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Characterization of Black Carbon Aerosols over Darjeeling - A High Altitude Himalayan Station in Eastern India

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ABSTRACT

A continuous monitoring of black carbon (BC) aerosols was carried over a high altitude station Darjeeling (27°01′N, 88°15′E; 2200 m a.s.l.) at eastern part of Himalaya in India during January 2010–December 2011. In this article, we have presented the results of our analysis of the data collected during this interval. This study is focused on the investigation of the temporal variations, potential sources, long-range transport of BC aerosols along with the meteorological impact on these aerosols. BC is found to exhibit strong seasonal variation with the maximum concentration during premonsoon (5.0 ± 1.1 μg/m^3) followed by winter (3.9 ± 2.2 μg/m^3), postmonsoon (2.9 ± 1.0 μg/m^3) and minimum during monsoon (1.7 ± 0.7 μg/m^3). BC concentration varied between 0.2–12.8 μg/m^3 with an average of 3.4 ± 1.9 μg/m^3 over the entire period of study. The diurnal variation of BC aerosol shows sharp morning and evening peaks associated to the local anthropogenic activities as well as the impact of up-slope mountain wind. Amongst the long distant sources, BC concentration associated with the transport from Middle East countries and passing over South West countries like Pakistan, Afghanistan and Indo-Gangetic Plain is found to be comparatively higher. It is observed that the contributions of local emissions, long-range transport and mountain wind transport are approximately 56%, 27% and 17%, respectively, towards the total BC loading over Darjeeling during premonsoon. Fossil fuel emissions during premonsoon and biomass burning during winter are the major sources of BC with the overall dominance of fossil fuel burning throughout the entire study period. The BC concentration over Darjeeling is found to be much higher than any other high altitude stations in India and Nepal and even higher and comparable with some of the metro-cities in India.

Keywords: Black carbon; Himalaya; Darjeeling; Fossil fuel; Biomass.

INTRODUCTION

The studies of Black carbon (BC) aerosol has drawn a special attention as it can alter the radiation budget of the Earth-atmosphere system directly by absorbing the solar and terrestrial radiation (Charlson *et al.*, 1992) and thereby contributing to global warming significantly (Ramanathan and Carmichael, 2008). BC aerosols not only enhance the warming of the lower atmosphere but also the atmospheric instability which in turn affect the vertical motion, large scale circulation and hydrological cycles with significant regional climate effects (Meehl *et al.*, 2008). BC aerosols have strong influence on the summer monsoon circulation and the development of the convective precipitation in India (Wang *et al.*, 2009).

Characterization of BC aerosol over high-altitude sites at Himalaya has a special importance for understanding the role of BC on radiative forcing, deposition and melting of Himalayan glaciers (Kumar *et al.*, 2011). BC aerosols can modify the snow albedo significantly over the Himalayan regions contributing to the retreat of Himalayan glaciers (Mikhailov *et al.*, 2006; IPCC, 2007; Ramanathan and Carmichael, 2008). In addition to the local emissions, a significant amount of BC aerosols over Himalaya is associated with the long range transport from Indo-Gangetic basin and beyond as observed by Ram *et al.* (2008); Dumka *et al.* (2010); Kumar *et al.* (2011); Babu *et al.* (2011) etc. Considerable amount of BC aerosol is also associated with the mountain breeze circulations over different parts of the Himalaya (Decesari *et al.*, 2010). These transported BC aerosols would enhance the warming over the Himalayan regions significantly before the onset of summer monsoon

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(Ramanathan, 2007). One of the international programs called Joint Aerosol Monsoon Experiment (JAMEX) is being conducted to understand the characteristics of absorbing aerosols especially BC over the Himalayas with a special attention to address the aerosol-monsoon interface problem (Lau *et al.*, 2008).

The number of studies on BC aerosols over high altitude Himalayan stations is less as compared to studies over plain land cities and other urban atmospheres in India. These studies were mostly carried out at an altitude of 1958 m a.s.l. at Manora Peak (Pant *et al.*, 2006; Ram *et al.*, 2008; Dumka *et al.*, 2010; Ram *et al.*, 2010; Srivastava *et al.*, 2012a, b etc.) and at ~2200 m a.s.l. at Mukteshwar (Hyvärinen *et al.*, 2009) over central Himalaya; at Hanle (~4520 m a.s.l.) over western part of the Himalaya (Babu *et al.*, 2011; Nair *et al.*, 2013 etc.) and at Nepal Climate Observatory Pyramid (NCOP, 5079 m a.s.l.) over the eastern part of Himalaya (Bonasoni *et al.*, 2010; Marinoni *et al.*, 2010; Decesari *et al.*, 2010; Marcq *et al.*, 2010; Yasunari *et al.*, 2010; Nair *et al.*, 2013 etc.). Studies were also conducted over some other high altitude stations like Khumbu valley, Namche Bazar at eastern Himalayan stations in Nepal under the framework of Stations at High Altitude for Research on the Environment (SHARE) project (Lau *et al.*, 2008) which falls under the program JAMEX. Earlier, we had carried out a study (Chatterjee *et al.*, 2012a) on the effect of BC along with other anthropogenic and dust aerosols on aerosol optical depth over the same location, Darjeeling, at eastern Himalaya. However, studies on long term characterization of BC aerosols have not been performed so far over eastern part of Himalayan in India.

The present study is performed on the characterization of BC aerosols over Darjeeling (2200 m a.s.l.), a high altitude hill station at eastern Himalaya, India. The measurement of BC was done for a period of two years (January 2010–December 2011) to investigate 1) the seasonal and diurnal heterogeneity of BC aerosols, 2) the relative dominance of the biomass and fossil fuel sources resulting to seasonal heterogeneity, 3) the role of long range and mountain wind transport and the overall meteorological influences on BC pollution and 4) to compare BC concentration over Darjeeling with other Himalayan high altitude stations in India and Nepal and several metro-cities/urban atmospheres in India.

The two-year data, presented here, would certainly provide substantial improvements to the heretofore poor knowledge regarding black carbon aerosols over eastern Himalayas. This study would also help in evolving climatological models for radiation budget studies and proper understanding of the local atmospheric radiative balance over eastern Himalaya.

STUDY SITE AND SYNOPTIC METEOROLOGY

The study has been carried out at a high altitude hill station Darjeeling (27°01′N, 88°15′E, 2200 m a.s.l.) at eastern Himalaya in India. The map showing geographical location of the measurement site and adjacent regions in Darjeeling has been given in our earlier study (Adak *et al.*, 2014). The main town center in Darjeeling is situated at the north-eastern side and 200 m below the sampling station and 250 m below the ridge top. The area in and around the main town center is populated mainly by several hotels and resorts, offices, shopping malls, bus and car stands, schools and colleges and hence is the main source of anthropogenic aerosols. Most of the tourist spots fall under this area. The road with high vehicular density is National Highway (NH55) which starts from plain land city called Siliguri (~2000 m below and south of Darjeeling) and ends at Darjeeling. Darjeeling Himalayan Railway (also known as "Toy Train" and a world heritage site) is a narrow gauge railway which runs by coal and diesel and follows the highway, NH55. The Darjeeling railway station and the railway workshop are situated in the area of the main town center. The densely populated Indo-Gangetic Plain (IGP) is located ~50 km south of Darjeeling. Another two hill stations Kurseong (1450 m a.s.l.) and Kalimpong (1250 m a.s.l.) are situated south of Darjeeling.

