2018). Likewise, the absorption Ångström exponent (AAE) value, which is used to express the wavelength dependence of light absorption, widely ranged from 1.5 to 12 in different studies (Alexander *et al.*, 2008; Lewis *et al.*, 2008). The extensive spectrum of refractive index and AAE values suggest the complex chemical nature of BrC that depends on various mechanisms and types of sources resulting in its formation with diverse spectral characteristics. As a result, choosing any arbitrarily selected reference material as a substitute is unlikely to adequately represent the diverse range of BrC present in the atmosphere. Moreover, the optical characteristics of atmospheric BrC species



might undergo alterations with aging/processing in the atmosphere.

In this work, we reviewed the present understanding of the chemistry related to the formation of light-absorbing organic compounds in the atmosphere, particularly those pertinent to BrC formation, and it is structured as follows: in [Sections 2 and 3](#), we provide an overview of the main sources of BrC as deduced from field observations, both primary and secondary, and examine the factors that influence the changes in its physicochemical characteristics in urban study locations. [Section 4](#) presents recent studies involving changes in light absorption properties of BrC, with atmospheric processing highlighting important factors responsible for these alterations. In [Section 5](#) we discuss the wide range of compounds detected in BrC across different research studies emphasizing the complex composition of BrC. It highlights the need for a more comprehensive approach related to BrC measurements and development of reference material to establish the relationship between BrC's optical properties and its chemical composition. [Section 6](#) summarises various approaches used for determination of BrC in the published studies with the view to stress the need for more optimised measurement methods for accurate evaluation of BrC. [Section 7](#) reviews global model and field studies constraining radiative forcing effect of BrC aerosols. [Section 8](#) presents a summary of field observations describing trends in air pollution and climate forcing attributed to BrC at different cryogenic locations. [Section 9](#) summarizes the established facets of BrC chemistry and offers the authors' insights and views on specific areas for future research in this field.

2 BrC FROM PRIMARY SOURCES

Biomass burning (BB) stands out as the primary origin of atmospheric BrC emissions ([Lukács *et al.*, 2007](#); [Chen and Bond, 2010](#); [Chakrabarty *et al.*, 2010](#)), typically associated with smoldering or pyrolysis processes occurring at relatively low temperatures. These conditions are more conducive to BrC formation than the combustion of fossil fuels for energy generation. Nevertheless, there are also sources of BrC related to fossil fuel usage. The sources of BrC in the atmosphere have been comprehensively reviewed by [Laskin *et al.* \(2015\)](#). Here, we will discuss recent studies related to the primary BrC sources (summarised in [Table 1](#)).

In a recent study by [Tang *et al.* \(2020\)](#), the molecular compositions and optical properties were examined for organic carbon aerosols emitted from combustion of coal, BB, and vehicular emissions using ultraviolet-visible (UV-Vis) spectroscopy, excitation-emission matrix (EEM) spectroscopy, and Fourier transform ion cyclotron resonance mass spectrometry (FT-ICR MS) coupled with electrospray ionization (ESI). The results revealed that light absorption (at 365 nm) by BB and coal combustion (CC) aerosol samples was higher than that in vehicular emissions. In addition, the FT-ICR mass spectra indicated higher fraction of CHO and CHON species in BB aerosols (88–93%) compared to CC and vehicle emission aerosols. While S-containing species exhibited greater prevalence in CC (38–75%) and vehicle emissions (41–46%) than BB. This demonstrates the significant contributions of various sources to light-absorbing BrC and molecular compositions.

In order to identify the potential sources of light-absorbing carbonaceous aerosols (LACs), [Targino *et al.* \(2023\)](#) measured the real-time concentrations of BrC (at $\lambda = 370$ nm) and black carbon (BC) ($\lambda = 880$ nm) at urban and suburban sites in Brazil using seven-wavelength (λ) Aethalometers ($\lambda = 370, 470, 520, 590, 660, 880, \text{ and } 950$ nm) operated with a PM_{2.5} cyclone. Traffic emission is considered an important source as suggested by concomitant increase in BrC and BC concentrations with traffic volume. Furthermore, observation of nocturnal LAC peaks outside the traffic peak hours in addition to relatively large Å_{370/880} (Ångstrom absorption exponent) values in the evening center suggest biomass burning (residential waste and grass) to be the predominant source.

Recently, [Hossen *et al.* \(2023\)](#) examined the optical properties and chemical functional groups using a UV-Vis spectrophotometer at a wavelength of 365 nm and attenuated total reflection-Fourier transform infrared spectroscopy (ATR-FTIR) in aerosols derived from specific combustion sources such as biomass, biofuels, and mosquito coils. Results revealed nitro compounds linked with aromatic ring to be the major component of atmospheric BrC responsible for enhanced optical properties based on FTIR band spectra. In addition, presence of heterocyclic aromatic carbon compounds, carbonyl group (C=O), and nitrogen-containing groups such as -NO₂, and C≡N were also detected.

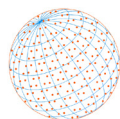


Table 1. Literature values of light absorption parameters, their measurement techniques, major chromophore species and potential sources in recent years.

Location	Measurement	Technique	BrC/HULIS-light absorption parameter concentration			Major chromophores	Sources	References	
			A _{AE}	MAE (m ² g ⁻¹)	b _{abs} (Mm ⁻¹)				k
Delhi, India	H ₂ O, MeOH, and hexane soluble BrC and HULIS (H ₂ O soluble) extracts from urban filter samples	SPE method, UV-Vis spectrophotometer (λ = 365 nm)	Mean = 4.88, 5.86 (BrC); 7.7 (HULIS)	Mean = 0.75 ± 0.2, 1.18 ± 0.5	Mean = 22.5 ± 9, 29 ± 21	Mean = 0.04 ± 0.02, 0.03 ± 0.01	BBE from regional transport, open burning of wood, cow dung cake, roadside trash	Alang <i>et al.</i> , 2023	
Dhaka, Bangladesh	WSOC of urban filter samples	UV-Vis spectrometer (λ = 370 nm), ATR-FTIR	Mean A _{AE} = 5.13 ± 1.45 (BB), 2.85 ± 0.32 (FF), 5.45 ± 0.82 (mosquito coil)	Range = 0.07 ± 0.01–0.99 ± 0.15	Range =	Nitro aromatics, heterocyclic aromatic, carbonyl, nitrogen-containing groups	Mosquito coils, biomass, biofuel combustion	Hossen <i>et al.</i> , 2023	
Londrina, Southern Brazil	WSOC of urban filter samples	Aethalometer (λ = 370 nm)	(i) A _{370/880} range = 1.05–1.40	–	–	–	Vehicular emissions, BBE, residential open waste burning	Targino <i>et al.</i> , 2023	
Western US	H ₂ O and MeOH soluble BrC	UV-Vis spectrophotometer (λ = 405 nm), HR ToF-MS	–	–	–	–	Polycyclic aromatic hydrocarbons, oxygenated aromatics, nitrogen-containing organics	Wildfire smoke emissions	Shetty <i>et al.</i> , 2023
Kochi, India	H ₂ O and MeOH soluble BrC fraction of urban filter samples	UV-Vis spectrometer (λ = 365 nm)	Range = 3.05–4.55	Mean = 2.12 ± 0.25, 2.18 ± 0.41	Mean = 0.09 ± 0.01	–	BBE, combustion emissions, secondary formation	Boreddy <i>et al.</i> , 2022	
Delhi, India	H ₂ O soluble BrC fraction of urban filter samples	UV-Vis spectrophotometer (λ = 365 nm)	Range = 4.2–8.0	Range = 0.01–3.4	Range = 0.05–0.65	–	Biomass/coal/trash burning, secondary formation	Rastogi <i>et al.</i> , 2021	
Xinjiang, China	WSOC extracts in snow	Spectrofluorometer, HPLC-ESI-HRMS ¹	Range = 5.4–6.0	Mean = 0.94 ± 0.31, 0.39 ± 0.11	–	–	Unsaturated aromatic humic and protein like species	Combustion sources	Zhou <i>et al.</i> , 2021
Xi'an, Guan Zhong Basin	H ₂ O and acetonitrile soluble BrC fraction of rural filter samples	UV-Vis spectrophotometer (λ = 365 nm)	Range = 5.01–6.04	Range = 0.93–1.82	Range = 2.95–21.9	–	PAHs, Nitrophenols	BBE, secondary formation	Li <i>et al.</i> , 2020b

