

Dear Editor:

Thanks a lot for your suggestion and positive evaluation on this article. We have answered all the questions of editor and reviewer, please check them. If you have any more suggestion or questions please let us know. Meanwhile, we also made some other minor modifications of expression and updated the acknowledgement part in the revised MS. Our answers and modifications in the revised MS were marked out.

"Seze" has been changed to "size" in Fig.5 of revised article.

Thanks a lot for your work and best wishes.

Chaoliu Li on behalf of all the co-authors

2017/8/30

Suggestions for revision or reasons for rejection (will be published if the paper is accepted for final publication)

- Line 74-77 :It would be good to say what is really discussed, namely the real subjects of the paper.

Answer: Thank for the suggestion. This part was modified to "Therefore, in this article, we discussed the actual concentrations and deposition of BC in the HTP based on data of aerosols collected at two remote stations and previously reported BC deposition data."

- Line 111: why a reference to Fig. 1?

Answer: Fig.1 was deleted and the sentence was modified to "although neither of these two studies have directly discussed this issue or evaluated the effects of IC".

- Lines 130-136: Why the review paper of Ming et al. (An overview of black carbon deposition in High Asia glaciers and its impacts on radiation balance, Jing Ming, Cunde Xiao, Zhencai Duc, Xingguo Yang, Advance in water resources, <http://dx.doi.org/10.1016/j.advwatres.2012.05.015>) about the BC deposition in Asia Glacier is not cited ?

Answer: Thanks for the suggestion. We double checked this article and found although the title of this article was related to "black carbon deposition", this article only reported the black carbon concentration at glacier region but not black carbon deposition. Because this is an important article on black carbon at glacier region in the HTP, we added this reference at introduction part (lines 116-117) in the revised article.

- Lines 169-170: what is the reason for which this comparison remains reliable ?

Answers: Thanks a lot for the suggestion. This sentence was modified to "the comparison between data of lake core and snowpit is still reliable because BC deposition of the former was much higher (approximately 20 times) than that of the latter, far above up to 7 times among different methods (Watson et al., 2005)."

(Watson et al., 2005)

- Lines 258-259: revise the language

Answers: This sentence was modified to "In this study, it was shown that carbonate carbon significantly contributes to the BC, TC and OC concentrations of the TSP samples of Nam Co and Everest stations after comparing BC and OC concentrations between original and acid treated samples."

- Lines 268-270: This sentence does not say why BC is more influenced than OC and TC. I

45 suppose that BC decompose at higher T than OC and is therefore more influenced.

Answers: Thanks for the suggestion. This sentence was modified to "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 decompose at high temperatures along with BC during analyses."

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

Chaoliu Li<sup>1,3,4\*</sup>, Fangping Yan<sup>3</sup>, Shichang Kang<sup>2,4</sup>, Pengfei Chen<sup>2</sup>, Xiaowen Han<sup>1,5</sup>,  
55 Zhaofu Hu<sup>2,5</sup>, Guoshuai Zhang<sup>1</sup>, Ye Hong<sup>6</sup>, Shaopeng Gao<sup>1</sup>, Bin Qu<sup>3</sup>, Zhejing Zhu<sup>7</sup>,  
Jiwei Li<sup>7</sup>, Bing Chen<sup>7</sup>, Mika Sillanpää<sup>3,8</sup>

<sup>1</sup>Key Laboratory of Tibetan Environment Changes and Land Surface Processes, Institute of Tibetan Plateau Research, Chinese Academy of Sciences, Beijing 100101, China;

<sup>2</sup>State Key Laboratory of Cryospheric Sciences, Northwest Institute of Eco-Environment and  
60 Resources, Chinese Academy of Sciences, Lanzhou, 730000, China;

<sup>3</sup>Laboratory of Green Chemistry, Lappeenranta University of Technology, Sammonkatu 12, Mikkeli 50130, Finland

<sup>4</sup>CAS Center for Excellence in Tibetan Plateau Earth Sciences, Beijing 100101, China;

<sup>5</sup>University of Chinese Academy of Sciences, Beijing 100049, China

65 <sup>6</sup>Institute of Atmospheric Environment, China Meteorological Administration, Shenyang 110166, China

<sup>7</sup>Environmental Research Institute, Shandong University, Jinan 250100, China

