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Carbonaceous aerosols recorded in a Southeastern Tibetan glacier: variations, sources and radiative forcing

M. Wang[1,2], B. Xu[1], J. Cao[3], X. Tie[3,4], H. Wang[2], R. Zhang[5,2], Y. Qian[2], P. J. Rasch[2], S. Zhao[3], G. Wu[1], H. Zhao[1], D. R. Joswiak[1], J. Li[1], and Y. Xie[1]

[1]Key Laboratory of Tibetan Environment Changes and Land Surface Processes, Institute of Tibetan Plateau Research, Chinese Academy of Sciences, Beijing 100101, China
[2]Atmospheric Sciences and Global Change Division, Pacific Northwest National Laboratory (PNNL), Richland, WA 99352, USA
[3]State Key Laboratory of Loess and Quaternary Geology, Institute of Earth Environment, Chinese Academy of Sciences, Beijing 100085, China
[4]National Center for Atmospheric Research, Boulder, CO, 80303, USA
[5]Key Laboratory for Semi-Arid Climate Change of the Ministry of Education, College of Atmospheric Sciences, Lanzhou University, Lanzhou 730000, Gansu, China

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Correspondence to: M. Wang (wangmo@itpcas.ac.cn)

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Abstract

High temporal resolution measurements of black carbon (BC) and organic carbon (OC) covering the time period of 1956–2006 in an ice core over the southeastern Tibetan Plateau show a distinct seasonal dependence of OC / BC ratio with higher values in the non-monsoon season than during the summer monsoon. We use a global aerosol-climate model, in which BC emitted from different source regions can be explicitly tracked, to quantify BC source-receptor relationships between four Asian source regions and the southeastern Tibetan Plateau as a receptor. The model results show that South Asia is a primary contributor during the non-monsoon season (October to May) (81 %) and on an annual basis (74 %), followed by East Asia (14 % and 21 %, respectively). The ice-core record also indicates stable and relatively low BC and OC deposition fluxes from late 1950s to 1980, followed by an overall increase to recent years. This trend is consistent with the BC and OC emission inventories and the fuel consumption of South Asia as the primary contributor. Moreover, the increasing trend of OC / BC ratio since the early 1990s indicates a growing contribution of coal combustion and biomass burning to the emissions. The estimated radiative forcing induced by BC and OC impurities in snow has increased since 1980, suggesting an increasing influence of carbonaceous aerosols on the Tibetan glacier melting and the availability of water resources in the surrounding regions. Our study indicates that more attention to OC is merited because of its non-negligible light absorption and the recent rapid increases evident in the ice core record.

1 Introduction

Carbonaceous aerosol, released from fossil fuel, biofuel and/or biomass combustion, contains both black carbon (BC, a.k.a, elemental carbon, EC), a strong light absorber, and organic carbon (OC), which also absorbs in the near infrared, but more weakly than BC. Often mixed with other aerosol species, BC impacts human health, crop yields and

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regional climate (Auffhammer et al., 2006; Tie et al., 2009), and is believed to be the second strongest climate warming forcing agent after carbon dioxide (Jacobson, 2001).

Because of their high population density and relatively low combustion efficiency, developing countries in South and East Asia such as India and China are hotspots of carbonaceous aerosol emissions (Ramanathan and Carmichael, 2008). During the cold and dry winter monsoon seasons, haze (heavily loaded with carbonaceous aerosols) builds up over South Asia, and exerts profound influences on regional radiative forcing (Ramanathan et al., 2007; Ramanathan and Carmichael, 2008), hydrologic cycles (Menon et al., 2002; Ramanathan et al., 2005), and likely Himalaya-Tibetan glacier melting that could be accelerated by the absorption of sunlight induced by BC in the air and deposited on the ice and snow surfaces (Ramanathan et al., 2007; Hansen and Nazarenko, 2004).

Due to the lack of long-term observations of emissions and concentrations of atmospheric carbonaceous aerosols, it is difficult to evaluate the effects of BC and OC on historical regional climate and environment before the satellite era. Some studies have evaluated historical anthropogenic emissions based on the consumption of fossil fuels and biofuels (Novakov et al., 2003; Ito and Penner, 2005; Bond et al., 2007; Fernandes et al., 2007). While fossil fuel is the major energy source in the urban areas of South Asia and East Asia, biomass combustion, such as fuel wood, agricultural residue and dung cake, is prevalent in rural areas (Revelle, 1976; Venkataraman et al., 2010; Street and Waldhoff, 1998). Biomass is considered as the major source of black carbon emissions (Reddy and Venkataraman, 2002; Venkataraman et al., 2005). However, as reliable biofuel consumption data are hard to obtain, estimates of BC and OC emissions from biomass burning are ambiguous and incomplete.

Measurements of carbonaceous aerosol concentrations in glacier ice are an ideal means to reconstruct historical emissions and reveal long-term trends of anthropogenic aerosol impacts on local climate. Greenland ice core measurements were previously used to reconstruct the North American BC emission history and its effects on surface radiative forcing back to the 1880s (McConnell et al., 2007). Himalayan ice cores

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retrieved from the Tibetan Plateau have revealed the mixed historical emissions from South Asia, Central Asia and the Middle East and also been used to evaluate radiative forcing from BC in snow (Ming et al., 2008; Kaspari et al., 2011). Using the Snow, Ice, and Aerosol Radiative (SNICAR) model, Flanner et al. (2007) estimated an instantaneous regional forcing of exceeding 20 W m^{-2} by BC in snow/glaciers over the Tibetan Plateau during spring.

