Nat Hazards (2013) 65:1745–1764
DOI 10.1007/s11069-012-0449-1

ORIGINAL PAPER

Assessment of carbonaceous aerosol over Delhi in the Indo-Gangetic Basin: characterization, sources and temporal variability

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Received: 13 January 2012 / Accepted: 1 October 2012 / Published online: 14 October 2012

Abstract Semi-continuous measurements of organic carbon (OC) and elemental carbon (EC) and continuous measurements of black carbon (BC) and $PM_{2.5}$ aerosols were conducted simultaneously during the winter period of 2010–2011 at Delhi, one of the polluted urban megacities in western part of the Indo-Gangetic Basin region. The average mass concentrations of OC, EC, BC and $PM_{2.5}$ were about 54 ± 39, 10 ± 5, 12 ± 5 and 210 ± 146 μg m^{-3}, respectively. Contribution of total carbonaceous aerosol mass to $PM_{2.5}$ mass was found to be ~46 %. Average OC/EC ratio was found to be 5 ± 2 during the study period, suggesting the presence of secondary organic aerosols in the atmosphere over Delhi. Estimated mean secondary organic aerosol mass concentration was found to be 25 μg m^{-3} and varied between 14.6 (February) and 37.0 μg m^{-3} (December). A diurnal variation of OC and EC shows lower values during the day time and higher during the morning and night, which are highly associated with the corresponding variability in mixing layer heights. OC and EC were also found to be significantly correlated ($r = 0.71$) to each other, indicating their common sources. Concentrations of OC and EC were about 45 and 13 % higher during weekdays than weekends, respectively. Higher OC (67 %) and EC (53 %) were observed in the late evening during weekdays than those on weekends, which could be due to different emission sources during these two periods. The night/day ratio of EC and OC was found to be larger than 1.0, suggesting the relative accumulation of EC and OC near the surface at night hours.

Keywords Carbonaceous aerosol · Organic carbon · Elemental carbon · Black carbon · $PM_{2.5}$ · Indo-Gangetic Basin

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1 Introduction

Anthropogenic aerosols are typically composed of various inorganic and organic species including carbonaceous component. Carbonaceous aerosol, which consists of organic carbon (OC) and black carbon (BC) or elemental carbon (EC), are of the major research interest due to their ubiquitous nature and abundance, particularly in fine-mode (<2.5 μm) (Lonati et al. 2007; Dutkiewicz et al. 2009). On the other hand, OC is either emitted directly from sources (primary) or formed from the condensation of low volatility products of the oxidation of hydrocarbons (secondary). BC (or EC), produced by incomplete combustion of carbonaceous fuels, is an important component of the atmospheric aerosol because of its light absorbing characteristics (Bond and Bergstrom 2006) and possible health effects (Kim et al. 2003). EC (measured with thermal methods) or BC (measured with optical methods) can differ in inter-comparisons by factors of three or four depending on aerosol characteristics (Reisinger et al. 2008). Being highly absorbing and warming nature of these carbonaceous aerosols in the lower atmosphere, they play crucial role in the estimation of optical properties and direct radiative forcing on the regional to global scale (Haywood and Shine 1997). Furthermore, the relative amounts of OC and EC in the atmosphere and OC/EC ratios are the important parameters for the assessment of direct/indirect impacts of aerosols on the regional scale radiative forcing (Novakov et al. 2005).

Quantification of the carbonaceous content of the ambient aerosols on the basis of single species is a difficult task due to their chemical and physical properties, numerous individual molecular species and the complex aerosol matrix. Thus, operational concept of bulk carbonaceous material, such as OC and EC, has been established for their quantification in the ambient air. They scatter and absorb the short wave and long wave solar radiation (direct effect), thereby influencing the radiative balance of the Earth-atmosphere system (IPCC 2007). Nevertheless, EC could be the next most important contributor to global warming, in terms of direct forcing, after CO_2 (Jacobson 2001; Oen et al. 2006; Ramanathan and Xu 2010). The surface forcing due to absorbing aerosols is about 2–3 times larger than the forcing at the top of the atmosphere, thus producing a large atmospheric warming (Ramanathan and Carmichael 2008).

In India, the relative contributions of fossil fuel combustion, open biomass burning and bio-fuel combustion to BC were estimated to be 25, 33 and 42 %, whereas those to OC were to be 13, 43 and 44 %, respectively (Venkataraman et al. 2005). Although BC was studied variously by using optical methods (Babu and Moorthy 2001, 2002; Latha and Badrinath 2005; Tripathi et al. 2005; Ramachandran and Rajesh, 2007; Safai et al. 2007; Sreekanth et al. 2007; Dumka et al. 2010; Bano et al. 2011), very less measurements of carbonaceous components have been reported in the Indian subcontinent, particularly over the Indo-Gangetic Basin (IGB) region (Satsangi et al. 2010; Ram and Sarin 2010; Ram et al. 2010a). The region is of great research interest due to its unique topography and high population, as well as industrial density (Srivastava et al. 2011). Aerosols over the IGB region are highly associated with the emissions from various anthropogenic and natural sources (Reddy and Venketaraman 2002a, b; Rengarajan et al. 2007; Tiwari et al. 2009; Ram and Sarin 2010), which are found to have considerable radiative impacts over the region (Srivastava et al. 2012a) and are also identified as being comprised of different types (Srivastava et al. 2012b). Recently, Rehman et al. (2011) have conducted simultaneous atmospheric measurements inside rural households, ambient air and vehicular emissions from highways in a rural area in the IGB region under the project "Surya". They found that cooking with solid biomass fuels is a major source of ambient BC over IGB. Also, they reported that BC peaked during early morning hours (0500–0800 h) and early

evening hours (1700–1900 h) with the peak values of about 100 μg m^{-3}, which is about a factor of 10–30 larger than the day time values.

