Black carbon aerosol in winter northeastern Qinghai-Tibetan Plateau, China: the source, mixing state and optical property

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Abstract

Black carbon (BC) aerosol at high-altitude Qinghai-Tibetan Plateau has potential effects on the regional climate and hydrological cycle. An intensive measurement campaign was conducted at Qinghai Lake (~3200 ~~ASL~~metres above sea level) at the edge of the northeastern Qinghai-Tibetan Plateau during winter using a ground-based single particle soot photometer (SP2) and a photoacoustic extinctiometer (PAX). The average concentration of refractory BC (rBC)~~ concentration~~ and number fraction of coated rBC were found to be 160 ± 190 ng m^{-3} and 59~~.3~~% for the entire campaign, respectively. Significant enhancements of rBC loadings

and number fraction of coated rBC were observed during pollution episode, with an average value of 390 ng m^{-3} and ~~64.6~~65%, respectively. The mass size distribution of rBC particles showed lognormal distribution with a peak diameter of ~187 nm regardless of the pollution level. Five-day backward trajectory analysis ~~combined with the fire counts map~~ suggests that the ~~biomass burning~~ air masses from North India contributed~~ing~~ to the increased rBC loadings during the campaign. The potential source contribution function (PSCF) model combined with the fire counts map further proves that biomass burning from North India is an important potential ~~region~~ source influencing northeastern Qinghai-Tibetan Plateau during the pollution episode. The rBC mass absorption cross section (MAC_{rBC}) at λ = 532 nm was slightly larger in clean days (14.9 m^2 g^{-1}) than during pollution episode (9.3 m^2 g^{-1}), likely due to the effects of brown carbon and the uncertainty of the MAC_{rBC} calculation. The MAC_{rBC} was positively correlated with number fraction of coated rBC during pollution episode with an increasing rate of 0.18 (m^2 g^{-1}) $\%^{-1}$. The number fraction of coated rBC particles showed positive correlation with light absorption, suggesting that the increase of coated rBC particles will enhance the light absorption. Compared to rBC mass concentration, rBC mixing sate is more important in determining absorption during pollution episode, estimated from the same percentagewise increment of either rBC mass concentration or the number fraction of coated rBC. The estimated BC direct radiative forcing was + 0.93 W m^{-2} for pollution episode, which is 2 times larger than that in clean days. Our study provides insight into the potential climatic impacts of rBC aerosol transported to the Qinghai-Tibetan Plateau from South Asian regions, and is also useful for future modeling studies.

1 Introduction

Black carbon (BC) aerosol has received worldwide concern due to its effects on climate and human health (Anenberg et al., 2012; Bond et al., 2013). BC shows an overall warming effect by either absorbing incoming solar radiation in the atmosphere or by reducing the albedo of surface (i.e., snow and ice) (Jacobson, 2001; Ramanathan and Carmichael, 2008; Kühn et al., 2014). A total climate forcing of BC particles is estimated to be + 1.1 W m^{-2}, which is ranked as the second largest contributor to anthropogenic radiative forcing after carbon dioxide in the present-day atmosphere (Bond et al., 2013). BC particles, derived from incomplete combustion of fossil fuels or biomass, are mainly hydrophobic when emitted, but become hygroscopic over time due to atmospheric aging processes. When BC particles are mixed with

water-soluble aerosol composition, they can serve as cloud condensation nuclei and therefore affect microphysical properties of clouds leading to indirect effect on climate (Hansen et al., 2005; Lohmann and Feichter, 2005). BC also shows semi-direct effect through interaction with cloud processes (Koch and Del Genio, 2010). Moreover, the impacts of BC aerosols on the radiative balance may lead to far-reaching consequences, such as global dimming (Wild et al., 2007), lower crop yields (Tollefsen et al., 2009), and negative impacts on terrestrial and aquatic ecosystems (Forbes et al., 2006).

The Qinghai-Tibetan Plateau is known as the "Third Pole" of the Earth because of its immense area and high elevation. It covers the area of 27–45 °N, 70–105 °E with an average elevation >4000 m ASL (above sea level). Due to the special landform, ecosystem and monsoon circulation, the Qinghai-Tibetan Plateau exerts profound effects on regional and global radiative budget and climate (Kopacz et al., 2011; Su et al., 2013; Yang et al., 2014). The Qinghai-Tibetan Plateau is surrounded by many important anthropogenic BC aerosol source areas (Zhang et al., 2009), such as South Asia (e.g., India) and East Asia (e.g., China). Inventory study suggests that the BC emissions in China and India have increased by 40% and 54% from 2000 to 2008, respectively (Kurokawa et al., 2013). Due to the general circulation patterns, the Qinghai-Tibetan Plateau becomes a strong receptor of these high BC source areas (Cao et al., 2010; Xia et al., 2011; Cong et al., 2013; Zhao et al., 2013). Lu et al. (2012) shows that South Asia and East Asia are the main source regions, accounting for 67% and 17% of BC transported to the Himalayas and Qinghai-Tibetan Plateau on an annual basis, followed by former USSR (~8%), Middle East (~4%), Europe (~2%), and Northern Africa (~1%). Deposition of BC on snow and ice at the Qinghai-Tibetan Plateau would decrease the snow surface albedo (Xu et al., 2012; Ming et al., 2013). The Qinghai-Tibetan Plateau Glaciers, which are the largest glaciers outside of the Polar Regions, have shown signs of retreat (Xu et al., 2009). The snowmelt from Qinghai-Tibetan Plateau vitally affects the sustaining seasonal water availability leading to agriculture security in South, East, and Southeast Asia (Immerzeel et al., 2010).

