Reevaluating black carbon in the Himalayas and the Tibetan Plateau: concentrations and deposition

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Abstract: Black carbon (BC) is the second most important warming component in the atmosphere after CO_2. The BC in the Himalayas and the Tibetan Plateau (HTP) has influenced the Indian Monsoon and accelerated the retreat of glaciers, resulting in serious consequences for billions of Asian residents. Although a number of related studies have been conducted in this region, the BC concentrations and deposition rates remain poorly constrained. Because of the presence of arid environments and the potential influence of carbonates in mineral dust (MD), the reported BC concentrations in the HTP are overestimated. In addition, large discrepancies have been reported among the BC deposition derived from lake cores, ice cores, snowpits and models.

Therefore, the actual BC concentration and deposition values in this sensitive region must be determined. A comparison between the BC concentrations in acid (HCl)-treated and untreated total suspected particle samples from the HTP showed that the BC concentrations previously reported for the Nam Co station (central part of the HTP) and the Everest station (northern slope of the central Himalayas) were overestimated by approximately 52±35% and 39±24%, respectively, because of the influence of carbonates in MD. Additionally, the organic carbon (OC) levels were overestimated by approximately 22 ± 10% and 22 ± 12% for the same reason. Based on previously reported values from the study region, we propose that the actual BC concentrations at the Nam Co and Everest stations are 61 ng m^{-3} and 154 ng m^{-3}, respectively. Furthermore, a comprehensive comparison of the BC deposition rates obtained via different methods indicated that the deposition of BC in HTP lake cores was mainly related to river sediment transport from the lake basin as a result of climate change (e.g., increases in temperature and precipitation) and that relatively little BC deposition occurred via atmospheric deposition. Therefore, previously reported BC deposition rates from lake cores overestimated the atmospheric deposition of BC in the HTP. Correspondingly, BC deposition derived from snowpits and ice cores agreed well with those derived from models, implying that the BC depositions of these two methods reflect the actual values in the HTP. Therefore, based on reported values from snowpits and ice cores, we propose that the BC deposition in the HTP is 17.9±5.3 mg m^{-2} a^{-1}, with higher and lower values appearing along the fringes and central areas of the HTP, respectively. These adjusted BC concentrations and deposition values in the HTP are critical for performing accurate evaluations of other BC factors, such as atmospheric distribution, radiative forcing and chemical transport in the HTP.

Key words: the Himalayas and the Tibetan Plateau; black carbon; concentration; deposition; glacier region

1 Introduction

The Himalayas and the Tibetan Plateau (HTP) region is the highest mountain-plateau system in the world and is the source of approximately ten large rivers in Asia.

This region is also sensitive to climate change (Bolch et al., 2012; Kang et al., 2010; You et al., 2010). Black carbon (BC) in and around the HTP have been found to play key roles in climate change patterns in the HTP and Asia, including causing atmospheric warming (Lau et al., 2010; Ramanathan and Carmichael, 2008; Xu et al., 2016), promoting HTP glacial retreat (Qu et al., 2014; Xu et al., 2009b), altering monsoon system evolution (Bollasina et al., 2008) and affecting the fresh water supplies of billions of residents across Asia. To date, numerous studies have been conducted on the BC concentrations in the atmosphere (Cong et al., 2015; Marinoni et al., 2010; Ming et al., 2010; Wan et al., 2015; Zhao et al., 2013b) and atmospheric BC deposition as determined from lake core sediments (Cong et al., 2013; Han et al., 2015). However, all of these studies exhibit limitations because of certain special environmental factors in the HTP (e.g., high concentrations of mineral dust (MD) in aerosols and catchment inputs to lake core sediment). Therefore, the above studies should be reinvestigated to better define the actual BC values in the HTP. In this article, we discuss the actual concentrations and deposition of BC in the HTP in order to present basic input data for other important studies on the sources, radiative forcing patterns and chemical transport of BC in this region.

At present, the thermal-optical method is a widely used method for measuring BC concentrations in aerosols from the HTP (e.g., Cong et al., 2015; Li et al., 2016d; Ming et al., 2010; Zhao et al., 2013b). An important factor influencing the accurate measurement of BC concentrations via this method is the presence of carbonates (inorganic carbon (IC)) in MD. IC can also emit CO_2 in response to increasing temperature during measurements, thus causing an overestimation of the total carbon (TC) in carbonaceous aerosols (CAs) (Karanasiou et al., 2010). Hence, IC is generally excluded in CA studies (Bond et al., 2013). However, few studies of the HTP have considered the contributions of IC to TC and BC because one study concluded that IC can be neglected in studies of the TC and BC in mid-latitude aerosols because the IC exists at far lower concentrations relative to TC and BC (Chow and Watson, 2002).

This conclusion cannot be blindly applied to other areas because of the complexities of mid-latitude environments around the world (e.g., arid areas and deserts

with intense dust storm events). For example, previous studies in Xi'an, Middle west and Northeast China showed that IC accounts for approximately 8% (Cao et al., 2005) to 10% (Ho et al., 2011) of the TC in particles with diameters less than 2.5 μm ($PM_{2.5}$) during dust storm events. Similar phenomena have also been found for both $PM_{2.5}$ and total suspended particle (TSP) samples in southern Europe (Perrone et al., 2011; Sillanpää et al., 2005). Because TSP samples contain more MD and carbonates than $PM_{2.5}$, they should have higher concentrations of IC.

The above phenomenon should also be taken into consideration in the study of CAs of the HTP. Similar to northern China, large sand dunes and deserts are widely distributed across the western HTP (Liu et al., 2005), and dust storms occur frequently in winter and spring (Wang et al., 2005). Thus, IC may account for a large portion of the CAs in the HTP. Unfortunately, the potential contributions of IC to the TC and BC in HTP aerosols have been overlooked (e.g., Aiken et al., 2014; Cao et al., 2010; Cong et al., 2015; Li et al., 2016b; Ming et al., 2010; Wan et al., 2015; Zhao et al., 2013b). Additionally, IC contributions may be high because almost all of the reported data on CAs are based on the TSP content, which includes large volumes of coarse particles derived directly from MD. Therefore, the TC and BC concentrations in the HTP are likely overestimated. In fact, some published articles on aerosols collected from remote areas of the HTP have identified MD components (Cong et al., 2015; Zhao et al., 2013b), although none of these studies have directly discussed this issue or evaluated the effects of IC (Fig. 1).

Because MD has lower influences on light than BC in the atmosphere (Bond and Bergstrom, 2006; Clarke et al., 2004) and on glacier surfaces (Qu et al., 2014), considering IC as BC will overestimate the BC-driven climate forcing. Organic carbon (OC) is generally considered to scatter sunlight. However, some components of OC also absorb sunlight and warm the atmosphere (Andreae and Gelencser, 2006). Therefore, the contributions of IC to the OC and BC values in the HTP aerosols must be quantitatively evaluated. In this study, TSP samples from two remote stations in the HTP were collected to evaluate the contributions of IC to the TC and BC. Additionally, seasonal variations in the extent of the overestimations of TC and BC and possible

causes were also examined. Finally, previously published TC and BC concentrations at these two stations were adjusted (Cong et al., 2015; Zhao et al., 2013a).