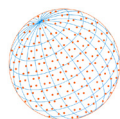


Table 1. (continued).

Location	Measurement	Technique	BrC/HULIS-light absorption parameter concentration		Major chromophores	Sources	References
			AAE	b_{abs} (Mm ⁻¹)			
Mt. Yulong, Tibetan Plateau	WSOC extracts of snow/ice, meltwater and aerosol samples	UV-Vis spectrophotometer ($\lambda = 365$ nm)	Mean = 0.61 ± 0.07 (meltwater), 0.55 ± 0.01 (snow-pit)	Mean = 4.07 ± 0.33 (surface-aged snow), 5.71 ± 1.14, snow-pit (1.92 ± 0.25), aerosols (0.54 ± 0.07)	–	Local vehicular emissions, long-range transport BBE	Niu <i>et al.</i> , 2020
Mumbai, India	H ₂ O and MeOH soluble BrC fraction of urban filter samples	UV-Vis spectrometer ($\lambda = 365$ nm), ¹ H-NMR	Range = 4.1–8.9	Mean = 1.1 ± 0.3, 1.5 ± 0.5	–	BBE, vehicular emission, secondary processing	Sarkar <i>et al.</i> , 2019
Kharagpur, India	H ₂ O soluble BrC fraction of urban filter samples	USB-4000 Spectrophotometer ($\lambda = 365$ nm)	Mean = 8.4 ± 2.6	Mean = 0.8 ± 0.2	Mean = 11 ± 5	wood burning emissions	Srinivas <i>et al.</i> , 2016; Bikkina and Sarin, 2019
Patiala, India	H ₂ O soluble BrC fraction of urban filter samples	USB-4000 Spectrophotometer ($\lambda = 365$ nm)	Mean = 5.1 ± 1.9, 5.1 ± 2.0	Mean = 1.3 ± 0.7	Mean = 40 ± 18, 52 ± 27	Open crop - residue burning, FF combustion	Srinivas <i>et al.</i> , 2016; Bikkina and Sarin, 2019
Kanpur, India	H ₂ O soluble BrC fraction of urban filter samples	Mean ~	Mean ~2.8	Mean ~1.8	Mean ~53.5, 69.3	BBE, secondary formation	Choudhary <i>et al.</i> , 2018
Nanjing, China	H ₂ O soluble BrC fraction of urban filter samples	UV-Vis spectrophotometer ($\lambda = 365$ nm)	Mean ~6.89	Mean ~0.76	Mean ~5.7	BBE, secondary formation/aging	Chen <i>et al.</i> , 2018
Xian, China	H ₂ O and MeOH soluble BrC fraction of urban filter samples	UV-Vis spectrophotometer ($\lambda = 365$ nm)	Mean = 6 ± 0.2	Mean = 0.95 ± 0.27, 1.65 ± 0.36	Range = 5 ± 1.3–46.3 ± 20.3	Domestic BB	Huang <i>et al.</i> , 2018

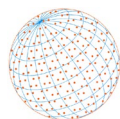
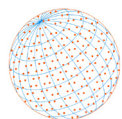


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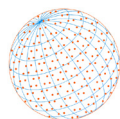
Location	Measurement	Technique	BrC/HULIS-light absorption parameter concentration			Major chromophores	Sources	References
			AAE	MAE (m ² g ⁻¹)	b _{abs} (Mm ⁻¹)			
Mt. Yulong, Tibetan Plateau	WSOC extracts of surface aged-snow and ice samples	UV-Vis spectrophotometer (λ = 365 nm)	Mean = 0.63 ± 0.22 (snow pit), 0.73 ± 0.21 (granular ice), 0.77 ± 0.46 (surface aged snow)	Mean = 5.23 ± 0.34 (snow pit), 8.59 ± 0.27 (granular ice), 5.10 ± 0.41 (surface aged snow)	–	–	Local vehicular exhaust, long-range transport of BBE	Niu <i>et al.</i> , 2018
Kanpur, India	H ₂ O soluble HULIS	UV-Vis spectrophotometer (λ = 365 nm), FTIR, ¹ H NMR, XPS	Mean = 4.9 ± 1.4 and 5.1 ± 1.3 (day and night, resp.)	mean = 13.6 and 28.8 (day and night, resp.)	–	–	Alcohols, ketones aldehydes, carboxylic acids, unsaturated and saturated carbon bonds	Kumar <i>et al.</i> , 2017
Rehovot, Israel	H ₂ O, acetonitrile, and orgmix soluble BrC fraction of nationwide bonfire festival samples	UV-Vis spectrometer (λ = 404 nm), HPLC-PDA-HRMS	Range = 2.9–12.5	Upper limit = ~4 (at 300 nm), 1.5 (at 400 nm)	–	–	Nitroaromatics	Lin <i>et al.</i> , 2017
Anmyeon Island, Korea	Water soluble HULIS fraction of coastal filter samples	SPE method, model/simulation studies	–	–	Range = 0.09–11.64	Range = 0.006–0.3	Fresh BB, secondary formation, incomplete combustion	Lee <i>et al.</i> , 2017
Kanpur, India	MeOH soluble fraction of urban filter samples	Photo acoustic soot spectrometer (PASS 3) (λ = 365 nm)	–	–	–	Mean ~0.042	BBE, secondary formation	Shamjad <i>et al.</i> , 2016
Delhi, India	WSOC extracts from urban filter samples	UV-Vis spectrophotometer, (λ = 365 nm)	Range = 3.1–9.3	Range = 1.1–2.7	–	–	BBE, secondary processing	Kirilova <i>et al.</i> , 2014



Shetty *et al.* (2023) examined how the optical characteristics of BrC, derived from smoke emissions from three wildfires in the western United States, were influenced by the chemical composition and volatility of organic aerosols (OA). They observed a robust connection between low and extremely low volatility organic compounds and elemental carbon (EC) as well as with the light-absorption coefficients of water-insoluble (and methanol soluble) fraction of BrC from real-world fresh-near source wildfire emissions. This suggests that organics (1) emitted during flaming-dominant combustion, (2) close to the fire source, and (3) subjected to limited atmospheric processing, are probably water-insoluble (indicated by strong correlation with EC). In other words, this suggests co-emission of strongly light absorbing chromophores with EC during high-temperature combustion, particularly near fire source conditions. It may be noted here that in addition to the variations in optical characteristics between BC and BrC, a fundamental difference exists in the life cycle of these two types of light-absorbing aerosols. BC is exclusively a primary component generated by combustion processes and is eliminated through wet and dry deposition mechanisms. In contrast, BrC can have common primary sources with BC and can also be produced through chemical reactions taking place in the atmosphere (as indicated by Hecobian *et al.* (2010) and Laskin *et al.* (2015)). Thus, distinguishing the impact of BrC on direct radiative forcing from that of BC presents a complex challenge. In fact, the secondary BrC formation pathways are known to significantly contribute to the enhanced light absorption and high chromophore concentration in the atmosphere. This point is discussed in the subsequent sections.