The effect of BC transported from surroundings on Qinghai-Tibetan's environment and climate is of great significance. However, BC studies are still very scarce to date in the Qinghai-Tibetan Plateau (e.g., Cao et al., 2010; Zhao et al., 2013; Wang et al., 2014a). In these limited studies, online and offline filter-based techniques are often used. Due to inherent systematic limitations, direct examination of BC size distribution and mixing state with filter-

based measurements is not feasible (Watson et al., 2005; Slowik et al., 2007; Collaud Coen et al., 2010; Bond et al., 2013). The BC optical properties are dependent on its physical (e.g., size and shape) and chemical (e.g., mixing with other materials) features. For example, the degree of enhancement in mass absorption cross section from internal mixture of BC with other aerosol components can lead to large difference in the prediction of global radiative budget (Bond et al., 2006; Chung et al., 2012; Zhuang et al., 2013). Consequently, accurate characterization of BC particles is crucial for a precise estimate of the impacts of BC on the atmospheric radiative forcing, human health, and air quality. In this study, a single particle soot photometer (SP2) and a photoacoustic extinctiometer (PAX) were used to investigate the refractory black carbon (rBC) mass concentrations, size distribution, mixing state, and aerosol light absorption properties in northeastern Qinghai-Tibetan Plateau. The primary objectives of this study were (1) to investigate the important potential rBC source regions responsible for the high wintertime rBC concentration in the Northeastern Qinghai-Tibetan Plateau, (2) to study the effect of rBC mixing state on light absorption properties, (3) to estimate the direct radiative forcing during rBC pollution episode.

2 Methodology

2.1 Measurement site

Qinghai Lake (36.53–37.25 °N and 99.6–100.78 °E), the largest saline lake in China, is located ~3200 m ASL in a drainage closed intermountain basin on the Northeast Qinghai-Tibetan Plateau with an area of ~4400 km^2 (Figure 1). This region is highly sensitive to global climate change, because it is situated in the sensitive semi-arid zone between the Asian monsoon-controlled area and the westerly jet stream-influenced area (An et al., 2012). Intensive measurements were taken from 16–27 November, 2012 from the rooftop (~15 m above ground level) of a sampling tower at the “Bird Island” peninsula (36.98 °N, 99.88 °E), which is located at the northwest section of the Qinghai Lake shore as shown in Figure 1.

2.2 rBC mass and mixing state measurements

The commercially available SP2 instrument (Droplet Measurement Technology, Boulder, CO, USA) has proven useful for measuring rBC mass, size, and mixing state (e.g., Gao et al., 2007; Moteki and Kondo, 2007; Schwarz et al., 2010; Wang et al., 2015). The operating principles of the SP2 are described elsewhere (Stephens et al., 2003; Schwarz et al., 2006). Briefly, the

SP2 relies on laser-induced incandescence to quantify the rBC mass of individual particles. Continuous intracavity Nd:YAG laser light at 1064 nm is used to heat rBC-containing particles to their vaporization point. The peak incandescence signal is linearly related to the rBC mass in the particle irrespective of the particle morphology or mixing state; this holds true over most of the rBC mass range typically observed in the accumulation mode (Slowik et al., 2007). In this work, the rBC mass in the range of ~0.4–1000 fg, equivalent to ~70–1000 nm volume equivalent diameter (VED), is quantified, assuming a void-free density of 2.0 g cm^{-3} (Schwarz et al., 2008). This range covers >90% of the rBC mass in the accumulation mode. The incandescence signal was calibrated using a standard fullerene soot sample (Lot F12S011, Alpha Aesar, Inc., Ward Hill, Massachusetts). The total uncertainty in the rBC mass determination was ~25%. More details about the SP2 calibration and uncertainty can be found in our previous work (Wang et al., 2014a). Note that the SP2 only quantifies the most refractory and most efficient light-absorbing component of combustion aerosol. The rBC concentration is adjusted to standard temperature and pressure (STP, $T_{standard}$ = 273.15 K and $P_{standard}$ = 1013.25 hPa).

The SP2 is capable of determining the rBC mixing state. The time delay between the peaks from the scattered light and incandescence signals is an indicator of the amount of non-rBC material mixed internally with individual rBC particles (Schwarz et al., 2006; McMeeking et al., 2011; Perring et al., 2011; Wang et al., 2014a). This method is sensitive to optically significant amounts of non-rBC material. The time delay occurs because the coatings must be removed from the rBC particle before the onset of incandescence. Because the scattering measurement is rather noisy for small particles and become saturation for large particles, the mixing state was studied for rBC core between ~70 and ~275 nm VED, which constitute the majority of rBC particle numbers (Wang et al., 2014a). The limitation of SP2 instrument is discussed in Taylor et al. (2015) when considering leading-edge scattering. The number fraction of coated rBC particles which is calculated from the distribution of time delay is an indicator of the degree to which the rBC particles are coated with other substances. This number fraction is higher for more aged rBC particles due to the formation of coating from atmospheric physical and chemical processes including coagulation, condensation, and heterogeneous reactions (Liu et al., 2013; Browne et al., 2015).

2.3 Particle light absorption measurements

The PAX (Droplet Measurement Technology, Boulder, CO, USA) measures light absorption and scattering coefficients simultaneously using a modulated diode laser. The light absorption coefficient is measured based on the intracavity photoacoustic technology. A laser beam in the acoustic chamber of the instrument heats suspended absorbing particles, by which a pressure wave is produced and detected with a sensitive microphone. A wide-angle integrating reciprocal nephelometer in the acoustic chamber measures the light scattering coefficient regardless of the particles' chemical makeup, mixing state, or morphology. In this study, the light absorption at λ = 532 nm is measured. Before sampling, nitrogen dioxide (NO_2) and ammonium sulfate are used for the calibration of light absorption and scattering, respectively. The PAX can provide the light extinction coefficient independently using the laser power. NO_2 was used to produce an absorption reading of ~500–30000 Mm^{-1}. A correction factor was then established from the relationship between the calculated light extinction coefficient using laser power and the measured light absorption. The uncertainty of the PAX is estimated to be ~10%. Like SP2 measurement, the absorption measurement reported here is also corrected for the standard temperature and pressure.