<sup>8</sup>Department of Civil and Environmental Engineering, Florida International University, Miami, FL-33174, USA

70 *Correspondence to:* C. Li ([lichao Liu@itpcas.ac.cn](mailto:lichao Liu@itpcas.ac.cn))

**Abstract:** Black carbon (BC) is the second most important warming component in the atmosphere after CO<sub>2</sub>. The BC in the Himalayas and the Tibetan Plateau (HTP) has  
75 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  
80 concentrations in the HTP are overestimated. In addition, large discrepancies have

been ~~existed~~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  $52\pm 35\%$  and  $39\pm 24\%$ , respectively, because of the influence of carbonates in MD. Additionally, the organic carbon (OC) levels were overestimated by approximately  $22\pm 10\%$  and  $22\pm 12\%$  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  $61 \text{ ng m}^{-3}$  and  $154 \text{ ng m}^{-3}$ , 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  $17.9\pm 5.3 \text{ mg m}^{-2} \text{ a}^{-1}$ , 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

## 110 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 (Xu et al., 2016;Ramanathan and Carmichael, 2008;Lau et al., 2010;Ji et al., 2015), promoting HTP glacial retreat (Xu et al., 2009;Qu et al., 2014;Li et al., 2017;Zhang et al., 2017b;Ming et al., 2009;Ming et al., 2013), 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 (Zhao et al., 2013b;Ming et al., 2010;Cong et al., 2015;Marinoni et al., 2010;Wan et al., 2015) and atmospheric BC deposition as determined from lake core sediments (Han et al., 2015;Cong et al., 2013). 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. Therefore, in this article, we discussed the actual concentrations and deposition of BC in the HTP based on data of aerosols collected at two remote stations and previously reported BC deposition data.~~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.~~

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At present, the thermal-optical method is a widely used method for measuring BC concentrations in aerosols from the HTP (Zhao et al., 2013b;Ming et al., 2010;Cong et al., 2015;Li et al., 2016d). 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<sub>2</sub> in response to increasing temperature during measurements, thus causing an overestimation of the total carbon (TC) in carbonaceous aerosols (CAs) (Karanasiou et al., 2011). 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).

145 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  
150 than 2.5  $\mu\text{m}$  ( $\text{PM}_{2.5}$ ) during dust storm events. Similar phenomena have also been found for both  $\text{PM}_{2.5}$  and total suspended particle (TSP) samples in southern Europe (Sillanpää et al., 2005; Perrone et al., 2011). Because TSP samples contain more MD and carbonates than  $\text{PM}_{2.5}$ , they should have higher concentrations of IC.

The above phenomenon should also be taken into consideration in the study of  
155 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 (Cao et al., 2010; Cong et al., 2015; Li et al.,  
160 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  
165 areas of the HTP have identified MD components (Cong et al., 2015; Zhao et al., 2013b). although neither of these two studies have directly discussed this issue or evaluated the effects of IC.

Because MD has lower influences on light than BC in the atmosphere (Clarke et al., 2004; Bond and Bergstrom, 2006) and on glacier surfaces (Qu et al., 2014),  
170 considering IC as BC will overestimate the BC-driven climate forcing. Organic

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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 HTP aerosols must be quantitatively evaluated. In this study, TSP samples from two remote stations in the  
175 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  
180 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  
185 terrain and dynamic regimes (Bond et al., 2013;Bauer 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  $9 \text{ mg m}^{-2} \text{ a}^{-1}$  (Zhang et al., 2015), which is approximately thirty times lower than the values measured in lake cores at Nam Co and Qinghai lakes ( $270\text{-}390 \text{ mg m}^{-2} \text{ a}^{-1}$ ) (Fig. 1) (Cong et al.,  
190 2013;Han et al., 2011). Although considerable uncertainties exist in atmospheric BC deposition estimated from models (Koch et al., 2009;Bond et al., 2013) and lake core sediments (Yang, 2015;Cohen, 2003), these large differences need to be thoroughly investigated.