The measurement was carried out at a height of ~10 m above ground in the campus of National Facility on Astroparticle Physics and Space Science, Bose Institute. The road adjacent to the campus starts from the main town centre and ends at a tourist spot which is ~300 m west of our campus with minimum traffic density. Biomass burning is the common feature in Darjeeling especially during night-time in winter.

The seasonal average along with minimum and maximum of surface meteorological parameters; temperature (T) in °C, wind speed (WS) in m/s, relative humidity (RH) in % are given in Table 1 and diurnal variation of those meteorological parameters are shown in Fig. 1. The entire study period is divided into four seasons; winter (December–February), premonsoon (March–May), monsoon (June–September) and postmonsoon (October–November). Table 1 shows that the temperature and relative humidity were highest during monsoon and minimum in winter whereas wind speed was highest in premonsoon followed by monsoon, postmonsoon and minimum in winter. The temperature was maximum at 1300–1400 hrs and minimum at 0600 hrs irrespective of the seasons. WS was very low during late evening to early morning whereas it was higher during early afternoon to early evening in all the seasons. The relative humidity was higher during late afternoon to late night except monsoon. The total amount of rainfall over the entire period was 5036 mm. However, ~90% rain occurred during monsoon (4522 mm) only whereas 476 mm rainfall was recorded during premonsoon in two-year study period.

INSTRUMENTATION AND DATA ANALYSIS

Continuous and real-time measurements of the mass concentration of black carbon (BC) were done using a seven-wavelength Aethalometer (Model: AE 42, Magee Scientific, USA; http://www.mageesci.com). BC was measured continuously (time resolution of 5 min) from January 2010 till December 2011. Aethalometer measures the optical attenuation at seven wavelengths; 370, 470, 520, 590, 660, 880 and 950 nm, flow rate being maintained at 2l p.m. The instrument uses the filter-based technique and measures light attenuation due to particles deposited

Table 1. Meteorological parameters over different seasons during the entire study period (January 2010–December 2011).

Seasons	Temperature (°C)			Wind Speed (m/s)			Relative Humidity (%)			Rainfall (mm)
	Min	Max	Mean ± SD	Min	Max	Mean ± SD	Min	Max	Mean ± SD	Total
Winter	3.8	11.9	6.4 ± 1.1	0.32	1.12	0.72 ± 0.11	65.3	82.1	75.2 ± 4.8	45.5
Premonsoon	8.7	18.3	13.2 ± 2.1	0.71	2.73	1.31 ± 0.21	75.5	83.5	78.3 ± 6.4	475.9
Monsoon	13.4	19.2	16.1 ± 1.1	0.51	2.21	1.02 ± 0.22	90.5	97.2	92.8 ± 1.6	4521.6
Postmonsoon	9.5	15.2	11.8 ± 2.5	0.44	1.48	0.82 ± 0.12	78.2	89.3	81.8 ± 4.7	148.1

on to a quartz filter. Further details of the instrument and the measurement techniques can be found elsewhere (Hansen *et al.*, 1984). The light absorption measured at 880 nm is considered to represent the effect of BC, since BC is the principal light absorber at that wavelength (Bodhaine, 1995). The uncertainty in the estimation of BC mass concentration by Aethalometry may arise due to the reduction in the optical path in the Aethalometer filter with an increased filter load, referred as shadowing effect (R-factor) and due to multiple scattering in the quartz fibre matrix of the filter tape (C-factor) (Weingartner *et al.*, 2003). Shadowing effect is almost negligible for mixture of several aerosols as reported by Weingartner *et al.* (2003) which is the probable case at Darjeeling for the present study. Weingartner *et al.* (2003) suggested a correction factor of 2.14 for multiple scattering corrections. We have used a factor of 1.9 in this study as suggested by the manufacturer based on Bodhaine (1995). According to Bodhaine (1995), the value of C = 1.9 remains fairly constants over a wide range of environmental conditions because in one sense it represents the ratio of the Aethalometer instrumental specific absorption to the in situ atmospheric specific absorption. Several earlier studies (Safai *et al.*, 2007; Dumka *et al.*, 2010; Srivastava *et al.*, 2012a, b etc.) on BC aerosols using Aethalometer (AE 42) have used 1.9 as the value of C factor.

The observational site in the present study is situated at an altitude of 2200 meter above mean sea level. As the ambient pressure decreases with altitude, the instrument samples particles with higher pumping speed in order to maintain the set mass flow rate, and hence more volume of air gets aspirated. The actual volume (V) of ambient air aspirated at an ambient pressure P and temperature T is thus,

$$V = V_0 \times (P_0/P) \times (T/T_0) \quad (1)$$

where V_0 is the standard mass flow rate.

Moorthy *et al.* (2004) suggested that, since the measured BC concentration ($[BC]_{measured}$) by Aethalometer is calculated based upon the standard mass flow rate, the actual/corrected BC concentration ($[BC]_{true}$) after correcting for the change in flow rate should be as follows:

$$[BC]_{true} = [BC]_{measured} \times [P_0T_1/P_1T_0]^{-1} \quad (2)$$

where P_1 and T_1 are the ambient pressure and temperature respectively. The instrument was run continuously during January, 2010 till December, 2011 and the total number of sampling days was 702 over the entire two-year period of study. The rest of the days were used for the maintenance purpose of the instrument.

RESULTS AND DISCUSSION

Seasonal Variability in BC Concentration

BC concentrations ([BC]) over Darjeeling varied between 0.2 μg/m^3 (during monsoon) and 12.8 μg/m^3 (during premonsoon) with the average of 3.4 ± 1.9 μg/m^3 over the entire period of study. The seasonal variation in [BC] is shown in Fig. 2. [BC] is found to be maximum during