BC deposition is closely related to the BC transport processes, lifetime and radiative forcing. Depositional value can be measured from historical media, such as sediments (Gustafsson and Gschwend, 1998; Han et al., 2016) and ice cores (Ming et al., 2007; Ruppel et al., 2014), estimated from BC concentrations in the atmosphere (Jurado et al., 2008) or calculated using models (Zhang et al., 2015). At present, the BC deposition process remains poorly quantified in the HTP because of its complex terrain and dynamic regimes (Bauer et al., 2013; Bond et al., 2013). Thus far, only three studies have directly reported on BC deposition in the HTP. One model indicated that the BC deposition in the central HTP was 9 mg m^{-2} a^{-1} (Zhang et al., 2015), which is approximately thirty times lower than the values measured in lake cores at Nam Co and Qinghai lakes (270-390 mg m^{-2} a^{-1}) (Fig. 1) (Cong et al., 2013; Han et al., 2011). Although considerable uncertainties exist in atmospheric BC deposition estimated from models (Bond et al., 2013; Koch et al., 2009) and lake core sediments (Cohen, 2003; Yang, 2015), these large differences need to be thoroughly investigated.

For instance, although the influence of sediment focusing on BC deposition in lake cores has been noted in other areas (Blais and Kalff, 1995; Yang, 2015), it has not been pointed out and evaluated in the HTP. Consequently, adopting and correcting for this process might result in incorrect data and explanations. Therefore, additional studies must be performed to provide more reliable BC deposition values. For instance, other researchers have reported BC concentrations and water accumulation rates in ice cores and snowpits from the HTP (Fig. 1) (Li et al., 2016a; Li et al., 2016c; Ming et al., 2008; Xu et al., 2009b). Although these studies did not report BC deposition values directly, BC deposition rates could be easily calculated from the data reported in those articles. Because the cols of glaciers where the snow and ice samples were collected are generally located at the highest altitudes of a given region, BC is only deposited via wet and dry deposition from the atmosphere. Therefore, these data need to be comprehensively evaluated.

Notably, some uncertainties exist in the comparison of BC data among different

studies. Despite recent technological achievements, accurately measuring BC concentrations in ambient samples remains a challenge in atmospheric chemistry research (Andreae and Gelencser, 2006; Bond et al., 2013; Lim et al., 2014). Because the methods used to measure BC concentrations and determine BC deposition levels are not the same, uncertainties will be introduced when directly comparing the results from different studies. For instance, different thermal-optical methods with different temperature increase protocols (e.g., NIOSH vs. IMPROVE vs. EUSAAR_2) will produce different BC concentrations for the same sample (Andreae and Gelencser, 2006; Karanasiou et al., 2015). In general, BC concentrations derived from the IMPROVE method are 1.2-1.5 times higher than those derived from the NIOSH method (Chow et al., 2001; Reisinger et al., 2008), and BC concentrations from the EUSAAR_2 temperature protocol are approximately twice as high as those derived from the NIOSH protocol (Cavalli et al., 2010). Furthermore, lake core samples need to be pretreated with HCl and HF several times prior to measurements with the thermal-optical methods (Han et al., 2015). However, because of the complex chemical properties of ambient samples, the "best" thermal-optical protocol has not been identified (Karanasiou et al., 2015), and an exact ratio for BC produced from different methods is difficult to determine. Therefore, although the direct comparison of BC concentrations and deposition levels across different studies presents certain uncertainties in this study, the comparison between lake core and snowpit data is still reliable. For instance, although large uncertainties exist for BC concentrations within the same environmental matrix (Hammes et al., 2007; Han et al., 2011; Watson et al., 2005), the similarity of the BC deposition values among different glaciers (Table 1) in different studies implies that comparing BC deposition data is feasible for the glacial region in the HTP. In addition, because BC concentrations measured via the SP2 method are far lower than those measured via thermal–optical methods (Lim et al., 2014) (the former can only measure BC in grain size finer than 500 nm (Kaspari et al., 2011)), SP2-based BC data were avoided in this study. Furthermore, BC concentrations among different methods have been found to vary by up to a factor of 7 (Watson et al., 2005). Accordingly, the BC deposition in the studied lake core (Nam Co) in this study is estimated to be 20 times

higher than those in snowpit and ice core studies in the HTP, providing strong evidence of their differences.

2 Methods

2.1 Collection of aerosols, surface soils and river sediments

TSP samples were collected from the Nam Co Station for Multisphere Observation and Research and the Qomolangma Station for Atmospheric and Environmental Observation and Research (Everest station) (Fig. 1) from 2014 to 2016. The Nam Co station is located in the center of the HTP. The Everest station is located on the northern slopes of the Himalayas. Both of these two stations are generally considered to be located in remote areas of the HTP that receive BC transported over long distances from South Asia, and several BC studies have been conducted in these areas (Chen et al., 2015; Cong et al., 2015; Li et al., 2016a; Ming et al., 2010). In detail, TSP samples were collected using 90 mm pre-combusted (550°C, 6 hours) quartz fiber filters (Whatman Corp) with a vacuum pump (VT 4.8, Germany). Because the pump was not equipped with a flow meter, the air volumes passing through each filter could not be determined (Li et al., 2016d); however, this did not influence the objectives of this study (e.g., relative concentrations of TC and BC in the original and acid-treated samples). Four field blank filters were also collected from each station by exposing the filters in each sampler without pumping.

To compare the BC concentrations of the Nam Co Lake cores, two surface soil samples and four suspended particle samples from four rivers in the Nam Co Basin were collected during a period of peak river flow in 2015. The <20 μm fraction of these samples was extracted (Li et al., 2009) and treated (Han et al., 2015) to measure the BC concentrations. In addition, ten surface soil samples around the Everest station were collected to study the pH values.