The aforementioned studies and facts, motivated us to present the results of our intensive EC–OC measurements along with BC and $PM_{2.5}$, conducted during the winter season (November 2010 to February 2011) at a representative urban site at Delhi in the IGB region. For the first time, near real-time semi-continuous measurements of OC and EC and continuous measurements of BC and $PM_{2.5}$ mass concentrations were conducted simultaneously at Delhi. The present study is focused on characterization of carbonaceous aerosols and to examine their temporal variability, aiming to identify the influence of meteorological versus general source emission variations. Also, the present study is focused on to establish a relationship between OC and EC, as well as secondary organic carbon (SOC) formation.

2 Experimental site and instrumentation

All measurements were conducted in the premises of Indian Institute of Tropical Meteorology, which is located in central Delhi (28.6°N, 77.2°E, ~240 above mean sea level) and represents a residential urban location. The population density of Delhi is ~11,297 km^{-2} in 2011, having 16.7 million inhabitants spread over 1,484 km^2. It has a sub-tropical climate with extremely hot temperatures during summer (maximum up to 44 °C) and moderately cold (minimum up to 2 °C) in winter. The relative humidity (RH) varies from 40 % in April to 80 % in August (Tiwari et al. 2009). The station is situated on western part of the Gangetic plains in northern India, which is covered by the Himalayan ranges in the north and Thar Desert in the west and influenced the climatic conditions of Delhi. The area around Delhi falls on the border zone lying between the rich rain-washed Ganges plains to the east and the semi-arid tracts of Rajasthan about 160 km to the west and southwest. During most of the year, a veil of white or brown haze hangs in the air blurring the views. Scenario becomes worst during frequent dust storm in summer time and also during foggy conditions in winter.

Sampling of OC, EC, BC and $PM_{2.5}$ were carried out during the winter period from November 2010 to February 2011. The OC and EC analysis was performed with a custom made thermo-optical transmission instrument by Sunset Laboratory Inc., USA (Model 4L) as per the standard method (Bauer et al. 2009). The particulate samples were collected using a predetermined sample schedule and analyzed by a thermal-optical method. A pair of quartz fiber membrane filters (Whatman-1851-047QMA) stacked back to back, located inside a quartz glass oven chamber. The filter surface area was 17 mm with flow path from the cyclone to the instrument was via 3/8 inch stainless steel tubing. The cyclone is installed vertically with the rain hat up to prevent any water entering into the sample lines. A charcoal impregnated filter strip denuder or carbon monolith denuder was installed in the flow path to remove (or reduce) organic vapors from the sampled air. The aerosol carbon analyzer was operated continuously on hourly cycles, which consisted of about 45-min aerosol collection period followed by 15-min analysis period. During the analysis, particulate carbon on the quartz filter was thermally converted to CO_2 using MnO_2 and detected downstream by non-dispersive infrared spectroscopy (Rattigan et al. 2010). An aliquot of sample filter was stepwise heated in a furnace up to 820 °C in a non-oxidizing atmosphere (100 % He); furnace is then cooled to 550 °C and filter is stepwise heated to 870 °C in an oxidizing atmosphere (98 % He, 2 % O_2). During each temperature step, evolved carbon is converted to methane and detected by a flame ionization detector.

Continuous monitoring of laser light transmission at 660 nm through the loaded filter was used to determine the split between the EC and OC fractions. The laser light transmission also provided optical measurements of EC (optical EC) on the quartz filter. Each hourly analysis of EC and OC was calibrated by automatically injecting a fixed volume of an external gas standard consisting of 5 % CH_4 in helium through the hot oven. The calculated detection limit for OC was 0.26 μg m^{-3}, and the detection limit for EC was 0.04 μg m^{-3} (using EC signal of 0.2 μg cm^{-2} as the instrumental detection limit).

BC mass concentrations were measured simultaneously using Aethalometer (Model AE-31-ER, Magee Scientific, USA), with a temporal resolution of 5 min. In this method, atmospheric air is pumped through an inlet at the desired flow rate, which impinges on a quartz micro fiber strip. A light beam from a high-intensity LED lamp is transmitted through the sample deposit on the filter strip, at 880 nm. The measurement of the attenuation of light beam is linearly proportional to the amount of BC deposited on filter strip. The precision of BC measurement is greatly dependant on the stability of the flow rate, which was kept within a small range of 3.8–4.1 l min^{-1} during the study period (Snyder and Schauer 2007). As the BC particles are in the fine-mode size, 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.

