Review

Brown carbon in the cryosphere: Current knowledge and perspective

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Abstract

Recently, the light-absorbing organic carbon, i.e., brown carbon (BrC), has received an increasing attention, because they could significantly absorb the solar radiation in the range of short wavelengths rather than the purely scattering effect. BrC is ubiquitous in the troposphere. It could undergo long range transport within the atmospheric circulation. After the deposition on the surface of snow or ice in the cryospheric region, as the major light absorbing impurities with black carbon and dust, BrC could reduce the snow albedo and accelerate the glacier melting. In this context, this paper summarized the current knowledge of BrC (in aerosols and snow) in the cryospheric regions including the Arctic, Antarctic, and Alpines. Although some works have been conducted in those region, the current dataset on the optical properties of BrC like Absorption Ångström Exponent (*AAE*) and Mass Absorption Efficiency (*MAE*) is still limited, which hampers stimulating an accurate evaluation of its climate effects. Especially in the Himalayas and Tibetan Plateau, where very limited information concerning BrC is available. Considering biomass burning as a dominant source of BrC, a large amount of emissions from biomass burning in South Asia could reach the Himalayas and Tibetan Plateau, where the climate effect of BrC merits more investigation in the future.

Keywords: Brown carbon; Black carbon; Atmospheric aerosol; Snow; Glacier

1. Introduction

Carbonaceous components in the atmospheric aerosols play an important role in the climate system, mainly due to their solar absorption and scattering properties (Seinfeld and Pandis, 2012). It is well established that black carbon (BC) could strongly absorb the solar radiation in visible bands (Ramanathan and Carmichael, 2008; Xu et al., 2009), resulting in a direct radiative forcing ranging from 0.17 to 1.48 W m^{-2} (Bond et al., 2013). In contrast to BC, organic carbon in the atmospheric aerosols is traditionally considered to be purely scattering by climate models. However, recent research

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demonstrated that substantial fraction of organic carbon in aerosols could absorb the light in the range of short wavelengths. Such kinds of organic substances with wavelength-dependent absorption are defined as brown carbon (BrC) (Andreae and Gelencser, 2006). BrC has multiple primary sources such as biomass burning (Saleh et al., 2014; Washenfelder et al., 2015), fossil fuel (e.g., coal) combustion (Bond, 2001; Yang et al., 2009), biogenic aerosols (e.g., plant debris and fungi) and soil humic matters. Meanwhile BrC could also be secondarily formed from anthropogenic or biogenic precursors (Lack et al., 2013; Zhang et al., 2011). Such precursors like isoprene (Limbeck et al., 2003) and, lignin pyrolysis products (Gelencser et al., 2003; Hoffer et al., 2004) could yield BrC through heterogeneous or multiphase reactions in the presence of sulfuric acid or hydroxyl radicals. Although the source of BrC may varied with different locations and environment (e.g., urban, rural, forest, ocean, mountain), globally, biomass burning was identified as the most important source of BrC (Chung et al., 2012; Lack et al., 2012; Laskin et al., 2015).

BC, BrC, and mineral dust are the dominant light-absorbing substances in atmospheric aerosols. After deposited on the surface of snow and glaciers through dry/wet deposition processes, those light-absorbing substances could efficiently reduce the snow albedo (surface darkening), decrease the upwelling radiation, thereby accelerating snow melting (Flanner et al., 2007; Kaspari et al., 2015; Yang et al., 2015). A brief illustration of the sources, transport and deposition of BrC, as well its effects on the climate system is presented in Fig. 1.