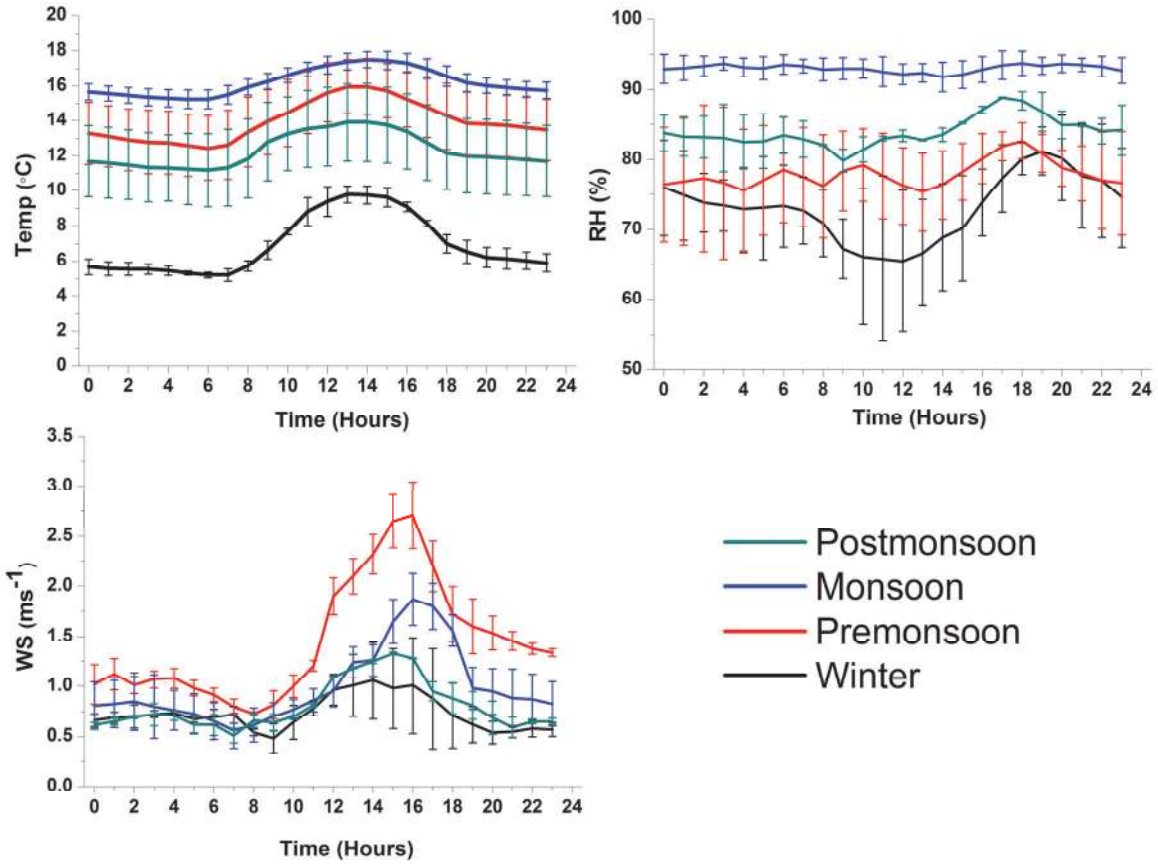

Fig. 1. Diurnal variation of micro-meteorological parameters.

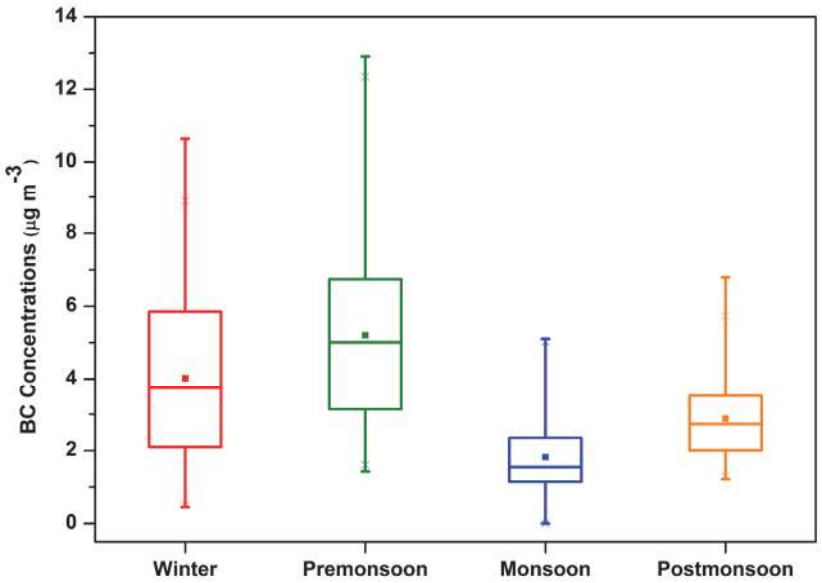

Fig. 2. Seasonal variation of BC concentration shown in box-whisker plot. The lower boundary of the box, the horizontal line inside the box and upper boundary of the box represent 25th percentile, median and 75th percentile respectively. The whiskers below and above represent minimum and maximum respectively.

premonsoon (Range: 1.4–12.8 µg/m^3; Av: 5.0 ± 1.1 µg/m^3; N = 177) followed by winter (Range: 0.4–8.9 µg/m^3; Av: 3.9 ± 2.2 µg/m^3; N = 174), postmonsoon (Range: 1.2–6.8 µg/m^3; Av: 2.9 ± 1.0 µg/m^3; N = 119) and minimum during monsoon (Range: 0.2–5.1 µg/m^3; Av: 1.7 ± 0.7 µg/m^3; N = 232). Premonsoon is the best season over Darjeeling for the tourists from all over India and abroad. The huge number of tourist vehicles (mostly diesel driven) is the primary and

most important reason for higher [BC] during premonsoon. Moreover, the "Toy Train" of Darjeeling Himalayan Railway runs more frequently in this tourist season and emits considerable amount of BC especially when runs on coal engine. In addition to the local emissions, high [BC] during premonsoon could also be associated with the transport from long distances (discussed later). Dust and carbonaceous aerosols from western and north-western India and beyond reach the foothills of Himalayas driven by premonsoon westerlies and are vertically advected to higher altitudes (Lau *et al.*, 2006). Having a very long continental overpass, these air masses could pick-up carbonaceous aerosols while passing over densely populated IGP. Biomass burning smokes from agricultural fires over north-west India was also identified as one of the sources of carbonaceous aerosols over Himalayan region during summer/premonsoon (Kumar *et al.*, 2011; Srivastava *et al.*, 2012b; Vadrevu *et al.*, 2012). Our earlier studies (Chatterjee *et al.*, 2010, 2012a) showed that significant amount of aerosols over Darjeeling are the transported aerosols from western part of India and beyond during premonsoon. High [BC] during premonsoon associated to the transport from long distances was also observed at Manora Peak (1958 m a.s.l.), western Himalaya, (Dumka *et al.*, 2010) and Hanle (4259 m a.s.l.), western trans-Himalaya (Babu *et al.*, 2011) in India. The boundary layer deepens due to higher temperature and convection during premonsoon which could flush up BC aerosols from lower altitude hill stations and plain land regions and thus get accumulated over the high altitude stations (Babu *et al.*, 2011). Massive biomass burning in addition to the vehicular emissions over Darjeeling is the major source of BC aerosols during winter. Our earlier studies (Chatterjee *et al.*, 2010, 2012a) showed high loading of fine mode K^+ (tracer of biomass burning) and significant increase in [BC] due to biomass burning during winter over Darjeeling. The boundary layer associated to the plain land IGP regions becomes shallow and remains well below our observational site which could inhibit the aerosol transport from low-level valley or plain land regions which could decrease [BC] over Darjeeling in winter. Hyvarinen *et al.* (2009) observed very low aerosol scattering coefficient over Mukteshwar (2180 m a.s.l.) at central Himalaya during winter as the boundary layer remains well below the site and the pollutants accumulated in the IGP regions could not reach the station. Another study conducted by Raatikaenen *et al.* (2014) over the same station, Mukteshwar, showed that the average altitude of airmass trajectories remained higher than the maximum boundary layer height of the plains during winter and thus Mukteshwar receives minimum pollutants being situated well above the pollution layer. Dumka and Kaskaoutis (2014) also showed that the uplift of aerosols from IGP regions gets prevented during colder winter months over Nainital, another high altitude central Himalayan station. Thus, local sources (biomass burning, vehicular emissions etc.) play major role over Darjeeling and other high altitude Himalayan station during winter or colder months. Long range transport by westerly wind flow was observed also in winter (discussed later). During monsoon, the significant drop in [BC] is due to below-cloud scavenging or wash-out of BC aerosols associated to the high rainfall amount (Table 1) over Darjeeling. Unlike premonsoon, the decrease in solar radiation and hence the weakening of convection due to cloudy skies do not favour the uplift of aerosols from low-level valley regions and thus the contribution of up-slope valley wind decreases significantly resulting in the reduction of BC concentrations during monsoon. The minimum BC concentration due to rain wash out during monsoon was also observed by earlier studies over several Himalayan high altitude stations (Pant *et al.*, 2006; Ram *et al.*, 2008; Hyvärinen *et al.*, 2009 over central Himalaya; Dumka *et al.*, 2010; Babu *et al.*, 2011; Raatikaenen *et al.*, 2014 etc.). Dumka and Kaskaoutis (2014) also showed minimum absorption and scattering coefficients of aerosols during monsoon due to rain scavenging. The monsoon winds originating from Bay of Bengal and Arabian Sea have significant continental overpass before reaching Darjeeling. But these air masses bring lesser amount of BC aerosols over Darjeeling due to aerosol wash out by frequent and intense rain along their transport pathways. Postmonsoon is also found to be the favourable season for the tourists but the influx of tourists during postmonsoon is comparatively less than premonsoon. In addition to the local emissions, transported carbonaceous aerosols could also contribute significantly in enhancing BC concentration over eastern part of Himalaya during postmonsoon. Bonasoni *et al.* (2010), Marinoni *et al.* (2010) and Dumka *et al.* (2010), through ground based observation, have shown BC influence over Himalayas due to transported plumes associated to crop residue burning over Punjab and adjacent IGP regions during postmonsoon seasons. Kaskaoutis *et al.* (2014) conducted a study on the influence of crop residue burning over northern India and its transport to eastern India and Himalayas during postmonsoon in 2012. They showed the transport of biomass burning plumes from western IGP (mainly Punjab) to eastern Himalayas and other regions. They found high Aerosol Index (> 2.0, retrieved from OMI) over western IGP regions during late burning period and over eastern part of Himalaya and IGP regions during post burning period in postmonson season.