By using five ice core records, Xu et al. (2009a) elucidated an important contribution of BC to the retreat of Tibetan glaciers in addition to greenhouse gases. Due to the short atmospheric lifetime of carbonaceous aerosols compared to greenhouse gases, emission reductions may be an effective way to mitigate their warming effects. Thus it is particularly important to identify the source regions and the combustion sources of carbonaceous aerosols observed in Tibetan glaciers. Xu et al. (2009a) suggested that BC deposited on Tibetan Plateau was broadly from Europe and Asia. However, they did not discuss on more specific source regions and the combustion sources. In this study, we use the ice core retrieved from the southeastern Tibetan Plateau, also known as the Zuoqiupu ice core in Xu et al. (2009a), to reconstruct the history of atmospheric deposition of carbonaceous aerosols in this region, and to characterize emissions and source-receptor relationships with the help of a global climate model in which BC emitted from different source regions can be explicitly tracked. We also estimate the respective contributions from BC and OC to radiative forcing in glaciers using the ice core measurements and the SNICAR model.

2 Methods

2.1 Measurements of carbonaceous aerosols in ice core

Zuoqiupu glacier is in the southeastern Kangri Karpo Mountains, located at the southeastern margin of the Tibetan Plateau (Fig. 1). In 2007, an ice core of 97 m in depth (9.5 cm in diameter) was retrieved within the accumulation zone of Zuoqiupu glacier at

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96.92° E, 29.21° N, 5600 m a.s.l. The ice core was kept frozen and transported to laboratory facilities at the Institute of Tibetan Plateau Research (Lhasa branch) for analysis. BC and OC concentrations were measured by using a Desert Research Institute (DRI) Model 2001 Thermal/Optical Carbon Analyzer following the IMPROVE TOR protocol (Chow et al., 1993; Chow and Watson 2002; Cao et al., 2008). Note that according to the thermal/optical measurement method, the analytical result is technically called "EC". Herein we use "BC" to be consistent with the notation in our model simulations and in the literature. The reported OC concentrations from the ice-core measurements can only account for water-insoluble part of OC from the aerosol emissions. Further details on the analysis methods, ice core dating and calculation of BC and OC seasonal deposition fluxes can be found in Xu et al. (2009a).

2.2 Model and experimental setup

We use the Community Atmosphere Model version 5 (CAM5; Neale et al., 2010) to help understand the emissions, transport and dry/wet deposition of carbonaceous aerosols in the atmosphere. In the default aerosol scheme of CAM5, BC and primary OC are emitted into an accumulation size mode, where they immediately mix with co-existing hygroscopic species such as sulfate and sea salt (Liu et al., 2012). Hygroscopic aerosol particles in the accumulation mode are subject to wet scavenging and removal by precipitation. Recent model improvements to the representation of aerosol transport and wet removal in CAM5 by Wang et al. (2013) have substantially improved the global distribution of aerosols, particularly, over remote regions away from major sources. To minimize the model biases in simulating meteorological conditions and, particularly, circulations that are critical to aerosol transport, we configure the CAM5 model to run in an offline mode (Ma et al., 2013) with wind, temperature, surface fluxes and pressure fields constrained by observations. However, cloud/precipitation fields and interactions between aerosol and clouds are allowed to evolve freely. A source tagging technique has been recently implemented in the CAM5 model to allow for explicitly tracking aerosols emitted from individual source regions and, therefore, assists in quantitatively charac-

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terizing source-receptor relationships (Wang et al., 2014). This capability is used in the present study to do source attribution for carbonaceous aerosols deposited to the Zuoqiupu glacier.

We conducted an 11 yr (1995–2005) CAM5 simulation at horizontal grid spacing of $1.9^{\circ} \times 2.5^{\circ}$ and 56 vertical levels, with prescribed sea surface temperatures and sea ice distribution. Reanalysis products from NASA Modern Era Retrospective-Analysis for Research and Applications (MERRA) (Rienecker et al., 2011) are used to constrain the meteorological fields of CAM5. For aerosols (including OC, BC and other important species), we use the year-2000 monthly mean emissions described by Lamarque et al. (2010) that have been used in many global climate models for present-day climate simulations, included in the fifth assessment report (AR5) by the Intergovernmental Panel on Climate Change (IPCC). The monthly mean emissions are repeatedly used for the 11 yr simulation.

3 Results and discussion

3.1 Seasonal dependence of carbonaceous aerosols

BC and OC concentrations in the Zuoqiupu ice core both exhibit statistically significant seasonal variations at the 0.05 level corresponding to the stable oxygen isotope variability, which shows high values during the winter and low values during the summer (Xu et al., 2009a). This seasonal dependence of BC and OC in ice core is consistent with available observations of atmospheric aerosols in the south slope of the Himalayas and the southeastern Tibetan Plateau, and the high concentration of carbonaceous aerosols during the cold and dry season was suggested to associate with the South Asian haze (Cong et al., 2009; Marinoni et al., 2010; Kaspari et al., 2011; Zhao et al., 2013).

