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Light absorbing organic aerosols (brown carbon) over the tropical Indian Ocean: impact of biomass burning emissions

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Abstract
The first field measurements of light absorbing water-soluble organic carbon (WSOC), referred as brown carbon (BrC), have been made in the marine atmospheric boundary layer (MABL) during the continental outflow to the Bay of Bengal (BoB) and the Arabian Sea (ARS). The absorption signal measured at 365 nm in aqueous extracts of aerosols shows a systematic linear increase with WSOC concentration, suggesting a significant contribution from BrC to the absorption properties of organic aerosols. The mass absorption coefficient (b_{abs}) of BrC shows an inverse hyperbolic relation with wavelength (from ~300 to 700 nm), providing an estimate of the Angstrom exponent (α_P, range: 3–19; Av: 9 ± 3). The mass absorption efficiency of brown carbon ($\sigma_{abs-BrC}$) in the MABL varies from 0.17 to 0.72 $m^2\ g^{-1}$ (Av: 0.45 ± 0.14 $m^2\ g^{-1}$). The α_P and $\sigma_{abs-BrC}$ over the BoB are quite similar to that studied from a sampling site in the Indo-Gangetic Plain (IGP), suggesting the dominant impact of organic aerosols associated with the continental outflow. A comparison of the mass absorption efficiency of BrC and elemental carbon (EC) brings to focus the significant role of light absorbing organic aerosols (from biomass burning emissions) in atmospheric radiative forcing over oceanic regions located downwind of the pollution sources.

Keywords: water-soluble organic carbon, brown carbon, elemental carbon, mass absorption efficiency, Angstrom exponent, biomass burning emissions

S Online supplementary data available from stacks.iop.org/ERL/8/044042/mmedia

1. Introduction

The rapid increase in anthropogenic activities and enhanced emissions of trace gases and particulate matter (organic and inorganic species) to the atmosphere over south and south-east Asia has been a subject of major debate in recent years (Lawrence and Lelieveld 2010, Ramanathan *et al* 2001). The emission and subsequent long-range transport of pollutants to the remote marine regions can significantly alter the chemical composition of the atmosphere (namely, cloud activation, enhancement in the fractional solubility of toxic trace metals). The relevance of the chemical composition of ambient particulate matter in a changing climate scenario is also of prime concern (Fuzzi *et al* 2006). In particular, a considerable mass of organic aerosols remains unidentified, leading to a large degree of uncertainty in assessing the climate change scenario (Huebert and Charlson 2000). In this context, recent studies have highlighted the importance of light absorbing organic aerosols, referred as brown carbon, ubiquitous and abundant over rural, urban and remote continental and marine locations (Alexander *et al* 2008, Andreae and Gelencsér 2006, Yang *et al* 2009).

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Several studies have made reference to the presence of atmospheric brown carbon (BrC) based on the spectral absorption properties of aqueous extracts of continental aerosols (Bahadur *et al* 2012, Chakrabarty *et al* 2010, Hoffer *et al* 2004, Kirchstetter *et al* 2004, Limbeck *et al* 2003). Likewise, the presence of BrC has been studied using spectrally resolved light absorption measurements on aerosols from specific combustion sources (Bond 2001, Chen and Bond 2010). A significant overlap in the absorption proprieties of BrC and humic-like substance (HULIS) is noteworthy in the ambient aerosols derived from biomass burning emissions during the LBA–SMOCC (Large scale Biosphere atmosphere experiment in Amazonia—SMOke aerosols, Clouds, rainfall and Climate) experiment ((Hoffer *et al* 2004, 2006)). It has been inferred that biomass burning is a primary source of HULIS and, thus, implicitly of brown carbon (Hoffer *et al* 2004, Lack *et al* 2012, Limbeck *et al* 2003, Lukács *et al* 2007). The light absorbing organic aerosols may also originate from biogenic material; their oxidation and polymerization at low temperature were first addressed by Andreae and Crutzen (1997). A detailed understanding of the sources of light absorbing organic species is essential in order to assess their contribution to climate forcing. In this paper, we have studied the absorption properties of the water-soluble organic fraction (BrC) of aerosols from the marine atmospheric boundary layer (MABL) of the Northern Indian Ocean (Bay of Bengal: BoB and Arabian Sea: ARS).