2.2 Measurement of BC and elemental concentrations

The carbonates of the collected aerosol samples were removed via a fumigation process involving exposing a subset of samples to a vapor of 37% hydrochloric acid (HCl) for 24 hours. Then, the treated samples were held at 60°C for over 1 hour to remove any acid remaining on the filter (Bosch et al., 2014; Chen et al., 2013; Li et al.,

2016a; Pio et al., 2007). The OC and elemental carbon (EC, the common chemical/mass definition of BC) concentrations of both the original and treated samples were measured using a Desert Research Institute (DRI) Model 2001 Thermal/Optical Carbon Analyzer (Atmoslytic Inc., Calabasas, CA, USA) following the IMPROVE-A protocol (Chow and Watson, 2002). The OC and BC concentrations were determined based on varying transmission signals. To investigate the BC concentration measured by different methods, sixteen acid-fumigated aerosol samples were measured following the EUSAAR_2 and NIOSH protocols for comparison with the results of the IMPROVE protocol. The results showed that the TC concentrations of three methods for the same sample were similar, as suggested by previous research (Chow et al., 2001). The ratios of $BC_{(IMPROVE)}/BC_{(NIOSH)}$ and $BC_{(EUSAAR\_2)}/BC_{(NIOSH)}$ for the studied samples were 1.36 ± 0.35 and 1.88 ± 0.60, respectively, both of which agreed with the previously proposed ratios of 1.2-1.5 (Chow et al., 2001; Reisinger et al., 2008) and 2 (Cavalli et al., 2010), respectively. To evaluate the concentrations of MD, the concentrations of Ca, Fe, Al, and Ti in the aerosol samples were measured by inductively coupled plasma optical emission spectroscopy (ICP-OES) following method of Li et al., (2009). All the reported values in this study were corrected based on the values of the blanks. The contributions of MD (Maenhaut et al., 2002) and CA (Ram et al., 2010) of the collected samples were calculated using the following equations:

$$MD = (1.41 \times Ca + 2.09 \times Fe + 1.9 \times Al + 2.15 \times Si + 1.67 \times Ti) \times 1.16 \quad (1)$$

where Si is calculated from Al assuming an average ratio of Si/Al=2.5 (Carrico et al., 2003), and

$$CA = OC \times 1.6 + BC \quad (2)$$

2.3 Adoption and calculation of BC deposition data

To determine the actual BC deposition in the HTP, previously reported data were compiled and evaluated (Table 1). In addition, BC deposition rates from the Nam Co station and Qinghai Lake Basin were estimated from the average BC concentrations in the atmosphere and average precipitation levels using the method described in detail in other studies (Fang et al., 2015; Jurado et al., 2008) (Table 2). In brief, the annual atmospheric deposition rate of BC (μg m^{-2} a^{-1}) was calculated as follows:

$$F_{BC} = F_{DD} + F_{WD} \quad (3)$$

$$F_{DD} = 7.78 \times 10^4 \cdot V_D \cdot C_{BC\text{-}TSP} \quad (4)$$

$$F_{WD} = 10^{-3} \cdot P_0 \cdot W_p \cdot C_{BC\text{-}TSP} \quad (5)$$

where F_{DD} and F_{WD} are the seasonal dry and wet deposition (μg m^{-2}), respectively; V_D, P_0 and W_p are the dry deposition velocity of aerosol (0.15 cm s^{-1}), the precipitation amount (mm) in a given season and the particle washout ratio (2.0×10^5), respectively (Fang et al., 2015); and $C_{BC\text{-}TSP}$ is the BC concentration of the TSPs (μg m^{-3}). The seasonal BC concentrations at the Nam Co station were monitored with an AE-31, and the average precipitation levels at the station were recorded from 2014-2015. The BC concentrations in Qinghai Lake are reported in Zhao et al., (2015), and the average 1961-2010 precipitation levels recorded by the China Meteorological Administration from the Huangyuan station in the lake basin were used. The values used in the BC deposition calculations for these two areas are shown in Table 2.

3 Results and discussion

3.1 Actual BC concentrations in the atmosphere over the HTP

3.1.1 Contribution of carbonate carbon to both TC and BC

In this study, after compared with BC and OC concentrations of original and acid treated TSP samples, we found that carbonate carbon significantly contributes to the BC, TC and OC concentrations of the aerosols at the Nam Co and Everest stations, especially during non-monsoon periods (winter and spring) when dust storms occur frequently at the Nam Co station. The ratios of the TC, OC and BC levels of the aerosols treated with acid (TC_A, OC_A and BC_A) to those of the original samples (TC_O, OC_O and BC_O) were 0.81 ± 0.13, 0.78 ± 0.10 and 0.48 ± 0.35, respectively, for the Nam Co station and 0.76 ± 0.12, 0.78 ± 0.12 and 0.61 ± 0.24, respectively, for the Everest station. Meanwhile, because of heavy precipitation during monsoon period, influence of IC to both BC and TC during this time were lower than those of non-monsoon period at two studied stations (Fig. 2). As proposed in previous work (Chow and Watson, 2002), BC concentrations are more heavily influenced than OC and TC concentrations because carbonates are more prone to decomposition at high temperatures during OC and BC

analyses. The OC concentrations in the treated samples used in this study also decreased, indicating that carbonates can also decompose at low temperatures (Karanasiou et al., 2010). Clear seasonal variations, i.e., low TC_A/TCo ratios during non-monsoon periods and high TC_A/TCo ratios during monsoon periods, were observed in the aerosols at the Nam Co station (Fig. 2). This pattern is consistent with the intense dust storms that occur during non-monsoon periods. However, clear seasonal patterns in the TC_A/TCo ratio at the Everest station were not observed, in accordance with the relatively stable seasonal variations in the Ca^{2+} content recorded at this station (Cong et al., 2015). To evaluate the relative ratio of MD and CA, MD/(MD+CA) values were calculated (Fig. 3). The MD/(MD+ CA) levels recorded at the Nam Co station during non-monsoon periods were significantly higher than those recorded during monsoon periods ($p<0.01$), whereas the corresponding values at the Everest station were not significantly different between the two periods ($p>0.05$) (Fig. 3). Compared with those of other areas, the MD/(MD+ CA) values recorded at the two stations were higher than those recorded at the NCO-P station (27.95°N, 86.82°E, 5079 m.a.s.l) (70% and 73% for the pre-monsoon and monsoon periods, respectively) located on the southern slope of the Himalayas (Decesari et al., 2010). This difference may be related to the serious levels of South Asian pollutants at the NCO-P station and the relative ease with which polluted clouds are transported to this station. However, because the measured particle size (PM_{10}) and the measurement methods of Ca, Mg and EC at the NCO-P station differed from those in this study, uncertainties exist in such a direct comparison.

The Everest station is located in a dry river valley with sparse vegetation cover (a typical barren site), and the MD derived from the local surface soil contributes considerably to aerosols collected during monsoon periods (Liu et al., 2017). However, the Nam Co station is located in a typical grassland region with limited amounts of locally sourced dust during monsoon periods. Additionally, the Everest station is located in the rain shadow of the Himalayas; thus, the precipitation level recorded at the Everest station (172 mm during the monsoon period between 2014 and 2015) is much lower than that of the Nam Co station (258 mm), causing high MD concentrations in the atmosphere of the Everest station during that period. Potential carbonate-induced

biasing of aerosol samples has been proposed to occur in arid areas with alkaline soils (Chow and Watson, 2002). Because of the dry weather conditions, the pH values of the soil around the Nam Co and Everest stations are as high as 8 (Li et al., 2008) and 8.3, respectively, implying considerable carbonate contributions. During non-monsoon periods, MD is mainly transported by westerlies from the arid western HTP, where MD is distributed across large deserts with sand dunes; thus, the aerosol samples were influenced by MD with high concentrations of carbonates. Finally, the significant positive relationship ($p<0.01$) between Ca and IC (TC_O-TC_A) for the aerosols of these two stations further demonstrates the contributions of $CaCO_3$ to aerosol IC (Fig. 4). The ratio of Ca/IC was higher in the Everest station samples than that of Nam Co station, possibly reflecting different types of carbonate at these two stations.