For instance, although the influence of sediment focusing on BC deposition in  
195 lake cores has been noted in other areas (Yang, 2015;Blais and Kalff, 1995), 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  
200 rates in ice cores and snowpits from the HTP (Fig. 1) (Xu et al., 2009;Li et al.,

2016c;Li et al., 2016a;Ming et al., 2008). 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 (Karanasiou et al., 2015;Andreae and Gelencser, 2006). 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.~~ the comparison between data of lake core and snowpit is still reliable because BC deposition of the former was much higher (approximately 20 times) than that of the latter, far above up to 7 times among different methods (Watson et al.,

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2005). For instance, although large uncertainties exist for BC concentrations within the same environmental matrix (Watson et al., 2005; Hammes et al., 2007; Han et al., 2011), 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.~~

**Table 1.** Monitored or recovered BC deposition ( $\text{mg m}^{-2} \text{a}^{-1}$ ) 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
East China	Aerosol of Nam Co station	10.5	2005-2007	8
	Aerosol of Qinghai Lake	92.7	2011-2012	8
	Chaohu lake core, East China	1160	1980-2012	9
	Northern China	1660	Around 2010	10
	Northern China Plain	1500	2008-2009	11

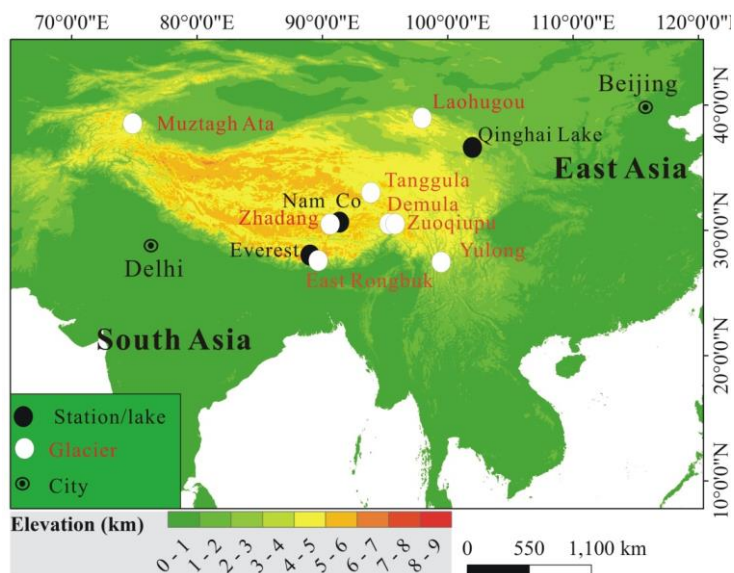
Note: 1: (Bauer et al., 2013); 2: BC concentration ( $20.3 \text{ ng g}^{-1}$ ) and snow accumulation (500 mm) were adopted from (Ming et al., 2008) and (Li et al., 2016c), respectively; 3: (Li et al., 2016c); 4: (Li et al., 2016a); 5: (Zhang et al., 2015); 6: (Cong et al., 2013); 7: (Han et al., 2015);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

## 2 Methods

250 **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  
255 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 ~~therein~~  
~~these areas~~ (Chen et al., 2015; Cong et al., 2015; Ming et al., 2010; Li et al., 2016a). In detail, TSP samples were collected using 90 mm pre-combusted (550°C, 6 hours)  
260 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  
265 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  
270 the BC concentrations. In addition, ten surface soil samples around the Everest station were collected to study the pH values.



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

## 2.2 Measurement of BC and elemental concentrations

275 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 (Li et al., 2016a; Pio et al., 2007; Chen et al., 2013; Bosch et al., 2014). The OC and elemental carbon (EC, the common

280 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

285 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  $BC_{(EUSAAR_2)}/BC_{(NIOSH)}$

290 for the studied samples were  $1.36 \pm 0.35$  and  $1.88 \pm 0.60$ , 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  
 295 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:

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

300 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  
 305 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 (Jurado et al., 2008;Fang et al., 2015) (Table 2). In brief, the annual atmospheric deposition rate of BC ( $\mu\text{g m}^{-2} \text{a}^{-1}$ ) was calculated as follows:

$$310 \quad F_{BC} = F_{DD} + F_{WD} \quad (3)$$

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

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

where  $F_{DD}$  and  $F_{WD}$  are the seasonal dry and wet deposition ( $\mu\text{g m}^{-2}$ ),  
 respectively;  $V_D$ ,  $P_0$  and  $W_p$  are the dry deposition velocity of aerosol ( $0.15 \text{ cm s}^{-1}$ ),  
 315 the precipitation amount (mm) in a given season and the particle washout ratio ( $2.0 \times 10^5$ ), respectively (Fang et al., 2015); and  $C_{BC-TSP}$  is the BC concentration of the TSPs ( $\mu\text{g m}^{-3}$ ). 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),  
 320 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.