The cryosphere, comprising snow, river and lake ice, sea ice, glaciers, ice shelves and ice sheets, and frozen ground, is crucial in the Earth's climate system (IPCC, 2014). The cryosphere is very sensitive to climate changes and anthropogenic activities. Currently, most efforts dedicated to the light-absorbing substances were focused on BC and dust, which were well documented in recent review literatures (Bond et al., 2013; Gertler et al., 2016; Wang et al., 2014). However, in this review, the state of art on the BrC in the cryosphere will be specifically summarized. Section 2 introduces the basic optical parameters of BrC, as well the current dataset available for the occurrence of BrC in aerosol and snow in the Artic, Antarctic, and mountain glaciers. Section 3 presents the current knowledge about the chemical speciation of BrC. Section 4 describes the radiative forcing of BrC in the atmosphere and snow, i.e., its impact on the cryosphere. Finally, research perspectives for the future research are proposed in Section 5.

2. Optical properties of BrC

The basic theory used to describe light absorption properties of BrC is the well-known power-law relationship. It can be defined as the following equation:

$$Abs_\lambda = K * \lambda^{-AAE}, \tag{1}$$

where Abs_λ (unit: Mm^{-1} or M^{-1}) refers to the absorption coefficient of BrC at wavelength λ (unit: nm). K is a constant. *AAE* indicates the wavelength dependence of light absorption.
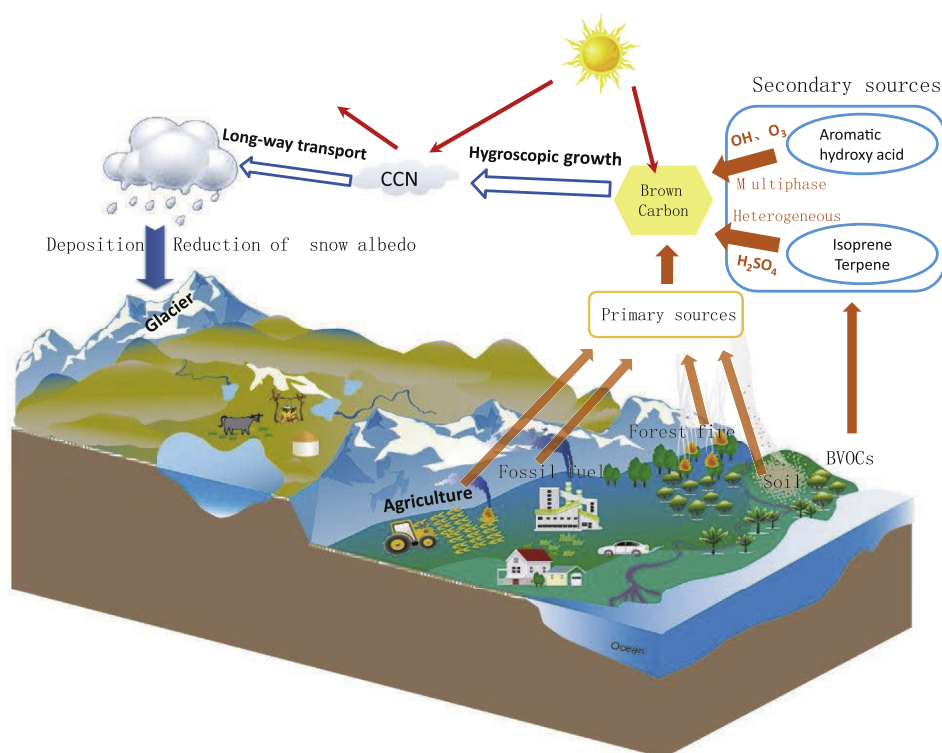