3.1.2 Actual BC concentrations at the two stations and implications

In summary, we clearly showed that the presence of carbonates in MD led to the TC levels in TSP samples in the HTP being overestimated by approximately 19±13% and 24±12% at the Nam Co and Everest stations, respectively. These overestimates were higher than the corresponding value of 10% found for coarse particles in the central Mediterranean region of Europe (Perrone et al., 2011). In addition, the related BC values were overestimated by approximately 52±35% and 39±24%, respectively, thus implying that the actual BC concentrations at these two stations were lower than previously reported values. Although fumigation with HCl can cause the loss of volatile organic acids in treated samples (Chow et al., 1993), this potential influence is not important because of the significant relationship between TC_O-TC_A and Ca (Fig. 4). Moreover, because of the large variations in the above values, the corrected BC concentrations at the two stations have large uncertainties. Therefore, based on previously reported BC concentrations measured via the same method as in this study (Cong et al., 2015; Zhao et al., 2013a), the actual BC concentrations at the Nam Co and Everest stations were estimated to be 61 ng m^{-3} and 154 ng m^{-3}, respectively.

Carbonates can decompose at relatively low temperatures during measurement, leading to overestimation of both BC and OC concentrations (Karanasiou et al., 2010). In addition, sometimes the acid-treated ambient samples transfer some components of

OC to BC, leading to higher BC concentrations (Jankowski et al., 2008). However, this phenomenon was not common in the aerosol samples examined in this study, although several samples from both stations showed higher BC concentrations in the acid-treated samples (Fig. 2). Because BC_A cannot be higher than BC_O, the samples with BC_A/BC_O values greater than one were not included in the above calculations. Nevertheless, the ratio of BC_A/BC_O was considered to be slightly overestimated as some portion of OC was considered BC in the acid-treated samples (Jankowski et al., 2008).

Since the influence of carbonate carbon on TC has been observed in $PM_{2.5}$ samples from Qinghai Lake, Northwest China (Zhao et al., 2015), this phenomenon should be clear in the TSP samples in this study. Because dust storms in the northern and western parts of the HTP are more severe than those near the two studied stations during the non-monsoon periods, the effect of carbonates on the concentrations of OC and BC should be more pronounced in such areas and must be seriously considered in future studies. Therefore, the overestimation of BC values is likely greater in the northern and western parts of the HTP than near Nam Co, as we noted previously. MD concentrations have been shown to be much higher than BC concentrations in snow and ice core samples from the HTP (Li et al., 2017; Qu et al., 2014). However, numerous studies have measured BC concentrations without using an acid pretreatment step (e.g., Ming et al., 2009; Qu et al., 2014; Wang et al., 2015; Xu et al., 2009a; Xu et al., 2009b). Therefore, the contribution of carbonates in MD to the BC concentrations in snow and ice core samples is likely considerable and needs to be quantitatively evaluated in a future study. Similarly, related HTP studies on other issues, such as BC radiative forcing and atmospheric transport models, based on in situ BC concentrations must be adjusted.

3.2 Actual BC deposition in the HTP

3.2.1 Overestimated BC deposition in lake cores from the HTP

In general, the BC deposition levels measured via different methods should be consistent for a given region. For instance, in the severely polluted region of eastern China (Chen et al., 2013; Yan et al., 2015), the BC deposition rate recovered from a Chaohu lake core was 1,660 mg m^{-2} a^{-1} (Han et al., 2016), which was close to the values of northern China calculated from the BC concentrations in aerosols (Fang et al., 2015)

and determined via in situ monitoring on the Northern China Plain (Tang et al., 2014) (Table 1). However, this consistency was not the case in the HTP, where large discrepancies were found among the reported HTP BC deposition values. Catchment inputs have been shown to significantly influence the chemical deposition values reconstructed from lake cores (Yang, 2015). For instance, BC deposition rates derived from lake cores of Nam Co Lake (NMC09) and Qinghai Lake were 260 mg m^{-2} a^{-1} and 270-390 mg m^{-2} a^{-1}, respectively, which were much higher than those derived from ice core and snowpit samples from the HTP (Fig. 5, Table 1). We proposed that the BC deposition in the lake cores of Qinghai Lake mainly reflected atmospheric deposition followed by catchment inputs. However, the NMC09 value of Nam Co Lake was mainly influenced by catchment inputs.

Lake core-derived BC deposition in Qinghai Lake was only 2-3 times higher than that estimated from the BC concentrations of $PM_{2.5}$ in the atmosphere (Zhao et al., 2015). Because $PM_{2.5}$ does not include all particles in the atmosphere, the actual BC concentration in the atmosphere should be higher than that of $PM_{2.5}$ (Li et al., 2016b; Viidanoja et al., 2002); therefore, the atmospheric BC deposition should be more similar to that of a lake core. In addition, a previous study showed that approximately 65% and 22% of the surface sediments in Qinghai Lake result from atmospheric deposition and catchment inputs (Wan et al., 2012), respectively, further demonstrating the significant effects of atmospheric deposition on lake core sediments. Therefore, if the BC deposition from atmospheric particles and that of lake core are the same, then the atmospheric BC deposition based on Qinghai Lake core data is overestimated by approximately 35%.

Correspondingly, catchment inputs account for a large proportion of the NMC09 samples. BC is widely distributed throughout environmental materials (e.g., soil and river sediments) because of its inert characteristics (Bucheli et al., 2004; Cornelissen et al., 2005). Therefore, river inputs contribute sediments as well as BC to lakes. For instance, in the Nam Co Basin, BC concentrations within the <20 μm fraction of surface soil and sediment reach 0.78±0.48 mg g^{-1}, which is close to the Nam Co Lake core concentration of 0.74 mg g^{-1} (Cong et al., 2013). In addition, several findings have

demonstrated the contributions of catchment inputs to Nam Co Lake cores because of focusing factor, which was shown in the following sections.