As shown in Fig. 2, concentrations of OC and BC have distinct differences between the monsoon and non-monsoon seasons. The ratio of OC to BC also shows clear

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seasonal dependence. The slope of the fitted line to measured OC vs. BC concentrations during non-monsoon season is ~6.3, which is twice the slope for monsoon season (~3.2). The analysis of covariance (ANCOVA) for slope differences of single linear regressions of OC against BC between monsoon and non-monsoon seasons indicates that the seasonal dependence of the relationship between the concentrations of OC and BC is significant (at the 0.05 significance level). This agrees with measurements derived from the ice core drilled from the Palong-Zanbu No. 4 Glacier (Xu et al., 2009b) and in atmospheric samples collected from Lulang, southeastern Tibetan Plateau (Zhao et al., 2013). The seasonal dependence of the relationship between OC and BC can be derived from the seasonal sources of carbonaceous particles and/or atmospheric sink processes. For instance, the emissions from forest fires and biomass burning for heating are likely to increase in cold and dry non-monsoon seasons, and/or wet deposition of atmospheric carbonaceous aerosols and biogenic matter emitted from lush vegetation in the hot and wet monsoon seasons may increase. High concentrations of carbonaceous aerosols recorded in the ice core suggest an increased solar radiative forcing in non-monsoon seasons.

3.2 Source attribution

As the ice core drilling site was located at a remote and elevated area over the southeastern Tibetan Plateau, where local emissions are minimal. Deposition of carbonaceous aerosols is most likely contributed by the non-local major emission sources (e.g., BC emissions shown in Fig. 3) in South Asia and East Asia. The 10 yr average wind fields (at the surface and 500 hPa from MERRA reanalysis datasets), as shown in Fig. 3, indicate distinct circulation patterns during summer monsoon (June–September) and non-monsoon (October–May) seasons, which in part determine the seasonal dependence of transport of aerosols emitted from the different major sources.

To quantitatively attribute the source of BC at the drilling site (as a receptor region), we use the CAM5 model with the BC source tagging capability to conduct an 11 yr simulation (Wang et al., 2014), with the last 10 yr (1996–2005) used for analysis. The

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surrounding area is divided into four source regions (see Table 1 and Fig. 4): South Asia, East Asia, Southeast Asia and Central Asia. BC emissions from each of the four regions and the rest of the world are explicitly tracked, so that the fractional contributions by emissions from the individual source regions to BC deposition at the receptor region can be explicitly calculated. Figure 4 shows the spatial distribution of fractional contribution from the four source regions. BC deposition at the drilling site (indicated by the pink box in Fig. 4), which has a consistent seasonal dependence (i.e., more during the non-monsoon season; Fig. 5) with ice core measurements, is predominately (over 95 %) from South Asia and East Asia. The seasonal dependence of BC deposition is also consistent with a recent regional climate modeling study on BC deposition on the Himalayan snow cover from 1998 to 2008 (Ménégoz et al., 2013).

During the non-monsoon season, strong westerly dominates the transport from west to east at all levels. Emissions from northern India and central Asia can have influence on BC in the direct downwind receptor region over southeastern Tibetan Plateau. During the summer monsoon season, the westerly moves northward and the monsoon flow from Bay of Bengal at the surface and middle levels (e.g., 500 hPa), coupled with the monsoon from Indochina peninsula and South China Sea, exerts influence on BC in the receptor area. The strong monsoon precipitation removes BC from the atmosphere during the transport. The high Himalayas can partly block the further transport of emissions from South Asia to Tibetan Plateau, although small local topographical features such as the Yarlung Tsangpo River valley can provide a gate for the pollution to enter the inner Tibetan Plateau (Cao et al., 2010). Elevated emissions from the west (or northern part of South Asia) can take the pathways at middle and upper levels but they have minimal contribution to deposition. Therefore, BC emissions from East Asia play a relatively more important role affecting deposition at the Zuoqiupu site during the monsoon season.

The fractional contributions to 10 yr mean BC deposition at the drilling site from the four tagged regions are summarized in Table 1. Results show that South Asia is the dominant contributor (~81 %) during the non-monsoon season with ~14 % from East

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Asia, while the contribution of East Asia (~56%) is larger than that of South Asia (~39%) during the monsoon season. For the annual mean BC deposition, South Asia (~75%) is the biggest contributor, followed by East Asia (~21%). Emissions from the central Asia and Southeast Asia regions have much smaller contributions (< 3%) for all seasons. These results agree well with the short-term source attribution study by Lu et al. (2012) using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model. Note that the BC emission inventory (Lamarque et al., 2010) used in CAM5 does not consider seasonal variations in anthropogenic emissions, which is likely to have introduced biases in the quantitative estimates by the model, but the relative importance of source regions should be robust.

3.3 Interannual variations and long-term trend

Based on annual snow accumulation and BC and OC concentrations derived from the ice record, the annual BC and OC deposition fluxes can be estimated, which are then used to examine the interannual variations and long-term trend in the fluxes and the ratio of OC / BC, as well as the relationship with emissions from the major contributor. As illustrated in Fig. 6, from late 1950s to 1980, the BC and OC fluxes in the Zuoqiupu ice core are relatively low and stable in comparison to those after 1980. During the period 1956 to 1979, average fluxes are 9.1 and 28.7 $mg\,m^{-2}\,a^{-1}$ for BC and OC, respectively. Both BC and OC fluxes began to show increasing trends from early 1980s. These trends continued in the early 1990s but started to drop in the mid-1990s, reaching a minimum in 2002 followed by a rapid increase. In 2006, BC and OC fluxes are 19.2 and 93.9 $mg\,m^{-2}\,a^{-1}$, respectively, which are two and three times the respective average fluxes before 1980. The 5 yr average OC / BC flux ratio is steady before 1990; however, it shows a continual increase afterwards and has been higher than the average value (3.2) for the period of 1956–1979 since mid-1990s (Fig. 6). The 10 yr CAM5 model simulation, in which emissions are fixed but meteorological conditions vary, shows no increasing trend in BC and OC deposition fluxes (Fig. 5), indicating that the increasing trend seen in the observations was not due to changes in meteorology.