2. Methodology

2.1. Sample collection and analysis

Bulk aerosol (TSP) samples ($N = 14$ from the ARS and $N = 11$ from the BoB) were collected using the STAPLEX high volume air sampler, installed onboard FORV Sagar Sampada, from 8 to 30 November 2008 (figure 1). Subsequently, aerosols in two size fractions ($N = 31$ of $PM_{2.5}$ and $N = 33$ of PM_{10}) were collected from the MABL of the BoB during 27 December 2008–28 January 2009 (figure 1). These samples have been used to study the absorption properties of the water-soluble organic fraction (BrC). A portion of the aerosol filter (one quarter) was extracted with Milli-Q water (specific resistivity ≈ 18.2 MΩ cm) and filtered through pre-combusted (at 400 °C for 6 h) glass fiber filters (Whatman Co., GF/F type). An aliquot of filtered extract was used for the measurement of water-soluble organic carbon (WSOC) on a total organic carbon analyzer (TOC-5000A, Shimadzu Inc.). Working standard solutions of potassium hydrogen phthalate (KHP), made from a 1000 ppm aqueous solution, were used for the assay of WSOC. Measured concentrations of WSOC in the samples were suitably corrected for blanks (contribution of the blank varying from 2 to 40% of the maximum and minimum signal). All aerosol samples were analyzed for elemental and organic carbon (EC & OC) on a Sunset EC–OC analyzer using the NIOSH protocol (Birch and Cary 1996). For analytical details reference is made to our earlier publications (Ram *et al* 2010 and reference therein; Rengarajan *et al* 2007). Another aliquot of water

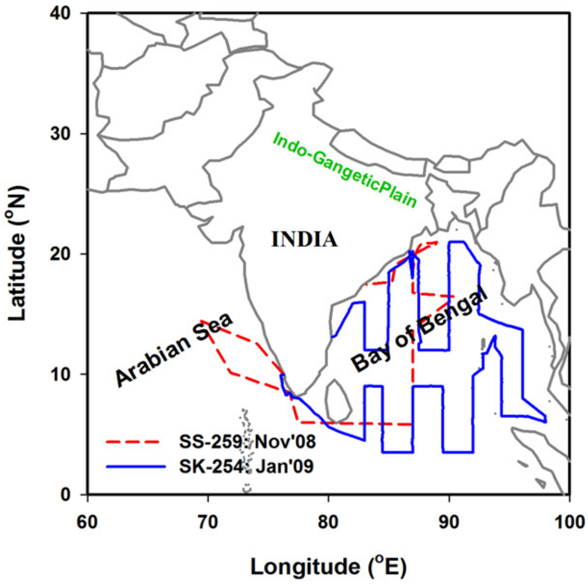

Figure 1. Cruise tracks undertaken in the Northern Indian Ocean during November 2008 and January 2009.

extract was used for a spectral scan from 300 to 700 nm in wavelength, using a 2 m long liquid-core waveguide capillary cell (LWCC from WPI Inc.) connected in line with a USB-4000 (from Ocean Optics Inc.) spectrophotometer. Based on the repeat number of samples ($N = 10$), the overall analytical reproducibility in the absorption measurement was within 10%.

2.2. Absorption properties of brown carbon

The measured absorbance at 365 nm relative to 700 nm is used to assess the mass absorption coefficient of BrC ($b_{\text{abs-365}}$, expressed in $\text{M m}^{-1} = 10^{-6}\ \text{m}^{-1}$), mass absorption efficiency (MAE) of BrC (i.e., $\sigma_{\text{abs-BrC}}$, expressed in $\text{m}^2\ \text{g}^{-1}$) and the Angstrom exponent (α_P) as per the equations provided by Hecobian *et al* (2010). Briefly, for each sample, the mass absorption coefficient and efficiency ($b_{\text{abs-365}}$ and σ_{abs}) of BrC are estimated using equations (1) and (2):

$$b_{\text{abs-365}}\ (\text{M m}^{-1}) = \left[\frac{(A_{365} - A_{700}) f_{\text{dil}} V_{\text{ext}} \times 4 \ln(10)}{V_{\text{air}} \times 2}\right]. \quad (1)$$