First, a large glacial area (141.88 km^2) is present within the Nam Co Basin (Fig. 5), and large volumes of glacier meltwater and sediment flow into the lake annually (Wu et al., 2007). Due to recent increasing temperatures and precipitation in the Nam Co Basin, glacier meltwater accounts for approximately 50.6% of the lake's volume, which has increased over the last 30 years (Zhu et al., 2010). Originating at high-elevation glacier terminal, these rivers flow are at a steep angle, and large volumes of suspended allochthonous sediments are transported into Nam Co Lake annually (Doberschütz et al., 2014). A similar phenomenon was also observed in lake cores of a glacier-fed lake as a result of glacier meltwater effects (Bogdal et al., 2011). Second, previous studies on the accumulation rates in lake cores have revealed significant contributions of riverine particles. The accumulation rates in a Nam Co Lake core (NMC 08-1) are consistent with the precipitation variations recorded in the Nam Co Basin during the last 60 years (Fig. 5A) (Wang et al., 2011), indicating that heavy precipitation promotes the transport of large riverine particles to the lake, thus increasing the accumulation rates in the lake cores. Interestingly, the mean grain size of the lake core (NMC09) that reported BC atmospheric deposition showed a significant positive relationship with precipitation (Fig. 5B), thus reflecting the same relationship between catchment inputs and lake core accumulation rates (Li et al., 2014). Because these two lake cores were drilled from different sites (Fig. 5), their similar catchment input characteristics reflect a common feature of Nam Co sediment. As shown above, the BC concentrations in the fine fraction of the river sediments are nearly equivalent to those in the lake cores; thus, additional catchment inputs will increase the BC deposition rates within lake cores. Third, the atmospheric BC deposition rate calculated from BC concentrations in the atmosphere is much lower than the BC deposition rate recorded in the Nam Co Lake cores (Fig. 6), further reflecting the dominant contributions of catchment inputs relative to atmospheric inputs in lake cores.

The above evidences demonstrate that variations in the BC deposition in Nam Co Lake mainly reflect variations in catchment inputs rather than in atmospheric inputs;

thus, atmospheric deposition plays a minor role relative to catchment inputs. Because most lakes in the HTP have increased in area over the last 20 years (Zhang et al., 2016), this phenomenon likely occurs in many other lakes in the HTP.

3.2.2 Actual atmospheric BC deposition and potential uncertainties

BC deposition rates derived from ice cores and snowpits are proposed to be closer to the actual atmospheric values in the HTP. This hypothesis is supported by two lines of evidence. First, BC deposition levels in the snowpits of different glaciers are consistent. For example, the estimated BC deposition rates of Laohugou, Tanggula, Zhadang, Demula and Yulong are 25, 21.3, 20, 14.5 and 20.2 mg m^{-2} a^{-1}, respectively (Table 1), which reflects a homogeneous spatial distribution in BC deposition. The above values are also similar to those of ice cores described in other articles (e.g., 18, 12, and 10.1 mg m^{-2} a^{-1} for the Muztagh Ata, Zuoqiupu, and East Rongbuk glaciers, respectively (Bauer et al., 2013; Ming et al., 2008; Xu et al., 2009b) (Table 1). Second, these values are nearly equivalent to those of atmospheric BC deposition rates derived from completely different methods (e.g., Community Atmosphere Model version 5 (Zhang et al., 2015) and other models (Bauer et al., 2013) (Table 1). In summary, despite some uncertainties associated with the remote study area, the atmospheric BC deposition rate of 17.9±5.3 mg m^{-2} a^{-1} in the glacial region of the HTP is proposed.

4 Conclusions

The BC concentration and deposition in the HTP region, which features the largest glacial area in the middle latitudes, were investigated and reevaluated in this article. Our findings indicated that carbonate carbon contributions from MD have led to overestimations of approximately 52±35% and 39±24% in previously reported BC concentrations in TSP samples at the remote Nam Co and Everest stations, respectively in the central and southern HTP. After omitting the contributions of carbonate carbon, the actual BC concentrations at the Nam Co and Everest stations should be 61 ng m^{-3} and 154 ng m^{-3}, respectively. In addition, the levels of OC and TC in TSP samples were also overestimated by 22±10% and 22±12%, respectively, at the Nam Co station and by 19±13% and 24±12%, respectively, at the Everest station. Large arid areas that receive little precipitation are distributed across the western and northern HTP; thus,

the effects of carbonates on BC measurements are expected to be greater in these areas and must be considered in future related studies. In addition, TSP samples must be treated with acid to eliminate the effects of carbonates prior to measuring BC. A comparison among BC deposition values based on different methods and materials showed that, because of catchment inputs, the BC deposition rates derived from HTP lake cores were higher than the actual atmospheric deposition values. Correspondingly, the BC deposition values measured from snowpits and ice cores in glacial regions were similar to those obtained via models; thus, these data reflect the actual atmospheric BC deposition values. Although the HTP is located adjacent to seriously polluted regions in South Asia and East China, the HTP BC deposition rates are relatively low because of the high elevation. Finally, our results indicate that the atmospheric BC deposition rate in the HTP is approximately 17.9±5.3 mg m^{-2} a^{-1}, with lower and higher values appearing in the central and peripheral areas of the HTP, respectively.

Acknowledgements

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Table 1 Monitored or recovered BC deposition (mg m^{-2} a^{-1}) from the HTP and other regions of the world.

Region	Sites	Deposition	Period	References
Tibet	Zuoqiupu glacier	12	1970-2005	1
	Muztagh Ata	18	1970-2005	1
	East Rongbuk ice core	10.2	1995-2002	2
	Laohugou glacier	25	2013-2014	3,4
	Tanggula glacier	21.2	2013-2014	3,4
	Zhangdang glacier	22.8	2013-2014	3,4
	Demula glacier	14.4	2013-2014	3,4
	Yulong glacier	20.3	2013-2014	3,4
	Model results of central TP	9	2013-2014	5
	Nam Co Lake core	260	1960-2009	6
	Qinghai Lake core	270-390	1770s-2011	7
	Aerosol of Nam Co station	10.5	2005-2007	8
	Aerosol of Qinghai Lake	92.7	2011-2012	8
East China	Chaohu lake core, East China	1160	1980-2012	9
	Northern China	1660	Around 2010	10
	Northern China Plain	1500	2008-2009	11

Note: 1: (Bauer et al., 2013); 2: BC concentration (20.3 ng g^{-1}) and snow accumulation (500 mm) were adopted from (Ming et al., 2008) and (Li et al., 2016c), respectively; 3: (Li et al., 2016c); 4: (Li et al., 2016a); 5: (Zhang et al., 2015); 6: (Cong et al., 2013); 7: (Han et al., 2015); 8: calculated in this study; 9: (Han et al., 2016); 10: (Fang et al., 2015); 11: (Tang et al., 2014).

Table 2 Precipitation (mm) and BC concentration (ng m^{-3}) values used for the BC deposition calculations for Nam Co Lake and Qinghai Lake.

	Nam Co Lake		Qinghai Lake	
	precipitation	BC concentration	precipitation	BC concentration
Spring	29.65	135.86	77.51	1000
Summer	190.05	90.97	244.02	530
Autumn	79.72	86.58	89.78	690
Winter	2.95	93.55	3.81	1050

485

Figure Captions

Figure 1 Selected study sites, including the HTP stations, lakes and glaciers.

Figure 2 Seasonal variations in the BC and TC concentrations in the original and acid-treated aerosol samples collected at the Nam Co and Everest stations.

Figure 3 Percentage of MD and CA relative to their sum during both non-monsoon and monsoon periods at the Nam Co and Everest stations.

Figure 4 Relationship between aerosol IC and Ca at the Nam Co and Everest stations.