Diurnal Variability in BC Concentrations: Effect of Local Activities and Mountain/Valley Wind

The diurnal variability of [BC] is highly related to the combined effect of variations in the anthropogenic emissions, surface meteorology and associated boundary layer dynamics (Ramachandran and Rajesh, 2007). Fig. 3 shows the average diurnal variations of [BC] for different seasons over Darjeeling. [BC] is found to exhibit strong diurnal variability with sharp morning and evening peaks in all the seasons. The gradual build-up in morning [BC] (at ~0500–0600 hrs in premonsoon and monsoon and at ~0700–0800 hrs in winter and postmonsoon), as shown, may be due to the fumigation effect of the boundary layer (Babu *et al.*, 2002) which brings aerosols from the nocturnal residual layer when it evolves after the sunrise and the strengthened thermal lift breaks the night-time inversion (Stull, 1989). The pollutants accumulated within the residual layer thus get uplifted to the higher altitudes and reach our observational site. The

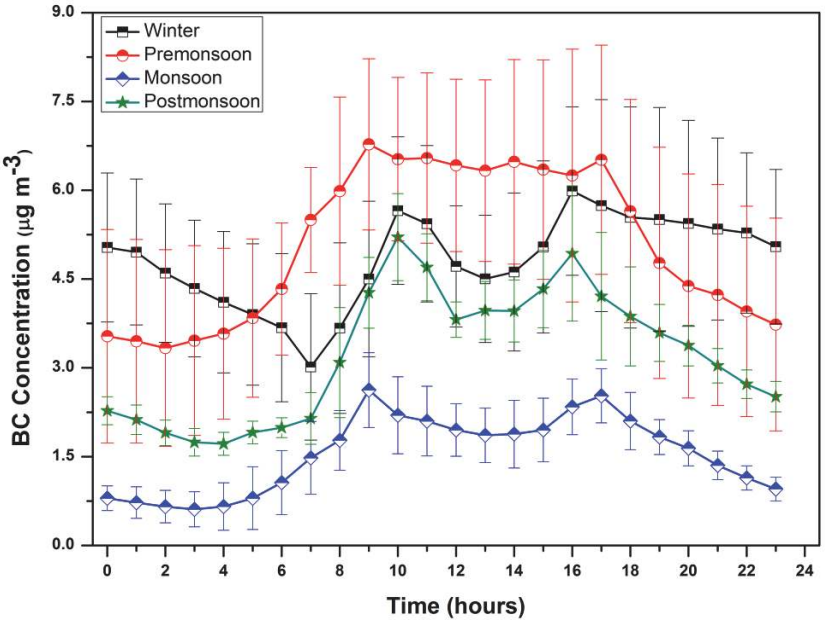

Fig. 3. Diurnal variation of BC concentrations over different seasons.

morning peak in premonsoon and monsoon was at 0900 hrs whereas that in postmonsoon and winter is found to be delayed by 1 hour i.e., at 1000 hrs. Similarly the evening peak in postmonsoon and winter (at 1600 hrs) is found to appear 1 hour earlier from that in premonsoon and monsoon (at 1700 hrs). After the morning peak, the [BC] is found to be decreasing gradually till an afternoon minima which could be due to the dispersion of BC aerosols caused by higher wind speed (Fig. 1) and subdued vehicular activities in afternoon. After the evening peak, the [BC] is found to fall gradually due to decrease in vehicular and local anthropogenic activities. Also, due to very low temperature during night-time, the atmospheric boundary layer (stable/nocturnal boundary layer) associated to the plain land IGP regions could remain well below the observational site (> 2000 m a.s.l.) and thus the ambient conditions correspond more to the free tropospheric conditions leading to the lowering in [BC]. The drop in [BC] after the evening peak, as shown, is very sharp and rapid in all the seasons except in winter where it is very slow and steady. In winter, the massive biomass burning (by local people/villagers on the road side and also in the fire places in most of the hotels/resorts to get warmth against cold) just after the sunset emits a large amount of BC aerosols which enhances night-time [BC]. The ratio of night-time (1800–0500 LST) to daytime (0600–1700 LST) [BC] over Darjeeling was higher than 1.0 during winter (1.2) whereas less than 1.0 during postmonsoon (~0.8), premonsoon (~0.7) and monsoon (~0.5). During winter, the night-time biomass burning was found to be so massive that it dominates over the usual day-time anthropogenic activities which increased the ratio (> 1.0). As discussed above, the boundary layer associated to IGP regions remains well below the observational site in winter inhibiting uplift of polluted aerosols and thus local/regional emissions play major role.