It is noteworthy that BC and BrC are not the only light absorbing aerosols in the atmosphere, as hematite and goethite in dust are also important absorbers at ultraviolet and mid visible wavelengths (Kerker *et al.*, 1979; Chen and Bond, 2010), especially at 550 nm where all these species show light absorption. It is therefore crucial to distinguish one type of absorbing aerosol from another to better understand distinct optical properties of each aerosol type. In a study by Yang *et al.* (2009), the overall light absorption among BC, BrC, and dust was distinguished based on wavelength-dependence, resulting in mass absorption efficiencies of 9.5, 0.5, and 0.03 m² g⁻¹ at 550 nm, respectively. In addition, the results showed that lowest scattering Ångstrom exponent and high absorption Ångstrom exponent imply the presence of dust, which is attributed to its large particle size and presence of ferric oxides. Recently, Schuster *et al.* (2016) utilised Aerosol Robotic Network (AERONET) size distributions and complex refractive indices to determine the relative proportion of carbonaceous aerosols and dust minerals. The authors proposed that separation between carbonaceous aerosols and dust occurs in the imaginary refractive index space, as 95% of biomass burning aerosols possess imaginary indices greater than 0.0042 at the 675–1020 nm wavelengths, whereas 95% of dust exhibits imaginary refractive indices less than 0.0042 at those wavelengths. Thus, it can be emphasised that although black carbon is the primary light absorber in the mid-visible range, brown carbon and dust also contribute significantly to absorption, particularly at shorter wavelengths.

To summarise, biomass burning is a significant source of BrC in the atmosphere, originating from uncontrolled sources like wildfires (Shetty *et al.*, 2023; Chakrabarty *et al.*, 2023; Palm *et al.*, 2020) and controlled sources such as cookstoves and domestic wood burning (Yan *et al.*, 2022; Targino *et al.*, 2023). In South and Southeast Asia, agricultural crop-residue burning is carried out on a large scale, emitting huge amounts of carbonaceous aerosols and trace gases in the atmosphere (Sahu and Sheel, 2014; Shaik *et al.*, 2019). In our earlier work (Alang *et al.*, 2023), we evaluated the influence of biomass burning activities on the light absorption properties of aerosols collected in Delhi, Indo-Gangetic Plain (IGP) in different seasons. Comparison of the seasonal trends showed relatively higher values of mass absorption coefficient in both polar and non-polar BrC extracts during post-monsoon season when there is intense post-harvest crop residue burning in the north-western IGP, along with air mass back trajectories and fire count data suggest that BrC aerosols over Delhi are significantly associated with biomass-burning emissions. Potential sources, measurement techniques, light absorption parameters, and chromophore compositions of several recently published results are summarised in Table 1. The emissions from biomass burning are expected to become increasingly important in the future due to the growing number and geographical spread of wildfires (Flannigan *et al.*, 2009). These emissions contain various chemical constituents resulting from the pyrolysis of lignin and cellulose, including phenolic compounds and organic acids. Pyrolysis products from lignin, which contain aromatic structures that absorb



visible light, are believed to contribute to BrC. However, a considerable portion of the BrC chromophores remains unidentified.

3 SECONDARY PATHWAYS OF BrC FORMATION

In addition to primary sources, atmospheric processing comprising multiple phases particularly when nitrogen oxides are present, significantly contribute to the generation of BrC chromophores in aerosol species. Several studies have pinpointed that nitrophenols/nitroaromatic compounds (NACs) are the potential representative BrC chromophores formed during daytime photo-oxidation reactions in both gas and aqueous phase reactions (Wang *et al.*, 2019b; Hems *et al.*, 2020; Li *et al.*, 2020c). In a recent study by Wang *et al.* (2019b), it has been proposed that the process of photo-oxidation involving toluene and benzene in the presence of NO₂ may have a greater significance in the secondary formation of nitrophenol and its related compounds which significantly contribute to the light absorption by BrC, surpassing the contribution from primary emissions of biomass burning. The rise in NACs was generally associated with higher levels of toluene and benzene, suggesting that the nitration of aromatic hydrocarbons, such as benzene and toluene, may constitute the significant secondary origin of NACs in the atmosphere.

Liu *et al.* (2023) examined the enhancing effect of ammonia in the secondary aqueous phase formation of BrC during humid haze period, resulting in the increased light absorption in PM_{2.5} aerosols collected during winter, 2019 in China. This inference was based on exponential increase in particulate imidazoles (IMs) (BrC chromophore) concentration with increasing aerosol water content and increasing pH (ammonia-rich conditions) observed in the humid haze events. On the contrary, in dry haze periods, primary biomass burning sources of IMs were more prominent as indicated by strong positive correlations with levoglucosan and mass ratio of EC/PM_{2.5}. The two types of BrC chromophores i.e., nitro-aromatic compounds (NACs) and IMs were detected using gas chromatography coupled with mass spectroscopy and high-performance liquid chromatography coupled with orbitrap-mass spectrometry, respectively. Seven NACs were detected in PM_{2.5} among which two gas phase nitrophenols (4-nitrophenol (4NP) and 3-methyl-4-nitrophenol (3M4NP)) were most abundant compared to aqueous phase products during the campaign. Additionally, a total of eight imidazole compounds were identified in the samples out of which 4(5)-methylimidazole (4MI) was the most abundant. IMs in the atmosphere can be generated through two pathways: either directly from emissions resulting from biomass burning or secondarily through aqueous reactions (Gao *et al.*, 2021). Fig. 1 depicts that in aerosol aqueous phase, NH₄⁺ undergoes dissociation, leading to the formation of NH₃(aq), which then engages in reactions with carbonyl compounds to generate primary and secondary imines and amines. Subsequently, the imine group and carbonyl groups undergo intramolecular cyclization to form nitrogen-containing heterocyclic compounds like IMs. Thereby, the authors proposed that formation of BrC chromophores such as IMs (N-heterocyclic chromophores) (in aqueous phase reaction) and nitroaromatic (in gas phase reaction) compounds is promoted in favourable conditions such as increasing pH, owing to increased NH₃ in the atmosphere.

Another study conducted by Zhou *et al.* (2023) demonstrated that the aqueous phase reaction of carbonyls (glyoxal and methylglyoxal) with reduced nitrogen compounds (ammonium sulfate, glycine, and dimethylamine) is an important formation mechanism of secondary brown carbon in the atmosphere. This inference is based on the presence of carbonyls, ammonium, amines, and imidazole in BrC aerosol species (Lian *et al.*, 2021). Results revealed that properties of BrC (optical, cloud condensation nuclei activity and effective particle density) measured at 532 nm wavelength varied with different reaction conditions such as reactant concentration, type, and reaction time.