Fig. 1. Schematic overview on the sources, transport, and radiative forcing of BrC, as well as its impact on the snow and glaciers after deposition.
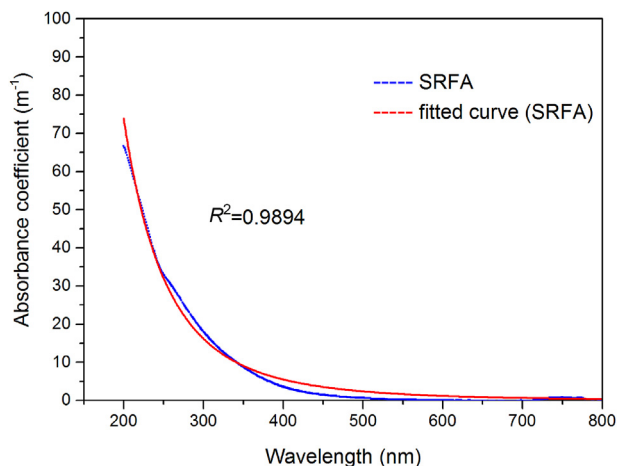
Fig. 2. The absorption spectra of a typical BrC compound (Suwannee River Fulvic Acid (SRFA) from International Humic Substance Society) measured with a Hitachi U-3900H.

It could be derived by fitting the power-law relationship Eq. (1) to the scanned spectra or calculated for a given range based on the following equation (Hoffer et al., 2006):

$$AAE = -\frac{\ln(A_{\lambda_1}/A_{\lambda_2})}{\ln(\lambda_1/\lambda_2)}, \tag{2}$$

where A_λ is the absorbance of BrC at wavelength λ. It can be directly measured from the spectrophotometer as the attenuation of incident light intensity.

In the cryosphere, McNaughton et al. (2011) found that AAE of BrC in the western Arctic aerosols vary between 1.5 and 3, while Corr et al. (2012) reported much larger AAE of BrC (3−7) in the same location. The AAE of BrC is larger than that of BC (typically unity), which means the light absorptivity of BrC will increase more dramatically as the wavelength become shorter (an example is provided in Fig. 2). Furthermore, the magnitude of AAE is closely relevant to the particle size and composition (Bahadur et al., 2012; Moosmueller et al., 2011), implying that the difference in AAE of BrC may be due to the various sources.

In addition to AAE, another crucial parameter for the definition of BrC's light absorption property is MAE (unit: $m^2 \ g^{-1}$), which demonstrate the light absorptivity for a specific mass unit of BrC. It could be derived from the following equation:

$$MAE = \frac{Abs_\lambda}{C}, \tag{3}$$

where C is the concentration of BrC. The MAE of BrC decreases sharply with an increasing wavelength, which means it has much stronger absorption in the shorter wavelength (Alexander et al., 2008; Andreae and Gelencser, 2006). To our best knowledge, there are very limited studies available on the MAE of BrC in the cryosphere. The MAE and AAE of BrC reported or used in the published literatures are listed in Table 1. McNaughton et al. (2011) reported that MAE of BrC in the western Arctic aerosols is $(0.83 \pm 0.5) \ m^2 \ g^{-1}$ at 470 nm and $(0.27 \pm 0.08) \ m^2 \ g^{-1}$ at 530 nm, which are comparable to the MAE of BrC in some rural or even urban aerosols, for instance, $(0.7 \pm 0.2) \ m^2 \ g^{-1}$ at 365 nm in Gosan, Korea (Kirillova et al., 2014) and $0.60 \ m^2 \ g^{-1}$ at 365 nm in DeKalb, USA (Hecobian et al., 2010). While the MAE of BC and dust in snow across Northern China were $6.3 \ m^2 \ g^{-1}$ and $0.9 \ m^2 \ g^{-1}$, respectively (Wang et al., 2013). Similarly, the MAE of BC in Arctic snow was $6.0 \ m^2 \ g^{-1}$ (Hegg et al., 2010). Although the MAE of BrC is lower than that of BC, the global burden of BrC aerosols was simulated to be $0.65 \ mg \ m^{-2}$, nearly 3-fold of the BC concentration $(0.19 \ mg \ m^{-2})$ in the atmosphere (Feng et al., 2013). Given the large abundance of BrC in the environment, a contribution of light absorption by BrC may not be ignored.

3. Chemical properties of BrC

The chemical composition of BrC is extremely complex. Nevertheless, in terms of chemical speciation, BrC could be generally divided into humic-like substances (HULIS) and tar ball (Chakrabarty et al., 2010; Cheng et al., 2016; Hoffer et al., 2016).