3 Results and discussion

3.1 Mass, size and mixing state of rBC aerosol

The time series of hourly averaged rBC mass concentrations and the mixing state obtained during the campaign are shown in Figure 2, and a statistical summary of the data is presented in Table 1. The mean concentration of rBC aerosol (±standard deviation) was 160 ± 190 ng m^{-3} during the entire campaign period, ranging from 6 ng m^{-3} to 1040 ng m^{-3}. The mean number fraction of coated rBC is found to be 59~~.3~~ ± 7~~6.9~~% (range ~~39.8~~40–73~~.2~~%), suggesting the majority of aged rBC particles in wintertime in the Qinghai Lake region. It is found that ~~30~~25% of the rBC values are higher than the ~~mean~~ 75th value, and the variation coefficient (defined by SD/mean) of rBC values reaches as high as 120%, suggesting a large rBC burden even at the free tropospheric altitude. ~~It is found that ~30% of the BC values are higher than the mean value, and the variation coefficient (defined by SD/mean) of BC values reaches as high as 120%, suggesting a large BC burden even at the free tropospheric altitude.~~ Elevated rBC concentration was observed from 19 to 21 November (defined as a pollution episode hereafter) with an average rBC loading of 390 ng m^{-3}, which is about 4 times higher than that

from the rest of measurement period (86 ng m^{-3}, defined as clean days). The mean number fraction of coated rBC also increases to ~~64.6~~65% during the pollution episode, higher than that in the clean days (~~57.7~~58%). Given that local rBC emissions in the Qinghai Lake region and even the entire Qinghai-Tibetan Plateau are very limited, the enhanced rBC concentrations observed during this campaign are most likely from regional transport as discussed below.

Figure 3 shows the mass size distribution of rBC particles for the entire campaign period. A lognormal size distribution pattern in VED for the size of rBC core of a particle is found, with a very close peak diameter for rBC pollution episode (188 nm) and clean days (187 nm). The size distributions of rBC core in the ambient atmosphere are affected by the size of fresh rBC particles and the subsequent atmospheric processing (Bond et al., 2013). The growth of rBC particles is a complex process, including water accretion, coagulation, condensation, and the accumulation of other materials through heterogeneous reactions. However, only the process of coagulation can lead to the increase of rBC core in VED. The coagulation of particles in ambient air is dominated by Brownian motion, a slow process for particles in the accumulation mode (Seinfeld and Pandis, 1998). Therefore, the similarity in VED size distribution for rBC core between clean and pollution episode indicates that the measured rBC particles are likely from biomass burning emissions, given that fossil fuel and biomass burning tend to have different rBC size distributions and that the peak diameter measured in this study is similar to the reported rBC peak diameter from biomass burning plumes (range ~187–193 nm, see Kondo et al., 2011; Sahu et al., 2012; Taylor et al., 2014).

3.2 rBC potential pollution source areas

To examine the contribution of regional rBC transport, five-day back trajectories were calculated using the hybrid single-particle Lagrangian integrated trajectories (HYSPLIT) model (http://ready.arl.noaa.gov/HYSPLIT.php~~www.arl.noaa.gov/reday.html~~). The HYSPLIT model was driven with full vertical dynamics using gridded meteorological data (Global Data Assimilation System, GDAS1). ~~was used to compute the five-day back trajectories using BC as a marker. The five-day period is chosen because the atmospheric lifetime of BC is typically in the order of one week (Chung and Seinfeld, 2005; Cape et al., 2012).~~ Figure 4a shows the hourly results of backward trajectories calculated with the arrival height of 100, 500 and 1000 m above ground level. The rBC data were averaged to 1 hr in order to match the timestep of the trajectories. The different arrival height of trajectories shows similar transport directions,

suggesting the air masses mixed well at different altitude. During rBC pollution episode, the air masses were mainly originated from the regions of high rBC emissions in North India (Sahu et al., 2008), which then passed over the rather clean western Qinghai-Tibetan Plateau (Zhang et al., 2009). In contrast, the air masses were originated from Europe and passed through the western part of China during clean days. ~~An aerosol optical depth (AOD) map, retrieved from the measurements of Moderate Resolution Imaging Spectroradiometer (MODIS) on the Terra satellite, describes the mean atmospheric aerosol loading around Qinghai-Tibetan Plateau (Figure 5a). Since 5-day back trajectory analysis shows that the polluting air masses arriving the measurement site on 19–21 November were from North India, the pollution status of North India on 14–16 November (i.e., 5 days backward) was examined. As shown in Figure 5a, high AOD values can be found along the Indo-Gangetic Basin in India and South Pakistan, indicating heavy pollution in this region. The fire counts map obtained from MODIS observation on NASA satellites also shows a large number of biomass burning activities in North India during 14–16 November, indicating large biomass burning aerosol (including BC aerosol) emissions. Although the high altitude of the Himalayas was thought to be a physical wall for atmospheric pollutants, previous studies indicate that the high Himalayan valleys can act as a "direct channel" for the transport of air pollutants up to 5000 m ASL (e.g., Bonasoni et al., 2010). After reaching the north of the Himalayan, the air pollutants can further transport to the central Qinghai-Tibetan Plateau (Hindman and Upadhyay, 2002; Xia et al., 2011). Therefore, the BC pollution episode observed in the Qinghai Lake measurement site is most likely derived from the biomass burning emissions in North India.~~