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As shown in the CAM5 model simulation, the annual mean atmospheric deposition of BC over southeastern Tibetan Plateau is mostly contributed by emissions from South Asia, particularly, in the non-monsoon season. The BC and OC deposition fluxes derived from the ice-core measurements may reflect changes in South Asian emissions to some extent. The temporal variations of BC and OC deposition fluxes (see Fig. 6) are compared with the primary BC and OC emissions from fossil fuel and biofuel combustion in South Asia from 1955–2000 (Bond et al., 2007). More recent (1996–2010) BC and OC emissions (Lu et al., 2011) are also illustrated in Fig. 6 to extend the emission data to cover the entire time period having ice core data, but it is only for India, the largest energy consumer and carbonaceous aerosol-emitting country in South Asia. There are differences between the emissions of Bond et al. and Lu et al. during the overlap time period (1996–2000). However, good agreements on the increasing trend can be found in the respective deposition fluxes and emissions of BC and OC (Fig. 6). The OC / BC emission ratio also shows an increasing trend from the late 1990s to 2003, which is consistent with that of OC / BC ratio in the ice core record. The annual mean aerosol index over industrial and populated cities in the northern part of India increased from 1982–1993 and more significantly from 2000–2003 (Sarka et al., 2006). This trend is similar to that of carbonaceous aerosols in the ice core record, and it indicates a causal relationship between BC and OC over southeastern Tibetan Plateau and emissions from north part of South Asia.

3.4 Emission source analyses

BC and OC in the atmosphere are co-emitted from a variety of natural and anthropogenic sources, including combustion of fossil fuel, biofuel and biomass burning. In general, open biomass burning typically produces more abundant OC (i.e., larger OC / BC ratio) compared to fossil fuel combustion due to a lower process temperature (Ducret and Cachier, 1992). The OC / BC ratio has often been used to discriminate fossil fuel combustion and biomass burning emissions in the atmosphere and in precipitation (Novakov et al., 2000; Stone et al., 2007; Ducret and Cachier, 1992; Xu et al.,

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2009b). For example, Cao et al. (2005) collected particulate matter samples from the plumes of residential biomass burning, coal combustion, and motor-vehicle exhaust sources, and analyzed OC and BC with DRI Thermal/Optical Carbon Analyzer (Model 2001). They reported average OC / BC ratios of 60.3, 12.0, and 4.1 for biomass burning, coal-combustion and vehicle exhaust, respectively. The increasing OC / BC ratios based on the ice core measurements since the early 1990s (Fig. 6) suggest an expanded coal consumption and usage of biomass fuel although the ratios might have low bias because water-soluble OC was not captured in the sample analyses. However, such bias would have occurred to all the samples and had little impact on the trend, unless including water-soluble OC could dominate the temporal variation of OC / BC ratio. Otherwise, our results indicate that the relative contribution of coal combustion and biomass burning to the carbonaceous particles deposited into the ice core in southeastern Tibetan Plateau has been increasing faster than the contribution of fossil fuel combustion since early 1990s. Improved combustion technologies could also increase the OC / BC ratio, which was less likely to dominate the increasing trend in OC / BC ratio because BC emissions had the same trend.

The temporal variations of BC and OC in the Zuoqiupu ice core, along with the source attribution analysis of the CAM5 model results, suggest an increasing trend in emissions and altered emission sources in South Asia during the late 20th century. Coal has been the primary energy source in South Asia. For example, in India coal accounted for 41 % of the total primary energy demand in 2007, followed by biomass (27 %) and oil (24 %) (IEA, 2009). The consumption data of coal and crude oil in South Asia (BP Group, 2009) is compared with the BC and OC fluxes in Fig. 6 (bottom right). Coal consumption increased from 1965–2008, in particular from 1980–1995 and from 2003–2008 after a level off during 1996–2002. This trend is consistent with the variations of BC and OC deposition fluxes in the Zuoqiupu ice core. The correlations between coal consumption and BC ($R^2 = 0.43$, $p < 0.001$) and OC ($R^2 = 0.62$, $p < 0.001$) in the ice core are both statistically significant. The oil consumption had a comparable increasing trend as coal before it slowed down during 2000–2006.