3.1. HULIS

HULIS are termed as their similarity with terrestrial/aquatic humic substances. They are mixtures of various macromolecular organic compounds and ubiquitous in atmospheric environment (Graber and Rudich, 2006; Lin et al., 2010a; Mukai and Ambe, 1986; Zheng et al., 2013). HULIS mainly consist of aromatic and aliphatic structure bond with hydrocarbon side chains like hydroxyl, carbonyl, carboxyl and

Table 1
Summary of light-absorbing properties of BrC reported or used in the published literatures.

Site	Sampling period	Sample type	MAE (W)[a] ($m^2 \ g^{-1}$)	AAE	Reference
Western Arctic	31 March−19 April, 2008	Aerosol	0.83 ± 0.15 (470 nm); 0.27 ± 0.08 (530 nm)	1.5−3	McNaughton et al., 2011
Western Arctic	Spring and summer of 2008	Aerosol		3−7	Corr et al., 2012
Artic	1998 and 2005−2009	Snow		5	Doherty et al., 2010
Northern China	January and February of 2010	Snow	0.3 (550 nm)	6	Wang et al., 2013
North America	10 January and 28 January−21 March, 2013	Snow		5	Doherty et al., 2014

[a] W refers to the wavelength at which the MAE (mass absorption efficiency) is calculated; AAE (Absorption Ångström Exponent).

organosulfate functional groups (Chakrabarty et al., 2010; Graber and Rudich, 2006; Mukai and Ambe, 1986).

3.1.1. Experimental methods for extraction and determination of HULIS

Given the unclear physical-chemical characteristics of HULIS, there is no uniform extract method applicable in all conditions. It is even hard to evaluate whether an extract method valid to isolate HULIS from atmospheric media (Graber and Rudich, 2006). Table 2 summarizes the isolation and quantification method applied in published literatures. It is obvious that the solid phase extraction (SPE) method is the most frequently employed in isolating HULIS from aerosols and snow in cryospheric regions for its simple operation and high extract efficiency (Facchini et al., 2000; Fan et al., 2012). Specifically, there are four different SPE methods, owing to the diverse sorbents: ENVI-18 (Fan et al., 2012), HLB (Varga et al., 2001), XAD (Duarte and Duarte, 2005) and DEAE (diethylaminoethyl) (Baduel et al., 2009). They are competent to the exaction of HULIS with low limit of detection and high recovery yield. However, they also have their particular preference and disadvantage in the isolation of HULIS (Fan et al., 2012, 2013). HLB method favors polar organic matters with high recovery. It also can preserve the nature of samples while not change the properties of isolated HULIS. XAD tends to isolate hydrophobic and aromatic components due to its sorbent (non-ionic macro porous resin) used. Acidizing of solute is not necessary for DEAE and it prefers the compounds containing more aromatic signatures. However, large amount of salts could present in the eluents with DEAE extraction, which will interfere the characterization of HULIS.

For the quantification of HULIS followed by its isolation, there are total organic carbon (TOC) measurement (Kiss et al., 2002; Krivacsy et al., 2008; Salma et al., 2007), UV−Vis detection (Krivacsy et al., 2008; Samburova et al., 2005), evaporative light-scattering detection (Emmenegger et al., 2007; Lin et al., 2010b) and gravimetric method (Kiss et al., 2002). TOC method is an easy and common approach to analyze the amount of HULIS-C. Hoffer et al. (2006) assumed the conversion factor is 1.90 between HULIS and HULIS-C for the aerosol samples collected in the Amazon basin, while 1.81 was used by Salma et al. (2007) for the quantity of HULIS at Budapest, Hungary. The choosing of proper conversion factor is critical during such calculation, which may arise large uncertainties.