Several reports are available in the recent literature on the uncertainties involved in the Aethalometer estimated BC (e.g., Weingartner et al. 2003; Arnott et al. 2005; Schmid et al. 2006; Virkkula et al. 2007). The uncertainty in BC may arise due to shadowing effect, which is reduction in the optical path in the Aethalometer filter with an increased filter load, and also due to multiple scattering of light in the quartz fiber matrix of the filter tape (Weingartner et al. 2003). In the recent study, Collaud et al. (2010) have proposed a new correction scheme based on previously reported methods, which accounts for the optical properties of the aerosol particles embedded in the filter. While the multiple scattering effect tends to overestimate the BC concentration, the shadowing effect may lead to an under estimation in the BC concentration. Based on several experiments, Weingartner et al. (2003) have found that the shadowing effect is quite significant for pure soot particles (or BC) while almost negligible for aged aerosols (i.e., mixture of different aerosols). Considering all the above corrections, the uncertainty in the estimation of BC mass concentration was given by 2–5 % of the measured values (Babu et al. 2004; Dumka et al. 2010).

Further, the data of $PM_{2.5}$ were carried out using real-time beta attenuation particulate monitor, manufactured by a sampler from Thermo Andersen, USA, Inc., Series FH 62 C14 (C14 BETA). The FH 62 C14 particulate sample collection area is located between both the C14 source and the proportional detector. While ambient particulate matter is being deposited onto a filter tape sample spot, the dynamic filter loading is measured continuously by the attenuation of the C14 source beta rays. It is not necessary to move the filter spot from the sample position to the detector position for zero and mass determination. The more details of the above instruments are given elsewhere (Hyvärinen et al. 2009).

3 Meteorological condition over the station

The meteorological condition of the station is discussed with the daily data obtained from the NOAA Air Resources Laboratory web server (http://www.arl.noaa.gov/ready.html). Day-to-day variations in temperature (Temp in °C), boundary layer mixing depth in terms of mixing height (MH in m) and wind speed (WS in ms^{-1}) over the station were shown in Fig. 1a–c during the study period from November 2010 to February 2011. A consistent

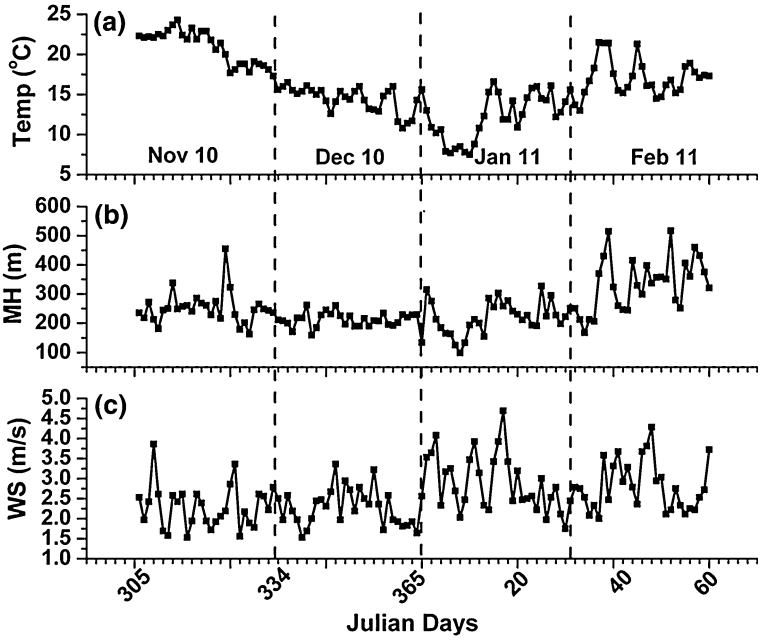

Fig. 1 Day-to-day variations of **a** Temperature; Temp, **b** mixing height; MH and **c** wind speed; WS over Delhi during the study period

decrease in temperature was observed from November to about mid-January with low day-to-day variability, from where it was found to increase with relatively large variability. Temperature was found to be varying from 7.5 to 24.3 °C during the entire study period over Delhi, with a mean ~16 (±4) °C. Mixing height over the station was positively correlated with the temperature (r ~0.4) and found to be varying from 99 to 517 m, with a mean ~250 (±77) m. It was found to be relatively more stable (with low day-to-day variability) during the month of December as compared to the other months. On contrary to the above parameters, wind speed over the station shows its variability from 1.5 to 4.7 ms^{-1}, with a mean of 2.6 (±0.65) ms^{-1}. The monthly mean values of Temp, MH and WS at Delhi during the study period are given in Table 1.

4 Results and discussion

4.1 Daily characteristics of carbonaceous species (OC, EC, BC) and $PM_{2.5}$

Summary statistics of mass concentrations of OC, EC, TC, OC/EC, BC and $PM_{2.5}$ are presented in Table 2 for the entire measurement period in winter. The daily mean concentrations of OC, EC, BC and $PM_{2.5}$ are shown in Fig. 2a–d, respectively which shows large day-to-day variations in all the species. The average concentrations of OC and EC were found to be 54.1 ± 39.4 (ranged from 13.2 to 247.8 μg m^{-3}) and 10.4 ± 4.6 μg m^{-3} (ranged from 2.6 to 25.4 μg m^{-3}), respectively. The general meteorology of the region during the winter is dominated by the high pressure usually centered over Western China, causing increased atmospheric stability, which in turn allows for less general circulation

Table 1 Monthly mean temp, MH and WS at Delhi during the study period

Months	Temp (°C)	MH (m)	WS (ms^{-1})
November 2010	20.7	249.7	2.3
December 2010	14.4	213.2	2.3
January 2011	12.2	219.0	2.9
February 2011	17.2	337.0	2.8