The diurnal feature of [BC] shows that in premonsoon, [BC] remained almost constant during afternoon (between morning and evening peaks) and does not show significant drop. The higher temperature and higher solar irradiance during premonsoon afternoon enhances the radiative heating of the mountain surface for which a stronger up-slope thermal mountain wind brings BC aerosols from the nearby lower altitude valley regions and plain land IGP regions. This probably inhibits a sharp drop in [BC] during premonsoon afternoon in spite of subdued vehicular activities and higher wind speed. The concentration of superfine particulate matters (< 1.0 μm) is found to be high due to their transport from low land regions driven by up-slope mountain wind during premonsoon afternoon as observed in our earlier study (Adak *et al.*, 2014) carried out over the same location, Darjeeling. This up-slope transport of superfine particles may be well correlated to the up-slope transport of BC aerosols as BC aerosols are confined mostly in this superfine size region of aerosols. Thus, upslope transport of BC aerosols by mountain wind is found to compensate the subdued vehicular activities during premonsoon afternoon. Thus we observed that the diurnal pattern of BC variation over Darjeeling was different from other Himalayan high altitude stations in India and Nepal. For example, the diurnal variation of [BC] over Manora Peak, Nainital (Pant *et al.*, 2006; Dumka *et al.*, 2010; Srivastava *et al.*, 2012 etc.), Mukteshwar (Hyvarinen *et al.*, 2009) and NCOP (Marinoni *et al.*, 2010) showed only afternoon peaks due to the uplift of BC aerosols from low land IGP regions as boundary layer deepens. Unlike Darjeeling, sharp morning and evening peaks were not observed over other high altitude stations. Rather, the diurnal variation of [BC] over Darjeeling shows similarity with that in various low altitude plain land regions in India with sharp morning peak followed by afternoon minima, sharp evening peak and minimum during night as also observed by Babu and Moorthy (2002) for Trivandrum, Tripathi *et al.* (2005) for Kanpur, Moorthy and Babu (2006) for Port Blair, Ganguly *et al.* (2006) for Delhi,

Safai *et al.* (2007) for Pune, Nair *et al.* (2007) for Kharagpur, Ramachandran and Rajesh (2007) for Ahmedabad, and Sreekanth *et al.* (2007) for Visakhapatnam and so on. This indicates that the impact of local emissions over Darjeeling is much higher than the other Himalayan high altitude stations and comparable to plain land urban atmospheres (also discussed later).

Long Range Transport of BC Aerosols

In order to investigate the transport of BC aerosols from long distances, we have computed 7 day air-mass back trajectories, arriving at an altitude of 500 m above ground level over Darjeeling for all the days on which BC were measured, using Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (http://www.arl.noaa.gov/ready/hysplit4.html). Over the entire period of study, we have identified six major source regions for long range transport as shown in Fig. 4. The frequency of transport from each of the source regions has also been shown in the figure. Region 1 corresponds to the transport from Nepal and Region 2 corresponds to IGP. The major source regions associated to IGP were northern states of India like Punjab and Uttar Pradesh. Region 3 corresponds to the transport from far western countries. The major and most frequent source regions under Region 3 are Middle East countries and the air masses are found to pass through south-west Asian countries like Afghanistan and Pakistan along with IGP. Some events have also been found when air masses originated from Pakistan and Afghanistan only and passed through IGP. Region 4 corresponds to the transport from eastern Indian states and the major region is found to be Assam. Region 5 corresponds to Bay of Bengal (BOB). Air masses in this case passed mainly through Bangladesh and sometimes West Bengal, India before reaching Darjeeling. Region 6 corresponds to Arabian Sea (AS), in which air masses passing through southern India, BOB and Bangladesh/West Bengal have longer marine overpass. Region 1–4 thus corresponds to the continental sources with the frequency of 61% and Region 5–6 corresponds to the mixed sources (marine + continental) with the frequency of 39%. Table 2 shows average [BC] associated to all these long range source regions for each seasons and the entire two-year study period. It is important to mention over here that the altitudes of the air masses were around

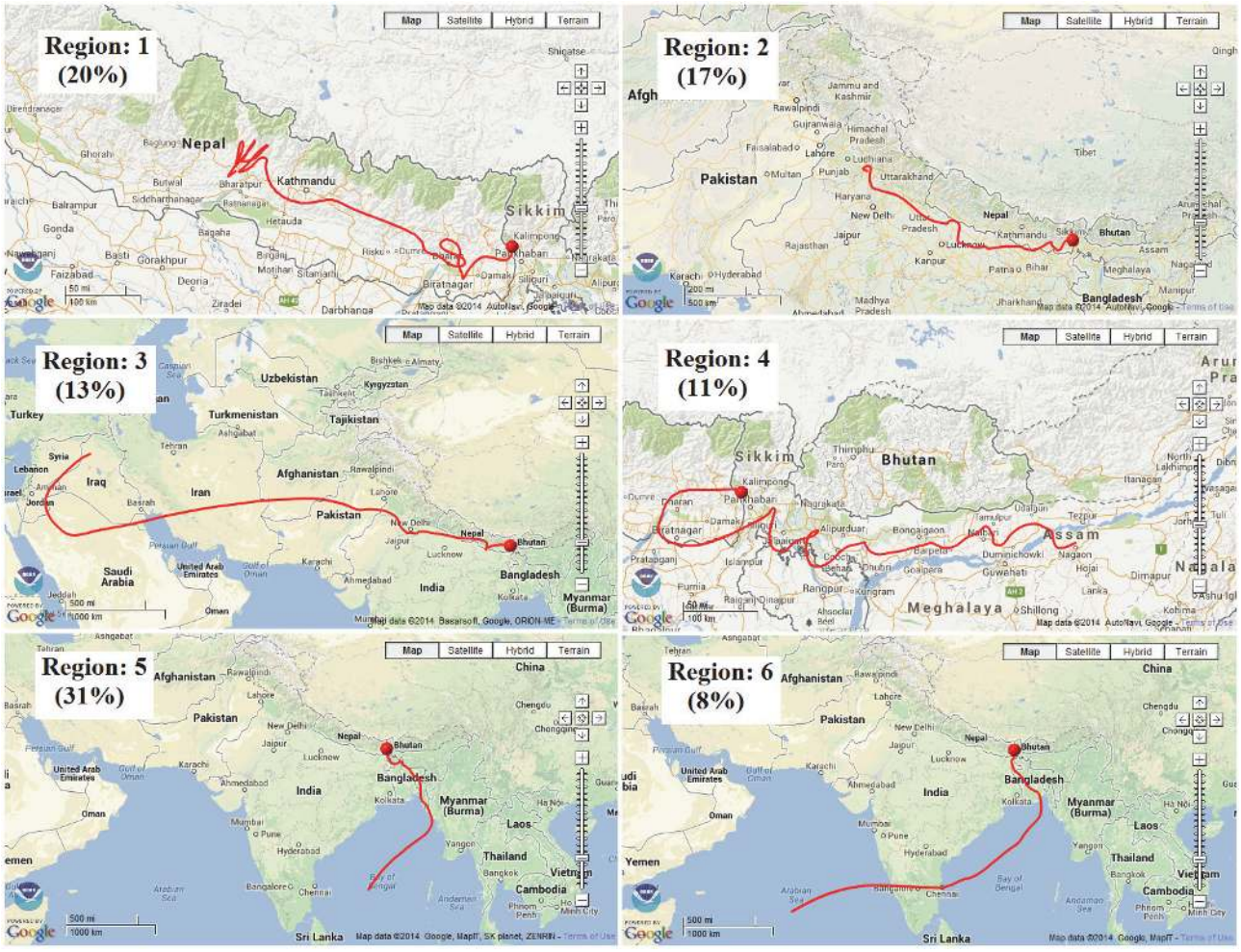

Fig. 4. Wind trajectories arriving at Darjeeling (500 m a.s.l) showing long distant source regions along with their frequencies over the entire two-year period of study. Region 1: Nepal; Region 2: IGP; Region 3: Middle East countries; Region 4: Eastern Indian states; Region 5: Bay of Bengal and Region 6: Arabian Sea.