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Biomass is the second largest energy resource in South Asia, and it is essential in rural areas. In India, 70 % of the population lives in rural areas, and depends substantially on solid fuels (i.e., firewood, animal dung, and agriculture residues) for cooking and heating (Heltberg et al., 2000). Even in urban areas, biomass contributes to 27 % of the household cooking fuel (Venkataraman et al., 2010). Previous studies have concluded that carbonaceous aerosol emissions from biomass burning are the largest source in South Asia (Venkataraman et al., 2005; Gustafsson et al., 2009). A general increase in energy-intensive life-styles associated with accelerated growth of population and economy puts pressure on energy resources, and induces energy transitions and use of non-sustainable biomass in South Asia (Sathaye and Tyler, 1991; Pachauri, 2004; Fernandes et al., 2007). For instance, biofuel consumption in South Asia increases by 21 % per decade on average during 1950–2000 (Bond et al., 2007; Fernandes et al., 2007). In addition, fuel wood, a more desirable biofuel option, contributed 68 % in 1978 to total energy demand by rural populations in India, and increased to 78 % in 2000 (Fernandes et al., 2007).

3.5 Radiative forcing induced by carbonaceous aerosols in Tibetan Glaciers

BC is often the most important light-absorbing impurity in surface snow because of its strong absorption of solar radiation. It can decrease snow surface albedo and results in positive radiative forcing (Warren and Wiscombe, 1980; Clarke and Noone, 1985; Hansen and Nazarenko, 2004; Hadley and Kirchstetter, 2012). We use the SNICAR-online model (available at http://snow.engin.umich.edu/; Flanner et al., 2007) to estimate radiative forcing induced by the observed BC and OC as if they were present in snow. Detailed description of the SNICAR model was given by Flanner and Zender (2005; 2006) and Flanner et al. (2007). A mass absorption cross-section (MAC) of 7.5 $m^2 g^{-1}$ at 550 nm for uncoated BC particle and 0.6 $m^2 g^{-1}$ for OC (Bond and Bergstrom, 2006; Kirchstetter et al., 2004) is assumed, and the MAC scaling factor of 1 within spectral broadband for BC and 0.08 for OC is used. According to the previous studies (Cuffey and Paterson, 2010; Wiscombe and Warren 1980) and measure-

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ments in Qiyi glacier and Zuoqiupu glacier, an effective radius of 100 µm with density of 60 kg m^{-3} for new snow, and the effective radius of 400 µm with density of 400 kg m^{-3} for aged snow are adopted for the forcing calculation. As we focus on the estimation of radiative forcing by carbonaceous particles, other impurity contents, such as dust and volcanic ash, are set to be zero. Based on the measured annual mean BC and OC concentrations, the in-snow BC and OC radiative forcing is calculated using SNICAR-online. The annual mean BC and OC concentrations during 1956–1979 is 4.4 and 13.8 ng g^{-1}, respectively, and they increase to 12.5 and 61.3 ng g^{-1} in 2006. As a consequence, the annual mean radiative forcing derived from BC (OC) increases from 0.75 (0.20) to 1.95 (0.84) W m^{-2}. Although BC concentration is one order of magnitude lower than OC, radiative forcing of BC is about two times larger than OC due to its strong absorption of solar radiation.

Since BC and OC concentrations increased rapidly from 1980, the radiative forcing rises as a consequence. The average BC radiative forcing had increased 43 % after 1980, and OC radiative forcing had an increase of 70 %. Because of the stronger increasing trend in OC than BC during 1990–2006 (Fig. 6), the contribution of OC to the total radiative forcing cannot be neglected.

4 Summary and conclusions

Light-absorbing carbonaceous aerosols can induce significant warming in the atmosphere and in snow and glaciers, which likely accelerates the melting of glaciers over Himalayas and Tibetan Plateau. Ice-core measurement of carbonaceous aerosols is a useful mechanism for challenging historical emission inventories and revealing long-term changes in anthropogenic aerosols and their impacts on local climate. In this study, we use an ice core (97 m in depth and 9.5 cm in diameter) retrieved from the Zuoqiupu glacier (96.92° E, 29.21° N, 5600 m a.s.l.) in the southeastern Tibetan Plateau to reconstruct the history of atmospheric deposition of carbonaceous aerosols in this region. The glacier has a unique geographical location that is in close proximity to ma-

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jor Asian emission sources. With the help of a global climate model (CAM5) in which black carbon (BC) emitted from different source regions can be explicitly tracked, we are able to characterize BC source-receptor relationships between four Asian source regions (i.e., South Asia, East Asia, Southeast Asia and Central Asia) and the southeastern Tibetan Plateau as a receptor. We also estimate the radiative forcing in snow due to BC and OC over the southeastern Tibetan Plateau using the ice core measurements and an offline snow-ice-aerosol-radiation model (called SNICAR).

BC and OC concentrations in small segments of the Zuoqiupu ice core were measured using a thermal-optical method. Ice core dating based on significant seasonal variations of oxygen isotope ratios ($\delta\ ^{18}O$) was used to construct the time series of BC and OC concentrations, which turned out to span the time period of 1956–2006. Not only do the concentrations of OC and BC in the ice core exhibit significant differences between the summer monsoon and non-monsoon seasons, but also the ratio of OC to BC shows a clear seasonal dependence that might be due to seasonal change in sources and/or atmospheric deposition processes. The CAM5 results show a similar seasonal dependence in the concentrations and the OC / BC ratio.

The CAM5 model simulation indicates distinct circulation patterns during summer monsoon (June–September) and non-monsoon (October–May) seasons. Both circulation patterns and seasonal changes in emissions influence the seasonal deposition of aerosol at the Zuoqiupu site. The CAM5 simulation with tagged BC regional sources shows that South Asia is the dominant contributor (~81 %) to the 10 yr mean BC deposition at the Zuoqiupu site during the non-monsoon season with ~14 % from East Asia, while the contribution of East Asia (~56 %) is larger than that of South Asia (~39 %) during the monsoon season. For the annual mean BC deposition, South Asia (~75 %) is the biggest contributor, followed by East Asia (~21 %).