Table 2 Daily statistics of OC, EC, TC, OC/EC, BC and $PM_{2.5}$ mass concentrations at an urban location Delhi during winter season of 2010–2011

	OC ($\mu g\ m^{-3}$)	EC ($\mu g\ m^{-3}$)	TC ($\mu g\ m^{-3}$)	OC/EC	BC ($\mu g\ m^{-3}$)	$PM_{2.5}$ ($\mu g\ m^{-3}$)
Mean	54.1	10.4	64.5	5.2	11.8	209.6
Std. deviation	38.7	4.6	42.5	2.2	5.3	145.5
Minimum	13.2	2.6	16.9	1.6	2.4	51.3
Maximum	247.8	25.4	273.1	10.8	25.0	1,257.9

and thus more stagnant air masses to results of more accumulation of pollutants. The atmospheric dispersion is typically at a minimum, and therefore, the pollutants will not be widely dispersed throughout the planetary boundary layer. The highest concentration (39.5 $\mu g\ m^{-3}$) of EC was found in the month of November, which could be due to burning of crackers during Diwali festival, celebrated on 05th November 2010.

Daily mean BC mass concentration was found to be varied from 2.4 to 25.0 $\mu g\ m^{-3}$ with a mean of 11.8 ± 5.3 $\mu g\ m^{-3}$ at Delhi during the winter period. Frequency distributions of BC from November 2010 to February 2011 were grouped into six different intervals as <5; 5–10; 11–15; 16–20; >21 $\mu g\ m^{-3}$ and found to be ~10, 39, 24, 24 and 4 %, respectively. Maximum BC frequency (~39 %) over the station was observed for BC mass concentration ranging between 5 and 10 $\mu g\ m^{-3}$ during the study period. Coal burning is major energy source in Delhi having three thermal power plants in and around Delhi (Ali et al. 2004). Apart from this, biomass burning is also a large carbon emitter in this region during the winter (Singh et al. 2008). In a recent study, Srivastava et al. (2012c) have analyzed BC aerosols at two different wavelengths (at 370 and 880 nm) coupled with air mass back-trajectory and fire count data to discriminate the potential BC sources from biomass and fossil fuel combustions. They have reported the major contribution of BC over Delhi is from fossil fuel combustion sources, whereas biomass burning is the dominant source during the winter period.

EC, which is primarily measured by the thermal–optical method whereas BC-a light absorbing carbon (LAC), is measured by optical method, often equated with EC due to its dependent on the chemical and physical properties utilized to measure each class of carbonaceous aerosols. BC represents the fraction of carbonaceous material that absorbs visible light; however, EC is graphitic carbon that absorbs in the visible range and is not reduced to CO_2 when heated to 800 °C in an inert atmosphere (Andreae and Gelencsér 2006). In principle, all EC is BC, but all BC is not necessarily EC. Aethalometer-based BC values were found to be ~17 % higher than those measured with EC–OC analyzer. The difference between BC and EC mass concentrations can be understood based on their definition and measurements using different analytical techniques. In a recent study at

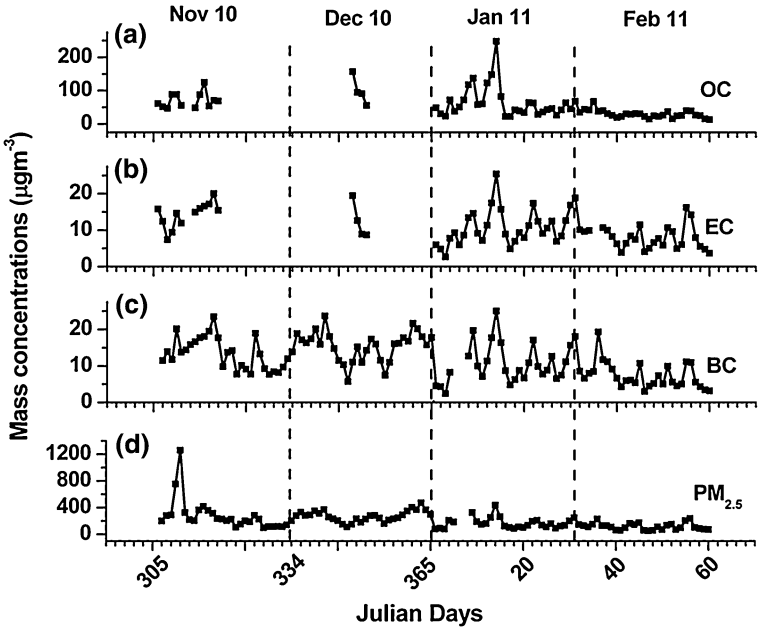

Fig. 2 Day-to-day variations of **a** OC, **b** EC **c**, BC and **d** $PM_{2.5}$ over Delhi during the study period from November 2010 to February 2011

Kanpur—another polluted urban station situated ~400 km south east of Delhi in the IGB region, Ram et al. (2010b) have found BC mass concentration ~20 % higher than that of EC. Further in the present study, the ratio and regression analysis (r) between BC and EC was found to be 1.13 and +0.90, respectively. Relatively large BC concentrations over the station may be attributed to the enhanced biomass burning activities during the period in and around the station (Srivastava et al. 2012c).

