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Characteristics of black carbon over Delhi and Manora Peak – a comparative study

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Abstract

The characteristics of aerosol black carbon (BC) were studied at two different climatic regimes, i.e. Delhi and Manora Peak during winter and spring of 2007. Spring BC was found to be ∼59% lower at Delhi and ∼23% higher at Manora Peak than their corresponding winter BC. Diurnal BC variation showed two enhanced peaks at Delhi (morning and night) whereas a single late afternoon peak at Manora Peak. Delhi BC showed a clear correlation with prevailing winds whereas no correlation was observed at Manora Peak. The major contribution of BC at Manora Peak can be expected from biomass burning while at Delhi fossil fuel dominates. Copyright © 2012 Royal Meteorological Society

Keywords: black carbon; emission sources; boundary layer; wind speed; urban; Himalayan foothills

1. Introduction

The climatic and environmental effects of atmospheric aerosols are the critical issues in current global science community. There is a large uncertainty in the estimation of aerosol radiative forcing and hence in the assessment of global climate change as atmospheric aerosols are the complex mixtures of absorbing and scattering particles (Intergovernmental Panel on Climate Change – IPCC, 2007). In this context, black carbon (BC), the optically absorbing component of carbonaceous aerosols, is the major anthropogenic component of atmospheric aerosol system, which has been found to be one of the important contributors to current global warming (Ramanathan and Carmichael, 2008). In addition, BC is found to affect the monsoon in Asian countries (Wang *et al*., 2009 and references therein) and also produce heating of the elevated regions of the Himalayan–Tibetan (Lau *et al*., 2010 and references therein). Although BC constitutes only a few percent of the aerosol mass, it can have a significant positive forcing in the atmosphere (Srivastava *et al*., 2012a and references therein).

BC emission from India constitutes a large fraction of the total global BC burden and exhibits large spatio-temporal variability in their sources (biomass and fossil fuel) as well as their emission strengths because of varying degrees of land-use, transportation and agricultural practices (Rehman *et al*., 2011 and references therein). The characterization of BC is attracting considerable interest in recent years because of its significant contribution to the environment and climate studies as well as anthropogenic nature of its origin. Considering the key role they play in radiative forcing (Srivastava *et al*., 2012a), studies on BC aerosols have become an important topic. However, there are less data available on BC aerosols from the Indian region though a few studies have shown the characteristics of BC at different locations in India (Ramachandran and Rajesh, 2007; Dumka *et al*., 2010; Bano *et al*., 2011).

This study shows a comparative BC aerosol characteristics at two different climatic regimes at Delhi and Manora Peak during the winter and spring periods. Also, the potential source identification of BC and their long-range transport at both these stations have been done using satellite-derived fire counts data along with the air mass back-trajectory analysis, which has been substantiated using the absorption due to BC at two different wavelengths.

2. Experimental details and database

2.1. Description of the measurement sites

Measurements were carried out at Delhi (28.6 °N, 77.2 °E, ∼300 m amsl) and Manora Peak (29.4 °N, 79.5 °E, ∼2000 m amsl) during the winter and spring periods of the year 2007. Delhi is one of the densely populated and industrialized urban megacities in Asia, typically represents the plains of Ganga basin in the northern part of India. Delhi, being in the proximity to the Thar Desert region in the western India, experiences the influence of dust aerosols mostly during the spring. The general weather condition over the station is reported elsewhere (Singh *et al*., 2010). On the other hand, Manora Peak is one of the high-altitude and sparsely inhabited clean site in the Indian Himalayan foothills situated in the central

Himalayas at the aerial distance of ∼300 km in the northeast direction of Delhi. It is surrounded by sharply undulating topography of Himalayan mountain ranges in the north and northeast sides and low-elevated plain land merging into the Ganga basin in the southwest. Recent studies over the station have demonstrated the considerable influence of long-range transported aerosols from variety of sources and their radiative impacts (Kumar *et al*., 2011; Srivastava *et al*., 2011a). The general weather condition over the station is reported elsewhere (Dumka *et al*., 2010; Kumar *et al*., 2011).