The annual mean BC and OC deposition fluxes into the ice core are also estimated to explore the interannual variations and long-term trends. Results show stable and relatively low BC and OC fluxes from late 1950s to 1979, followed by a steady increase through the mid-1990s. A more rapid increase occurred after the minimum in 2002.

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The BC and OC fluxes in 2006 were two and three times the respective average fluxes before 1980.

The overall increasing trend in deposition fluxes since 1980 is consistent with the BC and OC emissions in South Asia as the major contributor. Moreover, the increasing trend of OC / BC ratio since early 1990s indicates a growth of the contribution of coal combustion and biomass burning to the carbonaceous aerosol emissions in the major contributing source regions, which is also supported by the trends in the consumption of coal, oil and biofuels in South Asia.

Our offline calculation using the SNICAR model shows an increase of radiative forcing induced by the observed BC and OC in snow, with an average increase of 48 % since 1980. The rapid increase in radiative forcing has implications for the Tibetan glacier melting and availability of water resources in the surrounding regions, and more attention to OC is merited because of its non-negligible light absorption and the recent rapid increases evident in the ice core record.

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Table 1. Source regions (South Asia, East Asia, Southeast Asia, and Central Asia) and corresponding fractional contributions (%) to BC deposition flux at the Zuoqiupu site in monsoon (June–September), non-monsoon (October–May), and all months from 1996–2005.

Source Regions	Latitude	Longitude	Monsoon	Non-monsoon	Annual
South Asia	5–35° N	50–95° E	38.51	81.26	74.48
East Asia	15–50° N	95–150° E	56.24	13.91	20.66
Southeast Asia	0–15° N	95–130° E	0.05	0.16	0.15
Central Asia	35–50° N	50–95° E	2.62	0.86	1.14

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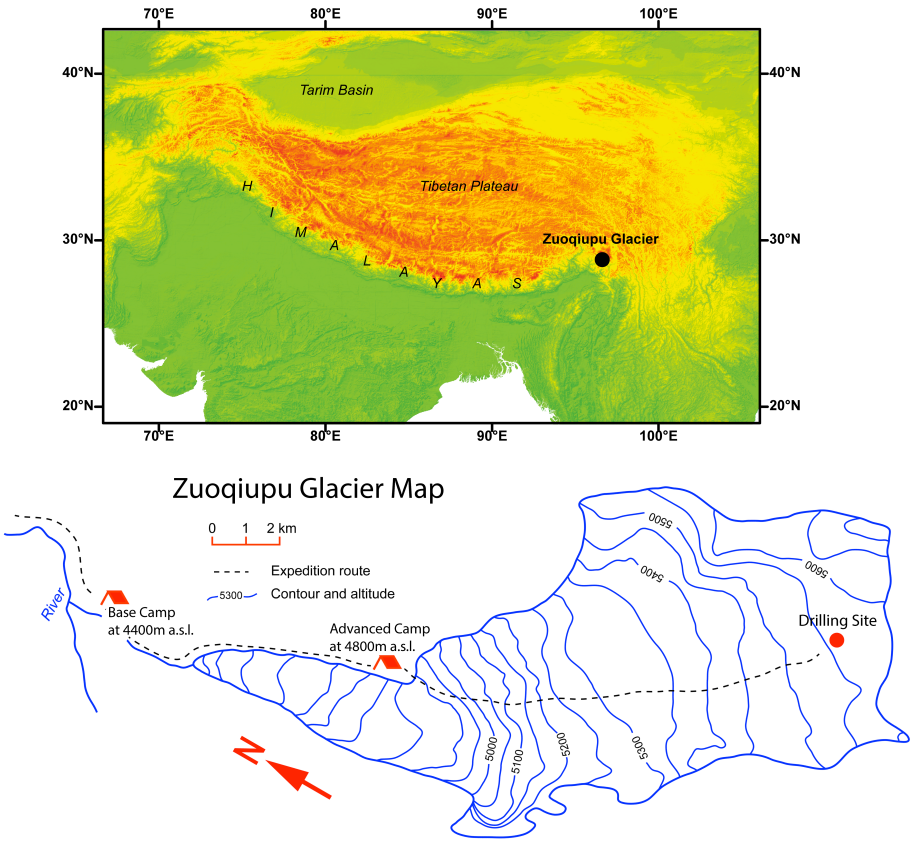

Figure 1. Site location of Zuoqiupu Glacier (top): black circle represents the location of Zuoqiupu Glacier and the warm colors illustrate the high elevations of Tibetan Plateau. Detailed elevation contours of the Zuoqiupu Glacier are shown in the bottom pannel. Red circle illustrates the ice core drill site.