Table 2. BC concentrations for various long distant source regions over different seasons and entire study period.

Seasons	Directions	Source regions	Frequency (%)	Average [BC]
Winter	W/NW	Nepal	48	4.6 ± 1.3
	W/NW	IGP	39	6.4 ± 1.1
	W/NW	Middle East and South West Asian Countries	13	6.2 ± 2.3
Premonsoon	W/NW	Nepal	13	6.4 ± 1.9
	W/NW	Middle East and South West Asian Countries	38	6.7 ± 1.6
	E	Eastern Indian States	22	2.9 ± 0.5
	S	Bay of Bengal	27	2.5 ± 0.4
Postmonsoon	W/NW	Nepal	45	3.5 ± 1.6
	W/NW	IGP	22	2.6 ± 0.8
	E	Eastern Indian States	33	2.1 ± 0.4
Monsoon	S	Bay of Bengal	75	2.9 ± 1.1
	SW	Arabian Sea	25	2.2 ± 1.2
Entire study	W/NW	Middle East and South West Asian Countries	13	6.5 ± 1.9
period	W/NW	IGP	17	4.5 ± 0.9
(2010–2011)	W/NW	Nepal	20	4.8 ± 1.6
	E	Eastern Indian States	11	2.5 ± 0.4
	S	Bay of Bengal	31	2.7 ± 0.7
	SW	Arabian Sea	8	2.2 ± 1.2

1000 m a.s.l. throughout their trajectories/pathways originating from their source regions. Thus the air masses could well interact with or pick up the boundary layer aerosols of the regions they passed over before reaching our observational site. It can be seen from the table that W/NW air masses (Region 1–3) dominated in all the seasons except monsoon (100% in winter, 67% in postmonsoon and 51% in premonsoon). Higher [BC] is found to be associated to IGP and Middle East countries in winter, Nepal and Middle East countries in premonsoon, Nepal in postmonsoon and BOB in monsoon. It was observed that during monsoon, [BC] associated to BOB was found to be higher than AS. The air masses originating from AS have much longer continental (Indian peninsula and Bangladesh/West Bengal) as well as marine (AS itself and BOB) overpass and should bring more continental polluted aerosols over Darjeeling. But aerosol wash out due to rain along these longer transport pathways could dilute the air mass and minimize BC loading. On the other hand, air masses originating from BOB comparatively have shorter continental and marine overpass where the effect of aerosol wash out is less and local/regional emissions are high. This could be the reason for slightly higher [BC] associated to BOB than AS. Overall our observation is that [BC] associated to the transport from W/NW directions (Middle East countries, Nepal, IGP) was much higher than E (eastern Indian states) and S/SW (marine/mixed sources; BOB and AS) directions. This could be due to the fact that the air masses originated from W/NW directions have much longer stretch of continental overpass and could pick up BC aerosols while passing over the south west Asian countries like Afghanistan, Pakistan and over most polluted IGP before reaching Darjeeling.

[BC] During a Strike Event: Assessment of the Contributions from Mountain Wind and Long Range Transport

A general strike was called by the local political party for 48 hours during 14–15 March, 2010 over Darjeeling. During the period, all the activities like vehicular, railway, schools, colleges, offices, hotels/resorts etc. had fully been ceased. Moreover, the night-time biomass burning is not a usual practice other than winter and postmonsoon season. Hence the strike event could be considered as a period of almost zero local emissions. We have used the [BC] during this strike event in order to investigate and estimate (roughly) the contributions of long-range and up-slope mountain wind.

It is observed that during premonsoon seasons, a total of 88 days including the two-day strike event are associated to the long range transport driven by W and NW winds. Thus, we got 86 days (excluding two strike days) with the contribution from long range transport (LRT), mountain wind transport (MWT) and local emissions (LE) and two (02) strike days with the contribution from LRT, MWT and no LE (LE = 0). We have compared the average diurnal variation of [BC] during strike event (MWT + LRT) and 86 normal days (LRT + MWT + LE) as shown in Fig. 5. It can be seen that morning and evening peaks are absent during strike event whereas those are present for other normal days. However, a broad peak during afternoon is present for both the normal days and strike event which could be associated to mountain wind transport as discussed earlier. The [BC] during the period 1300–1800 hrs (duration of broad peak) on strike event is due to the MWT and LRT contribution whereas [BC] during < 1300 hrs and > 1800 hrs is due to LRT only as LE is zero. Thus MWT contribution can be estimated as:

MWT = [BC](1300–1800 hrs) – [BC](< 1300 and > 1800 hrs) which gives us [BC] from MWT is 0.9 $\mu g/m^3$. The average [BC] during strike event is found to be 2.5 ± 0.5 $\mu g/m^3$ and thus contribution from LRT is (2.5–0.9) 1.6 $\mu g/m^3$. Now, we have used the [BC] from MWT and LRT on each of the 86 normal days to find out the contribution from LE. Thus [BC] from LE can be calculated as (([BC] from LRT + MWT + LE) from 86 individual days - (MWT + LRT)).

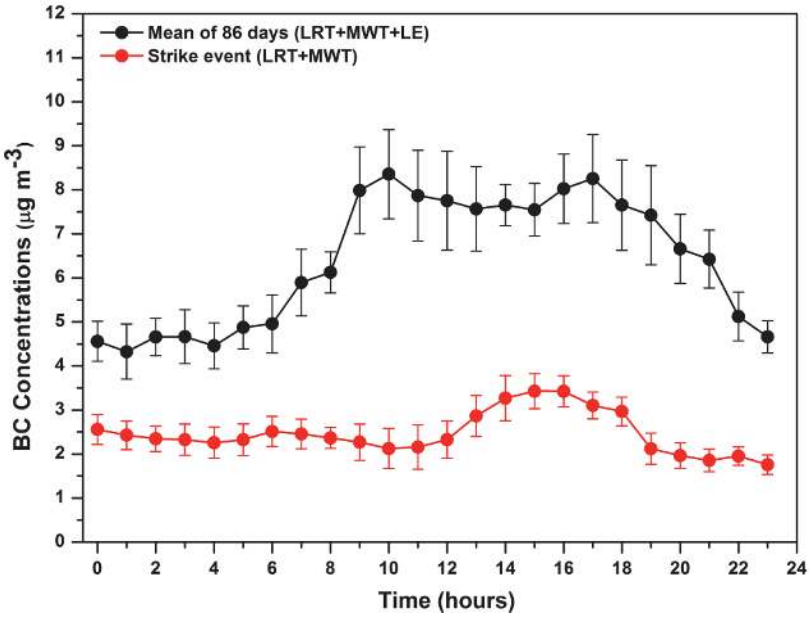

Fig. 5. Mean diurnal variation of BC concentrations during a two-day strike event (14-15 March, 2010) with long range and mountain wind transport and zero local emissions and 86 normal days with long range, mountain wind and local emissions during premonsoon 2010 and 2011.

It is observed that the average [BC] from LE was 3.9 ± 1.7 μg/m^3 (N = 86).