2.2. Instrumentation and data analysis

Simultaneous measurements of BC aerosol mass concentration were carried out at both the stations using a seven channel Aethalometer (Model AE-42, Magee Scientific Company, USA). A total 72 days simultaneous BC data (with temporal resolution of 5 min), for 4 months period from January to February (a representative of winter period) and from April to May (a representative of spring period) in the year 2007 is used in this study. As the BC particles are in the fine-mode size with typical aerodynamic diameter of < 2.5 µm, the ambient air was drawn through a glass impinger inlet tube to have very small losses, with one end opening at the free air and the other attached to a low power exhaust fan. The instrument was connected through a suitable pipe without any dryer or heater.

Aethalometer measures BC mass concentration in the wavelength, ranging from 370 to 950 nm. It is a filter-based technique that measures light attenuation due to particles deposited on to a quartz filter. The measurement of the attenuation of light beam is linearly proportional to the amount of BC deposited on filter strip. Further details of the instrument and the measurement techniques can be found elsewhere (Hansen *et al*., 1984). It has been found that certain organic aerosol components of wood combustion have enhanced optical absorption at 370 nm wavelength relative to 880 nm (Wang *et al*., 2011). Hence, a difference between the measured BC at 370 and 880 nm channels can provide useful information on the potential sources of BC. This can be used as a qualitative indicator, particularly for wood smoke particles from the local wood combustion sources (Wang *et al*., 2011). However, BC mass concentration measured at 880 nm wavelength is considered to represent a true value of BC in the atmosphere as at this wavelength, BC is the principal absorber of light while the other aerosol components have negligible absorption (Bodhaine, 1995).

The uncertainty in BC estimations by aethalometer may arise due to reduction in the optical path in the aethalometer filter with an increased filter load, referred as the shadowing effect, and also due to multiple scattering of light in the quartz fiber matrix of the filter tape (Weingartner *et al*., 2003; Collaud *et al*., 2010). On the basis of several experiments, Weingartner *et al*. (2003) have reported that the shadowing effect is quite significant only in the case of pure soot (or BC) particles and it is almost negligible for the aged aerosols (i.e. mixtures of different aerosols), which is the probable case at both the stations in the present study. They have further suggested a correction factor of 2.14 for multiple scattering corrections; however, a value of 1.9 is used in the present case (as suggested by the manufacture based upon Bodhaine *et al*. (1992) and Bodhaine (1995)). In addition, the other uncertainties due to changes in sampling conditions and instrument noise may also be present but this will be considerably reduced in the present case as we are using average of 1 h from every 5 min of data collected. Considering all the factors, the overall uncertainty in the BC measurements was given approximately 20% (Singh *et al*., 2010). As the pump speed varied with altitude, to maintain the set flow rate in Aethalometer, BC measurements are also corrected following Tripathi *et al*. (2007) for the ambient temperature and pressure measured at both the stations.

3. Local meteorology of the stations

Figure 1 shows the meteorological conditions prevailed over both the stations during the study period of winter and spring 2007. Relatively high surface temperature was observed at Delhi as compared to Manora Peak almost throughout the study period (Figure 1(a)). The temperature at Delhi was found to vary from 9.1 to 35.0 °C during the entire study

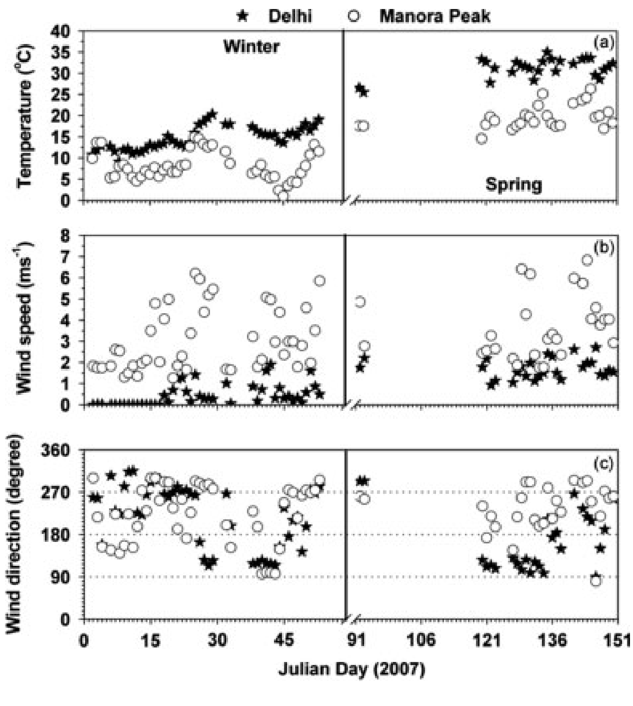

Figure 1. Julian day variations of (a) Temperature, (b) wind speed and (c) wind direction at Delhi and Manora Peak during the study period of winter and spring 2007.