Here, A_{365} and A_{700} correspond to the measured absorbance at 365 and 700 nm; f_{dil} is the dilution factor. V_{ext} is the volume of Mill-Q water used for the extraction of the one quarter aerosol filter whereas V_{air} refers to the volume of air filtered through the aerosol filter.

$$\sigma_{\text{abs-BrC}}\ (\text{m}^2\ \text{g}^{-1}) = \frac{b_{\text{abs-365}}}{\text{WSOC}}. \quad (2)$$

It has been suggested that the mass absorption coefficient, assessed based on the measured absorbance of aqueous extracts, varies as a function of wavelength in accordance with the following equation (3):

$$b_{\text{abs-}\lambda}\ (\text{M m}^{-1}) \approx \lambda^{-\alpha_P}. \quad (3)$$

In the above equation, α_P refers to the Angstrom exponent of BrC. Likewise; we have estimated the mass absorption of efficiency of EC as described in Ram and Sarin (2009).

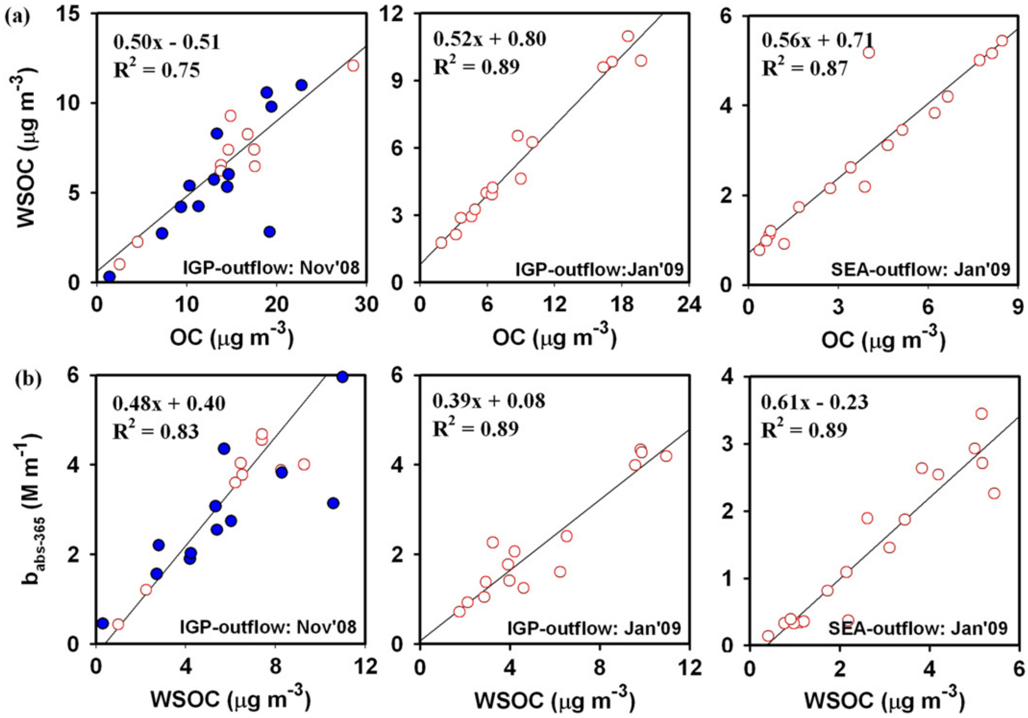

Figure 2. Scatter plot between (a) WSOC and OC, (b) mass absorption coefficient of brown carbon ($b_{\text{abs-365}}$) and WSOC for aerosols collected during November 2008 and January 2009 from the marine atmospheric boundary layer of the Bay of Bengal (open circles) and the Arabian Sea (filled circles).