The potential source contribution function (PSCF) model (e.g., Wang et al., 2006) was used to further explore the potential source regions which influence rBC concentration in the Qinghai Lake region. To do so, the geographic region covered by the trajectories was divided into an array of 0.5×0.5 degree grid cells. The PSCF values for the grid cells were calculated by counting the trajectory segment endpoints that terminate within each cell. The number of endpoints that fall in the *ij*th cell is designated as n_{ij}. The number of endpoints for the same cell corresponding to rBC concentrations higher than an arbitrarily set criterion is defined to be m_{ij}. Then, the PSCF value for the *ij*th cell is defined as: $PSCF_{ij}=m_{ij}/n_{ij}$. Because of the impact of small values of n_{ij}, an arbitrary weight function W_{ij} was used to better reflect the uncertainty in the values for these cells (Polissar et al., 1999). The weight function reduces the PSCF values when the total number of the endpoints in a particular cell is less than about

three times the average value of the end points per each cell. Here, W_{ij} is defined as (Polissar et al., 2001):

$$W_{ij} = \begin{cases} 1.00 & 80 < n_{ij} \\ 0.70 & 20 < n_{ij} \leq 80 \\ 0.42 & 10 < n_{ij} \leq 20 \\ 0.05 & n_{ij} \leq 10 \end{cases} \quad (1)$$

Although PSCF model is often used to determine the potential source regions (e.g., Wang et al., 2006; Heo et al., 2013; Zhang et al., 2013), a limitation of this model is that grid cells can have the same PSCF value when sample concentrations at the receptor site are either only slightly higher or extremely higher than the criterion. This may lead to difficulties in distinguishing moderate sources from strong ones. To compensate for this limitation, the PSCF result calculated from the 75th percentile of all the data is set as the criterion (170 ng m^{-3}) in this study. Figure 4b shows the map of PSCF results for the entire campaign period. High PSCF values are found at North India. The PSCF values are low in the Qinghai Lake and surrounding regions, indicating lower likelihood of high rBC emissions from local sources compared to regional transport from North India. An aerosol optical depth (AOD) map, retrieved from the measurements of Moderate Resolution Imaging Spectroradiometer (MODIS) on the Terra satellite, describes the mean atmospheric aerosol loading around Qinghai-Tibetan Plateau (Figure 5a). High AOD values can be found along the Indo-Gangetic Basin in India and South Pakistan, indicating heavy pollution in this region. The fire counts map (Figure 5b) obtained from MODIS observation on NASA satellites also shows a large number of biomass burning activities in North India, indicating large biomass burning aerosol (including rBC aerosol) emissions. Although the high altitude of the Himalayas was thought to be a physical wall for atmospheric pollutants, previous studies indicate that the high Himalayan valleys can act as a "direct channel" for the transport of air pollutants up to 5000 m ASL (e.g., Bonasoni et al., 2010). After reaching the north of the Himalayan, the air pollutants can further transport to the central Qinghai-Tibetan Plateau (Hindman and Upadhyay, 2002; Xia et al., 2011). Therefore, the rBC pollution episode observed in the Qinghai Lake measurement site is most likely derived from the biomass burning emissions in North India.

3.3 Optical properties of rBC aerosol

The hourly light absorption coefficient varied from 0.0 to 18.1 Mm^{-1} with an average value of 2.1 ± 2.4 Mm^{-1} for the entire campaign (Figure 2). The average value increased to 3.7 ± 2.9 Mm^{-1} during rBC pollution episode, which is ~3 times higher than the average value in clean days (1.3 ± 1.6 Mm^{-1}). The rBC mass absorption cross section (MAC_{rBC}, expressed here in m^2 g^{-1}) is one of the most important optical properties for rBC aerosol because this parameter links optical properties to rBC mass. The MAC_{rBC} can be calculated by dividing the absorption coefficient measured with the PAX by the rBC mass concentration from the SP2 (MAC_{rBC} = [Absorption]/[rBC]). Due to the difference in cutoff size for PAX (< 2.5 μm) and for SP2 (< 1.0 μm), the MAC_{rBC} may be overestimated by ~13% given that BC concentration in $PM_{1.0}$ accounted for ~85% of $PM_{2.5}$ in the Tibetan Plateau (Wan et al., 2015).

Figure 6a and b show histograms of the MAC_{rBC} values during clean days and pollution episode, respectively. The distribution of MAC_{rBC} in clean days tends to larger values than that during pollution episode, with an average value of 14.9 ± 8.9 m^2 g^{-1} for clean days and 9.3 ± 3.1 m^2 g^{-1} for pollution episode. These values are higher than the MAC_{rBC} of 7.8 m^2 g^{-1} for uncoated rBC particles (interpolated to 532 nm from 550 nm assuming an Absorption Ångström Exponent of 1.0) suggested by Bond and Bergstrom (2006). It is interesting that the MAC_{rBC} in clean days is ~60% larger than that during pollution episode, the reason for which is not clear. A possible explanation involves the interference from brown carbon. Previous studies demonstrate that brown carbon, like black carbon, is an important light-absorbing aerosol composition in the atmosphere which can absorb light at visible wavelength (e.g., λ = 532 nm) (Yang et al., 2009). In the rural areas of Qinghai, biofuels including yak and sheep dung, firewood, and crop residues account for ~80% of total household energy (Ping et al., 2011). Biofuel/biomass combustion emissions are considered as especially significant sources for brown carbon (Andreae and Gelencser, 2006). It may produce enough brown carbon (particularly during the smoldering combustion phase) influencing the light absorption when rBC loading is low. Thus, the MAC_{rBC} may be overestimated in clean days. In addition, the calculation method using the light absorption and rBC mass may also introduce uncertainty, especially when rBC concentration is low. The high MAC_{rBC} values always correspond to the very low rBC mass. The MAC_{rBC} calculation method can bring ~30% uncertainty estimated from the square root of uncertainties in the PAX (10%) and SP2 (25%) measurements.