The mean mass concentration of $PM_{2.5}$ was found to be 209.6 ± 146.1 μg m^{-3} (ranged from 51.3 to 1,257.9 μg m^{-3}) during the study period, which is substantially higher and far excess of its annual mean Indian National Ambient Air Quality Standards (NAAQS) ($PM_{2.5}$ = 40 μg m^{-3}) and also US-EPA NAAQS ($PM_{2.5}$ = 15 μg m^{-3}) levels. Further, if we compare the same with its daily NAAQS level (24-h) of India ($PM_{2.5}$ = 60 μg m^{-3}), approximately 98 % of $PM_{2.5}$ samples were found to exceed with the daily mean NAAQS. In a recent study, similar values were obtained for $PM_{2.5}$ during the winter (190 ± 109 μg m^{-3}) at Gual Pahari, close by station to New Delhi, where as much as 87 % of days were found to be exceeded the daily Indian NAAQS level (Hyvärinen et al. 2011). However, the level of $PM_{2.5}$ in Delhi is comparable to Beijing, China (115 μg m^{-3}), reported by He et al. (2001). In another study, Watson et al. (2002) have reported the $PM_{2.5}$ concentration of ~138 μg m^{-3} during wintertime at the California's San Joaquin Valley, USA which is lower than that of Delhi. Accumulation of particles especially in fine size due to thermal inversion is the major cause for these high concentrations of $PM_{2.5}$ over Delhi during the winter period, and the source for these particles could mostly be from the local emission sources such as combustion of biomass, bio and fossil fuels. Western disturbances were also identified as other contribution from the long range transported sources over the station during this period, which brings air masses rich with pollutants

from other parts of India (in the western side) and the surrounding western countries (Awasthi et al. 2011). The mean contribution of OC in $PM_{2.5}$ mass was found to be ~26 %, ranging from ~25 % (February) to ~37 % (January) during the study period, which is approximately five times higher than the EC mass fraction to $PM_{2.5}$ (i.e., ~5 %). The BC contribution to $PM_{2.5}$ mass was found to be similar to that of EC. The highest mean concentration of $PM_{2.5}$ (1,257 μg m^{-3}) was also observed on November 05, 2010 that can be attributed to the burning of fire crackers due to Diwali. On this day, the mixing height was found to be low (~150 m), accompanied with the surface temperature ~21 °C and wind speed ~2.0 ms^{-1}.

The BC content in $PM_{2.5}$ was affected by the mixture of pollution sources and meteorological conditions. Fugitive dust from neighboring areas in Delhi could also increase the concentrations of $PM_{2.5}$ with non-carbonaceous materials, thereby decreasing the BC/$PM_{2.5}$ ratio. However, higher BC/$PM_{2.5}$ ratio in autumn may be influenced by biomass burning (Cao et al. 2003). The contribution of BC in total suspended particulate (TSP) was reported ~7–15 % for Kanpur (Tripathi et al. 2005), ~7 % for Hyderabad (Latha and Badrinath 2005) and ~2.3 % for Pune (Safai et al. 2007), which are the other urban/sub-urban stations in India. However, Venkatachari et al. (2006) reported higher BC fractions (~13 and 11 %) of $PM_{2.5}$ mass at two different sites in New York.

4.2 Impact of meteorological parameters on BC

Impact of meteorological parameters such as MH and WS on BC aerosols over the station during the study period was also studied in the present study (Fig. 3a, b). On the basis of daily mean data, the correlation analysis has been done among BC, MH and WS. A significant negative correlations between BC and MH ($r = -0.45$) and WS ($r = -0.44$) were observed. The BC concentration was observed to be lower in the month of February due to dilution of soot particle in the atmosphere with the higher WS (2.8 ms^{-1}) and MH (317 m); however, higher BC in December was due to low WS (2.3 ms^{-1}) and MH (213 m). In the case of higher WS, it was suggested that increase in WS causes increase in ventilation and thus disperses the particles in the ambient air and consequently causes a decrease in the observed BC concentrations (Saha et al. 2009). Recently, Wang et al. (2011) observed a strong dependence between daytime BC concentration and average WS up to 2.5 ms^{-1}. Ramachandran and Rajesh (2007) also reported a negative correlation ($r = -0.54$) between BC and WS at urban sites in Ahmedabad, India. A sub-urban and an urban site in Canada showed negative BC correlations with WS, but this was not found at a rural location (Sharma et al. 2002). Such inverse relationships for these parameters have been reported for other urban locations in different places in India and abroad (Sharma et al. 2002; Babu and Moorthy 2002; Hussain et al. 2007).