3.1.2. Abundance of HULIS in aerosols and snow

We focus on the concentration of atmospheric HULIS at the regions where there are a wide range of distributions of glacier and ice sheets (i.e., Arctic, Alps). Considering the albedo modification, glaciers are very sensitive to the increase in light-absorbing impurities like HULIS. Meanwhile, the analysis of the HULIS trapped and accumulated in polar ice sheets and mid-latitude mountain glaciers provides a unique chance to reconstruct the changing atmospheric composition in the past (Legrand et al., 2013).

Nguyen et al. (2014) found the annual mass concentration of HULIS in Arctic aerosols is (0.02 ± 0.01) µg m^{-3} and the concentration of HULIS showed an obvious seasonal variation with the highest value occurred in biomass burning influenced periods. Guilhermet et al. (2013) estimated the concentration of water-soluble atmospheric HULIS was 0.07 µg m^{-3} in summer, basing on the analysis of HULIS trapped in the ice core at Col du Dôme (4250 m a.s.l.), Alpine.

Legrand et al. (2007) determined HULIS in an Alpine ice core by measuring the absorbance with a UV−VIS spectrophotometer at the wavelengths of 250 and 350 nm. They found that summertime mass concentrations of HULIS were generally below 150×10^{-9} before 1945, while the contributions tended to increase after World War II (ranging from 200×10^{-9} to 900×10^{-9}). The increased HULIS in Alpine glaciers could be attributed to the growing emission of anthropogenic precursors as well as the enhanced atmospheric oxidants (Legrand et al., 2007).

Using a DEAE resin, Legrand et al. (2013) extracted water-soluble HULIS in Antarctic snow samples, and then quantified it with a Shimadzu TOC analyzer. Water-soluble HULIS in central Antarctica snow was determined to be 2×10^{-9} (annual average), much lower than those in Alpine snow of 30×10^{-9}. In Arctic (Barrow, Alaska) snowpack, Beine et al. (2011) estimated that HULIS and other unknown chromophores could account for nearly half of the total absorption measured between 300 and 500 nm. Voisin et al. (2012) further calculated the *MAE* of HULIS in the snow from the same region as (26 ± 11) cm^2 mg^{-1} at 250 nm. And *AAE* varied from 4.5 (between 300 and 350 nm) to 7.7 (between 450 and 500 nm).

Table 2
Summary of isolation and quantification method for HULIS in cryosphere.

Site	Sample type	Isolat. Meth.	Quantif. Meth.	HULIS concentration (µgC L^{-1})			Reference
				Min	Max	Average	
Station Nord, Arctic	Aerosol (PM$_{10}$)	HLB	TOC			0.02[a]	Nguyen et al., 2014
Barrow, Alaska, Arctic	Snowpack	DEAE	TOC	1	16		Voisin et al., 2012
Alaska, Arctic	Snowpack		UV−Vis	1200[b]	1500[b]		France et al., 2012
Col du Dôme, Alpine	Ice core		UV−Vis	50	400		Legrand et al., 2007
Col du Dôme, Alpine	Ice core	DEAE	TOC	4	250		Guilhermet et al., 2013
Antarctic	Snow, coast	XAD-8	Gravimetry	16[c]	397[c]		Calace et al., 2005
Antarctic	Snow, inland	XAD-8	Gravimetry	25[c]	146[c]		Calace et al., 2005

[a] in the unit of µgC m^{-3}.
[b] the concentration of non-BC light-absorbers in snowpack.
[c] in the unit of µg L^{-1}.

The high values of *AAE* were the typical characteristics of BrC (Andreae and Gelencser, 2006), i.e., the HULIS in snow has strong absorption in the short wavelengths.