**Table 2.** Precipitation (mm) and BC concentration (ng m<sup>-3</sup>) 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

### 325 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

330 In this study, it was shown that carbonate carbon significantly contributes to the BC, TC and OC concentrations of the TSP samples of Nam Co and Everest stations after comparing BC and OC concentrations between original and acid treated samples. 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<sub>A</sub>, OC<sub>A</sub> and BC<sub>A</sub>) to those of the original samples (TC<sub>O</sub>, OC<sub>O</sub> and BC<sub>O</sub>) 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, influences 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 decompose at high temperatures along with BC during analyses. As proposed in previous work (Chow and Watson, 2002), BC concentrations are more heavily influenced than OC and TC concentrations

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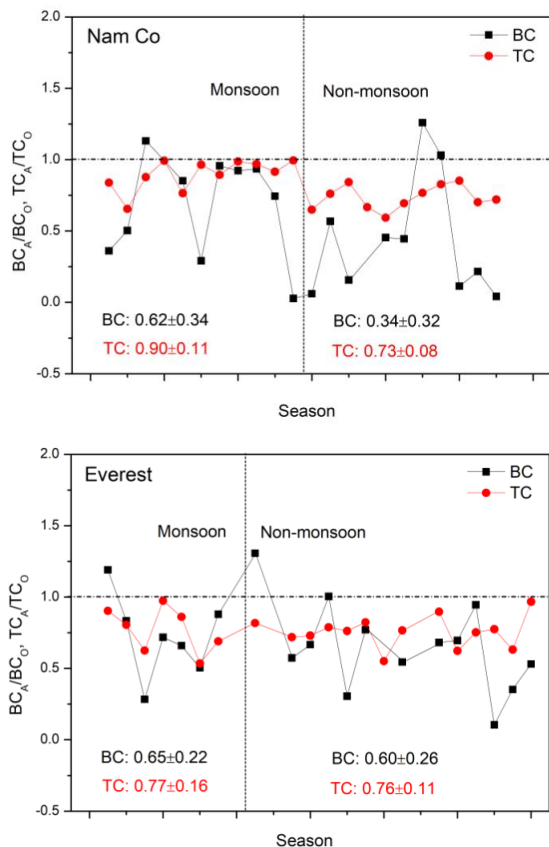
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~~because carbonates are more prone to decomposition at high temperatures during OC and BC analyses.~~ The OC concentrations in the treated samples used in this study also decreased, indicating that carbonates can also decompose at low temperatures (Karanasiou et al., 2011). Clear seasonal variations, i.e., low  $TC_A/TC_o$  ratios during non-monsoon periods and high  $TC_A/TC_o$  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  $TC_A/TC_o$  ratio at the Everest station were not observed, in accordance with the relatively stable seasonal variations in the  $Ca^{2+}$  content in aerosols 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.



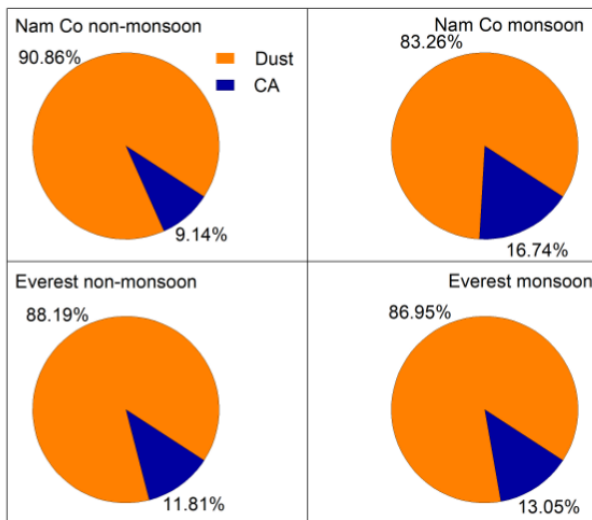
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**Figure 2.** Seasonal variations in the BC and TC concentrations in the original and acid-treated aerosol samples collected at the Nam Co and Everest stations.