During the cruise undertaken in the BoB (SK-254 cruise), we had sampled two wind regimes (Kumar *et al* 2010, Srinivas and Sarin 2013a, 2013b); air masses originating from (a) the Indo-Gangetic Plain (referred to hereafter as IGP-outflow: during 27 December 2008–10 January 2009) and those from (b) south-east Asia (referred to as SEA-outflow: from 11 to 28 January 2009). Likewise, during the SS-259 cruise in November 2008, air masses originated from the Indian subcontinent influenced by the source regions in the IGP and are also referred as IGP-outflow. Typical air mass back trajectories (AMBTs) for the sampling days along the cruise tracks are presented in supporting information (available at stacks.iop.org/ERL/8/044042/mmedia) (figure S1 available at stacks.iop.org/ERL/8/044042/mmedia). Back trajectories were computed using the hybrid single particle Lagrangian integrated trajectory model (HYSPLIT, version-4; Draxler 1999, Draxler and Rolph 2013, Rolph 2013) with a GDAS reanalysis data set from NOAA Air Resources Laboratory. A detailed description of the meteorological parameters is given in Kumar *et al* (2010). In this study, we have used PM_{10} samples (collected during December 2008–January 2009) to compare with bulk (TSP) aerosols sampled during November 2008.

3. Results and discussions

3.1. Brown carbon over the Indian Ocean and temporal variability

A significant linear relationship between mass concentrations of WSOC and OC is conspicuous during the two cruises (figure 2(a)). On average, WSOC contributes more than 50% of OC over the ARS and the BoB. The statistical description of the concentrations and absorption properties of carbonaceous species (namely, WSOC, OC, EC, $b_{\text{abs-EC}}$, $\sigma_{\text{abs-EC}}$, $b_{\text{abs-BrC}}$, $\sigma_{\text{abs-BrC}}$, and α_P) is presented in table 1. During both cruises (SS-259: November 2008 and SK-254: December 2008–January 2009), a prominent absorption at ~365 nm is noteworthy in the aqueous extracts of the aerosol samples. The mass absorption coefficient ($b_{\text{abs-365}}$), based on the measured absorbance at 365 nm relative to 700 nm, varied from 0.4 to 8.1 M m^{-1} (Av: 3.6 ± 2.0 M m^{-1}) for TSP and 0.2 to 3.9 M m^{-1} (Av: 1.5 ± 1.1 M m^{-1}) for PM_{10}.

A striking feature of the data pertains to the significant ($P < 0.05$) linear relationship between $b_{\text{abs-365}}$ and WSOC in aerosol samples collected from the MABL (figure 2(b)). The slope of the regression line provides a robust estimate of the mass absorption efficiency of BrC ($\sigma_{\text{abs-BrC}}$). The slope of the regression line for TSP and PM_{10} samples corresponds to 0.48 and 0.36, respectively, in the IGP-outflow, whereas it corresponds to 0.61 in the SEA-outflow (see section 2.2 for classification of the air masses). A significant increase in $\sigma_{\text{abs-BrC}}$ in the SEA-outflow relative to that for the IGP-outflow is attributed to the contribution of air masses influenced by forest fires (supporting figure S2 available at stacks.iop.org/ERL/8/044042/mmedia). A close match between the slope of the regression lines and the mean mass absorption efficiency of brown carbon is noteworthy for both IGP- and SEA-outflows sampled over the Northern Indian Ocean (table 1). Likewise, a close similarity in the temporal variability of nss-K^+ (a proxy for biomass burning emissions, BBE), WSOC and $b_{\text{abs-365}}$ in all samples collected