4.3 Effect of Diwali event on $PM_{2.5}$ and BC

Time series of hourly averaged concentrations of $PM_{2.5}$ and BC from 05th to 07th November, 2010 is shown in Fig. 4. Very high loading of $PM_{2.5}$ (8,227.3 μg m^{-3}) and BC (50.3 μg m^{-3}) was observed at 02:00 am on 05th November, which clearly indicates the effect of fireworks and add large amount of anthropogenic fine particulate pollutants into the lower atmosphere during festival period. Babu and Moorthy (2001) have also reported the similar results at Thiruvananthapuram, India which showed that the concentration of BC increased by a factor of 3 during the festival. Similarly, about threefold increase in PM_{10} and TSPM was attributed to fireworks at Hisar, India (Ravindra et al. 2003). In a

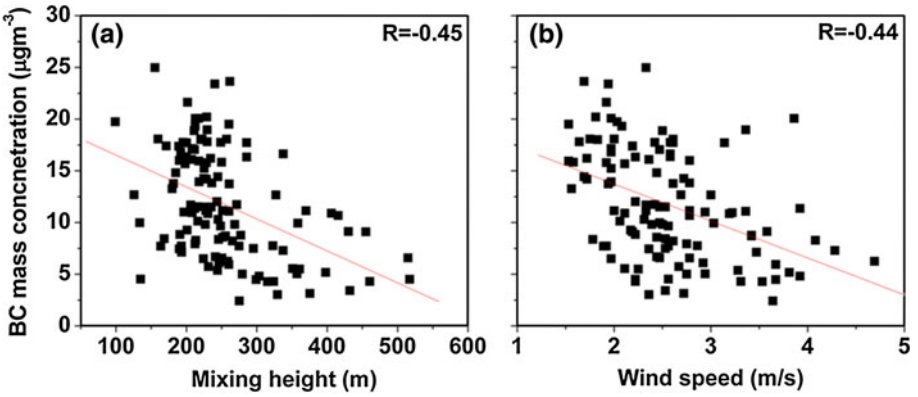

Fig. 3 Correlation between BC mass concentration with **a** mixing height and **b** wind speed during the study period

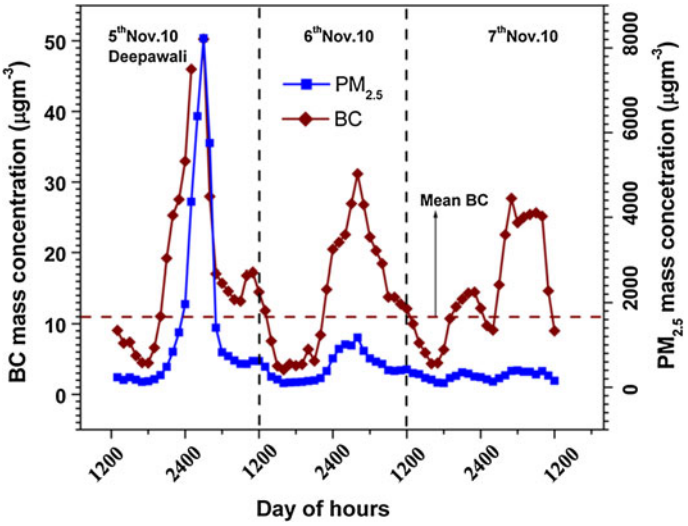

Fig. 4 Time series of hourly averaged concentrations of $PM_{2.5}$ and BC from 5 to 7 November 2010

recent study, Sarakar et al. (2010) have characterized PM_{10} concentration and reported high 24-h PM_{10} levels (317.2–616.8 μg m^{-3}, 6–12 times the WHO standard) and massive loading of EC (40.5 μg m^{-3}) during Diwali festival in Delhi.

4.4 Monthly characteristics of carbonaceous species and $PM_{2.5}$

Monthly mean values of OC, EC, BC and $PM_{2.5}$ are given in Table 3. OC concentrations ranked in the order of December > November > January > February whereas EC concentrations were in order of November > December > January > February. High contributions of OC and EC in the month of November and December are attributed to more incidences of domestic biomass burning and impact of late afternoon traffic loads. In addition, it also originated due to the use of fire crackers in the festivals during these

Table 3 Monthly mean concentration of OC, EC, BC, $PM_{2.5}$, $(OC/EC)_{Primary}$, SOC, SOC/OC at Delhi during winter season of 2010–2011

Months	OC ($\mu g\ m^{-3}$)	EC ($\mu g\ m^{-3}$)	BC ($\mu g\ m^{-3}$)	$PM_{2.5}$ ($\mu g\ m^{-3}$)	$(OC/EC)_{Primary}$	SOC ($\mu g\ m^{-3}$)	SOC/OC (%)
November 10	70.6	14.3	13.4	247.7	3.1	26.3	33
December 10	89.2	11.1	15.2	267.0	6.4	30.8	40
January 11	64.1	10.9	10.9	171.0	2.3	30.2	38
February 11	29.1	7.8	7.2	116.8	1.6	14.6	54

months, when a smoky atmosphere persists over Delhi (Attri et al. 2001). Also, relatively high OC concentrations are due to formation of secondary aerosols by condensation processes fostered by the low temperature and the low boundary layer height during the winter period. In fact, condensation of semi-volatile gaseous precursors is enhanced by low temperatures, whereas temperature inversions limit their dilution and allow for super-saturation conditions with the exceedance of the equilibrium vapor pressure. Other processes that can contribute to cold-season OC levels are the adsorption of semi-volatile OCs onto existing solid particles and the dissolution of soluble gases that can undergo reactions in particles (Pandis et al. 1992). The highest concentration of EC (14.3 $\mu g\ m^{-3}$) in the month of November is due to burning of crackers during Diwali, as discussed in the previous section. EC contributes 6 % (10.4 $\mu g\ m^{-3}$) to $PM_{2.5}$ mass during the study period. In a recent study at Kanpur, Ram et al. (2010a) have reported about one-third contributions of carbonaceous aerosols to the PM_{10} mass during the winter period. They further reported a three- to fourfold increase in the concentration of OC and suggested biomass burning (wood fuel and agricultural waste) emission as a dominant source over the station.