period, with a mean values of 14.6(±2.7) °C in winter and 31.2(±2.3) °C in spring. However, at Manora Peak, temperature was found to be in the range from 1 to 26.3 °C, with a mean of 8.1(±3.4) °C in winter and 19.6(±2.8) °C in spring. Figure 1(b) shows daily mean wind speed (WS), which varies from 0 to 2.7 m s^{-1} at Delhi and 1.3 to 6.8 m s^{-1} at Manora Peak. Relatively higher WS was observed at Manora Peak as compared to Delhi. At Delhi, the mean WS was 0.4(±0.5) and 1.7(±0.5) m s^{-1} during winter and spring respectively, while at Manora Peak, it was 3.0(±1.5) and 3.7(±1.5) m s^{-1} respectively. The corresponding wind direction (Figure 1(c)) depicts that winds at Manora Peak are mostly south-westerlies during the entire study period, which are more pronounced during the spring. However, winds at Delhi are prominently south-westerlies during the winter and south-easterlies during the spring. As the Ganga basin region particularly Delhi is situated in the southwest to the Manora Peak, there is a possibility of transport of BC aerosol emissions at the high-altitude station at Manora Peak from these regions.

4. Results and discussion

4.1. Daily variations of BC

Daily BC mass concentrations measured with a temporal resolution of 5 min are averaged (for 24 h) at both the stations to get the daily mean BC mass concentration, shown in Figure 2. The daily mean BC concentration for the entire study period was found to vary between 4.2 and 70.5 μg m^{-3} at Delhi (with a mean of 23.5 ± 13.2 μg m^{-3} during the winter and 9.5 ± 3.0 μg m^{-3} during the spring), whereas it was found to be vary between 0.3 and 3.1 μg m^{-3} at Manora Peak (with a mean of 1.2 ± 0.8 μg m^{-3} during the winter and 1.4 ± 0.6 μg m^{-3} during the spring). The seasonal mean BC mass concentration of the present observations may be compared with

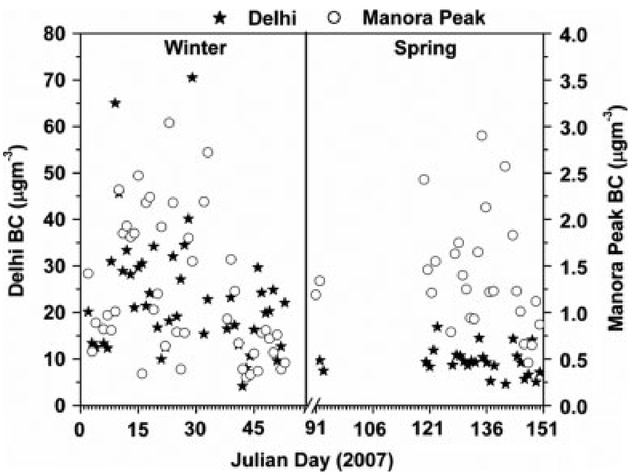

Figure 2. Julian day variation of BC mass concentrations at Delhi and Manora Peak during the study period of winter and spring 2007.

another observations at Delhi during the year 2006 made by Bano *et al.* (2011). They have observed the average BC concentration of 25.5 μg m^{-3} during the winter (December–February) and 9.4 μg m^{-3} during the summer (March–May). Although BC concentration at Manora Peak was observed to be much lower as compared with Delhi (as expected) during both the seasons, it was found to have significant contribution to the total aerosol optical depth and the resultant atmospheric forcing at Manora Peak (Srivastava *et al.*, 2012a). A clear decreasing trend in BC mass concentration was observed at Delhi from winter to spring period (Figure 2) where spring BC was found to be ~59% lower than the winter BC concentration. On the other hand at Manora Peak, spring BC concentration was found to increase by ~23% compared with the winter BC.