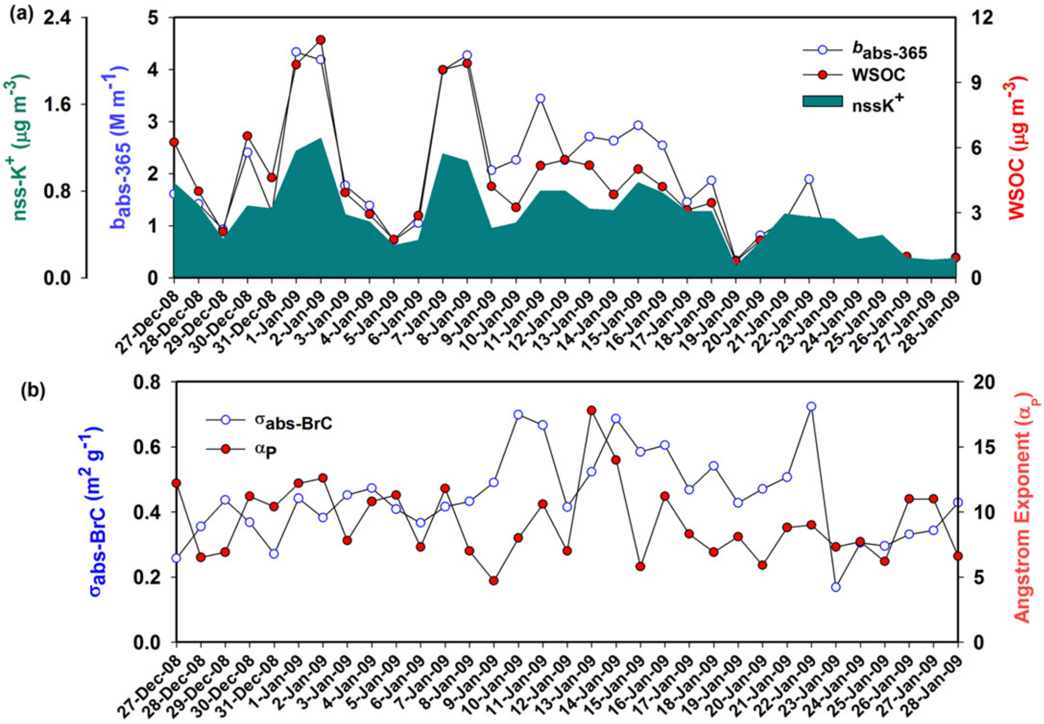

Figure 3. Temporal variability of (a) the mass absorption coefficient at 365 nm ($b_{\text{abs-365}}$), WSOC and non-sea-salt potassium (nss-K^+); (b) the mass absorption efficiency ($\sigma_{\text{abs-BrC}}$) and Angstrom exponent (α_P) of brown carbon (BrC) in PM_{10} sampled from the Bay of Bengal during January 2009.

Table 1. Statistical description of concentrations of carbonaceous species (WSOC, OC and EC) and their absorption properties ($\sigma_{\text{abs-EC}}$, $b_{\text{abs-365}}$, $\sigma_{\text{abs-BrC}}$ and α_P) over the Northern Indian Ocean.

	SS-259: November 2008	SK-254: December 2008–January 2009	
Parameter	IGP-outflow	IGP-outflow	SEA-outflow
WSOC (μg m^{-3})	0.3–17.4 (6.7 ± 3.9)	1.8–11.0 (5.5 ± 3.1)	0.4–5.4 (2.7 ± 1.7)
OC (μg m^{-3})	1.4–28.5 (14.9 ± 7.0)	1.9–19.7 (9.1 ± 6.0)	0.4–8.5 (3.9 ± 2.8)
EC (μg m^{-3})	0.3–5.6 (3.2 ± 1.4)	1.0–6.7 (2.7 ± 1.7)	0.2–2.3 (1.1 ± 0.5)
OC/EC	2.8–9.0 (4.7 ± 1.7)	1.5–5.1 (3.4 ± 1.1)	0.7–6.5 (3.2 ± 2.0)
WSOC/OC	0.15–0.65 (0.45 ± 0.12)	0.50–0.92 (0.65 ± 0.11)	0.56–1.02 (0.79 ± 0.17)
$\sigma_{\text{abs-EC}}$ (m^2 g^{-1})	1.0–5.6 (2.5 ± 1.1)	3.0–9.0 (6.1 ± 1.7)	5.2–10.7 (7.7 ± 1.6)
$b_{\text{abs-365}}$ (M m^{-1})	0.4–8.1 (3.6 ± 2.0)	0.7–4.3 (2.2 ± 1.3)	0.1–3.4 (1.4 ± 1.1)
$\sigma_{\text{abs-BrC}}$ (m^2 g^{-1})	0.3–1.5 (0.6 ± 0.2)	0.3–0.7 (0.4 ± 0.1)	0.2–0.7 (0.5 ± 0.2)
α_P	1.1–8.4 (5.8 ± 1.5)	5.9–14.0 (9.1 ± 2.5)	3.5–11.6 (6.9 ± 1.9)