Figure 5. Similar variations in precipitation and mass accumulation rates (A) (Wang et al., 2011) and significant relationships between mean precipitation and mean grain size (B) (Li et al., 2014) in the Nam Co Lake cores.

Figure 6 Comparison of atmospheric BC deposition rates derived from the glacial region, models, lake cores and values calculated from BC concentrations in the aerosols of the HTP.

Figure 1

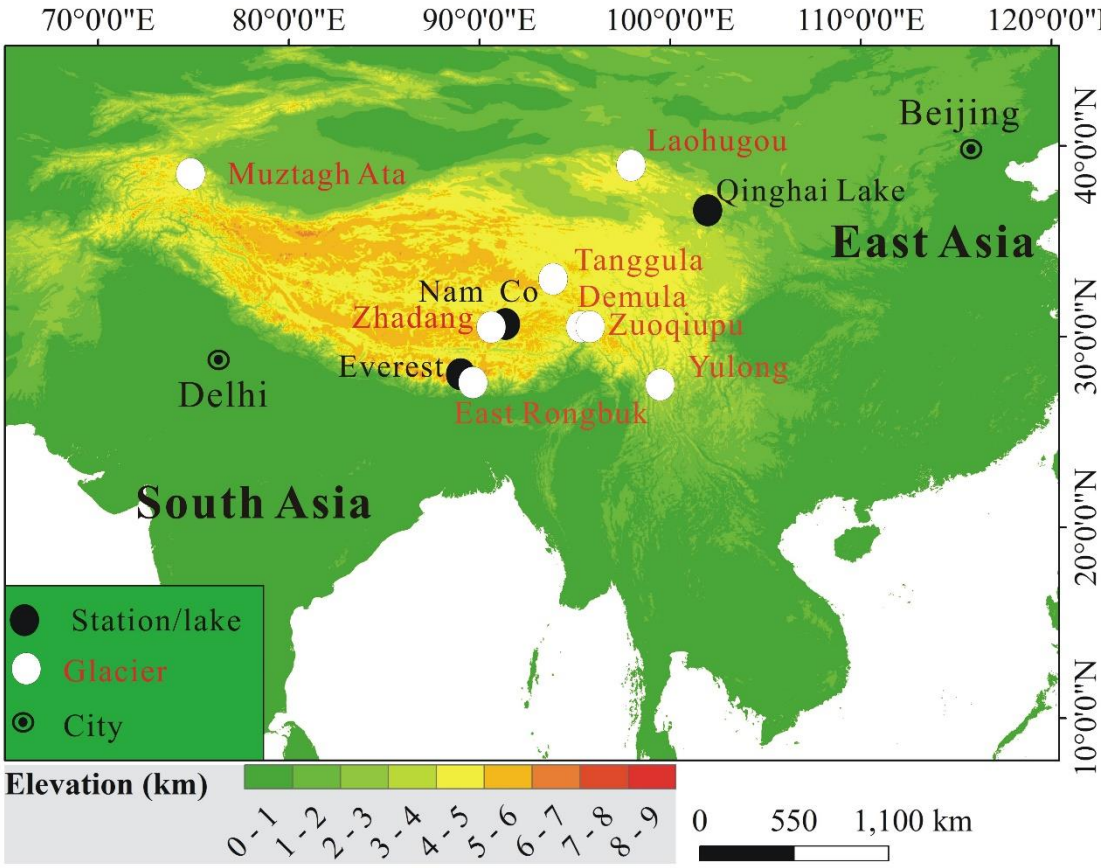
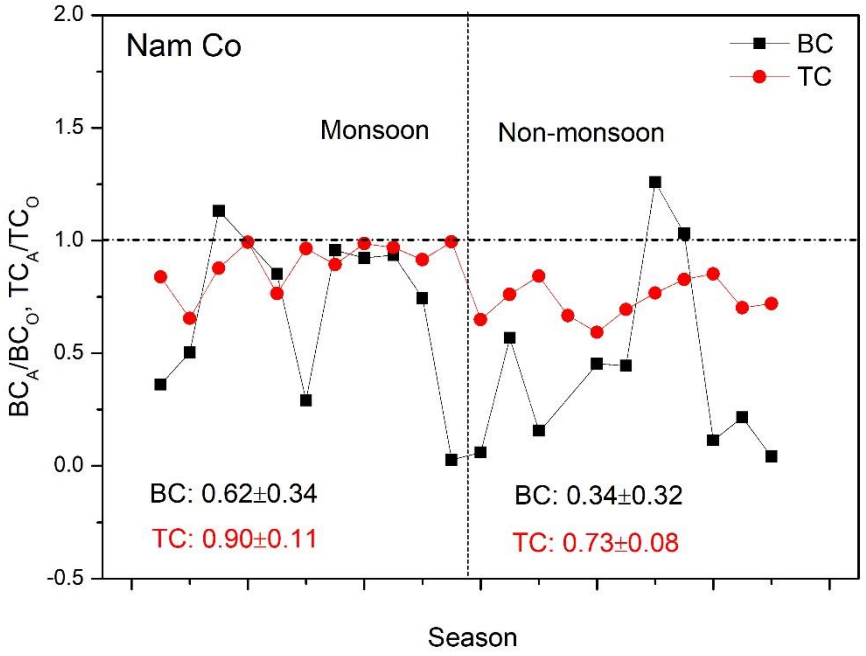

70°0'0"E
80°0'0"E
90°0'0"E
100°0'0"E
110°0'0"E
120°0'0"E
40°0'0"N
30°0'0"N
20°0'0"N
10°0'0"N
Beijing
Laohugou
Muztagh Ata
Qinghai Lake
East Asia
Tanggula
Nam Co
Demula
Zhadang
Zuoqiupu
Everest
Yulong
Delhi
East Rongbuk
South Asia
Station/lake
Glacier
City
Elevation (km)
0-1
1-2
2-3
3-4
4-5
5-6
6-7
7-8
8-9
0
550
1,100 km

Figure 2


Nam Co
BC
TC
Monsoon
Non-monsoon
2.0
1.5
1.0
0.5
0.0
-0.5
BC_A/BC_O, TC_A/TC_O
BC: 0.62±0.34
TC: 0.90±0.11
BC: 0.34±0.32
TC: 0.73±0.08
Season