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

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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 ~~demonstrats~~demonstrated 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.



**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.

### 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  $19 \pm 13\%$  and  $24 \pm 12\%$  at the Nam Co and Everest stations, respectively. These overestimates were higher than the corresponding value of 10% found for coarse

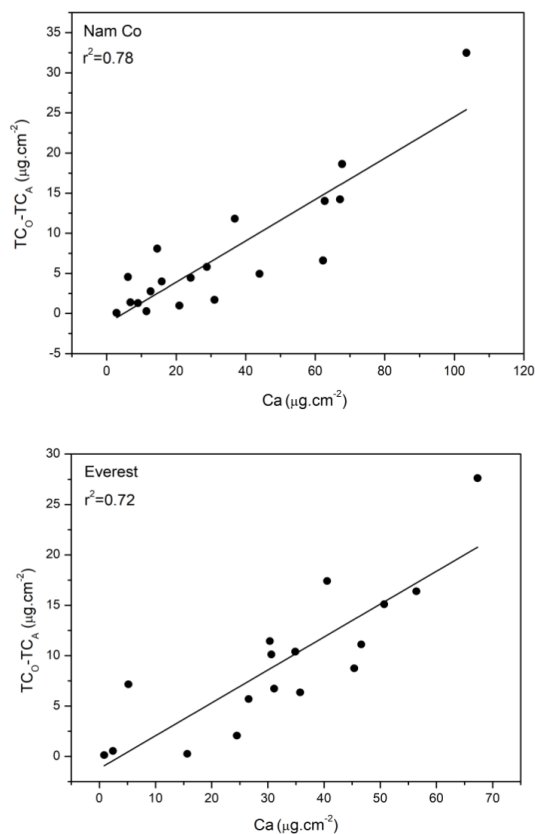


particles in the central Mediterranean region of Europe (Perrone et al., 2011). In addition, the related BC values were overestimated by approximately  $52\pm 35\%$  and  $39\pm 24\%$ , respectively, thus implying that the actual BC concentrations at these two  
405 stations were lower than previously 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.  
410 Therefore, based on previously reported BC concentrations measured via the same method as in this study (Zhao et al., 2013a; Cong et al., 2015), the actual BC concentrations at the Nam Co and Everest stations were estimated to be  $61 \text{ ng m}^{-3}$  and  $154 \text{ ng m}^{-3}$ , respectively.

Carbonates can decompose at relatively low temperatures during measurement,  
415 leading to overestimation of both BC and OC concentrations (Karanasiou et al., 2011). 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  
420 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).

425 Since the influence of carbonate carbon on TC has been observed in PM<sub>2.5</sub> samples from Qinghai Lake, Northwest China (Zhao et al., 2015), this phenomenon should be ~~obvious~~ 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  
430 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 (Qu et al., 2014; Li et al., 2017). However, numerous studies have measured BC concentrations without using an acid pretreatment step (Qu et al., 2014; Li et al., 2017; Zhang et al., 2017b). 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.

