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Spectral dependence of aerosol light absorption at an urban and a remote site over the Tibetan Plateau

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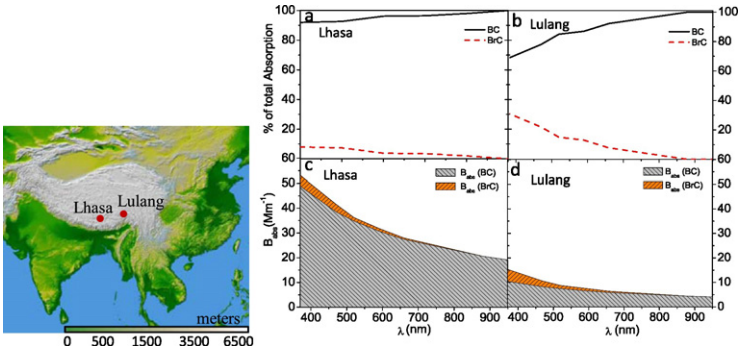
HIGHLIGHTS

- Aerosol light absorption is measured over the Tibetan Plateau.
- Spectral dependence of aerosol light absorption due to black and brown carbon is studied.
- The absorption Ångström exponent of brown carbon is directly extracted.

GRAPHICAL ABSTRACT



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ABSTRACT

We present a study of aerosol light absorption by using a 7-wavelength Aethalometer model AE33 at an urban site (Lhasa) and a remote site (Lulang) in the Tibetan Plateau. Approximately 5 times greater aerosol absorption values were observed at Lhasa (53 ± 46 Mm^{-1} at 370 nm and 20 ± 18 Mm^{-1} at 950 nm, respectively) in comparison to Lulang (15 ± 19 Mm^{-1} at 370 nm and 4 ± 5 Mm^{-1} at 950 nm, respectively). Black carbon (BC) was the dominant light absorbing aerosol component at all wavelengths. The brown carbon (BrC) absorption at 370 nm is $32 \pm 15\%$ of the total aerosol absorption at Lulang, whereas it is $8 \pm 6\%$ at Lhasa. Higher value of absorption Ångström exponent (AAE, 370–950 nm) was obtained for Lulang (1.18) than that for Lhasa (1.04) due to the presence of BrC. The AAEs (370–950 nm) of BrC were directly extracted at Lulang (3.8) and Lhasa (3.3). The loading compensation parameters (k) increased with wavelengths for both sites, and lower values were obtained at Lulang than those observed at Lhasa for all wavelengths. This study underlines the relatively high percentage of BrC absorption contribution in remote area compared to urban site over the Tibetan Plateau.

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1. Introduction

The dependence of light absorption on the wavelength can be of importance for research on the effects and source apportionment of carbonaceous aerosol (Sandradewi et al., 2008; Bond et al., 2013; IPCC, 2013). The major light absorbing components of aerosol are black carbon (BC), brown carbon (BrC) and mineral dust (Andreae and Gelencsér, 2006). BC is considered the most efficient light absorber in the visible spectrum with a weak dependence on wavelength (λ) (Moosmüller et al., 2009; Bond et al., 2013; Zhu et al., 2015). BrC is another light absorbing component of carbonaceous aerosol, with increased absorbance at the blue and ultraviolet (UV) wavelength of the solar spectrum (Pöschl, 2003; Andreae and Gelencsér, 2006; Moosmüller et al., 2011). BC and BrC are typically emitted by incomplete combustion of fossil fuels and biomass burning. Their optical properties are different and are distinctive functions of the wavelength of light. Typically, the 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 BC (Chen and Bond, 2010; Laskin et al., 2015). It is worthy to note that the uncertainty is hard to be quantified for these approaches.

The previous studies showed most absorption by BrC takes place at the UV wavelength and with variable contributions to total carbonaceous aerosol absorption (Bond et al., 2013; Wang et al., 2014; Saleh et al., 2015; Jo et al., 2016). The absorption Ångström exponent (AAE) describes the spectral dependence of absorption. AAE has been shown to be sensitive not only to aerosol chemical composition but also to particle size and morphology. The value of AAE differed significantly around 1 for BC and up to 9.5 for BrC (Kirchstetter et al., 2004; Lewis et al., 2008; Lack and Langridge, 2013; Wang et al., 2014; Laskin et al., 2015; Utry et al., 2015). In some studies, it has been used as a parameter to identify the origin of carbonaceous aerosols (Sandradewi et al., 2008; Flowers et al., 2010; Filep et al., 2013), with the validation performed recently using ^{14}C analysis (Zotter et al., 2016). The published absorption efficiencies and AAE for BrC are inconsistent, which can be attributed to the aerosol properties.

The effects of carbonaceous aerosol are pronounced in the Tibetan Plateau. These components accumulate on snow and ice, which reduce the surface albedo and consequently accelerate the melting of glaciers (Flanner et al., 2009; Menon et al., 2010; Xu et al., 2009, 2012). The studies of carbonaceous aerosols over the Tibetan Plateau have thus been the subject of increasing interest (Cao et al., 2009; Wang et al., 2012; Cong et al., 2013; Ming et al., 2013; Zhao et al., 2013; Zhu et al., 2016). However, the spectral dependence of light absorption of atmospheric BC and BrC are poorly understood in the Tibetan Plateau.