from the MABL suggests a contribution of BrC from BBEs. Figure 3 depicts temporal variability of nss-K^+, WSOC, $b_{\text{abs}-365}$, $\sigma_{\text{abs-BrC}}$ and α_P for PM_{10} samples collected from the MABL.

3.2. Mass absorption efficiency (MAE or σ_{abs}) of brown carbon over the Bay of Bengal

A comparison of MAE of BrC from a sampling site (Kharagpur: 22.3°N, 87.3°E) within the Indo-Gangetic Plain (Srinivas and Sarin 2013c) and in the IGP-outflow sampled over the Bay of Bengal (during January 2009) is presented in table 2. The MAE of BrC over the BoB is relatively lower (0.45 ± 0.14 m^2 g^{-1}) compared to that from the source region in the IGP (0.78 ± 0.24 m^2 g^{-1}). This can be explained by the relative decrease in the source strength together with the contribution of WSOC from sources other than BBEs. This argument is further corroborated by the relative increase in the WSOC/OC ratio for the IGP (Av: 0.52 ± 0.16) compared to that over the BoB (0.77 ± 0.22; table 2). The relative increase in the fractional contribution of WSOC to OC could be attributed to a shift in the source regions and source strength (BBEs and fossil-fuel combustion sources).

Although the MAE of BrC from the BoB (Av: 0.45 ± 0.14 m^2 g^{-1}) is somewhat lower than that observed for the continental site in the IGP (0.78 ± 0.24 m^2 g^{-1}), a remarkable similarity in the Angstrom exponent among the two data sets (BoB: 9 ± 2; IGP: 8 ± 3; table 2) suggests the contribution of BrC from biomass burning emission sources. It is relevant to state that α_P values for fossil-fuel combustion

Table 2. Spectral absorption properties of EC and BrC over the Indo-Gangetic Plain and IGP-outflow sampled from the Bay of Bengal (BoB).

Parameter	Indo-Gangetic Plain[a]	IGP-outflow: BoB
WSOC/OC	0.26–1.06 (0.52 ± 0.16)	0.60–0.96 (0.77 ± 0.10)
$\sigma_{abs\text{-}EC}$ ($m^2\ g^{-1}$)	0.7–6.6 (2.2 ± 1.2)	3.3–8.3 (6.6 ± 1.6)
$b_{abs\text{-}365}$ ($M\ m^{-1}$)	1.8–21.4 (11.4 ± 4.8)	0.4–3.9 (2.0 ± 1.1)
$\sigma_{abs\text{-}BrC}$ ($m^2\ g^{-1}$)	0.2–1.5 (0.8 ± 0.2)	0.2–0.8 (0.5 ± 0.1)
α_P	2.5–15.5 (8.4 ± 2.6)	4.7–17.8 (9.5 ± 2.9)

[a] From (Srinivas and Sarin 2013c).

sources are significantly lower (∼1–2; Cheng *et al* 2011; Kirchstetter *et al* 2004 and references therein) compared to that of biomass burning emissions (∼6–9; Cheng *et al* 2011; Hecobian *et al* 2010; Hoffer *et al* 2006). The α_P of light absorbing organic aerosols over the BoB (figure 4) together with the linear relationship between WSOC and $b_{abs\text{-}365}$ suggest their contribution from BBE. However, the lower MAE over the BoB could arise due to a decrease in the contribution of BrC to WSOC in comparison to that over the IGP. It is likely that a relative increase in the contribution of WSOC from sources other than BBE (for example, from fossil-fuel combustion) could also explain the lower MAE of BrC over the BoB.

