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Light absorption properties of brown carbon over the southeastern Tibetan Plateau

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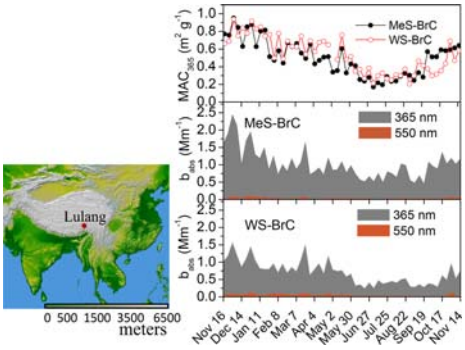
HIGHLIGHTS

- The light-absorbing properties of WS-BrC and MeS-BrC were investigated in the southeastern Tibetan Plateau.
- Mass absorption cross section of BrC in the southeastern Tibetan Plateau has a strong seasonal variability.
- BrC was the dominant absorption material in extracts.

GRAPHICAL ABSTRACT



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ABSTRACT

We present a study of the light-absorbing properties of water-soluble brown carbon (WS-BrC) and methanol-soluble brown carbon (MeS-BrC) at a remote site (Lulang, 3326 m above sea level) in the southeastern Tibetan Plateau during the period 2015–2016. The light absorption coefficients at 365 nm (b_{abs365}) of WS-BrC and MeS-BrC were the highest during winter and the lowest during monsoon season. MeS-BrC absorbs about 1.5 times higher at 365 nm compared to WS-BrC. The absorption at 550 nm appears lower compared to that of 365 nm for WS-BrC and MeS-BrC, respectively. Higher average value of the absorption Ångström exponent (AAE, 365–550 nm) was obtained for MeS-BrC (8.2) than that for WS-BrC (6.9). The values of the mass absorption cross section at 365 nm (MAC_{365}) indicated that BrC in winter absorbs UV–visible light more efficiently than in monsoon. The results confirm the importance of BrC in contributing to light-absorbing aerosols in this region. The understanding of the light absorption properties of BrC is of great importance, especially in modeling studies for the climate effects and transport of BrC in the Tibetan Plateau.

1. Introduction

Brown carbon (BrC) is a certain type of organic carbon (OC), which absorbs radiation efficiently at the blue and ultraviolet (UV) wavelength of the solar spectrum (Pöschl, 2003; Andreae and Gelencsér, 2006; Moosmüller et al., 2011). BrC may contribute substantially the total aerosol absorption at specific wavelengths, thereby affecting the accuracy of climate model results and satellite data retrieval. The light absorption of BrC produced from different types of biofuels burning under varied combustion conditions have been investigated (Ramanathan et al., 2007a, 2007b; Alexander et al., 2008; Andreae and Gelencsér, 2006). The previous studies showed that BrC (including primary and

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secondary BrC) is typically emitted during biomass burning, coal combustion, and the formation of secondary organic aerosol (SOA) (e.g., Hecobian et al., 2010; Lack and Langridge, 2013; Zhang et al., 2013; Laskin et al., 2015).

The light absorption of BrC is estimated by either measuring the absorption of organics extracted in water, acetone, methanol, or by calculating the difference between total absorption and that of black carbon (BC) (Chen and Bond, 2010; Laskin et al., 2015). At present, the transformations mechanisms and rates of BrC are not well characterized, and little is known about the relationship among the chemical components and its light absorption properties.

The Tibetan Plateau (TP), called as "the third pole" with the extremely high altitude, is an important and vulnerable region for the changes of global climate and regional environment. The TP glaciers determine the albedo of the area with snow cover, and provide the water of some Asian rivers. These rivers are important water resources for billions of local habitants in surrounding areas (Yao et al., 2012). The series effects of carbonaceous aerosol (e.g., BC and BrC) are attracting attention recently, because of their considerable role in the melting of glaciers and the albedo effects in the TP (e.g., Cao et al., 2009; Xu et al., 2009, 2012; Wang et al., 2012; Cong et al., 2013; Zhu et al., 2017). Long-range transport from upwind regions to the plateau is the major source of pollutants (Cao et al., 2010; Zhu et al., 2017). The abundant BrC in Indo-Gangetic Plain could enter the TP from the south side of the Himalayas, and may exert profound impacts on the climate and regional environment (Srinivas and Sarin, 2013, 2014; Zhao et al., 2013; Srinivas et al., 2016). The investigations for the chemical composition and absorption properties of BrC in the TP are needed. Until now, very few studies focused on the light-absorbing properties of water-soluble brown carbon (WS-BrC) and methanol-soluble brown carbon (MeS-BrC) were conducted in the TP.

In this work, an attempt has been made to investigate the light properties of WS-BrC and MeS-BrC in the southeastern TP. The relationships between BrC light absorption and carbonaceous fractions are also investigated during the sampling period.

