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Light absorption characteristics of carbonaceous aerosols in two remote stations of the southern fringe of the Tibetan Plateau, China

Chaoliu Li [a, c, *], Fangping Yan [b, e], Shichang Kang [b, c], Pengfei Chen [b], Zhaofu Hu [b, d], Shaopeng Gao [a], Bin Qu [e], Mika Sillanpää [e, f]

[a] Key Laboratory of Tibetan Environment Changes and Land Surface Processes, Institute of Tibetan Plateau Research, Chinese Academy of Sciences, Beijing 100101, China
[b] State Key Laboratory of Cryospheric Sciences, Northwest Institute of Eco-Environment and Resources, Chinese Academy of Sciences, Lanzhou 730000, China
[c] CAS Center for Excellence in Tibetan Plateau Earth Sciences, Chinese Academy of Sciences, Beijing 100101, China
[d] University of Chinese Academy of Sciences, Beijing 100049, China
[e] Laboratory of Green Chemistry, Lappeenranta University of Technology, Sammonkatu 12, FI-50130 Mikkeli, Finland
[f] Department of Civil and Environmental Engineering, Florida International University, Miami, FL 33174, USA

HIGHLIGHTS

- MAC of BC and WSOC at two remote areas on the Tibetan Plateau were studied.
- Remarkable seasonal fluctuation of MAC_{WSOC} related to photobleaching was found.
- Seasonal fluctuation of MAC_{WSOC} should be a general phenomenon at other remote areas.
- Contribution of direct radiative forcing (RDF) of WSOC to that of BC were calculated.
- RDF caused by WSOC was around 6% and 12% to that of BC at two areas.

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ABSTRACT

Light absorption characteristics of carbonaceous aerosols are key considerations in climate forcing research. However, in situ measurement data are limited, especially on the Tibetan Plateau (TP) – the Third Pole of the world. In this study, the mass absorption cross section (MAC) of elemental carbon (EC) and water soluble organic carbon (WSOC) of total suspended particles at two high-altitude stations (Lulang station and Everest station) in the Tibetan Plateau (TP) were investigated. The mean MAC_{EC} values at 632 nm were 6.85 ± 1.39 $m^2\ g^{-1}$ and 6.49 ± 2.81 $m^2\ g^{-1}$ at these two stations, both of which showed little seasonal variations and were slightly higher than those of EC of uncoated particles, indicating that the enhancement of MAC_{EC} by factors such as coating with organic aerosols was not significant. The mean MAC_{WSOC} values at 365 nm were 0.84 ± 0.40 $m^2\ g^{-1}$ and 1.18 ± 0.64 $m^2\ g^{-1}$ at the two stations. Obvious seasonal variations of high and low MAC_{WSOC} values appeared in winter and summer, respectively, mainly reflecting photobleaching of light absorption components of WSOC caused by fluctuations in sunlight intensity. Therefore, this phenomenon might also exists in other remote areas of the world. The relative contributions of radiative forcing of WSOC to EC were 6.03 ± 3.62% and 11.41 ± 7.08% at these two stations, with a higher ratio in winter. As a result, both the contribution of WSOC to radiative forcing of carbonaceous aerosols and its seasonal variation need to be considered in radiative forcing related study.

1. Introduction

Carbonaceous aerosols include elemental carbon (EC, a term used synonymously with black carbon (BC)) (Petzold et al., 2013) and organic carbon (OC). EC is emitted into the atmosphere solely

* Corresponding author. Key Laboratory of Tibetan Environment Changes and Land Surface Processes, Institute of Tibetan Plateau Research, Chinese Academy of Sciences, Beijing 100101, China.
E-mail address: lichaoliu@itpcas.ac.cn (C. Li).

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as primary particles from either fossil fuel combustion or biomass burning. OC is emitted either directly or is formed from gaseous precursors as a secondary organic carbon (SOC) (Jenk et al., 2006). The radiative forcing of carbonaceous aerosols influences the radiation budget of the atmosphere and climate forcing, which is one of the largest uncertainties in the climate system (Bond et al., 2013; Gustafsson and Ramanathan, 2016; IPCC, 2013; McComiskey et al., 2008). It is estimated that EC is the second atmospheric agent after carbon dioxide in terms of its climate warming effect in the present-day atmosphere (Bond et al., 2013; Jacobson, 2000). The mass absorption cross section of EC (MAC_{EC}) is a fundamental input to models of radiative transfer (Bond and Bergstrom, 2006; Bond et al., 2013; Cui et al., 2016). Measured MAC_{EC} values for EC of uncoated particles fall within a relatively narrow range of 7.5 ± 1.2 m^2 g^{-1} at 550 nm (Bond and Bergstrom, 2006). It has been shown recently by an increasing amount of evidences that some components of OC also absorb sunlight in addition to its scattering effects (Andreae and Gelencser, 2006; Saleh et al., 2014; Yan et al., 2015). This type of OC is called brown carbon (BrC), which originates mainly from low-temperature combustion processes (e.g., biomass burning) or secondary formation (Andreae and Gelencser, 2006; Chen and Bond, 2010; Kim et al., 2016). Water-soluble organic carbon (WSOC) accounts for large parts of the OC in the atmosphere, especially at remote areas. For instance, WSOC accounts for around 57% of total carbon (TC) in remote sites of Europe (Pio et al., 2007). Some component of WSOC shows strong light absorption in the blue to ultraviolet spectral range, which is called as water soluble organic carbon (WS-BrC) and also causes climate warming (Andreae and Gelencser, 2006; Chakrabarty et al., 2010). However, so far, compared to EC, knowledge of the atmospheric processing of WS-BrC species and variations in their light absorption abilities are still not well investigated, and less studies are conducted on in situ values and seasonal variations in their radiative impacts in remote areas with low aerosol loading and sensitive atmospheric conditions.