There are several reasons for the higher BC concentration over Delhi during the winter period, mainly associated with the prevailing meteorology over the station, which governs nature of the atmospheric boundary layer (ABL) and the WS, which is generally found to be low over Delhi during the winter period. The ABL also becomes highly stable in nature during the winter due to low surface convection. This does not allow aerosol particles to disperse much into the atmosphere and it gets trapped within the boundary layer near to the surface, as observed at Delhi. Apart from the prevailing meteorological conditions of the station, the potential sources of burning such as wood, waste and coal in and around the station are also responsible for the increased BC concentration over the station during the winter period (Khillare *et al.*, 2004). The concentration of soot also increases because of the transport of pollutants from thermal power plants by prevailing north westerlies over Delhi (Bano *et al.*, 2011). On the other hand, Manora Peak, being a high-altitude station, is mostly above the boundary layer during the winter when the temperature drops to a very low level and the thermal convection is weak (Pant *et al.*, 2006), resulting to a very low BC values. On the other hand, an increase in the vertical extent of ABL occurs because of strong convection during the summer, which tends to disperse the particulate matters up to the higher altitudes and reduce surface level aerosol concentrations (Srivastava *et al.*, 2012b). However, the observed high BC at Manora Peak during the spring may largely be associated with the long-range transport of the emissions from the Ganga basin (Kumar *et al.*, 2011).

The monthly mean BC along with the ambient temperature at both the stations is given in Table I. At Delhi, high BC concentration corresponds to the low surface temperature during the winter but reverse behavior is noticed during the spring. However, interestingly, different features were observed at the high-altitude station at Manora Peak where high BC corresponds to the high surface temperature during spring and relatively low BC corresponds to low surface

Table I. Monthly mean values of BC with corresponding ambient temperature and wind speed at Delhi and Manora Peak.

Months (2007)	Delhi			Manora Peak		
	BC (μg m^{-3})	Temperature (°C)	Wind speed (m s^{-1})	BC (μg m^{-3})	Temperature (°C)	Wind speed (m s^{-1})
January	27.61 ± 14.84	13.49 ± 2.72	0.27 ± 0.43	1.44 ± 0.72	8.89 ± 3.33	2.94 ± 1.58
February	17.25 ± 6.73	16.33 ± 1.48	0.68 ± 0.55	0.88 ± 0.67	6.90 ± 3.34	3.15 ± 1.31
April	8.81 ± 1.23	28.43 ± 4.25	1.91 ± 0.25	1.66 ± 0.68	16.56 ± 1.74	3.36 ± 1.31
May	9.55 ± 3.15	31.57 ± 1.83	1.68 ± 0.49	1.35 ± 0.59	20.02 ± 2.71	3.71 ± 1.55

temperature during the winter. Apart from the association of BC mass concentrations with the nature of ABL (associated with surface temperature), atmospheric winds may also play a key role in the transportation of BC from their various source regions.

4.2. Diurnal variations of BC and winds

Besides the daily variations, BC mass concentration also exhibits a pronounced diurnal variation, which is highly associated with the combined effect of variations in the production of BC, surface meteorology and the associated boundary layer dynamics (Ramachandran and Rajesh, 2007). Figure 3(a) shows mean diurnal variations of BC mass concentration at the urban station, Delhi, and at the high-altitude station, Manora Peak during the winter period. Two significant peaks in BC were observed at Delhi during the morning and night hours, which resembles with the previous measurements at other urban locations (Ramachandran and Rajesh, 2007 and reference therein). The morning peak, attaining at ~09:00 h local time (LT), is due to the morning build-up of local anthropogenic activities associated with the morning traffic activities. In addition to this, the morning peak might also be associated with the fumigation effect in the boundary layer, which brings aerosols from the nocturnal residual layer shortly after the sunrise (Stull, 1988). As the day advances, increased solar heating leads to an increase turbulent effect and a deeper boundary layer, leading to faster dispersion of aerosols and hence a dilution of BC concentration occurs near to the surface around the mid-afternoon (16 : 00 h LT). The BC mass concentration tends to increase from the mid-afternoon to reach the secondary peak at ~23:00 h LT, which can partly be attributed to the evening traffic rush (mainly heavy diesel vehicles). Also, it is highly associated with the shallow nocturnal boundary layer conditions and lower WS during night (Stull, 1988). An examination of meteorological data at Delhi shows that the diurnal variations of the WS reach a peak during the mid-afternoon, whereas WS during the morning and night hours is very low (Figure 3(b)). The well-developed boundary layer and increase in the WS are probably responsible for the observed low concentrations of BC in the mid-afternoon hours at Delhi. It is to be noted that BC is not totally lost from the atmosphere; it is only redistributed over a large spatial extent by the boundary layer dynamics.