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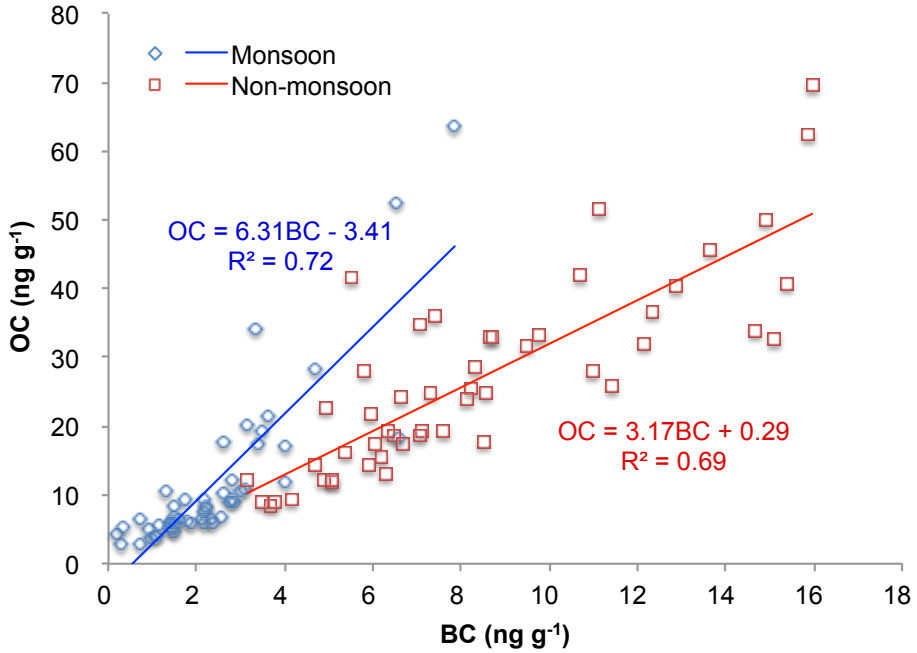

Figure 2. Scatter plots for OC and BC concentrations and corresponding linear regressions for the monsoon and non-monsoon seasons obtained from the ice core measurements

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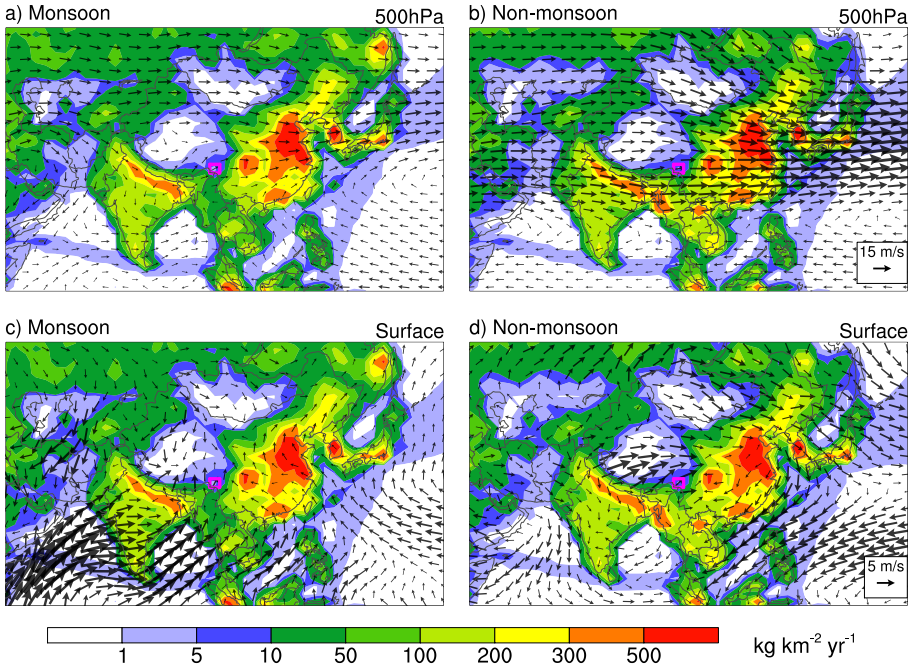

Figure 3. 10 yr (1996–2005) mean wind vectors (denoted by arrows) at 500 hPa (top) and the surface (bottom) during summer monsoon (June–September; left) and non-monsoon season (October–May; right) from MERRA reanalysis datasets being used in the CAM5 simulation. The background colors show mean BC emission rates based on the IPCC present-day scenario for the corresponding months. The pink square indicates the model gridcell in which the ice core drill site resides.

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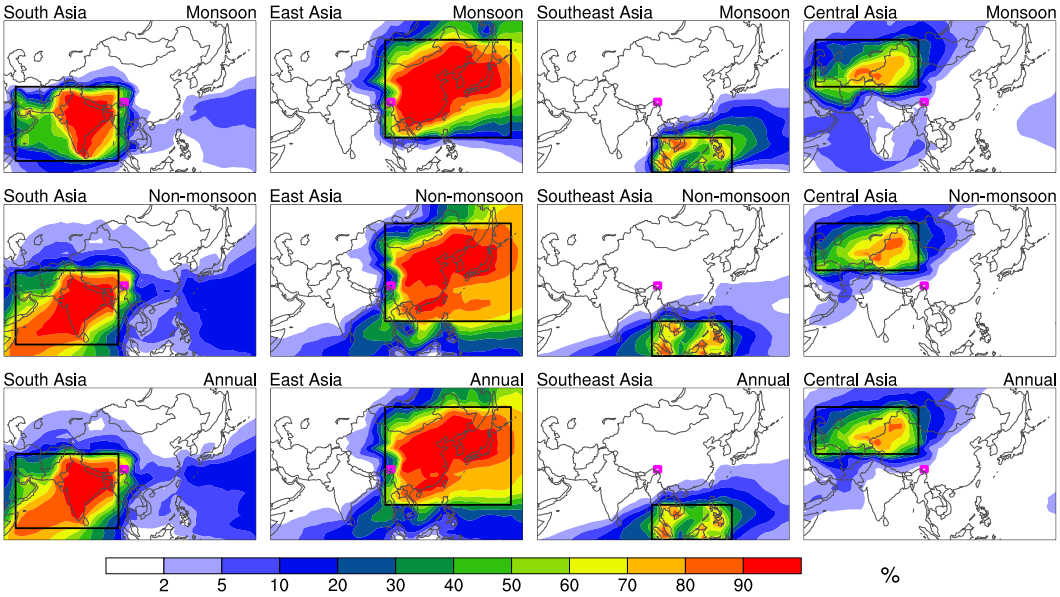