Overall, we have found that the MWT, LRT and LE contributed by 16.6 ± 6.4% (Range: 8.5–24%), 27.3 ± 7.9% (Range: 15–37%) and 55.5 ± 12.6% (Range: 38–68%) to the total [BC] respectively.

Effect of Wind Speed and Temperature on BC Concentration

In order to investigate the impact of wind speed on [BC], winds have been segregated into five categories based on the speed. They are 0–0.5 m/s, 0.5–1.0 m/s, 1.0–1.5 m/s, 1.5–2.0 m/s and above 2.0 m/s. The average [BC] corresponding to each of the wind categories have been estimated for two important seasons, premonsoon and winter, when the impact of wind speed on [BC] was very strong. In premonsoon, [BC] is found to gradually increase with wind speed and higher [BC] is observed at 1.5–2.0 m/s whereas in winter, [BC] is highest for 0–0.5 m/s and then gradually decreases with the wind speed. In winter, BC aerosols generated locally (vehicular and biomass burning) could blow away and disperse from the sampling site at higher wind speed and get accumulated under low and calm wind conditions. On the other hand, the reason for maximum [BC] at moderate wind speed during premonsoon could be due to the combined effect of local (favoured by low wind speed) and transported (favoured by higher wind speed) aerosols.

We observe a strong positive correlation (R^2 = 0.61) between ΔT (maximum – minimum temperature) and [BC] during premonsoon and weak correlation (R^2 = 0.13) during winter over Darjeeling. Chen *et al.* (2001) observed that the BC emissions, in particular, from heavy duty diesel engines, increase with increase in ambient temperatures. They suggested that this would become important during the dry months when the ambient temperature becomes higher. Lodommatos *et al.* (1998) demonstrated that increasing the temperature of the air entering diesel engines generally increases BC aerosols which could be due to the fact that hot air could favour diesel combustion. Human *et al.* (1990) showed that the air with low density over higher altitude stations increases particulate emission from diesel engines. The correlations between ΔT and [BC] over Darjeeling indicates the dominance of vehicular emission during premonsoon compared to winter. Higher correlation coefficient between [BC] and ΔT was also observed by several earlier studies in India (Babu and Moorthy, 2002; Ramachandran and Rajesh, 2007; Dumka *et al.*, 2010; Chatterjee *et al.*, 2012b).

Identification of Potential Sources of BC: Wavelength Dependence of BC

In order to investigate the relative influences of fossil fuel and biomass burning on the emission of BC aerosols over Darjeeling, we have performed a comparative analysis of [BC] measured at UV (370 nm) and near-IR (880) wavelengths. The ratio of [BC] at 370 nm to that at 880 nm ($R_{370/880}$) can be used as the indicator to find out the potential sources of BC qualitatively (Srivastava *et al.*, 2012b). The ratio greater than 1.0 indicates the dominance of biomass burning whereas less than 1.0 indicates the dominance of fossil fuel burning (Herich *et al.*, 2011). Some of the organic aerosol components from wood combustion could enhance the optical absorption at 370 nm wavelength relative to 880 nm (Park *et al.*, 2006). We observe that over Darjeeling, the ratio ($R_{370/880}$) was higher than 1.0 in winter (Range: 1.11–1.33; Av: 1.23 ± 0.07, N = 174), closed to 1.0 (Range: 0.87–1.05; Av: 0.96 ± 0.07; N = 119) in postmonsoon, less

than 1.0 in premonsoon (Range: 0.77–0.98; Av: 0.88 ± 0.08; N = 177) and monsoon (Range: 0.6–0.88; Av: 0.74 ± 0.11; N = 232). This indicates the dominance of biomass burning in winter and fossil fuel/vehicular emissions in premonsoon. As the biomass burning is inhibited under heavy precipitation, the ratio falls drastically during monsoon. Tiwari *et al.* (2013) found the ratio to be greater than 1.0 during winter and postmonsoon and less than 1.0 during summer (0.9) and monsoon (0.8) over New Delhi, India indicating the dominance of biomass burning during colder months. Srivastava *et al.* (2012b) estimated the percentage difference of [BC] at 370 and 880 nm wavelengths and found positive values during winter suggesting dominance of wood/biomass burning and negative values during spring suggesting dominance of fossil fuel burning over an urban atmosphere in Delhi and a high altitude station, Manora Peak at central Himalaya, India.

We have also calculated BC absorption coefficients at the operational wavelengths (σ_{abs}) using the following equation (Bodhaine, 1995; Weingartner *et al.*, 2003):

$$\sigma_{abs} = \frac{-1}{C.R}\left[\frac{A\ln\left(I_2 / I_1\right)}{\left(Q.\Delta t\right)}\right] \quad (3)$$

where I_1 and I_2 are the ratios of the intensities recorded by the detector for the sensing beam to the reference beam before and after each sampling interval of time Δt. The value of I_2/I_1 is given by $\Delta ATN = -100[\ln (I_2/I_1)]$, where ΔATN is the change in attenuation before and after the deposition of particles on the quartz filter tape. Q is volume of air sampled through the filter during interval Δt and A is the area of the spot where BC aerosols are collected. The wavelength dependent values C(λ) has been obtained from the study of Bodhaine (1995). R is an empirical correction factor which describes the change in instrumental response with the increase in particle loading. σ_{abs} is related to wavelengths (λ) by the following equation:

$$\sigma_{abs} = \beta.\lambda^{-\alpha_{BC}} \quad (4)$$

where α_{BC} is the wavelength exponent and β is particle loading.

Taking logarithm at both sides of the above equation, we get the following equation:

$$\ln(\sigma_{abs}) = \ln(\beta) - \alpha_{BC}\ln(\lambda) \quad (5)$$

α_{BC} is estimated from the slope by performing a linear regression of $\ln(\sigma_{abs})$ and $\ln(\lambda)$. Kirchstetter *et al.* (2004) has reported that the values of α_{BC} close to 1 represent BC from fossil fuel sources and those resulting from biomass burning would have much higher values (~1.8) and those mixed with dust would have still higher values (> 2). α_{BC} is found to be higher during winter (Range: 1.13–1.41; Av: 1.28 ± 0.1; N = 174) followed by postmonsoon (Range: 1.02–1.28; Av: 1.15 ± 0.09; N = 119), premonsoon (Range: 0.95–1.15; Av: 1.04 ± 0.07; N = 177) and monsoon (Range: 0.7–0.95; Av: 0.82 ± 0.09; N = 232). This could be taken as another indication of the relative dominance of fossil fuel emissions during premonsoon and biomass burning during winter. However the average value of α_{BC} is found to be 1.03 ± 0.09 over the entire study period. This suggests that BC aerosols over Darjeeling are mainly generated from fossil fuel sources. Dumka *et al.* (2010) found α_{BC} mostly around 1.0 (varying between 0.85 and 1.1) over Manora Peak at central Himalaya in India during November 2004 to December 2007. They suggested that BC aerosols observed at this site probably resulted from fossil fuel combustion.

Comparison of [BC] over Darjeeling with Other Stations

The [BC] over Darjeeling is compared with other high altitude stations in India and Nepal as well as several mainland cities in India. This is given in Table 3.