The Himalayas and the Tibetan Plateau (TP) are among the most remote and highest regions in the world. Warming of the atmosphere over the TP, partly caused by carbonaceous aerosol (i.e., EC), is an important trigger for the evolution of the Asian monsoon (Prell and Kutzbach, 1992; Ramanathan and Carmichael, 2008) and melting of TP glaciers (Menon et al., 2010). Carbonaceous aerosols deposited on glaciers also cause a decrease in glacier albedo and the retreat of glaciers (Qu et al., 2014; Xu et al., 2009), which is closely connected to the fresh water supply for billions of local residents in Asia (Ramanathan et al., 2007). Therefore, studies on carbonaceous aerosols in the TP have generated considerable concern during last several decades. At present, the consensus is that most carbonaceous aerosols on the TP are mainly transported from outside of the TP (e.g., South Asia). However, almost all the previous work has focused specifically on concentrations, sources and spatial and temporal variations of carbonaceous aerosols (especially on EC) in the atmosphere of the TP (Cao et al., 2010; Cong et al., 2015; Ming et al., 2010; Zhao et al., 2013), and few have examined the light absorption characteristics of EC and WS-BrC (Li et al., 2016a), which is an obstacle in precisely evaluating the radiative forcing of carbonaceous aerosols in the atmosphere of the TP. Due to the high elevation of the TP, it is assumed that the radiative forcing contributed by WS-BrC is high, as shown in a previous study (Liu et al., 2014), which revealed that BrC accounted for approximately 20% of the direct radiative forcing from aerosols at the top of the atmosphere. Similarly, another research even suggested that the primary organic aerosol (POA) absorptivity led to ~27% reduction in the amount of the net global average POA cooling (Lu et al., 2015).

In this study, therefore, total suspended particles (TSP) were collected at two remote stations on the TP. Values and seasonal variations of optical characteristics of EC and WSOC of collected aerosols were quantitatively investigated. In addition, the relative contributions of radiative forcing from WSOC and EC were evaluated to provide fundamental information on the optical characteristics of carbonaceous matter, especially WSOC at these two stations.

2. Methods

2.1. Sampling sites

TSP samples were collected at the Southeast Tibetan Station for Alpine Environment Observation and research in Lulang (Lulang station) (29°45′58.77″N, 94°44′17.68″E, 3330 m.a.s.l.) and the Qomolangma Station for Atmospheric and Environmental Observation and Research (Everest station) (28°21′40.52″N, 86°56′55.67″E, 4276 m.a.s.l.) from August 2014 to August 2015 (Fig. 1, Table 1). The Lulang station is located in a sub-valley of the Yarlung Tsangpo Grand Canyon, a corridor for the warm-humid Indian monsoon to penetrate into the inner part of the TP (Cao et al., 2010). The Everest station is located on the north slope of the middle Himalayas (Ma et al., 2011). Both of these two stations are located on the southern fringe of the TP, far from urban cities or industry centers, and are normally considered as typical remote areas of the Northern Hemisphere and the southern TP that easily receive pollutants transported mainly from South Asia (Fig. 1, Fig. S4).

2.2. Sample collection

Because aerosol sampler equipped with air flow meter is easily broken at this high altitude of TP and is hard to run continuously for long time, a stable vacuum pump (VT 4.8, Germany) was used to collect TSP samples on 90 mm pre-burned (550 °C, 6 h) quartz fiber filters (Whatman Corp). Although air volume, which passes through each filter, cannot be achieved, it does not influences the objectives of this research (e.g., the OC/EC ratio and relative contributions to radiative forcing between WSOC and EC). Two samples were collected every month, for approximately 7 and 20 days, respectively. Four field blank filters were also collected at each station by exposing the filters in the sampler for 7 and 20 days without pumping.

2.3. Analytical methods

The OC and EC concentrations of the samples were analyzed with the standard IMPROVE-A method at 632 nm using the Desert Research Institute (DRI) Model 2001 Thermal/Optical Carbon Analyzer (Atmoslytic Inc., Calabasas, CA, USA) (Chen et al., 2015; Chow et al., 2001). The DOC concentration was measured with a total organic carbon (TOC) −5000A (Shimadzu Corp, Kyoto, Japan) (Li et al., 2016b, 2016c). The major cations (e.g., K^+) and major anions (e.g., NO_3^-) were measured by Dionex-6000 Ion Chromatograph and Dionex-3000 Ion Chromatograph (Dionex, USA), respectively (Li et al., 2007). The light absorption of WSOC was measured with a spectrophotometer (SpectraMax M5, USA) from 200 nm to 800 nm (Li et al., 2016a). Field blank concentrations of all the measured indexes were far lower than those of samples (Table S1). Detailed information on the measurement method and calculation of light absorption of EC and WSOC and their relative contributions (Cheng et al., 2011; Kirillova et al., 2014a) are shown in the supporting information file.