Monthly average BC values varied twofold, from 7.2 (February) to 15.2 $\mu g\ m^{-3}$ (December). BC mass concentrations were found to be lower than the earlier reported values at Delhi during 2007 (Tiwari et al. 2009). In another study, Ganguly et al. (2006) have reported mean BC at Delhi, about 29.4 $\mu g\ m^{-3}$ and varied between 1 and 65 $\mu g\ m^{-3}$ during the winter period (December) of 2004. The BC abundance is affected by the mixture of pollution sources and meteorological conditions. Significant contribution of BC in the ambient air at Delhi during the winter is due to large emissions from fossil fuel and biomass burning from domestic cooking, traffic exhaust and industries; that are not easily dispersed due to low mixing heights and stable atmospheric boundary layer (ABL) conditions, prevailing over the station in winter. Shallowness of the boundary layer acts as a capping inversion, leading to the poor dispersal of atmospheric particulates and restricted mixing resulting in accumulation of BC aerosols in the near-surface region (Sreekanth et al. 2007). Table 4 shows a comparison of Aethalometer measured average BC mass concentrations at Delhi with the values reported at other megacities in India and elsewhere. The mean concentration of BC measured at New Delhi is equal to those measured at other metropolitan cities such as Mumbai and Kanpur but approximately three times higher than the values measured at Bangalore. On the other hand, Lahore (Pakistan) and Xi'am (China) have the higher concentrations of BC during the winter. According to Hussain et al. (2007), the high concentration of BC (21.7 $\mu g\ m^{-3}$; ranging from 5 to 110 $\mu g\ m^{-3}$) at Lahore is due to long range transport of pollutants apart from the local sources. Background level of atmospheric BC concentrations varies widely from 0.0011 $\mu g\ m^{-3}$ at a South Polar site

Table 4 Mean BC mass concentrations measured at different locations in south Asia during the winter period

Location	Period	BC (μg m^{-3})	References
Delhi, India	November 2010 to February 2011	11.8	Present study
Delhi, India	December 2004	29.4	Ganguly et al. (2006)
Delhi, India	January–December 2004	14.8	Bano et al. (2011)
Kanpur, India	December 2004	12.3	Tripathi et al. (2005)
Agara, India	December 2004	20.6	Safai et al. (2009)
Pune, India	January to March 2005	7.38	Safai et al. (2007)
Mumbai, India	January to March 1999	12.5	Venkataraman et al. (2005)
Bangalore, India	November 2001	4.2	Babu et al. (2002)
Lahore, Pakistan	November 2005 to January 2006	21.7	Hussain et al. (2007)
Xian, China	November 2003 to February 2004	21.4	Cao et al. (2009)

(Hansen et al. 1988) to ~10–20 μg m^{-3} in Paris, France (Ruellan and Cachier 2001), to 3–14 μg m^{-3} in Singapore (Balasubramanian et al. 2003).

4.5 OC/EC ratios and sources of carbonaceous species

OC/EC ratio during the study period is shown in Fig. 5, and their mean along with standard deviation and the maximum/minimum values are given in Table 2. The ratio over the station varies from 1.6 to 10.8 with an average of 5.2 ± 2.2. Several investigators have used the OC/EC (or OC/TC) ratio to identify the sources of carbonaceous species (Ram et al. 2008; Sudheer and Sarin 2008 and references therein). The higher OC/EC ratios (or higher OC/TC ratio) indicate biomass burning as a major source, whereas lower ratios are characteristics of emissions from fossil fuel (Ram and Sarin 2010). The average OC/EC

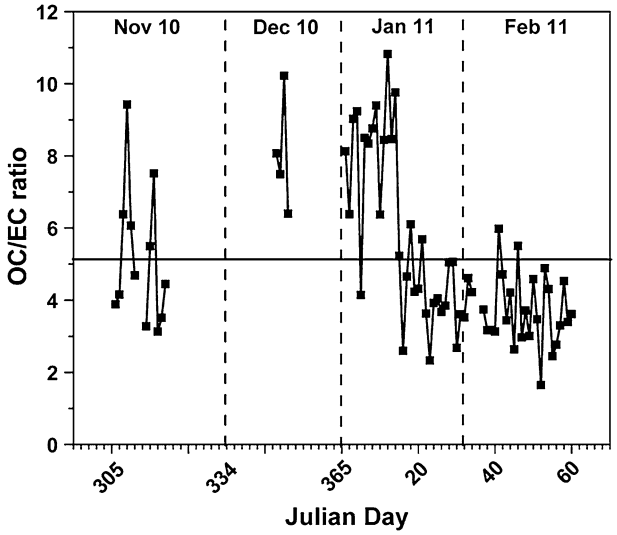

Fig. 5 Day-to-day variations of OC/EC ratio during the study period

ratio observed in the present case is comparable to those observed at other urban sites (Lonati et al. 2007; Rastogi and Sarin 2009). In general, OC/EC ratios are in the range of 2.0-3.0 for urban sites (Turpin and Huntzicker 1995). Particulate OC-to-EC ratios exceeding 2.0, have been used to identify secondary organic aerosol (SOA) formation (Chow et al. 1993). Similar higher ratio (5.8) was reported at Lahore (urban location), Pakistan (Hussain et al. 2007), and was suggested to be due to the formation of SOA. However, such high OC/EC ratios cannot be explained only in terms of enhanced contribution from SOA, rather it can be attributed to the predominance of biomass burning sources (Ram and Sarin 2010).