Another significant but insufficiently investigated atmospheric process that may have a potential impact on the ever-changing/dramatic characteristics of BrC light absorption, is the nighttime NO₃ radical mediated oxidation reactions. This heterogeneous oxidation in the atmosphere is also important due to (i) rapid production rate of NO₃ radicals, and (ii) their high ambient concentrations (Brown *et al.*, 2011; Ng *et al.*, 2017), which contribute to the significant formation of BrC chromophores. Recent work in this area was done by Li *et al.* (2023), who demonstrated that

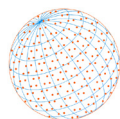
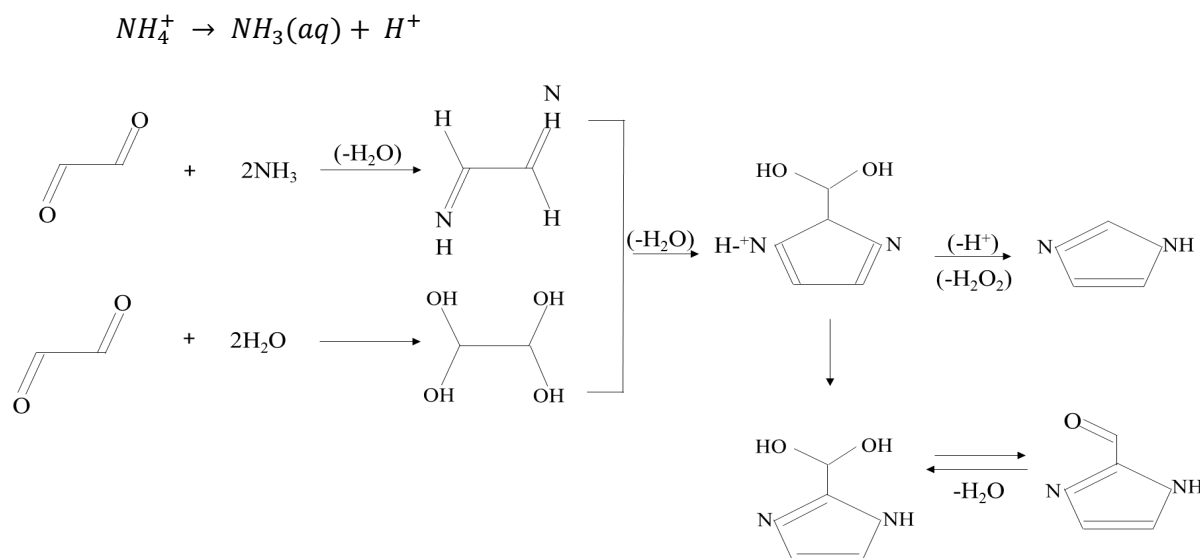
**Aqueous phase reactions**

Fig. 1. Secondary aqueous phase formation pathway of IMs (imidazoles) from reaction of carbonyls with ammonia in field study experiments by [Liu et al. \(2023\)](#).

Iron (Fe)-induced catalytic oxidation of ubiquitous phenolic compounds potentially results in BrC formation in the dark. Using Fourier transform ion cyclotron resonance mass spectrometry (FT-ICR MS), they were able to detect molecular components of the products, optical characteristics and reaction dynamics formed by reaction of 13 selected phenolic compounds with Fe^{3+} . The authors proposed structure-dependent formation of BrC from different phenolic precursors.

In one of the early studies by [Bolzacchini et al. \(2001\)](#), it has been demonstrated through laboratory chamber studies that nitration of phenols and cresols in the gas phase is facilitated by nighttime NO_3 radicals to yield 2-nitrophenol as the major product. These reactions serve as a significant mechanism for scavenging aromatic compounds at nighttime in gas as well as aqueous phase, and contribute to the formation of corresponding nitroderivatives. Phenols are recognized to be generated in the atmosphere through the oxidation of aromatic hydrocarbons initiated by OH radicals and have substantial atmospheric yields ([Rindone et al., 1999](#)). They can undergo subsequent transformation to form nitrophenols, which are also known to have detrimental effects on plants, often causing damage ([Lüttke et al., 1997](#)). This reaction is accelerated in the presence of NO_3 radicals which are formed in the atmosphere through the interaction of O_3 and NO_2 ([Finlayson-Pitts and Pitts, 1999](#)):



The nitration products obtained from gas phase reactions between phenol and NO_3 radicals were analysed using online FTIR and offline HPLC. The origin of nitrophenols in the atmosphere is elucidated based on the ratio of their product isomers.

A recent study by [Jiang et al. \(2019\)](#) provides evidence of increased formation of BrC during nighttime atmospheric processing of fire-derived heterocyclic compounds in the presence of NO_3 radicals. Since heterocyclic compounds (volatile organic compounds (VOCs) predominantly found in smoke from biomass burning) are known to be potential sources of atmospheric BrC, the authors examined the impact of nitrate radical initiated oxidation on three unsaturated heterocyclic compounds (pyrrole, furan, and thiophene). Results indicated substantially higher yield and enhanced light absorption of SOA generated from NO_3 oxidation of pyrrole (i.e., 2- and 3-nitropyrrole) compared to that of furan (2-nitrofuran) and thiophene (2-nitrothiophene). Furthermore, high-resolution mass spectrometry analysis was performed to evaluate the molecular

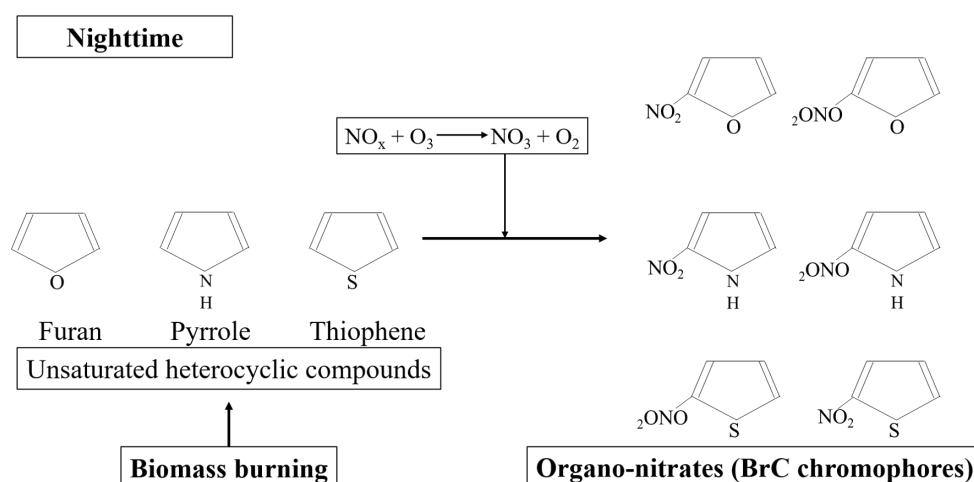
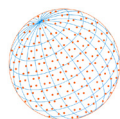


Fig. 2. A proposed reaction scheme for the production of brown carbon (BrC) from nighttime oxidation of unsaturated heterocyclic compounds mediated by NO_3 radicals in chamber studies by Jiang *et al.* (2019).

composition of SOA. Unsaturated heterocyclic nitro compounds or organonitrates were identified as potential light-absorbing constituents in BrC originating from each of the three precursor compounds as shown in Fig. 2.