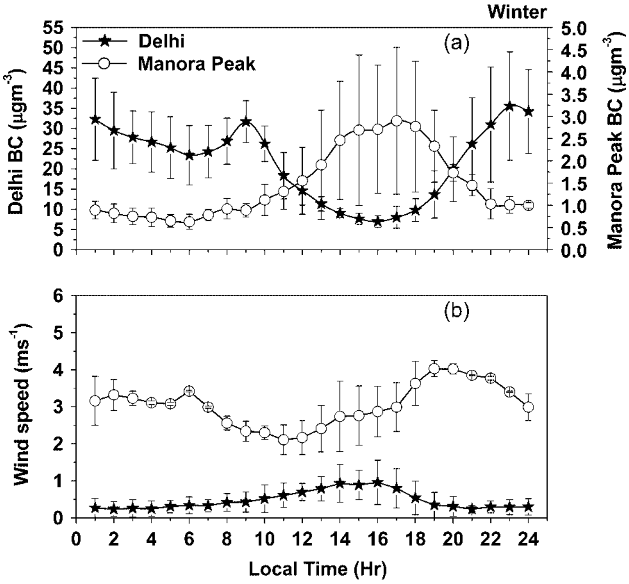

Figure 3. Diurnal variations of (a) BC mass concentration and (b) wind speed at Delhi and Manora Peak during the winter period.

On contrary to Delhi, a single enhanced peak was observed at Manora Peak in the late afternoon at around 17 : 00 h LT (Figure 3(a)). However, morning peaks are not apparent over the station, which are mainly due to the absence of locally generated pollution induced by anthropogenic activities. Similar feature in diurnal variation of BC has also been observed at other high-altitude site (Bhugwant *et al.*, 2001). In a previous study at Manora Peak, Pant *et al.* (2006) have also observed similar diurnal pattern of BC during the winter period of December 2004. Diurnality in the WS was also examined (Figure 3(b)); however, no clear association was observed with BC.

In order to examine the seasonal changes on the BC diurnal evolution at both the stations, the diurnal BC variations were also examined during the spring (Figure 4(a)) along with the corresponding WS (Figure 4(b)). Comparing Figures 3 and 4, it can be clearly seen that there are significant changes in the diurnal variations of BC and WS from winter to spring at both the stations. Although similar diurnal variations in BC were observed at Delhi during both the seasons, the magnitude of BC was found to be relatively lower during the spring as compared with the winter with less pronounced morning and night peaks. The reason could be (1) the well-developed boundary layer due to enhanced surface convection and

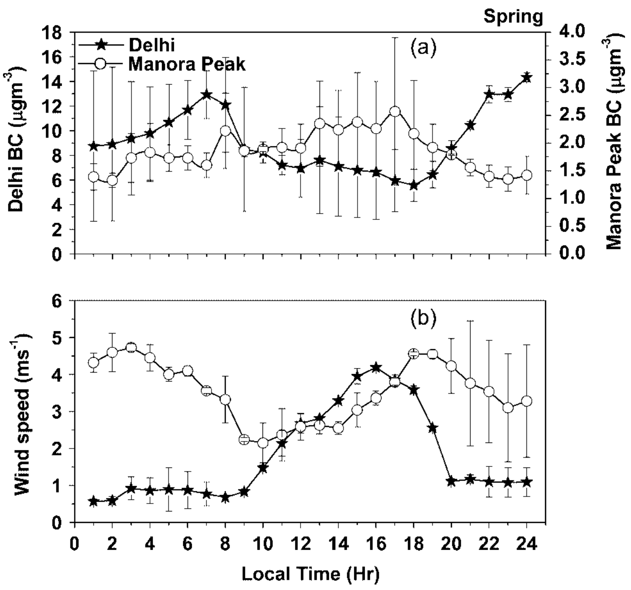

Figure 4. Same as Figure 3 except during the spring period.