To further investigate the effect of rBC mixing state on MAC_{rBC}, the MAC_{rBC} values were plotted against the number fraction of coated rBC. As shown in Figure 7, the MAC_{rBC} was not correlated with the number fraction of coated rBC during clean days, but positive correlation was observed during pollution episode suggesting that the mixing state leads to the increase of the MAC_{rBC}. The slope of 0.18 (m^2 g^{-1}) $\%^{-1}$ obtained from the linear regression is arguably representative of the rate of the mixing state effect on the MAC_{rBC}.

Both laboratory studies and field measurements have shown that the BC light absorption (related to its direct radiative effects) can be enhanced by a factor of 1.5–2.0 when BC particles are internally mixed with other non-light-absorbing aerosol components including sulfate, nitrate, organics and water (e.g., Bond et al., 2006; Shiraiwa et al., 2010; Wang et al., 2014b). This is because the non-absorbing materials act like a lens and therefore refract the light toward the absorbing BC core, leading to the enhancement of absorption on visible light. Figure 8 shows the relationship between light absorption and rBC mixing state during clean days and pollution episode. In clean days the light absorption shows no significant correlation with number fraction of coated rBC, which could be attributed to the influences of brown carbon. In contrast, during pollution episodes the light absorption coefficients generally increase with increasing number fraction of coated rBC with the latter being positively correlated with the rBC mass concentration during pollution episode (see Figure 8b). Such correlation indicates that the outflow from polluted south Asia would increase the rBC mass concentration leading to light absorption enhancement on the one hand, and the increased number fraction of coated rBC particles would further enhance the light absorption on the other hand. To further investigate whether rBC concentration or mixing state is more important for determining absorption, the increases in light absorption are compared based on the same percentagewise increment of either rBC mass concentration or number fraction of coated rBC. According to the regression function in Figure 8b and the correlation between absorption and rBC mass (Absorption = -0.38 + 10.17[rBC], r = 0.92), the increase of light absorption is larger for number fraction of coated rBC (e.g, Δ light absorption = 1.8 Mm^{-1}) than for the rBC mass (e.g, Δ light absorption = 0.5 Mm^{-1}), suggesting that, compared to rBC mass concentration, rBC mixing state is more important in determining absorption during pollution episode.

3.4 Implications for direct radiative forcing

The direct radiative forcing of BC particles (DRF_{BC}) refers to the change in energy balance at the top of the atmosphere due to absorption and scattering of sunlight by BC particles. Here the DRF_{BC} is estimated from a simple analytical solution derived from the following parameterization (Chylek and Wong, 1995):

$$DRF_{BC} = \frac{S_0}{4} T_{atm}^2 \times (1 - N) \times [4\alpha\delta_{ab} - 2 \times (1 - \alpha)^2 \beta\delta_{sc}] \qquad (2)$$

where S_0 is the solar irradiance (1370 W m^{-2}), T_{atm} is the atmospheric transmission (0.79), N is the cloud fraction (0.6), a is the surface albedo (i.e., 0.18 at rural region), β is the backscatter fraction, which is assumed to be 0.17 (Kim et al., 2012), and δ_{ab} and δ_{sc} are the absorption and scattering optical depth, respectively. The daily values of δ_{ab} and δ_{sc} are estimated from Aura-OMI satellite measurements (http://disc.sci.gsfc.nasa.gov/). More details about the assumption of this equation can be found in Kim et al. (2012). The average DRF_{BC} is estimated to be 0.6 ± 0.4 W m^{-2} for the entire campaign, ranging from 0.05 to 1.6 W m^{-2}. During rBC pollution episode, the DRF_{BC} was 0.93 ± 0.57 W m^{-2}, which is about two times higher than that in clean days (0.48 ± 0.29 W m^{-2}). It should be noted that the DRF_{BC} is calculated based on the assumption that BC particles are externally mixed with other non-light-absorbing particles. Given that a fraction of BC particles may be internally mixed with other aerosol compounds, the DRF_{BC} calculated here should be considered as the lower limit. Therefore, the BC mediated radiative forcing is of great importance for the local atmospheric radiative balance in the northeastern Qinghai-Tibetan Plateau. Given the much shorter lifetime of BC aerosol compared with greenhouse gases, mitigation of BC pollution could be an efficient control strategy for protecting the vulnerable environment in the Qinghai-Tibetan Plateau because it reduces the radiative forcing directly by reducing the BC particle concentration and indirectly by slowing down the melting of snowpack and ice that can reflect the sunlight. It is worth to note that the rBC concentration during pollution episode was 4 times higher than that in clean days, but the DRF_{BC} was only enhanced by a factor of two, suggesting the importance of other aerosol components which made negative contribution to DRF.

4 Conclusions

The mass concentration, size distribution, mixing state and optical properties of rBC particles in the Qinghai Lake region of the Qinghai-Tibetan Plateau are studied. The results show that average rBC concentration and number fraction of coated rBC are 160 ± 190 ng m^{-3} and 59~~.3~~%, respectively, for the entire campaign in November 2012. The average rBC mass concentration is about 4 times larger for pollution episode than for clean days; and the number fraction of coated rBC particles also increases from ~~57.7~~58% for clean days to ~~64.6~~65% for pollution episode. The mass size distribution of rBC particles shows lognormal distribution with a peak diameter of ~187 nm regardless of the pollution level. Back trajectory analysis and potential source contribution function (PSCF) model study show that North India is an important region influencing the rBC level in the northeastern Qinghai-Tibetan Plateau during the pollution episode. The fire counts map also suggests that the pollution episode is likely caused by biomass burning in North India.