Figure 4. Spatial distributions of fractional contribution from the four source regions (South Asia, East Asia, Southeast Asia, and Central Asia) to monsoon and non-monsoon, and annual mean BC deposition fluxes during 1996–2005. The black boxes are the boundary of source regions, and the small pink box is the model gridcell where the Zuoqiupu glacier sampling site is located.

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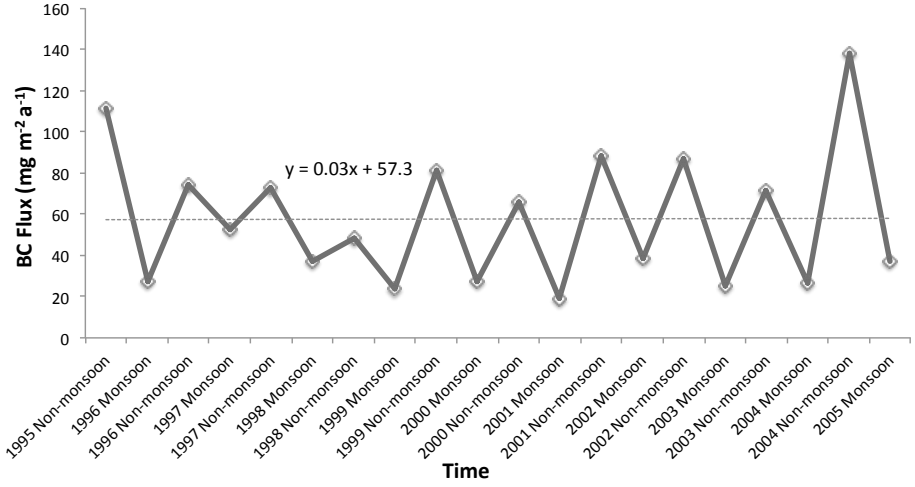

Figure 5. Seasonal dependence of BC deposition flux at the Zuoqiupu site from 1995 to 2005 simulated in CAM5. The dash line represents a linear regression of all data points.

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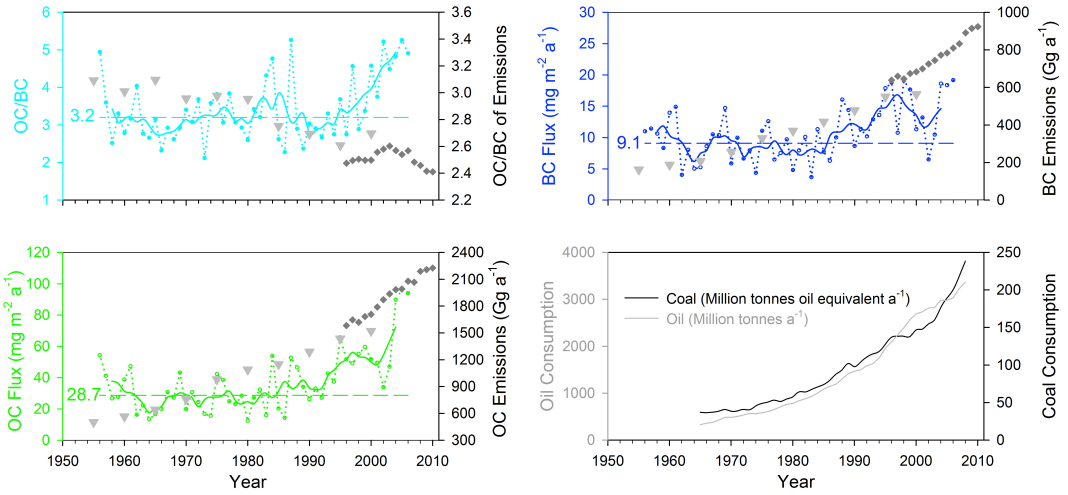

Figure 6. Time series of annual (dotted line with circles) and 5 yr averaged (solid line) OC / BC ratios (top, left), BC (top, right) and OC deposition fluxes (bottom, left) in the Zuoqiupu ice core for the time period of 1956–2006. The averages of OC / BC ratio, BC and OC during 1956–1979 are denoted by dashed lines with numbers. BC and OC emissions in South Asia (Bond et al., 2007) and corresponding BC/OC ratios are illustrated with gray triangles, and with gray diamonds for emissions in India (Lu et al., 2011). Coal and oil consumption data are also showed with black and gray lines respectively (bottom, right) (BP Group, 2009).

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