(2) enhanced WS throughout as compared to the winter period, which is more pronounced during the mid-afternoon (Figure 4(b)). On the other hand, at Manora Peak, diurnality in BC gets diminished during the spring (Figure 4(a)), with relatively large magnitude of BC throughout, as compared with that observed during winter. The reason may likely be due to strong wind throughout the period (Figure 4(b)), which is more pronounced during the morning and late evening hours.

4.3. Effect of winds on BC and source identification

In order to understand the effect of winds on BC concentration, a correlation analysis between BC and WS has been done at both the stations. BC was found to be negatively correlated with the WS during the entire measurement period at both the stations, which is more pronounced at Delhi ($r^2 = 0.70$) as compared with Manora Peak ($r^2 = 0.20$). Results suggest that due to high winds, BC aerosols produced from local sources are transported to other locations, which was observed to be more significantly at Delhi during the spring ($r^2 = 0.40$). However, no clear correlation ($r^2 = 0.10$) was observed at Manora Peak between these two parameters during the spring. When the sources of BC are more localized then it is possible that it will give more negative correlation with the WS due to dispersion, whereas a less significant correlation between WS and BC mass may indicate relatively distant sources. As there is no close by sources of BC at Manora Peak, it is showing insignificant correlation with winds. On the other hand, Delhi with comparatively more localized sources is showing strong negative correlation. Similar features have also been reported earlier by Ramachandran and Rajesh (2007).

Biomass (particularly agricultural fires) and bio-fuel burning have been recognized as one of the major sources of BC aerosols from the northern part of India, particularly over the Indo-Gangetic Basin (IGB) region (Venkataraman *et al*., 2006). It is also important to note that the above sources in this region are colocated in close proximity of fossil-fuel burning sources. The crop residues burning is a common practice over the IGB region during spring and due to this open burning an abundance of biomass aerosols are generated in this region (Badarinath *et al*., 2009; Kumar *et al*., 2011). To substantiate the above, spatial distribution of fire counts over the region obtained from the moderate resolution imaging spectroradiometer (MODIS) during the corresponding study periods of winter and spring are shown in Figures 5 and 6, respectively. More details of the MODIS fire product and fire detection algorithm are given elsewhere (Justice *et al*., 2002). Significantly large density of fire counts was observed during the spring as compared with the winter period, and found to be more concentrated at the northwest part of India, indicating dominance of agricultural fire activities.

BC, being fine size and chemically inert aerosols (having atmospheric lifetime of about a week), may subsequently be transported to the great distances. Therefore, even in the absence of local sources, significant amount of BC aerosols could be found over the regions like Manora Peak, far-off from the potential sources. In order to ascribe the possible transport pathways of BC aerosols from their potential sources of different origins to the experimental sites, corresponding day's air mass back-trajectories were also analyzed using the hybrid single particle Lagrangian integrated trajectory (HYSPLIT) model of the National Oceanic and Atmospheric Administration (NOAA), USA (Draxler and Rolph, 2003). Five-day air mass back-trajectories were analyzed for Delhi (at 1000 m amsl) and Manora Peak (at 2500 m amsl), and superimposed upon the respective fire counts plot (Figures 5 and 6). It is quite discernible from the figures that air masses reaching at both the stations during the winter are mostly from the western side (Figure 5(a) and (b)). However, during the spring, two significant branches of air masses were found to be reaching at Delhi (Figure 6(a)): (1) first branch of air masses comes from the Thar Desert, which is the single largest contributor to the mineral dust aerosols over the station (Srivastava *et al*., 2011b) and (2) second branch of air masses comes from the northwest part of India where density of the fire locations (mostly agricultural fires) was found to be highest during the spring, which may be one of the major sources of BC aerosols over the station apart from the other local emission sources (e.g. vehicular, various industries etc). On the other hand, at Manora Peak, air masses were also found to be mostly from the northwest side during the spring (Figure 6(b)). The majority of fire activities in the northwest side, associated with the biomass burning sources, during the spring periods have already been