The average light absorption (at λ = 532 nm) is 1.3 Mm^{-1} for the clean days and increases to 3.7 Mm^{-1} for pollution episode. The rBC mass absorption cross section (MAC_{rBC}) at λ = 532 nm was larger in clean days (14.9 m^2 g^{-1}) than during pollution episode (9.3 m^2 g^{-1}), likely due to the effects of brown carbon and the uncertainty of the MAC_{rBC} calculation. The MAC_{rBC} was positively correlated with number fraction of coated rBC during pollution episode with an increasing rate of 0.18 (m^2 g^{-1}) $\%^{-1}$. The number fraction of coated rBC particles shows positive correlation with light absorption, suggesting that the increase of aged rBC particles increases the light absorption. Compared to rBC mass concentration, rBC mixing sate is more important in determining absorption during pollution episode, estimated from the same percentagewise increment of either rBC mass concentration or the number fraction of coated rBC. The estimated BC direct radiative forcing is about 2 times higher for pollution episode (0.93 ± 0.57 W m^{-2}) than for clean days (0.48 ± 0.29 W m^{-2}).

This case study provides an insight into the sources, mixing state and optical properties of rBC particles in the northeast Qinghai-Tibetan Plateau.~~South Asia pollution impacting northeast Qinghai-Tibetan Plateau through long-range transport.~~ The enhancement of rBC absorption not only disturbs the energy budget of the atmosphere in this region, but also modifies the snow albedo by deposition. This in turn can accelerate the melting of the glaciers and snow-pack over Qinghai-Tibetan and, thus, affect the sustaining seasonal water availability leading to security of agriculture in downstream regions. More studies need to be

addressed on the basis of long-period investigations in the Qinghai-Tibetan Plateau region to improve our scientific understanding of the regional climate on the inter-annual as well as intra-seasonal scale.

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Table 1 Summary of rBC concentrations, number fraction of coated rBC, light absorption coefficient, and mass absorption cross section of rBC (MAC_{rBC}) during different sampling periods.

	rBC (mean ± SD, ng m^{-3})			Number fraction of coated rBC (%)			Absorption (Mm^{-1})			MAC_{rBC} (m^2 g^{-1})		
	PE*	CD*	All	PE	CD	All	PE	CD	All	PE	CD	All
Average	390±207	86±101	160±190	65±5	58±7	59±7	3.7±2.9	1.3±1.6	2.1±2.4	9.3±3.1	14.9±8.9	13.2±8.1
25th	219	40	50	63	53	54	1.4	0.7	0.8	7.4	9.0	8.3
50th	410	68	80	66	58	60	3.4	0.9	1.2	9.0	13.6	11.2
75th	489	103	170	68	63	65	4.4	1.4	2.0	10.7	18.7	16.7

*PE and CD represent the pollution episode and clean days, respectively.

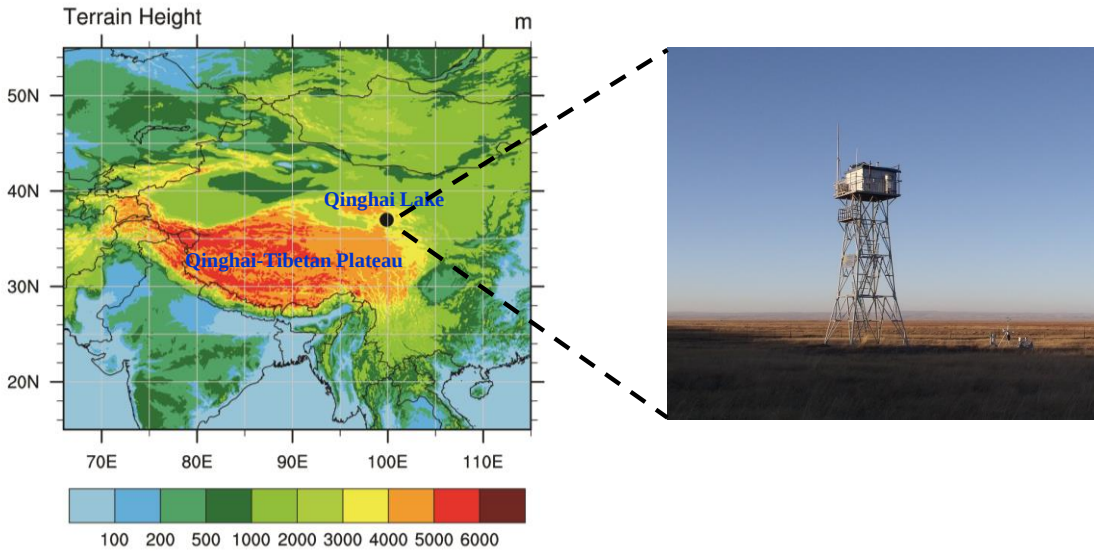

Figure 1. (left) Geographical location of Qinghai-Tibetan Plateau and surrounding areas. Color code represents topographical features (unit: m). (right) Observation tower at the "Bird Island" peninsula in Qinghai Lake, China.