3.2. Tar balls

Tar ball, emitted from biomass burning (especially in smoldering condition), is an important type of BrC (Chakrabarty et al., 2010; Hoffer et al., 2016). They are spherical (30−500 nm in diameter), amorphous, and typically not aggregated with other particles (Hand et al., 2005; Posfai et al., 2004), which are distinct from soot in terms of morphology (i.e., BC) (Fig. 3). According to Alexander et al. (2008), the light absorption of tar ball (carbon sphere) in East Asian outflow may be comparable to or even larger than soot at the individual particle level. The derived mean *MAE* of tar balls range from ~3.6 m^2 g^{-1} to 4.1 m^2 g^{-1} at 550 nm, approximating to that of BC (Alexander et al., 2008). For the laboratory-generated tar balls, their *MAE* values were determined to be 0.8−3.0 m^2 g^{-1} (λ = 550 nm), with average *AAE* of 2.9 in the wavelength band of 467−652 nm (Hoffer et al., 2016).

Tar ball is ubiquitous in the troposphere, especially in the regions receiving strong influence of biomass burning. According to our previous study in the Himalayas and Tibetan Plateau aerosols, tar balls accounted for 3%−27% of the particle number concentrations (Cong et al., 2009). Given the abundance and their optical properties, tar balls impart considerable effects in radiative forcing. However, few studies on the tar balls have been carried out in the snow surface and ice core. At the same time, the degree to which tar balls contribute to the solar absorption of BrC, especially relative to HULIS, is still unclear. Their hygroscopic properties, mixing states and aging processes urgently require more attentions to achieve accurate modeling of the radiative forcing.

4. Radiative forcing of BrC in the atmosphere and snow

Globally, the radiative forcing by BrC was estimated to be in the range of 0.1−0.25 W m^{-2}, close to 25% of that by BC (Feng et al., 2013). Chung et al. (2012) also highlighted the global significance of BrC. They reported that it could be responsible for 20% of carbonaceous aerosol solar absorption at 550 nm. Moreover, Lin et al. (2014) calculated the global absorption of BrC in the atmosphere ranging from +0.22 W m^{-2} to + 0.57 W m^{-2}, which is equal to 27%−70% of the BC absorption. Nevertheless, regarding the cryospheric area, such assessment in the regional scale is still absent.

BrC in the atmosphere could be transported to the surface of snow by dry or wet deposition, which will further lead to the enhancement of snow melting. For the radiative forcing of BrC in snow, most current studies focus on their contribution to the total light absorption relative to BC and dust, rather than providing an absolute value (in the unit of W m^{-2}). Dang and Hegg (2014) proposed that HULIS and polar organic carbon from the western North American snow samples account for 13% of the total light absorption. While for the Arctic snow samples, Doherty et al. (2010) proportioned about 40% of the visible and ultraviolet absorption to BrC. However, it should be kept in mind that this approach must base on a given *AAE* values of BrC, which might be variable with different sources and environment. Apart from these regional analysis, Lin et al. (2014) applied the IMPACT (Integrated Massively Parallel Atmospheric Chemical Transport) model to assess the global radiative forcing of organic aerosols deposited over in-land snow with the range of 1.1 × 10^{-3} W m^{-2} and 3.1 × 10^{-3} W m^{-2}. These values could account for as large as 24% of radiative forcing by BC. However, they can not separate the radiative forcing of BrC from total organics yet.