505

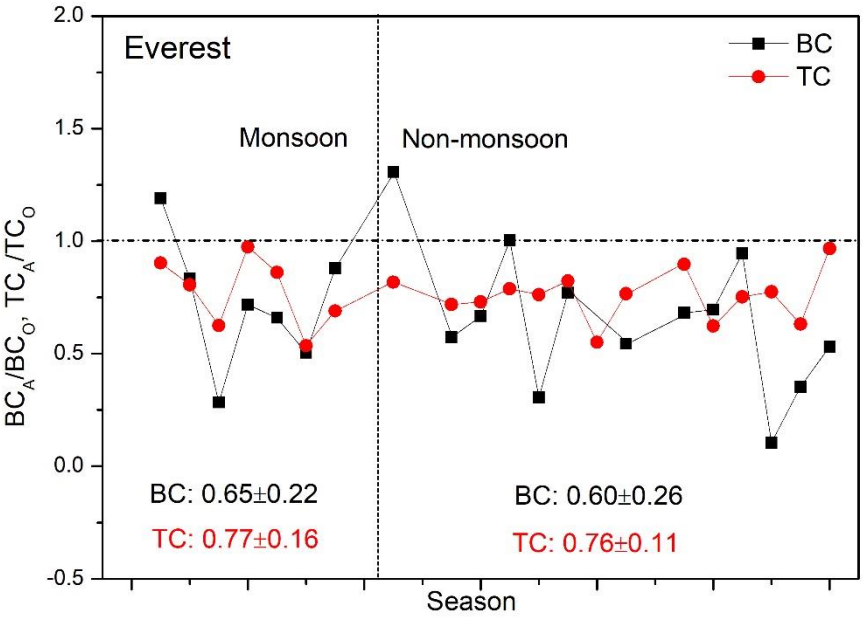
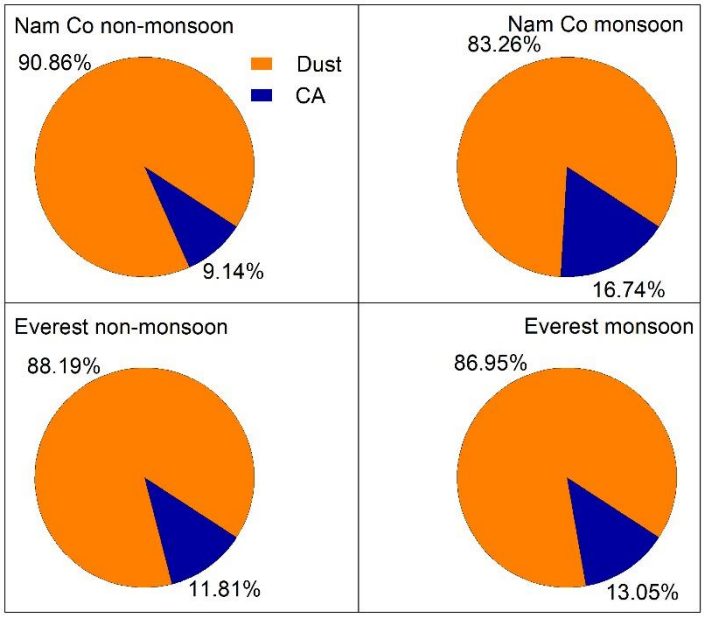
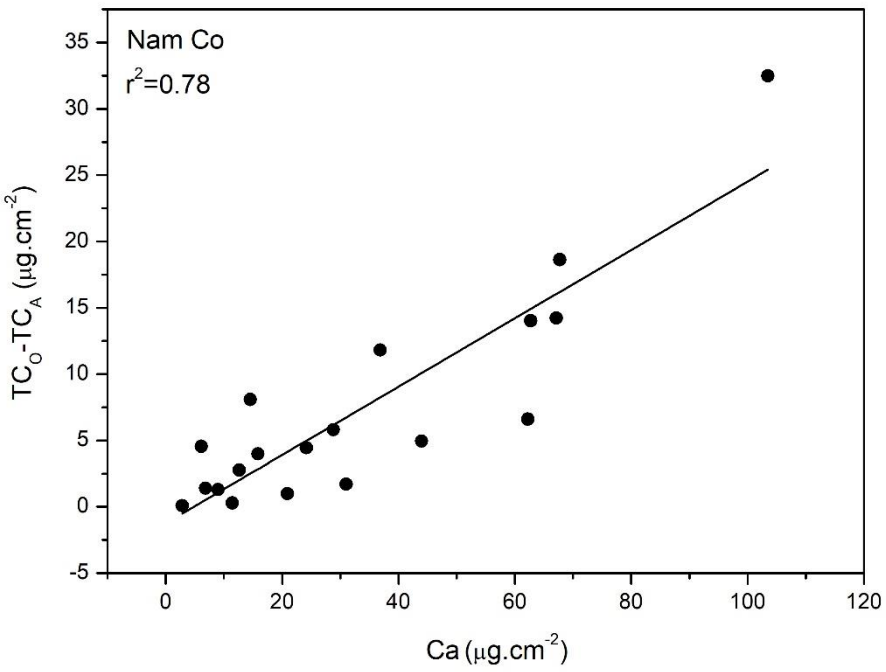

Everest
BC
TC
Monsoon
Non-monsoon
2.0
1.5
1.0
0.5
0.0
-0.5
BC_A/BC_O, TC_A/TC_O
BC: 0.65±0.22
TC: 0.77±0.16
BC: 0.60±0.26
TC: 0.76±0.11
Season

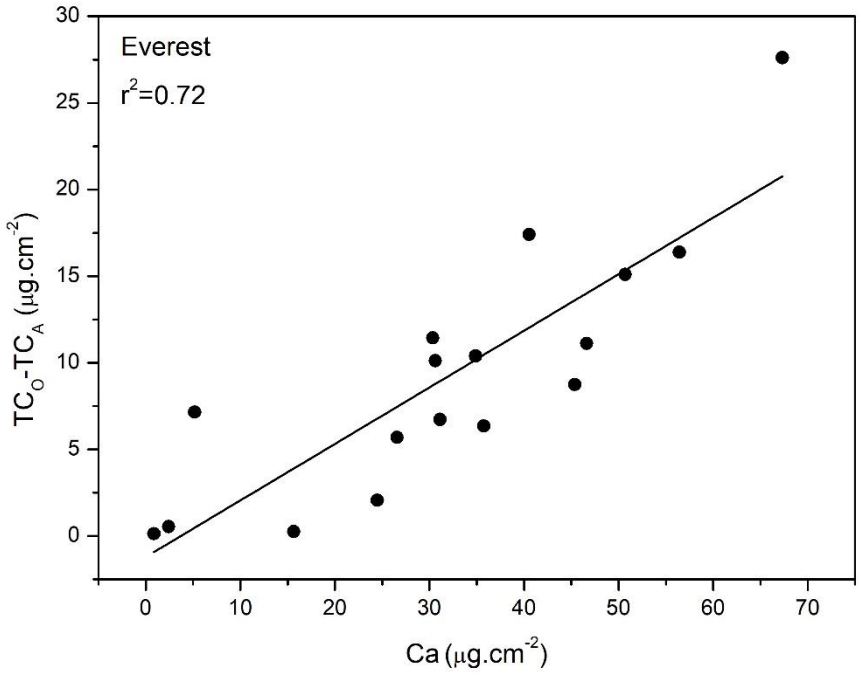
Figure 3


Nam Co non-monsoon
90.86%
Dust
CA
9.14%
Nam Co monsoon
83.26%
16.74%
Everest non-monsoon
88.19%
11.81%
Everest monsoon
86.95%
13.05%