In this work, an attempt has been made to investigate the aerosol light absorption for several wavelengths at Lhasa and Lulang. The contributions of BC and BrC to the light absorption can be distinguished based on their spectral absorption. The AAE of brown carbon is directly extracted for Tibetan urban and remote atmosphere.

2. Methodology

2.1. Measurement site

The measurements of light absorption at seven wavelengths ranging from UV to infrared part of the spectrum were conducted at an urban site (Lhasa) and a remote site (Lulang), respectively (Fig. 1). The urban site is located at the Lhasa branch of the Institute of Tibetan Plateau Research (91.1°E, 29.6°N, 3650 m a.s.l.) in the western part of Lhasa. The site is mainly influenced by emissions from anthropogenic sources including power plants, domestic heating, cement production facilities, vehicular traffic, and religious activities of local residents (Gong et al., 2011). The remote site is situated at Lulang (94.73°E, 29.76°N, 3326 m a.s.l.) in the southeastern margin of the Tibetan Plateau. There are extensive forests in Lulang without anthropogenic sources nearby. However, around the site, there are several small villages. Hence, local biomass burning for cooking and heating and long-range transport from upwind regions to the plateau are the major sources of pollutants (Cao et al., 2010; Zhao et al., 2013).

2.2. Instrumentation

The aerosol light absorption was measured continuously at 1-min averages by quartz-fiber filter tape transmission with a seven wavelengths Aethalometer model AE33 (Magee Scientific, λ = 370, 470, 520, 590, 660, 880 and 950 nm) (Drinovec et al., 2015). The Aethalometer was connected to an inlet and total suspended particles were measured at a flow rate of 4 L min^{-1}. Filter photometers are affected by non-linearities, which need to be compensated for (Weingartner et al., 2003; Arnott et al., 2005; Virkkula et al., 2007; Coen et al., 2010; Hyvärinen et al., 2013). The Aethalometer Model AE33 analyzes the sample on two parallel spots drawn from the same input stream, but collected at different rates of accumulation, i.e. at different values of attenuation (ATN) (Drinovec et al., 2015). It is therefore possible to eliminate the "loading effect" with the loading compensation parameter *k*, by making two simultaneous measurements of BC_1 and BC_2 at ATN_1 and ATN_2. This allows extrapolation to zero loading, and the accurate ambient BC concentration is obtained.

$$BC_i(\text{reported}) = BC(\text{ambient})^{*}\{1-k\cdot ATN\} \quad (1)$$

which holds for both spots $i = 1.2$:

$$BC_1 = BC^{*}\{1-k\cdot ATN_1\} \quad (2)$$

$$BC_2 = BC^{*}\{1-k\cdot ATN_2\} \quad (3)$$

From these two linear equations we may calculate the loading parameter k, and the ambient BC concentration. The mass absorption cross-section (MAC) values used in the Aethalometer model AE33 are 18.47, 14.54, 13.14, 11.58, 10.35, 7.77, and 7.19 $m^2\ g^{-1}$ for the wavelengths of 370, 470, 520, 590, 660, 880, and 950 nm, respectively. We have determined the absorption coefficient (B_{abs}) from the wavelength dependent BC concentrations: $B_{abs}(\lambda) = BC\ (\lambda)\cdot MAC\ (\lambda)$. The AAE values were calculated from the fit of B_{abs} over the measured spectral ranges. Consequently, real-time source apportionment has been implemented in the Aethalometer model AE33. A complete description of the method and the operating principles of the instrument can be found in Drinovec et al. (2015).

The diurnal variations of aerosol size distribution were obtained by using an optical particle sizer (OPS, Model 3330, TSI Inc., USA, 1.0 L/min aerosol flow rate, size range 0.3 μm–10.0 μm). The measurements were carried out in Autumn 2015: from 10 to 19 September 2015 at the urban site (Lhasa), and from 20 September to 21 November 2015 at the remote site (Lulang).

3. Results and discussion

3.1. Time series of light absorption and AAE

The variations of hourly B_{abs} (370 nm), B_{abs} (880 nm) and the AAE (calculated over all seven wavelengths) were shown for Lhasa and Lulang during the study period (Fig. 2). The hourly B_{abs} (370 nm) showed large variability, from 6 to 390 Mm^{-1} at Lhasa and from 0.7 to 160 Mm^{-1} at Lulang, respectively. B_{abs} (880 nm) ranged from 2 to 170 Mm^{-1} at Lhasa and from 0.3 to 46 Mm^{-1} at Lulang, respectively. The variations of B_{abs} (370 nm, 880 nm) showed events with large values of the absorption coefficient in Lhasa, which may be attributed to the local sources.

As expected, greater absorption values were observed at the urban site (Lhasa), with the average values approximately 53 ± 46 Mm^{-1} at