Moreover, BrC, BC and dust as the major light-absorbing matters in the cryosphere, they tend to mix with each other during the transport or aging process (Kaspari et al., 2014; Lack and Cappa, 2010; Liu et al., 2015), leading to another form of radiative forcing by acting as a lens, i.e., lensing effect. Lack et al. (2012) found that the absorption enhancement ascribes to internal mixing can be 1.4 times at 532 nm in biomass burning particles. The enhancement strongly dependent on the coating thickness, mixing state (Liu et al., 2015). Similar with BrC, dust also might mixed with BC to change its albedo reduction in snow (Kaspari et al., 2014). Thus BrC in the cryosphere not only absorbs solar radiation directly but also can leads to an extra radiative forcing by interaction with
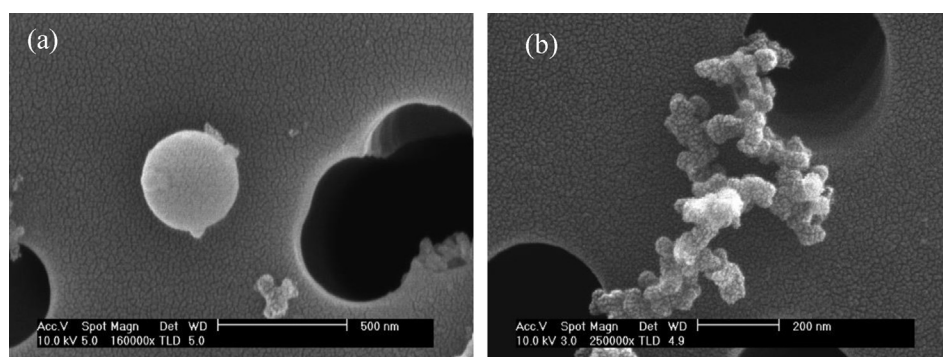

Fig. 3. Secondary electron images of (a) tar ball and (b) soot in the Himalayan aerosols (6500 m a.s.l.) observed by scanning electron microscopy (Cong et al., 2010).

BC. If the lensing effect was incorporated into the modeling, the current results on the radiative forcing of BrC may be modified considerably.

5. Perspective

As discussed above, unlike BC, the chemical and optical properties of BrC remains poorly understood especially in the cryospheric regions. The measurements of BrC still suffer from some problems. The light absorption characteristics of BrC were commonly determined in the water extracts. However, the remaining fraction of BrC (water insoluble) is usually ignored. In particular, the water-insoluble BrC may have higher mass-specific absorption than the water-soluble fraction. Another problem is that BrC were extracted by different chemical solvents like acetone and methanol besides water in different studies. Therefore, their abundances obtained by different pretreatment and determination methods are difficult to compare with each other. For many researches, BrC is defined as the light-absorbing carbon measured by optical instruments such as Aethalometer. The relationship between the specific chemical composition of BrC and optical properties also needs more research. Another uncertainty is associated with the determination of *MAE* of BrC in the mixing state. In order to derive reliable result from this method, more precise optical knowledge of BrC is needed. The hydroscopic properties of BrC need more attention, since it determines the role as cloud condensation nuclei for the formation of clouds.

Discrepancy between observations and model results in a substantial uncertainties in quantifying the effect of BrC. Accurate model simulation of BrC depends on our comprehensive knowledge regarding its sources, chemical composition, optical properties, and so on. More BrC measurements are needed not only in laboratory characterization, but also in field campaigns being carried out in surface air, total atmospheric column and snow. The mixing state, aging and transformation during wet scavenging and, dry deposition on the glacier surface are also important issues.

In terms of geographic distribution of research site, although there are several works have been conducted in Alpine, Arctic and Antarctic, the knowledge about the spatial distributions of BrC is still in deficit especially in the Himalayas and Tibetan Plateau. High levels of carbonaceous aerosol from biomass/biofuel burning exist over South Asia (Bonasoni et al., 2010; Gustafsson et al., 2009; Luthi et al., 2015). Increasing evidences have demonstrated that those air pollutants could reach the high altitude of Himalayas and even transport into the inland of the Tibetan Plateau (Cong et al., 2015). What's more, the strong solar radiation on the Himalayas and Tibetan Plateau may also accelerates the formation of BrC through the photochemical reactions. With the vast snow and ice cover (more than 100,000 km^2 (Yao et al., 2012)), the Himalayas and Tibetan Plateau have been proved to be very sensitive to climate change. Since biomass burning and secondary formation are the dominant sources of BrC, climate effects of BrC in the Himalayas and Tibetan Plateau

(not only the atmospheric warming but also the reduction of snow albedo) merit more in-depth investigation.

Acknowledgments

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