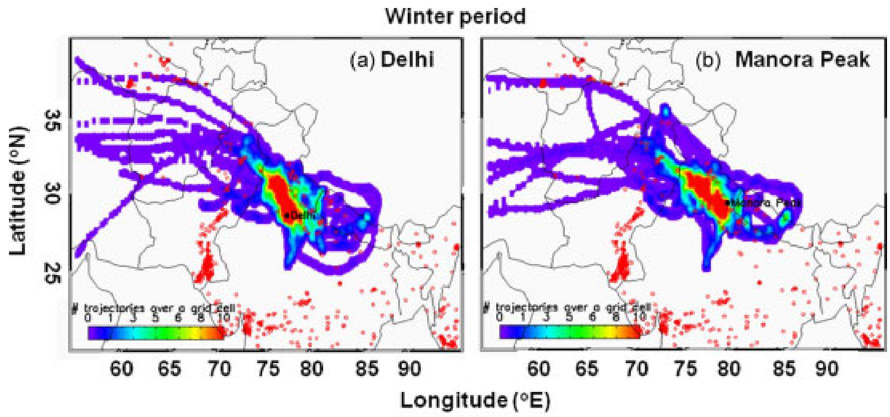

Figure 5. Spatial distribution of MODIS fire counts in the northern India during winter of 2007 for (a) Delhi and (b) Manora Peak superimposed along with the 5-day air mass back-trajectories. The locations of measurement are shown by dots over the region.

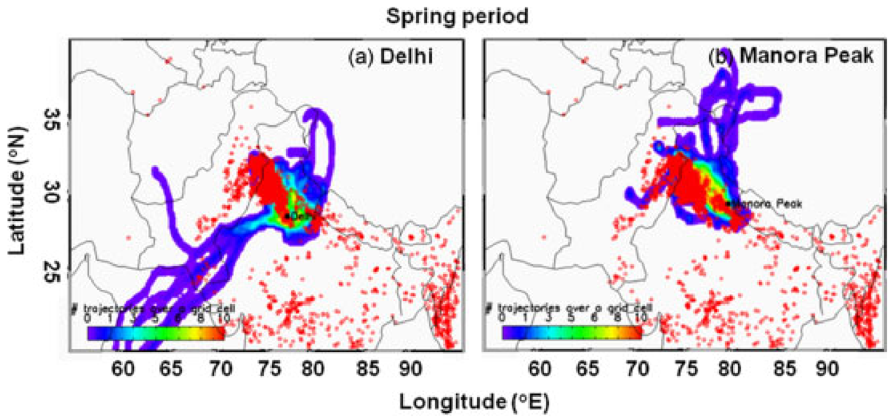

Figure 6. Same as Figure 5 except during the spring period.

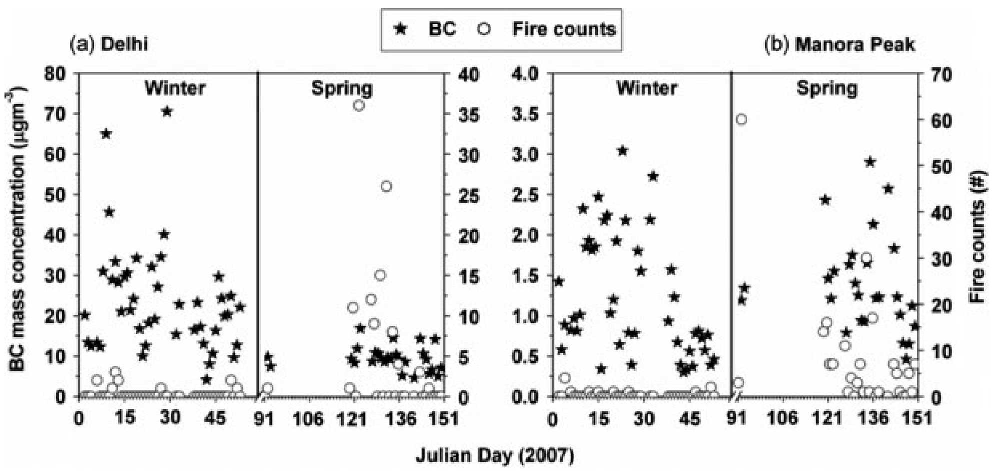

Figure 7. Variation of BC mass concentration and number of fire counts during different days of observations.

found to substantially impact the BC mass concentrations and various trace gases at Manora Peak (Kumar *et al*., 2011).