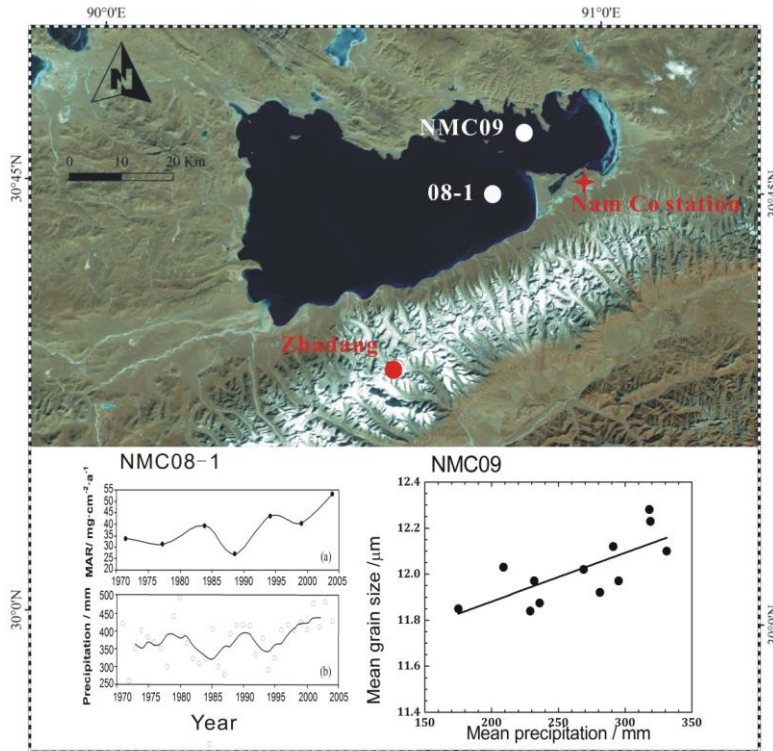


**Figure 4.** Relationship between aerosol IC and Ca at the Nam Co and Everest stations.

### 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<sup>-2</sup> a<sup>-1</sup> (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<sup>-2</sup> a<sup>-1</sup> and 270-390 mg m<sup>-2</sup> a<sup>-1</sup>, 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.



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

Lake core-derived BC deposition in Qinghai Lake was only 2-3 times higher  
 470 than that estimated from the BC concentrations of PM<sub>2.5</sub> in the atmosphere (Zhao et  
 al., 2015). Because PM<sub>2.5</sub> does not include all particles in the atmosphere, the actual  
 BC concentration in the atmosphere should be higher than that of PM<sub>2.5</sub> (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  
 475 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  
480 overestimated by approximately 35%.

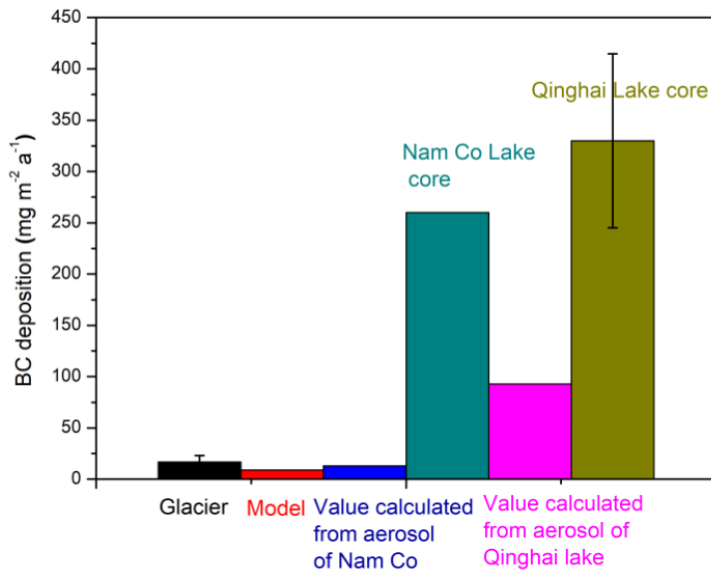
Correspondingly, catchment inputs account for a large proportion of the  
NMC09 samples. BC is widely distributed throughout environmental materials (e.g.,  
soil and river sediments) because of its inert characteristics (Cornelissen et al.,  
2005; Bucheli et al., 2004). Therefore, river inputs contribute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

490 First, a large glacial area ( $141.88 \text{ km}^2$ ) 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  
495 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  
500 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  
505 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.

510 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  
515 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  
520 most lakes in the HTP have increased in area over the last 20 years (Zhang et al., 2017a), this phenomenon likely occurs in many other lakes in the HTP.



**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.  
525

### 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  
530 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<sup>-2</sup> a<sup>-1</sup>, 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<sup>-2</sup> a<sup>-1</sup> for the Muztagh Ata, Zuoqiupu, and East Rongbuk glaciers,  
535 respectively (Xu et al., 2009; Bauer et al., 2013; Ming et al., 2008) (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  
540 deposition rate of 17.9±5.3 mg m<sup>-2</sup> a<sup>-1</sup> 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  
545 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  
550 should be 61 ng m<sup>-3</sup> and 154 ng m<sup>-3</sup>, 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. These values on TC were close to those of a study in the western HTP (Cao et al., 2009). Large arid areas that receive little precipitation are distributed across the  
555 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  $17.9 \pm 5.3 \text{ mg m}^{-2} \text{ a}^{-1}$ , with lower and higher values appearing in the central and peripheral areas of the HTP, respectively.

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