2. Materials and methods

2.1. Sampling

The field aerosol sampling campaign was conducted at Lulang (94.73°E, 29.76°N, 3326 m a.s.l.) in southeastern region of the TP (Fig. 1). The map presents the seasonal aerosol optical depth, retrieved from satellite (Terra/Modis) observations during the sampling period (http://www.nasa.gov). The detailed description about the sampling site has been reported elsewhere (Cao et al., 2010; Zhao et al., 2013).

Total suspended particulate (TSP) samples (47 mm Whatman quartz–fiber filters, with filter changing at 1000 local standard time) were collected on a weekly basis at 17 L/min with a custom-built sampler from November 2015 to November 2016. The field blank samples in southeastern TP were also collected during the sampling period. These quartz–fiber filters were pre-heated at 900 °C for 3 h to remove the residual carbon. The samples were stored in a refrigerator at about −20 °C to prevent the volatile components evaporation after sampling.

2.2. Measurement of carbonaceous fractions

All the filters (including field blank filters) were analyzed for carbon fractions by using a DRI Model 2001 Thermal/Optical Carbon Analyzer (Atmoslytic Inc., Calabasas, CA, USA). Carbon fractions were analyzed following the IMPROVE-A (Interagency Monitoring of Protected Visual Environments) thermal/optical reflectance (TOR) protocol (Chow et al., 2007). The concentrations of organic carbon (OC) and elemental carbon (EC) were obtained. The detailed procedures for quality assurance and quality control (QA/QC) have been reported elsewhere (Cao et al., 2003; Chow et al., 2011).

A part of the TSP sample was cut into pieces, and then extracted with Milli–Q water (30 mL) three times (60 min, 20 min for each cycle) under sonication, and filtered by using the Polytetrafluoroethylene (PTFE) filters. The combined water extracts were analyzed for water-soluble organic carbon (WSOC), water–soluble inorganic carbon (WSIC) and water-soluble total nitrogen (WSTN) by using the Shimadzu TOC–L CPH Total Carbon/Nitrogen Analyzer (Wang et al., 2010). Major water-soluble inorganic nitrogen (WSIN) species in atmospheric aerosols are NO_3^- and NH_4^+, thus the difference between WSTN and WSIN is defined as water-soluble organic nitrogen (WSON) (Wang et al., 2013). Temporal variability of WSOC and water-soluble organic nitrogen (WSON) concentrations for TSP during the sampling period was obtained.

2.3. Measurements of the light-absorbing properties of BrC

BrC was extracted from the filter subsamples by 1 h sonication in ultrapure Milli–Q water (>18.2 MΩ) or from the filter subsamples by 1 h sonication in methanol (J. T. Baker, HPLC Grade). The water and methanol extracts were then filtered to remove these insoluble material using 0.22 μm PES (polyether sulphone) and 0.45 μm PTFE pore syringe filter, respectively. Absorption spectra of the extracts (water or methanol) were measured using a Liquid Waveguide Capillary Cell (LWCC–3100, World Precision Instrument, Sarasota, FL, USA, 0.94 m in length) equipped with a UV–Vis spectrophotometer (300–700 nm) (Hecobian et al., 2010; Laskin et al., 2015; Kirillova et al., 2016). The light

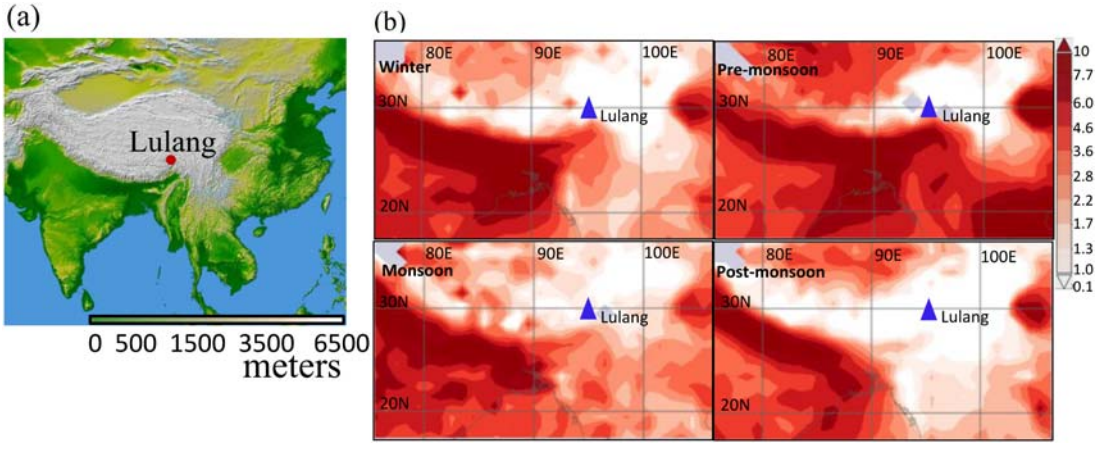

Fig. 1. Geographic location of the sampling site, Lulang (a). Time averaged maps of the seasonal AOD were produced with the Giovanni online data system, developed and maintained by the NASA GES DISC (b).