3.3. Mass absorption efficiency (MAE) of EC over the Bay of Bengal

The MAE of EC over the BoB during January 2009 centered on ∼6.1 ± 1.7 $m^2\ g^{-1}$ and 7.4 ± 2.0 $m^2\ g^{-1}$ for the IGP- and SEA-outflow, respectively (table 1). In contrast, the mean MAE of EC during the cruise undertaken in November 2008 corresponds to 2.5 ± 1.1 $m^2\ g^{-1}$. The mass absorption efficiency of ∼7 $m^2\ g^{-1}$ has been reported for the EC contribution from fossil-fuel combustion sources; whereas BBE is characterized by lower the MAE of EC (∼2–3 $m^2\ g^{-1}$; Cheng *et al* 2011). A significant contribution of carbonaceous aerosols from paddy-residue burning emissions in the IGP, during October–November, has been widely reported. Therefore, a lower MAE over the Northern Indian Ocean (during November 2008) is consistent with the dominance of emissions from biomass burning.

The MAE of EC in the MABL during January 2009 is relatively high for the IGP-outflow: 6.6±1.6 $m^2\ g^{-1}$ for $PM_{2.5}$ and 6.1±1.7 $m^2\ g^{-1}$ for PM_{10}. It is relevant to state that under favorable meteorological conditions, continental outflow from the Indo-Gangetic Plain to the Bay of Bengal persists only during the late NE monsoon (January–April). Therefore, the contribution of BBEs from the upwind source regions (in the IGP) to the Bay of Bengal (during the cruise undertaken in January 2009) could be lower than that from fossil-fuel (FF) combustion sources. This argument is further corroborated by the relative increase in the WSOC/OC fraction in the IGP source region (Av: 0.52 ± 0.16) than over the BoB (0.77 ± 0.22; see table 2). A relative increase in the fractional contribution of WSOC to OC over the BoB suggests a change in the type of source and source strength (BBE vis-à-vis FF combustion). The high MAE of EC, therefore, supports the dominant contribution of EC from FF combustion sources.

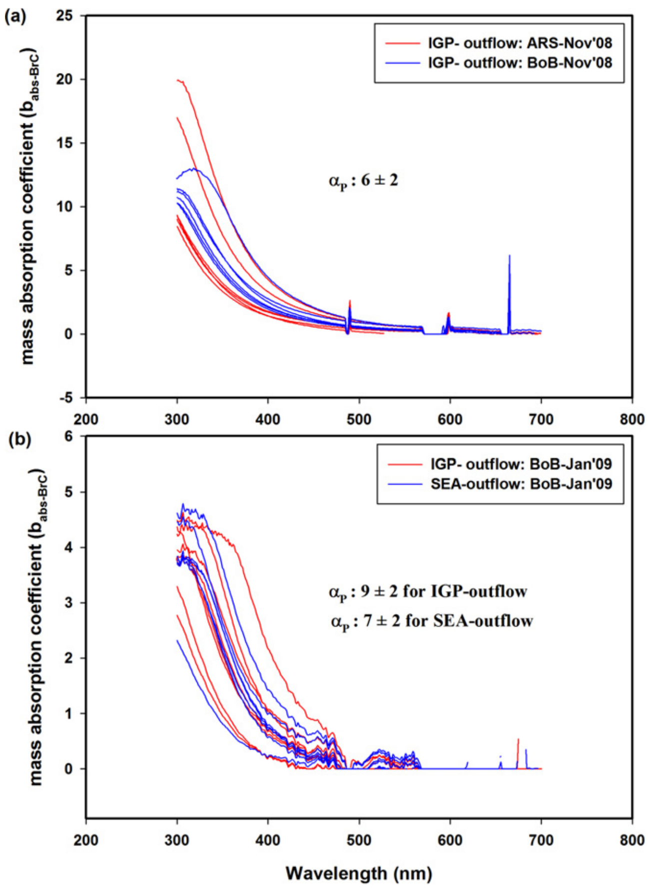

Figure 4. Typical wavelength dependent mass absorption coefficient ($b_{abs\text{-}Brc}$) of brown carbon in aerosol samples collected from the Northern Indian Ocean during (a) November 2008 and (b) December 2008–January 2009.