The influence of other emission sources such as bio- and fossil-fuel burning too cannot be neglected at these stations, particularly over Delhi during both the seasons. Figure 7(a) and (b) shows daily MODIS fire counts along with BC mass concentrations for Delhi and Manora Peak. The fire count data was obtained in a latitude and longitude bin of $2° \times 2°$, centered at Delhi and Manora Peak for both the seasons. The total fire counts of 12 were observed at both the stations during winter; however, much larger fire counts were observed at Delhi (127) and Manora Peak (209) during the spring. Figures clearly discriminate the possible BC sources at both the stations during these periods. A considerably high amount of BC concentration was observed at Delhi as compared with Manora Peak during both the seasons, which have been discussed in detailed in the previous section. However, BC concentration was found to be relatively lower at Delhi during the spring (when large fire counts observed) as compared with the winter. This may be possible because of the wind, which plays an important role in the transport of BC aerosols from the source region at Delhi, as also described in the previous section. On the other hand, opposite was observed during the winter at Delhi when very less fire counts were observed, and which could probably be due to the local emissions such as disposal of waste through burning of wood, dry leaves and some other solid materials by the population dwelling in the city slum areas to keep themselves warm during cold winter nights, combined with the ability of the region to trap these pollutions within the shallow and stable boundary layer.

To distinguish between the impact of local emissions from the above mentioned sources at both the stations, we have estimated the percentage difference of BC measured at two different channels [i.e. $(BC_{370} - BC_{880})/BC_{880}$]. Figure 8 shows daily variations of such fractional values for Delhi and Manora Peak. The positive values suggest significant contribution from wood burning emissions to BC (Wang *et al.*, 2011), which is the case observed at both the stations during winter, particularly in the month of January, with relatively higher contribution at Delhi as compared with Manora Peak. On the other hand, the negative fractional BC values suggest the dominant contribution from fossil-fuel combustion to BC (Herich *et al.*, 2011), which is the prominent case observed at Delhi during the late winter (February) and spring season. On the other hand, at Manora Peak, the fractional BC values are found to be near and very close to zero (mostly in positive side), which suggest that being a high-altitude and remote station, the contributions from fossil-fuel burning emissions to BC are relatively low. While the major contribution at Manora Peak can be expected from biomass burning particularly from agricultural fires, which has also been discussed earlier (Figure 7), it is also found to be one of the sources at Delhi during the spring period.

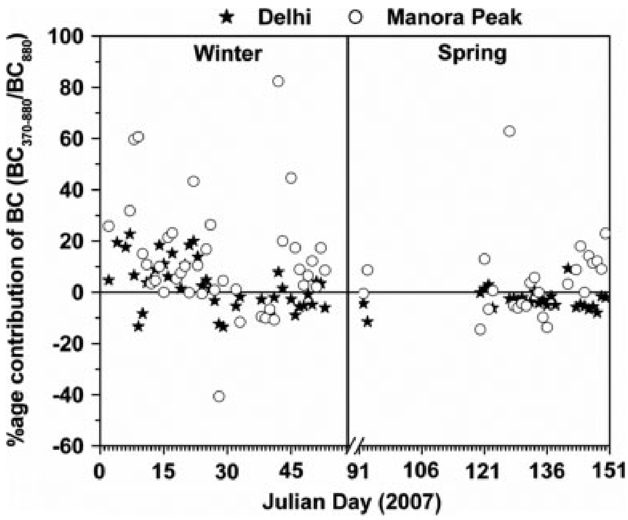

Figure 8. Fractional contribution of BC measured at 370 and 880 nm at Delhi and Manora Peak during winter and spring season of 2007.

5. Conclusions

A comparative study on the characteristics of BC aerosols was carried out at Delhi – an urban polluted station and Manora Peak – a clean high-altitude station in the Indian Himalayan foothills during the winter and spring seasons of 2007. The salient results are:

- Daily mean BC mass concentration at Delhi during the spring was found to be ~59% lower than the BC observed during the winter period. However, it was found to be ~23% higher at Manora Peak from spring to winter.
- Interestingly, different diurnal variations in BC were observed at both the stations, with two significant peaks (morning and night) at Delhi and a single late afternoon peak at Manora Peak, which are largely associated with the local boundary layer dynamics and the emission sources at both the stations.
- The wavelength dependence of BC observations at the two stations indicates that while the major contribution of BC at Manora Peak can be expected from biomass burning, the fossil fuel is the dominating contributor at Delhi.

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