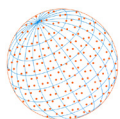
4 ATMOSPHERIC REACTIONS/INTERACTIONS OF BrC

The chemical composition and spectral characteristics of BrC, whether emitted from primary sources or formed secondarily, undergo changes as it progresses through its atmospheric lifespan. This transformation can lead to either a reduction or enhancement in its potential to absorb UV and visible light, a phenomenon referred to as “bleaching” (photochemical reactions initiated by OH radical and ozone during the daytime) or “darkening”, (oxidation induced by NO_3 radicals during nighttime), respectively.

Recently, Fang *et al.* (2023) studied the changes in light-absorption properties of BrC during atmospheric transport for 3 days from continental East Asia to a south-east Yellow Sea receptor, by evaluating optical properties and ageing-diagnostic $\delta^{13}\text{C}$ -BrC. The isotope signature indicates a robust negative association between the light-absorption of BrC and the age of its source material. In an important pioneer study by Aggarwal and Kawamura (2008), the enrichment in $\delta^{13}\text{C}$ isotope is linked with photochemical aging of aerosols. When combined with findings from South Asia, it becomes evident that there is a remarkable consistency between these two significant emission areas. The attenuation in BrC light absorption, is suggested to be primarily linked to photochemical processes/aging rather than its sources and this feature is probably a widespread occurrence. This consistency is characterized by the rapid photobleaching of BrC, which is attributed to its elevated intrinsic light-absorbing properties resulting from biomass burning.

Atmospheric BrC has light absorbing properties and simultaneously can undergo photolysis, which is anticipated as an important sink of BrC chromophores (Han *et al.*, 2020). It is assumed that co-photolysis mechanism occurs in complex BrC aerosol species. In this light, Wang *et al.* (2023) studied the effect of four BrC chromophores (two methoxyphenols (MPs) and two nitrophenols (NPs)), their concentration, and particle size on atmospheric lifetimes due to co-photolysis with a view to better constrain shadowing and sensitizing phenomenon. When estimating the duration of time particles remain in the atmosphere, it is proposed that the presence of MPs can enhance the degradation of NPs by a factor of up to 30. While in fine particles, the effect of shadowing between them is insignificant, it does have a noticeable impact on coarse particles.

On the other hand, recent studies have revealed that BrC light absorption is enhanced (darkening effect) as a result of interactions with NO_3 radicals, NO_x , mineral dust, and several other secondary atmospheric processing mechanisms (Laskin *et al.*, 2012; Cheng *et al.*, 2020; Li *et al.*, 2020a).



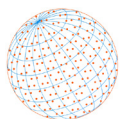
In a laboratory study by [Cheng *et al.* \(2020\)](#), authors examined the influence of NO₃ facilitated oxidation on the chemical composition and light absorption properties of BrC within an oxidation flow reactor. The BrC particles were generated from toluene combustion experiments which included a diverse blend of light-absorbing polycyclic aromatic hydrocarbons (PAHs). The results showed around 50% augmentation in BrC light absorption properties (quantified in terms of the imaginary part of the refractive index (*k*) measured at 532 nm) due to NO₃ exposure. This is ascribed to the introduction of chromophore functional groups through NO₃ induced heterogeneous oxidation. On the other hand, condensable products formed from the reactions of gas-phase PAHs with NO₃ radicals might somewhat counterbalance this improvement, because they were less absorbing than the particulate PAHs. Overall, these findings suggest that the heterogeneous oxidation induced by NO₃ leads to the darkening of atmospheric BrC which is more dominant and therefore this darkening effect can offset the bleaching effects caused by OH oxidation. The evidences from the above studies imply that NO₃ induced heterogeneous oxidation darkens atmospheric BrC.

In another recent study it was demonstrated that the chemical composition and light absorbing properties of toluene-SOA (Tol-SOA) (i.e., BrC chromophore formed by toluene photo-oxidation in presence of NO_x) rely significantly on the initial NO_x concentration as well as interaction with mineral dust/aerosols ([Lin *et al.*, 2015](#)). The results indicate that Tol-SOA produced when there is a high concentration of NO_x (defined by the authors in this case as an initial ratio of NO_x to toluene of 5/1) exhibits a yellowish appearance, and the mass absorption coefficient of the overall sample is approximately 80 times greater than what was observed for the Tol-SOA sample (i.e., colorless) generated under low-NO_x conditions (NO_x/toluene < 1/300). Furthermore, the BrC characteristics of Tol-SOA enhanced on interaction with mineral dust. It has been proposed that the acidic nature of SOA is neutralised on interaction with mineral dust or marine aerosols (which are mainly composed of alkaline components) ([Wang and Laskin, 2014](#)), leading to significant alterations in the light-absorption properties of nitroaromatic chromophores. This reaction proceeds the same way in which the acidity of rain water is buffered in the presence of mineral dust ([Fujita *et al.*, 2000](#)). A recent report indicated that a significant portion of adsorbed nitrophenols on the surfaces of mineral aerosols undergo deprotonation, resulting in a red-shift of the UV-Visible absorption cross-section extending up to 650 nm ([Hinrichs *et al.*, 2016](#)). This study suggests that the acidity of aerosols plays a crucial role in regulating the light-absorbing characteristics of BrC and therefore require comprehensive examination in forthcoming research.

5 POTENTIAL PROXY SPECIES OF BrC

The broad assortment of compounds identified in BrC in various research investigations highlights the intricate nature of BrC composition and underscores the necessity for a more comprehensive measure to establish the connection between BrC's optical characteristics and its chemical makeup.

Regarding its chemical composition, the extensive group of humic-like substances (HULIS) is often associated with atmospheric BrC as demonstrated in a series of investigations on atmospheric aerosols ([Mukai and Ambe, 1986](#); [Pöschl, 2003](#); [Krivácsy *et al.*, 2000](#); [Graber and Rudich, 2006](#); [Claeys *et al.*, 2012](#); [Utry *et al.*, 2013](#); [Wang *et al.*, 2019a](#); [Han *et al.*, 2020](#)). Conventionally, humic substances are typically defined as naturally produced, biologically originated, diverse organic compounds that exhibit colors ranging from yellow to black, possess high molecular weights, and are resistant to degradation ([Aiken *et al.*, 1985](#)). They are recognised as the important components responsible for light absorption in BrC at a wavelength of 365 nm, with their major source being emissions from biomass burning. Recently, [Wang *et al.* \(2019a\)](#) identified molecular composition of HULIS and found nitro-aromatic compounds (oxidised N-functional groups) and N-heterocyclic bases (reduced N-structures) as the major chromophore species in aged and fresh biomass burning organic aerosols, respectively. These inferences were based on mass spectra analysis of laboratory generated and field burning samples. Likewise, [Claeys *et al.* \(2012\)](#) chemically analysed HULIS from biomass burning aerosols and provided evidence that nitro-aromatic catecholic compounds constitute a significant portion of HULIS. In another study, species such as carbonyl oxygenated polycyclic aromatic hydrocarbons (OPAHs) with 2–3 aromatic rings in addition to nitrophenols were attributed to the light absorption by HULIS-BrC at 365 nm ([Huang *et al.*, 2020](#)).