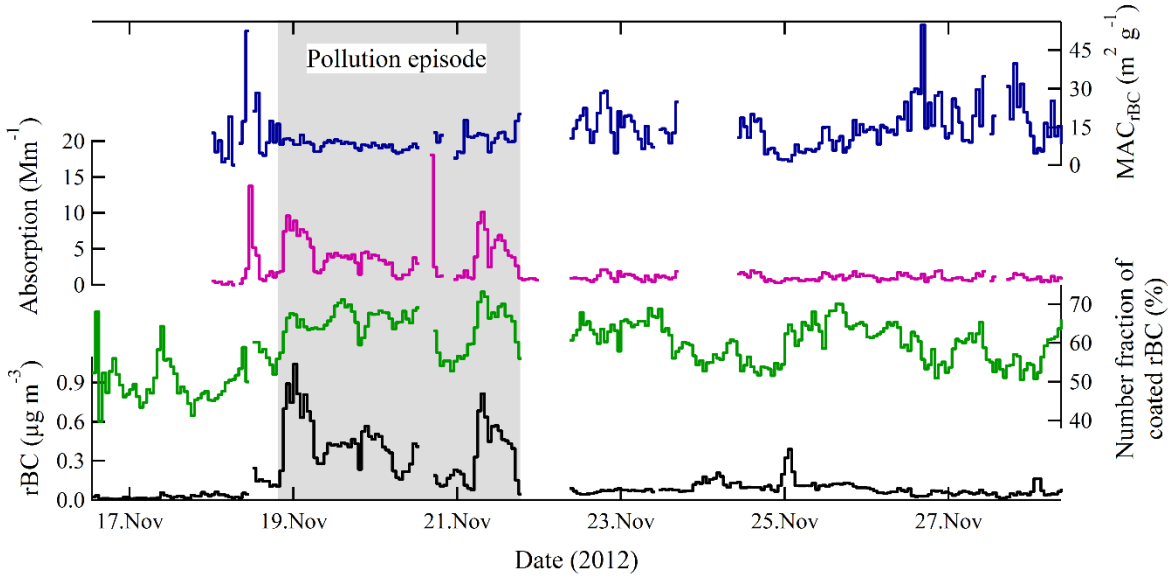

Figure 2. Time series of the rBC mass concentration, number fraction of coated rBC, light absorption at λ = 532 nm, and mass absorption cross section of rBC (MAC_{rBC}) during the entire campaign period. The pollution episode is highlighted with grey background.

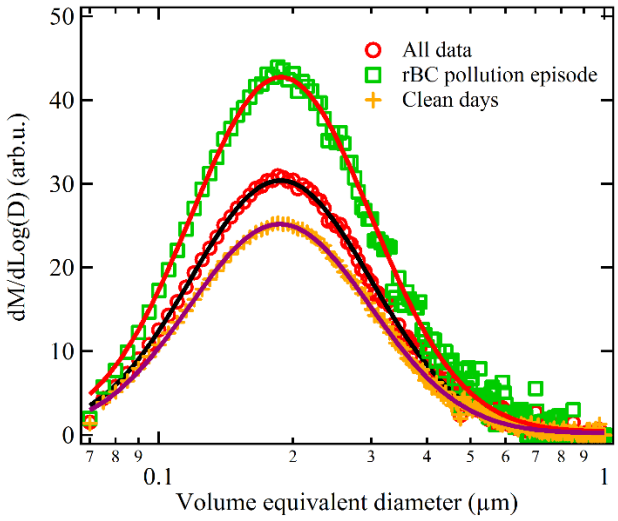

Figure 3. Mass size distribution of rBC in volume equivalent diameter during different sampling periods at Qinghai Lake. The solid lines represent lognormal fit. "M" and "D" in vertical label represent rBC mass and void free diameter (assuming 2 g cm^{-3} density), respectively.

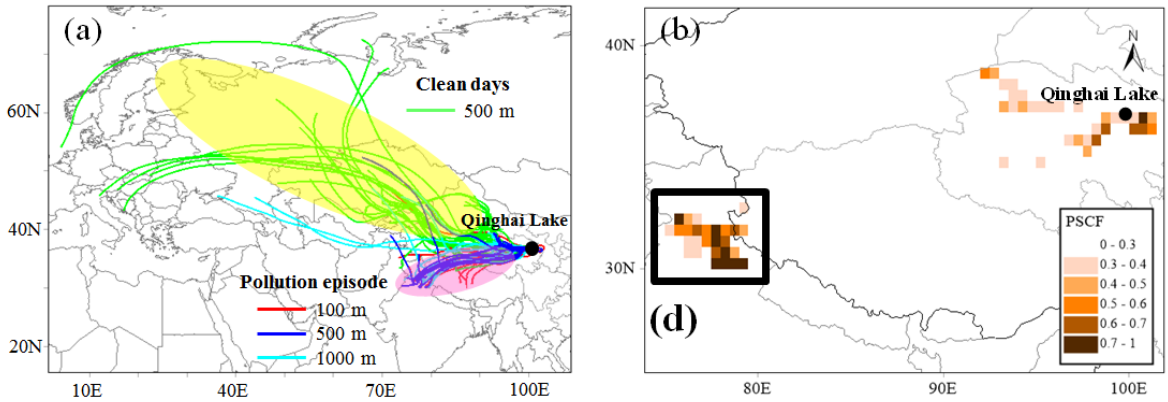

Figure 4. (a) Five-day backward air mass trajectories reaching at Qinghai Lake at 100, 500 and 1000 m above ground every six hours and (b) Likely source areas of rBC identified using potential source contribution function (PSCF) plots during the entire campaign.

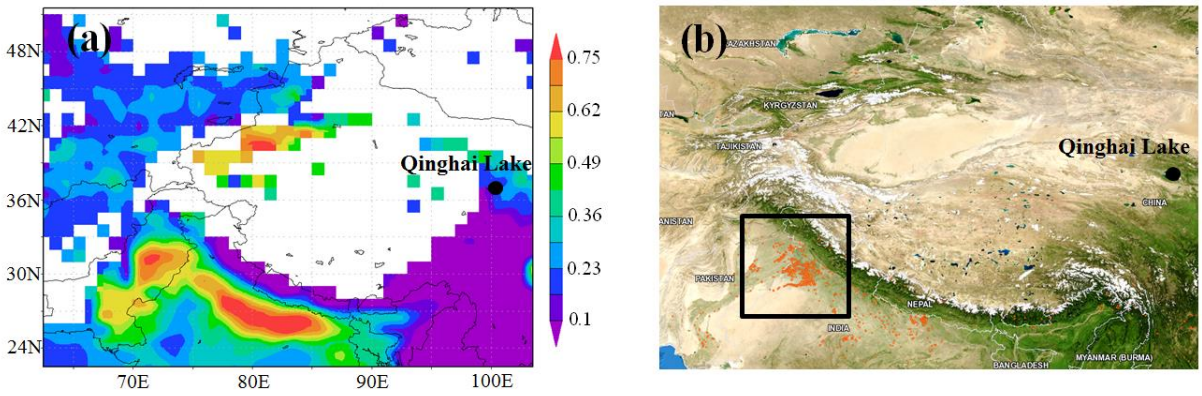

Figure 5. Regional distributions of (a) aerosol optical depth (AOD) and (b) fire counts map over Qinghai-Tibetan Plateau derived from MODIS observation during 16–27 November, 2012.