3.4. Comparison of MAE of BrC and EC over Bay of Bengal

In order to compare the MAE of BrC and EC, we have defined a parameter referred to as the 'absorbing potential (AP)', which accounts for the fractional contribution of species along with MAE, and is defined by equation (4):

$$AP_X = (X/PM_{2.5}\%)(\sigma_{abs\text{-}X}). \quad (4)$$

Here X is EC or BrC; X% is the fractional contribution of that species to $PM_{2.5}$ mass. Since we have not quantified the contribution of BrC, as a first-order approximation we have used the fractional contribution of WSOC to estimate the AP of BrC due to its significant linear relation with

$b_{abs\text{-}365}$. This approach has an inherent uncertainty (as all of the WSOC is not BrC); therefore, it yields a higher estimate for a scenario with a maximum contribution of WSOC from BEE. During the January 2009 cruise, the AP of EC and BrC averages around 27.5 ± 8.5 and 4.0 ± 0.8 m^2 g^{-1}, respectively. A comparison of AP of BrC with that of EC suggests that their relative proportion (i.e., $AP_{BrC}/AP_{EC} \times 100$) varies from 8 to 23% (Av: 15 ± 5%) in the IGP-outflow and 3–31% (Av: 16 ± 9%) for the SEA-outflow. Likewise, the mean AP corresponds to 9 ± 4 m^2 g^{-1} for EC and 4 ± 3 m^2 g^{-1} for BrC during November 2008. In these aerosol samples, the relative proportion of BrC varies from 23 to 100% (Av: 50 ± 23%). These observations highlight the significance of light absorbing organic aerosols (brown carbon) over the tropical Indian Ocean.

3.5. Implications to radiative forcing estimates

The results obtained in this study suggest reassessment of the radiative forcing estimates over the Northern Indian Ocean. Based on the chemical composition of ambient particulate matter, earlier studies have shown the dominant influence of anthropogenic sources on the MABL of the BoB compared to the ARS (Kumar *et al* 2008, 2010, Sarin *et al* 2010, Srinivas *et al* 2011, Srinivas and Sarin 2012, 2013a, 2013b). This is further supported by the radiative forcing estimates, suggesting a relative decrease at surface of the BoB compared to the ARS. Furthermore, it had been suggested that aerosols over the BoB are of a '*more absorbing*' type compared to that over the ARS (Nair *et al* 2008 and references therein). An earlier study by Babu *et al* (2004) had shown that aerosol direct radiative forcing increases with a rise in the atmospheric BC concentration. In this study, we demonstrate the ubiquitous presence of BrC in the MABL of the Northern Indian Ocean during the continental outflow. Thus, the presence of these light absorbing organics in the MABL would lead to a further decrease in incoming short-wave (solar) radiation and, therefore, would further downscale surface radiative forcing estimates. In this context, we suggest that the combined effect of BrC and BC needs reassessment for the projected estimates of aerosol radiative forcing over the Northern Indian Ocean.

4. Conclusions

Aerosol samples collected from the Bay of Bengal and the Arabian Sea, during the continental outflow, show unequivocal evidence for light absorbing brown carbon (BrC). The mass absorption coefficient (b_{abs}), measured at 365 nm, shows a linear relationship with WSOC and the Angstrom exponent is typical of that reported for emissions from biomass burning sources. The mass absorption efficiency and Angstrom exponent of BrC in the marine atmospheric boundary layer, during the continental outflow, are consistent with the downwind source regions in the Indo-Gangetic Plain. The absorption due to brown carbon over the Indian Ocean could account for ∼4–45% of that from EC. These results have implications to aerosol radiative forcing over oceanic regions influenced by continental pollution sources.

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