510

Figure 4


Nam Co
r²=0.78
TC_O-TC_A (µg.cm⁻²)
Ca (µg.cm⁻²)
35
30
25
20
15
10
5
0
-5
0
20
40
60
80
100
120


Everest
r²=0.72
TC_O-TC_A (µg.cm⁻²)
Ca (µg.cm⁻²)
30
25
20
15
10
5
0
0
10
20
30
40
50
60
70

515 Figure 5

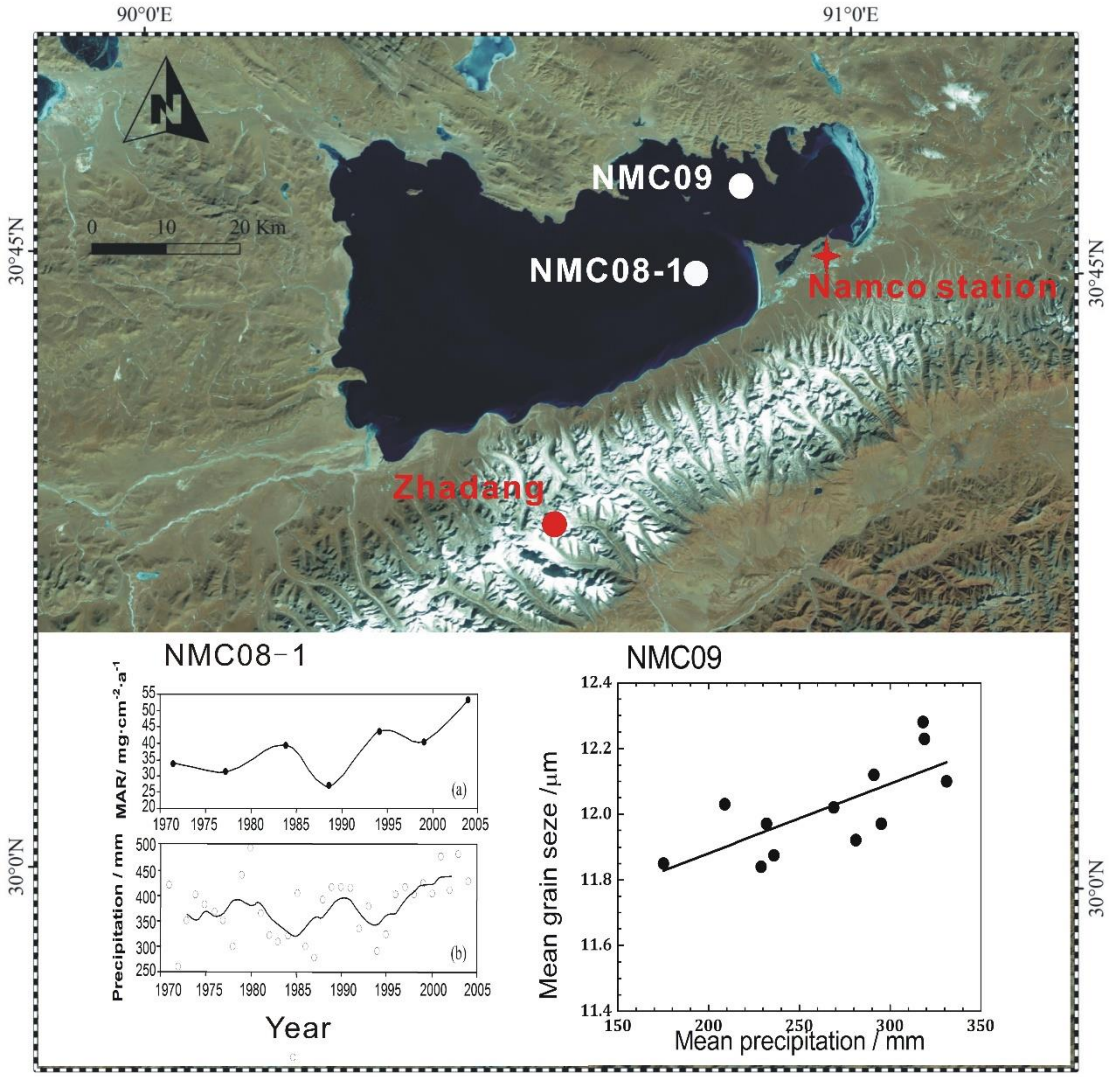
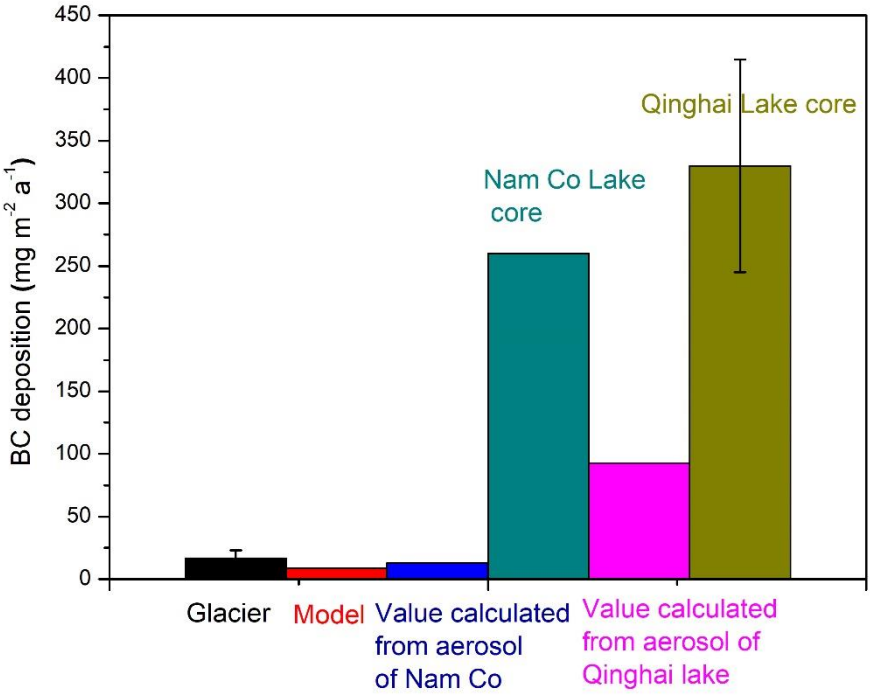

90°0'E
91°0'E
30°45'N
30°0'N
NMC09
NMC08-1
Namco station
Zhadang
0 10 20 Km
NMC08-1
MAR/ mg·cm⁻²·a⁻¹
Precipitation / mm
(a)
(b)
1970 1975 1980 1985 1990 1995 2000 2005
Year
NMC09
Mean grain seze /μm
Mean precipitation / mm
12.4
12.2
12.0
11.8
11.6
11.4
150 200 250 300 350

Figure 6


BC deposition (mg m-2 a-1)
450
400
350
300
250
200
150
100
50
0
Qinghai Lake core
Nam Co Lake core
Glacier
Model
Value calculated from aerosol of Nam Co
Value calculated from aerosol of Qinghai lake

520

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