Another comprehensive term, “tar balls” (Pósfai *et al.*, 2004), which are common products of wood pyrolysis, have been employed to classify a category of BrC and are released in copious amounts in fire emissions/biomass combustion (Hand *et al.*, 2005; Sedlacek *et al.*, 2018). Previous studies have recognised laboratory-generated wood tar aerosols as the suitable representative for BrC derived from biomass burning, since they strongly absorb light in UV-Vis region (Hoffer *et al.*, 2016; Li *et al.*, 2019; Li *et al.*, 2020a) and have highly similar physico-chemical properties compared to those of ambient tar aerosols (Pósfai *et al.*, 2004). In recent laboratory study by Li *et al.* (2020a), BrC light absorption is predominantly attributed to nitroaromatics/nitrophenols detected as major products in aged wood tar aerosols. The combustion derived tar products were reacted with NO₃ radicals in a flow reactor to identify new light absorbing species and study the influence of heterogeneous nighttime NO₃ mediated aging process on chemical composition and spectral properties of wood tar particles by online and offline mass spectrometry (315–650 nm). According to chemical characterisation, nitro-aromatic species are important chromophores in secondary BrC aerosols derived from biomass burning during nighttime NO₃ reactions which result in enhanced light absorption properties. Apart from aromatic and heteroatom (N- and O-) containing organic compounds, wood tar particles consist of high content of amides, conjugated species bearing oxygenated functional groups (such as hydroxy, methoxy, carbonyl and carboxyl), abiatic acid derivatives and lignin pyrolysis products (2-methoxyphenol). Similar BrC chromophores were detected in wood tar condensates produced in the laboratory through biomass pyrolysis and oxidative combustion conditions using high-performance liquid chromatography-photodiode array detection-high-resolution mass spectrometry (HPLC-PDA-HRMS) analysis method (Hettiyadura *et al.*, 2021).

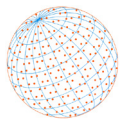
Lately, Mayorga *et al.* (2021) have identified new BrC chromophores generated from nighttime NO₃ oxidation of phenolic VOCs using electrospray ionization ion mobility spectrometry time-of-flight mass spectrometry (IMS-TOF). These new products encompass: multifunctional aromatic species consisting both -NO₂ and -OH groups, non-aromatic/ring opening nitro products, nitrated diphenyl ether dimers, and phenol and catechol products when C-containing substituents are present in precursor phenolic VOCs.

As discussed above, nitro-aromatics (particularly, nitrophenols), PAHs, and oxygenated aromatic species in addition to lignin pyrolysis products (methoxyphenols) and cellulose/hemi-cellulose decomposition products (Furan derivatives) are the major/important chromophores attributed to the light absorption by atmospheric BrC. It is important to note that among these species, particularly the aged/secondarily formed aerosols have high content of nitrophenols, which are also ascribed to the significant augmentation in BrC light absorption (Wang *et al.*, 2019a, 2019b; Li *et al.*, 2020a; Hems *et al.*, 2020; Li *et al.*, 2020c; Mayorga *et al.*, 2021; Rastogi *et al.*, 2021). Thus, it can be said that atmospheric secondary BrC formation processes are more important for studying spectral properties than primary sources, since the former source/pathway contributes significantly to the BrC light absorption. In addition, substantial high concentration of nitrophenols might potentially indicate aged/secondarily processed aerosols.

6 ADVANCEMENTS IN THE TECHNIQUES FOR MEASURING ATMOSPHERIC BROWN CARBON

Due to the diverse origins of atmospheric BrC, there are no universally recognized conventional techniques or reference materials for measuring BrC. In this section, we briefly review and summarise various commonly used approaches employed for determination of BrC in recent studies, highlighting their advantages and limitations, with a view to emphasise the need for developing/implementing standard characterization techniques.

Due to the ambiguous physical and chemical properties of HULIS, there is no standardized extraction method that can be universally applied under all circumstances. The most promising method for extraction of HULIS-C (consisting of optically active BrC) from water-soluble organic matter (WSOM) is the solid phase extraction (SPE) method developed by Varga *et al.* (2001). Their method involved applying acidified aqueous extracts (pH = 2) to HLB (Hydrophilic-Lipophilic Balanced polymer: Oasis HLB, waters, USA; 3 cc, 60 mg) cartridge, followed by methanol elution. Subsequently, the inorganic elements passed in the effluent, along with approximately 40% of



the total WSOC in the sample. The remaining 60% of WSOC, which was desorbed from the cartridge using methanol, held over 90% of the fluorescence and roughly 70% of the UV reactivity within the overall WSOC segment. The utilization of this technique offers several benefits, including efficient elimination of inorganic salts, and minimal, if any, irreversible adsorption on the column, allowing for the elution of enriched HULIS from ambient aerosols.

However, this method lacked validation with model low molecular weight (LMW) organic acids, fulvic acids, or other humic substances, making it uncertain regarding the distribution of hydrophilic and hydrophobic acids in the various fractions. Also, it is important to note that most of the investigations into HULIS focus on the water-soluble portion, encompassing solely the fulvic acid component of humic substances. This approach excludes the humic acid (soluble in base) and humin (insoluble) fractions of humic substances, thus undermining the total optical properties. Most of the pioneer as well as recent studies have followed SPE method developed by [Varga *et al.* \(2001\)](#), for extraction and separation of HULIS for subsequent chemical characterisation ([Salma *et al.*, 2007](#); [Feczko *et al.*, 2007](#); [Zheng *et al.*, 2013](#); [Zhao *et al.*, 2016](#); [Kumar *et al.*, 2017](#); [Afsana *et al.*, 2022](#); [Alang *et al.*, 2023](#)). There is a clear need for a more thorough and precise method for extracting and isolating atmospheric HULIS substances.

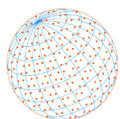
Several studies have derived the spectral properties of bulk BrC (and HULIS) using filter-based solvent extraction method ([Kumar *et al.*, 2017](#); [Lee *et al.*, 2021](#)) followed by direct measurement of spectrophotometric properties of filter extracts. The merit of this method is that it negates interferences from insoluble absorption material like BC ([Cheng *et al.*, 2016](#)) and provides a high-resolution spectrum over wide wavelength. So far, the majority of research utilize ultrapure water to extract organic matter from aerosols, allowing for the measurement of the optical properties of water-soluble BrC ([Srinivas *et al.*, 2016](#); [Wu *et al.*, 2019](#); [Chen *et al.*, 2020](#); [Deng *et al.*, 2022](#); [Zhang *et al.*, 2024](#)), while the non-polar or semi-polar BrC fraction has been generally ignored. However, it is important to examine the contribution of non-polar organic compounds to light absorption of BrC, since species like PAHs, PACs, and their derivatives are significant light absorbers and that their light absorption efficiency could be even greater than that of water-soluble BrC fraction ([Huang *et al.*, 2018](#); [Sengupta *et al.*, 2018](#); [Yan *et al.*, 2020](#); [Sun *et al.*, 2021](#)). In our previous work, we studied the light absorption properties of BrC in aerosol extracted with water, methanol, and n-hexane using filter-based extraction method followed by spectrophotometric analysis ([Alang *et al.*, 2023](#)). Results indicated that the mass absorption coefficient of methanol extracts at 365 nm was about 2 times higher than that of water extracts, and about 4 times higher than hexane extracts. Hence, for a thorough understanding of the sources and atmospheric processing of BrC it is important to extract water-soluble and water-insoluble organic components separately by employing solvents with varying polarities in a sequential manner.