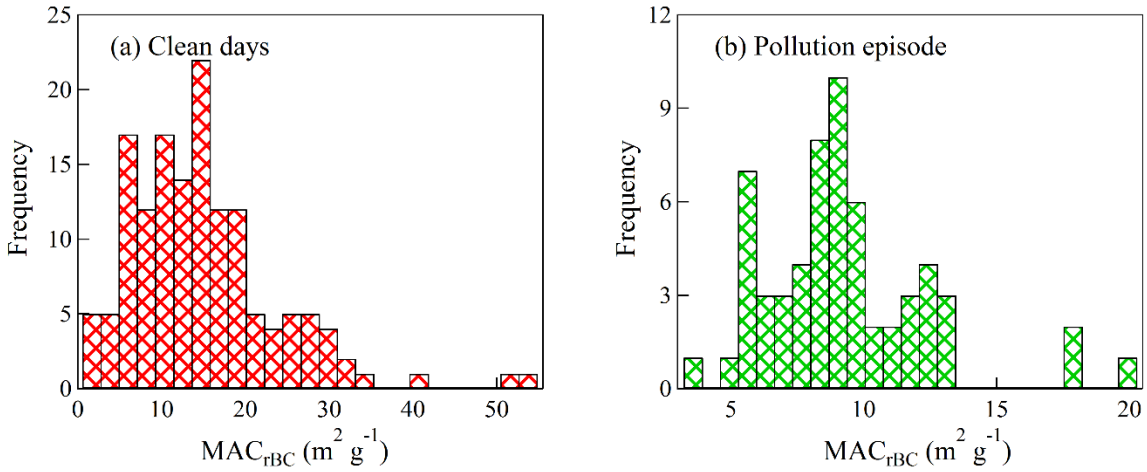

Figure 6. Frequency distributions of rBC mass absorption cross section (MAC_{rBC}) during (a) clean days and (b) pollution episode.

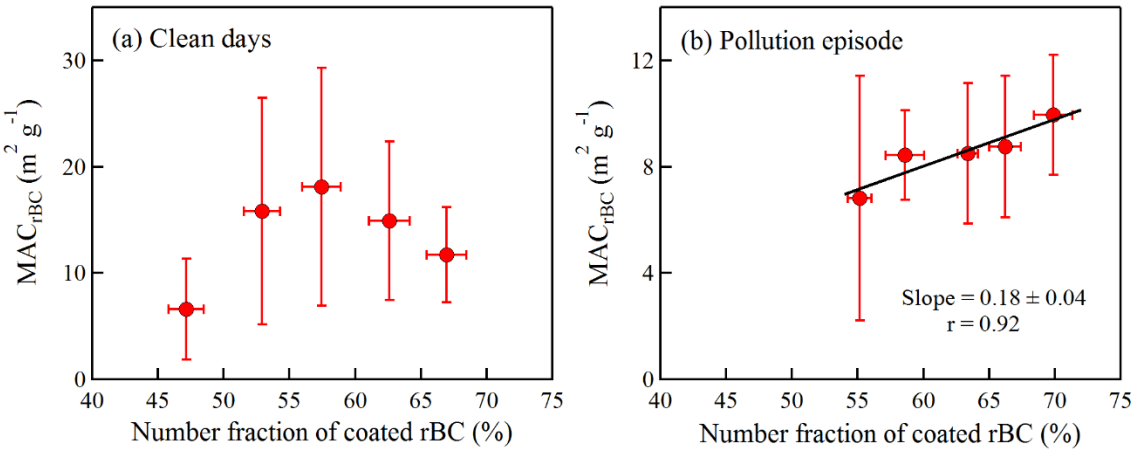

Figure 7. Mass absorption cross section of rBC (MAC_{rBC}) versus number fraction of coated rBC during (a) clean days and (b) pollution episode. The error bars correspond to the standard deviations of MAC_{rBC} and number fraction of coated rBC.

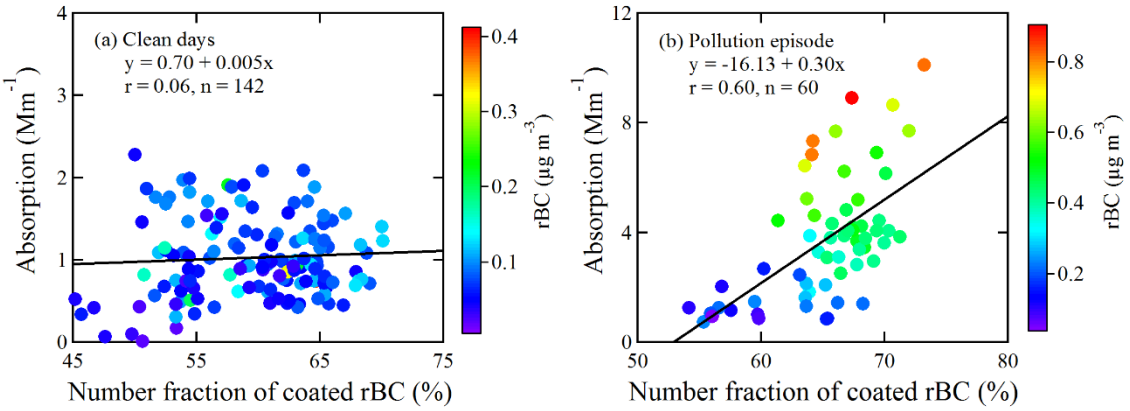

Figure 8. Light absorption as a function of number fraction of coated rBC during (a) clean days and (b) pollution episode. Data points are color coded for rBC mass concentration.