Comparison with Other High-Altitude Stations

The important feature emerging from Table 3 is that amongst all the high altitude stations, Darjeeling represents much higher polluted conditions. We observe that the average [BC] at Darjeeling is 20–40 times higher than Hanle, Nam Co and NCOP, Nepal. [BC] over Darjeeling is also found to be ~7 times higher than that in Langtang, Nepal. All these stations are situated at the higher elevations compared to Darjeeling (Table 3) and represent almost free tropospheric conditions with minimum or no anthropogenic activities. The most important feature is that Darjeeling shows 2–7 times higher [BC] than the hill stations with almost equal (Mukteswar and Nagarkot), slightly lower (Manora Peak, Nainital) and even much lower (Mt Abu and Sinhagad) altitudes (Table 3). This indicates the higher anthropogenic influences on BC pollution over Darjeeling compared to other high altitude stations which could be explained as follows:

1) The present study over Darjeeling and other earlier studies over western and central Himalaya have shown that aerosol transport from W/NW directions are most frequent during dry seasons. The impact of this W/NW transport is much higher over eastern Himalaya compared to other western and central Himalayan stations. Because, air masses arriving at eastern Himalaya from W/NW directions pass over almost all IGP regions covering west to east and thus could be enriched with more polluted aerosols. The air masses arriving at western and central Himalayan stations from W/NW directions pass only over western part of IGP and thus the influence of the central and eastern part of IGP regions is almost negligible.

2) The air masses from W/NW directions arriving at Darjeeling were also found to pass over densely populated Nepal and could pick up polluted aerosols from there. Earlier studies over central and western Himalaya (referred earlier) did not observe any air mass transport passing over Nepal.

3) The impact of local emissions over Darjeeling was also significantly higher than other Himalayan high altitude stations. This could be due to high anthropogenic emissions related to tourist activities, high population density and moreover it's unique orography and land use pattern with narrow roads, unplanned township and no open space/area which prevents ventilation and dispersion of aerosols.

Table 3. Comparison of BC concentrations between Darjeeling and other locations.

Station	Region	Altitude (m a.s.l.)	[BC] μg/m^3	References
High altitude stations				
Darjeeling	Eastern Himalaya	2200	3.45	Present study
NCOP, Nepal	Eastern Himalaya	5079	0.16	Marinoni *et al.*, 2010
Nam Co, Nepal	Eastern Himalaya	4730	0.08	Ming *et al.*, 2010
Hanle, India	Western Himalaya	4520	0.077	Babu *et al.*, 2011
Langtang, Nepal	Eastern Himalaya	3920	0.52	Carrico *et al.*, 2003
Nagarkot, Nepal	Eastern Himalaya	2150	1.0	Carrico *et al.*, 2003
Manora Peak, India	North-western Himalaya	1958	1.0	Dumka *et al.*, 2010
Mukteshwar, India	North-western Himalaya	2286	0.81	Hyvarinen *et al.*, 2009
Mt Abu, India	Western India	1680	0.5	Ram *et al.*, 2008
Sinhagad, India	Western India	1450	2.2	Safai *et al.*, 2007
Indian cities				
New Delhi,	Northern India		4–15	Singh *et al.*, 2010
Kanpur	Northern India		7.03	Nair *et al.*, 2007
Chandigarh	Northern India		3.7	Chowdhury *et al.*, 2007
Dibrugarh	North-eastern India		9.5	Pathak *et al.*, 2010
Kolkata	Eastern India		5.8	Chatterjee *et al.*, 2012(b)
Mumbai	Western India		12.4	Chowdhury *et al.*, 2007
Ahmedabad	Western India		2.1	Rastogi and Sarin, 2009
Pune	Western India		4.5	Safai *et al.*, 2007
Trivandrum	South-western India		3.44	Krishna Moorthy *et al.*, 2007
Bangalore	South-central India		3.21	Satheesh *et al.*, 2011

Comparison with Indian Cities

Darjeeling [BC] is found to be much lower (1.5–4.5 times) than [BC] observed over several cities like New Delhi, Mumbai, Dibrugarh, Kanpur, Kolkata and Pune. This is obvious as the all possible anthropogenic sources emitting BC aerosols are more pronounced over those urban atmospheres in mainland cities of India with much higher vehicular and industrial activities. But, the most interesting observation is that Darjeeling shows higher [BC] than the metro cities like Ahmedabad and Bangalore, whereas it shows almost equal and comparable concentration with cities like Trivandrum and Chandigarh.

SUMMARY AND CONCLUSIONS

Black carbon aerosol was measured over a high altitude station Darjeeling (2200 m a.s.l.) at eastern Himalaya for a period of two years (2010–2011) and the salient features and important conclusions of the present study are as follows:

1. BC concentration varied between 0.2–12.8 μg/m^3 with an average of 3.4 ± 1.9 μg/m^3 during the entire study period (January, 2010–December, 2011) over Darjeeling. BC exhibited strong seasonal variation with the maximum concentration during premonsoon (5.0 ± 1.1 μg/m^3) followed by winter (3.9 ± 2.2 μg/m^3), postmonsoon (2.9 ± 1.0 μg/m^3) and minimum during monsoon (1.7 ± 0.7 μg/m^3). The highest BC concentration during premonsoon was due to both the local (massive tourist activities) and long-range transported pollution. On the other hand, heavy rainfall resulted in minimum BC concentration during monsoon due to below-cloud scavenging of BC aerosols. BC showed strong diurnal heterogeneity with sharp morning and evening peaks during all the seasons which could be associated to local anthropogenic activities. The strong upslope thermal valley/mountain wind played a role in bringing BC aerosols from the nearby low land regions at the foothill of the Himalaya during premonsoon.

2. The major source of BC is found to be local fossil fuel burning (vehicular activities) during premonsoon and biomass burning during winter. This is supported by the ratio of [BC] at 370 nm to [BC] at 880 nm and wavelength exponent estimated from BC absorption coefficients at different wavelengths. However, their average values indicate the overall dominance of fossil fuel burning over Darjeeling throughout the year.

3. Six major regions were identified for BC transport from long distances; Nepal, IGP, Middle East countries, Eastern Indian states, Bay of Bengal and Arabian Sea. [BC] associated to the transport from Middle East countries has been found to be maximum followed by Nepal and IGP. We observed that mountain wind transport and long range transport have contributed significantly to the total BC concentration over Darjeeling in addition to the local emissions. During premonsoon, mountain wind transport, long range transport and local emissions were found to contribute ~17%, 27% and 56% respectively to the total BC concentrations. Overall, diurnal ambient temperature difference and wind speed were found to govern BC concentrations over different seasons. BC concentration during winter was favoured by low and calm winds (< 0.5 m/s) whereas that in premonsoon was favoured by moderate winds (1.5–2.0 m/s).

4. BC concentration over Darjeeling is found to be much higher than the other high altitude stations in India and Nepal and even higher and comparable with some of the Indian metro-cities like Ahmedabad, Bangalore, Trivandrum

and Chandigarh.

Thus we found that Darjeeling represents a typical urban atmosphere over eastern Himalaya in India. The high BC pollution over Darjeeling draws a serious attention as it could significantly increase the atmospheric heating rate over this part of Indian Himalaya. This two-year data set of BC can be used to estimate the direct and surface albedo radiative forcing, other aerosol optical properties and regional energy budget. This would, in turn, help us to make studies on the implications of BC aerosols for regional climate forcing over eastern Himalaya.

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