Combining spectrophotometric results with other analytical techniques such as HPLC and mass spectrometry ([Lin *et al.*, 2015](#); [Lin *et al.*, 2016](#); [Jiang *et al.*, 2019](#)) may help us for more comprehensive characterisation of BrC chromophores. However, the measured spectrophotometric signals might get superpositioned from each component present in complex heterogeneous atmospheric BrC samples ([Graber and Rudich, 2006](#)). Therefore, comparable spectroscopic findings do not inherently suggest similar compositions. Furthermore, other important characteristics of the particles like aerosol structure, size, phase, and mixing state, cannot be evaluated from this method ([Shetty *et al.*, 2019](#)).

The evaluation methods used for studying BrC spectral properties, can broadly be categorised into chemical and optical methods, which depend on highly sensitive spectroscopic measurements. These methods have not been validated due to the absence of BrC reference materials. So, to elucidate whether the different methods actually isolate and extract identical or comparable BrC fractions, inter-comparison between different online and offline measurement methods is very crucial. Subsequent research endeavours might utilize a blend of online and offline measurements to more accurately assess BrC emissions.

7 RADIATIVE FORCING OF BrC

Atmospheric carbonaceous aerosols consisting of BC and BrC influence the Earth's radiative balance by interaction with cloud and solar radiation. As per the Sixth Assessment Report of the



Intergovernmental panel on Climate Change (IPCC, 2013), the average global radiative forcing resulting from carbonaceous aerosols between 1750 and 2019 is estimated at -0.77 (-1.15 to -0.31) W m^{-2} , with a 95% confidence level (Forest, 2018). It is noteworthy that in regions where predominant biomass burning occurs, the absorption of BrC is similar to that of BC in the upper troposphere, and the resultant radiative heating tends to stabilize the atmosphere (Zhang *et al.*, 2017; Zeng *et al.*, 2020). Yet the current climate models do not account for the proper parametrisation of BrC (Brown *et al.*, 2018; Saleh, 2020); thereby, there are large uncertainties in observed and simulated net forcing estimates. Incorporating BrC absorption into climate models can lead to a more precise representation of the climatic impacts of aerosols.

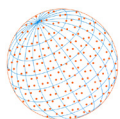
Global climate models have been applied to account for BrC absorption by estimating direct radiative forcing (DRF) and direct radiative effects (DREs). Aerosol DRE indicates the variation in the radiative balance with and without aerosols, while DRF denotes the disparity in DRE between the present day and pre-industrial eras (Heald *et al.*, 2014). Lin *et al.* (2014) calculated a BrC DRE ranging from $+0.22$ to $+0.57$ W m^{-2} , which represented 27% to 70% of the BC absorption in their model. The study by Brown *et al.* (2018) evaluated the global BrC DRE in terms of aerosol-radiation interaction ranging from 0 to $+0.13 \pm 0.01$ W m^{-2} during 2003–2011 using Community Atmosphere Model Version 5 (CAM5), taking into account the effect of emission of immediately bleached BrC to no bleaching BrC. They suggested that global low cloud cover decreases as a result of BrC semi-direct effects and proposed the inclusion of lensing effects and organic aerosol absorption in radiative transfer calculations. Drugé *et al.* (2022) estimated the annual global BrC radiative effect in terms of both aerosol-radiation interactions (0.029 ± 0.006 W m^{-2}) and aerosol-cloud interactions (-0.024 ± 0.066 W m^{-2}) over the period of 2000–2014. Furthermore, studies show that the spatial pattern of BrC radiative forcing exhibited significant heterogeneity (Zeng *et al.*, 2020). The DRE of BrC absorption is more pronounced in areas with intense biomass-burning activity, such as South America, Africa, and Southeast Asia, where it exceeds $+1$ W m^{-2} (Wang *et al.*, 2018; Zeng *et al.*, 2020; Zhu *et al.*, 2021; Sun *et al.*, 2023), markedly surpassing the global-average DRE. This emphasises that BrC from BB is a crucial component of the global radiative balance.

In addition to global model studies, several field and experimental studies have attempted to constrain DRF of BrC by estimating simple forcing efficiency (SFE_{BrC}) (Chylek and Wong, 1995; Bond and Bergstrom, 2006), which represents energy added to the atmosphere by a given mass of aerosols in watts per gram. The study by Bikkina and Sarin (2019) estimated SFE_{BrC} for the Indo-Gangetic Plain outflow with values ranging from 0.4 to 10.2 W g^{-1} (average = 2.5 ± 1.9 W g^{-1}), 0.3–1.9 W g^{-1} (0.7 ± 0.4 W g^{-1}), 0.3–1.9 W g^{-1} (1.0 ± 0.3 W g^{-1}), and 0.4–1.3 W g^{-1} (0.8 ± 0.2 W g^{-1}) over Patiala, Bay of Bengal, Kharagpur, and North Indian Ocean, respectively. Comparable values have been observed for laboratory biofuel-burning combustion aerosols (Lei *et al.*, 2018). Deng *et al.* (2022) analysed radiative effects of water-soluble BrC fraction in Northern China during 2016–2017 and found that DRF by BrC absorption relative to BC in 300–400 nm range was about 45 to 54%. In their study, the average SFE_{BrC} of aerosols was 4.6 ± 1.7 W g^{-1} to 6.2 ± 2.0 W g^{-1} during summer and winter season, respectively. Recently, Alang *et al.* (2023) estimated the average SFE_{BrC} in water-soluble (5 ± 2.2 W g^{-1}) and methanol-soluble BrC extracts (3.2 ± 1.1 W g^{-1}) in aerosol samples collected in Delhi during 2020–2021.

Despite a growing number of global model and field studies constraining radiative effects of BrC absorption, there exists a paucity of data on the parameterization of BrC sources, atmospheric processing of aerosols such as photobleaching, and long-range transport of air masses. Another challenge in global climate models is the limitation of clearly distinguishing BrC, OC, and BC emissions, which needs further study. More number of field measurements correlating molecular-level analysis with radiative effects of BrC especially over biomass burning dominant regions are crucial; however, so far these are lacking in the literature.

8 OPTICAL PROPERTIES OF BROWN CARBON IN THE CRYOSPHERE-RECENT STUDIES

Cryosphere is an important component of the Earth's climate/radiative system due to its high surface albedo (IPCC, 2013). Major light absorbing aerosols like BC, mineral dust, and BrC are



ubiquitous in the atmosphere and can undergo long-range transport. After being removed from the atmosphere by wet and dry deposition, they are deposited on snow and thereby effectively reduce the snow albedo (Hall, 2004) and also contribute to glacier's retreat processes (Ramanathan and Carmichael, 2008). Further it may be noted that, compared to BC, a considerable fraction of BrC is water-soluble (Hoffer *et al.*, 2006). This suggests the significant ability of BrC aerosols to dissolve within cloud droplets and affect cloud albedo, heating and evaporation of clouds. However, there is limited understanding regarding the impact of BrC deposition onto snow surfaces on the spectral albedo of snow, potentially leading to changes in radiative forcing and in-snow photochemical processes. In this section, we discuss the recent advancements in this field, particularly in high altitude Tibetan plateau and Himalayan region, where limited number of BrC studies have been conducted so far.

Zhou *et al.* (2022) identified major water-soluble BrC chromophores, their potential sources and light absorption contribution in snowpack samples collected from northern Xinjiang (northwest China) for the first time. The aqueous filtrates were extracted using SPE method followed by high-performance liquid chromatography-photodiode array detection-high-resolution mass spectrometry (HPLC-PDA-HRMS) analysis. Based on the obtained results, nitro-aromatics, oxygenated-aromatics, flavonoids, and phenol/lignin derived compounds were the major light absorbing species which attributed to approximately 23 to 64% of the total light absorption in the range 300–370 nm.

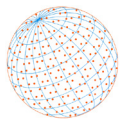
Niu *et al.* (2020) elucidated optical properties of WSOC in snow, river runoff, and atmospheric samples from Baishui glacier on Mt. Yulong (temperate glacier in southeast Tibetan Plateau). The light absorption spectra of the filtered water-soluble extracts were measured using an ultraviolet-visible spectrophotometer covering the spectral wavelength range of 200–800 nm. The authors observed the highest average mass absorption cross-section of WSOC value (MAC_{WSOC}) at $4.07 \pm 0.33 \text{ m}^2 \text{ g}^{-1}$ in the surface-aged snow samples. This result along with distribution of active fire spots indicated that glacier snow serves as a favourable medium for the deposition of biomass-derived light absorbing contaminants that have undergone long-distance transport in high-altitude regions. Furthermore, the aerosol samples exhibited the least MAC_{WSOC} value compared to the other sample types, and this is assumed to be linked to local emissions from private vehicles and tour buses.

In recent study by Li *et al.* (2022), authors examined the optical characteristics and the composition of WSOC in surface ice and snow from four glaciers on the Tibetan Plateau by utilizing fluorescence spectrometer and electrospray ionization-Fourier transform ion cyclotron resonance mass spectrometry. WSOC exhibited light absorption in the ultraviolet region (250–400 nm) and the visible region (400–600 nm), contributing approximately 60.42% and 27.17% of the overall organic light absorption, respectively. Four fluorescent components were detected in snow/ice samples collected at the campaign glaciers, among which two are associated with protein-like substances (PRLIS), one chromophore linked to HULIS and, one defined as undefined species.

The sensitivity of the Himalayan cryosphere to climate change is heightened majorly due to the long-range transport of anthropogenic emissions by upslope breezes from the Indo-Gangetic Plain (Bonasoni *et al.*, 2010; Reddy *et al.*, 2015), where light-absorbing aerosols contribute to glacier's melting process. Kirillova *et al.* (2016) studied light absorption properties of water-soluble (WS-BrC) and methanol-soluble BrC (MS-BrC) in PM_{10} aerosols at the Nepal Climate Observatory-Pyramid (NCO-P) station, located in the foothills of Mt. Everest. Results revealed that the light absorption coefficient of MS-BrC was two times higher than that of WS-BrC at 365 nm and about three times more at 550 nm, showing seasonal trend with maximum values during pre-monsoon season and minimum during monsoon.

Lately, Verma *et al.* (2023) evaluated the optical properties of fine aerosols collected in the western, central, and eastern Himalayan glacial region and observed highest light absorption in the near UV-Vis range (200–400 nm) attributing to BrC aerosols. Furthermore, a positive radiative forcing was observed, suggesting net warming in this region.

In a recent work by Choudhary *et al.* (2023), brown carbon lifetime in the Himalayan atmosphere and South Asia was studied from published datasets in past 15 years. Results showed that the decay rate of the water-soluble BrC imaginary refractive index ($k_{WS-BrC-365}$) in the Himalayan region ($0.09 \pm 0.02 \text{ day}^{-1}$) was about two times slower than the South Asian outflow ($0.17 \pm 0.04 \text{ day}^{-1}$).



This suggests their longer atmospheric half-life in the Himalayan atmosphere compared to the South Asian outflow with approximately 8 days and 4 days, respectively. Slower decay in the cryosphere atmosphere is due to low temperature and relative humidity. Thus, considering the delayed removal of light absorbing aerosols and their heightened climatic effects particularly in cryosphere environment underscore the necessity and pressing demand for conducting measurements and employing more advanced model simulations.

The recognition of specific chromophores and the measurement of their optical characteristics are valuable in understanding how BrC influences the radiative equilibrium and photochemical processes in snow. There exists a huge knowledge gap concerning molecular identification and optical properties of BrC chromophores in hotspot areas of cryosphere. From the above studies we deduce predominant light absorption ability of BrC in the UV-Vis region, proving its non-negligible presence even pristine/remote locations in cryosphere. Furthermore, water-insoluble fraction of BrC has substantial light absorption properties compared to water-soluble fractions in cryosphere regions, similar to that in low latitude regions.

9 CONCLUSION

As alluded above, there are several challenges in the accurate measurement of atmospheric BrC. Light absorption properties of bulk organic fractions are often assessed in water extracts, but the unaccounted portion of BrC, the water-insoluble fraction, is typically overlooked. Notably, the semi-polar, and non-polar fractions may exhibit a higher mass-specific absorption compared to the water-soluble fraction.

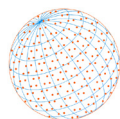
Another challenge arises from the lack of universally recognized standardized methods or reference materials for BrC determination attributing to its wide range of sources and complex chemical composition. We reviewed some recent studies to identify newly identified BrC chromophores and linked them to their potential sources and formation pathways in the atmosphere. Specific candidate substances that contribute to BrC absorption have been recognized. These categories encompass humic-like substances, aromatic/phenolic compounds, high-molecular-weight organic compounds containing heteroatoms, and oligomeric variations of these substances as discussed above. Since light absorption is directly associated with BrC in aerosols, it is imperative for the scientific community to reach a consensus on a standard reference material for quantifying BrC in carbonaceous aerosols.

Furthermore, various measurement methods for the determination of distinct polarity BrC fractions have been used in different studies. Consequently, comparing their abundances, optical properties, chemical, physical, and molecular composition obtained through different pretreatment and analytical methods becomes a complex task. It was also observed that nearly all the methods currently employed to quantitatively determine BrC had few limitations. Optimization measures are required to address the limitations of individual methods and making them somewhat inter-comparable to each other.

Regarding the geographical locations of research sites, even though numerous studies have been carried out in Alpine, Arctic, and Antarctic regions, there remains a lack of information regarding the spatial distribution of BrC, particularly in the Himalayan and Tibetan Plateau areas. Given that biomass burning and secondary formation are the primary contributors to BrC, further exploration is warranted to understand the climatic impacts of BrC in these pristine areas. This entails not only the assessment of atmospheric warming but also the evaluation of its effects on reducing snow albedo.

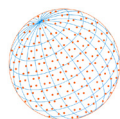
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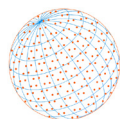


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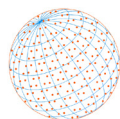
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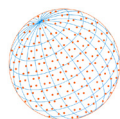
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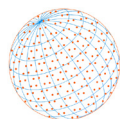
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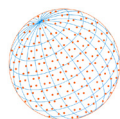
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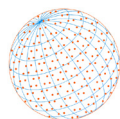
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