The correlation between OC and EC concentrations reflects the origin of carbonaceous particles. A significant correlation ($r = 0.71$) was observed between OC and EC in the present study (Fig. 6) which implies similar sources of emission contributing to ambient carbonaceous particles and also, indicating day-to-day fluctuations of emissions along with SOA formation in Delhi. Cao et al. (2003) have reported that motor vehicles played a crucial role in the contribution to OC and EC in atmosphere during cold seasons. EC is predominately emitted from the combustion sources and has often been used as a tracer of primary OC (Turpin and Huntzicker 1995). In the present study, the OC/EC ratio (hourly data) was found to be less than 2.0 ($\sim$15 %), indicating the influence of motor vehicle exhaust and burning of coal, whereas it was found to be more than 2.0 ($\sim$85 %) suggesting the presence of SOA. Vehicles with two- stroke engines are still common in India, so the presence of un-combusted gasoline and oil on the aerosols seems quite likely and this is likely to contribute to the higher OC. The observed minimum value of OC/EC ratio in this study was 1.6 during the study period. These values were in the range of 1.1–2.4, reported for primary aerosols but equal to the minimum OC/EC ratios of 1.7 in Beijing (Yang et al. 2005), 1.3 in the PRDR (Cao et al. 2003), 1.5 at Kaohsiung City (Lin and Tai 2001) and 1.1 in Birmingham (Castro et al. 1999). Sawant et al. (2004) reported that the high OC/EC ratio (5.2) ranged from 1.6 to 12.8 at California, USA.

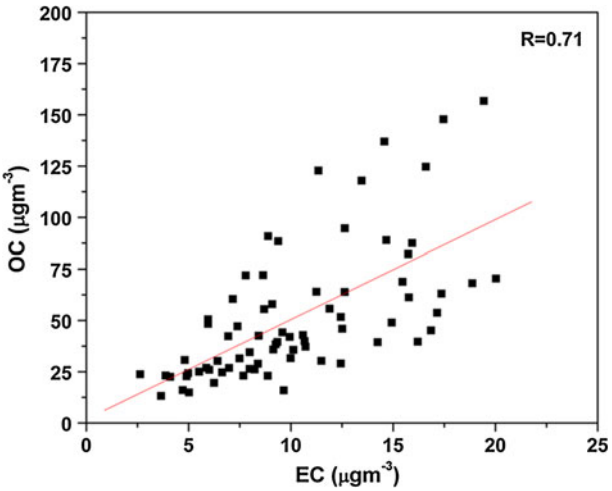

Fig. 6 Correlation between OC and EC concentrations during the study period

4.6 Estimation of secondary organic aerosol

Since there are oxygen, nitrogen and hydrogen associated with organic aerosol (OA), OC-to-OA conversion factor of 1.6 ± 0.2 and 2.1 ± 0.2 have been recommended for urban and non-urban atmosphere (Turpin and Lim 2001). OC-to-OA conversion factor is defined as the average ratio of the molecular weight of organic compounds to the molecular weight of carbon in these organic compounds. In the present study, a factor of 1.6 is used to evaluate OA concentration. TCA was calculated by the summation of OA (OC*1.6) and EC, which contributes to $PM_{2.5}$ mass of ~46 % during the study period over Delhi. This may be attributed to the rapidly increasing automobile, industrial and biomass burning emissions at Delhi. National Capital Region of Delhi is a rapidly growing industrial region in northern India with coal as the main source for energy. However, heavy-duty diesel trucks are also the source of TCA. High (69 %) carbonaceous aerosols were reported by Hussain et al. (2007) at Lahore, Pakistan which is located in the NW direction of Delhi. Recently, Perrino et al. (2011) have reported that the high contribution of secondary pollutants and organics during the post-monsoon and winter periods at Delhi. Khillare et al. (2004) have also presented the level of OC–EC at Delhi and found that the high concentrations of OC and EC during winter were due to atmospheric boundary layer conditions and low wind speeds and entrain the pollutants emitted in the atmosphere from burning wood, waste and coal during winter.

OC consists of a complicated mixture of species from both primary and secondary sources. The separation and quantification of primary and secondary OC is of great importance in understanding secondary aerosol formation as well as in controlling particulate carbon pollution. Since EC is predominantly emitted from the primary combustion sources, it has often been used as a tracer of primary OC in evaluation of the SOC concentrations. The ratio of OC/EC in source emissions when compared to the same ratio in atmospheric samples will be indicative of the presence of SOA formation. In the EC tracer method (Turpin and Huntzicker 1995), SOC is estimated by means of the following equation:

$$OC_{sec} = OC_{tot} - EC\left(\frac{OC}{EC}\right)_{pri} \quad (1)$$

where OC_{sec} is the secondary OC and OC_{tot} is the measured total OC. The primary organic carbon (POC) could be calculated from the formula EC $(OC/EC)_{pri}$. However, it is challenging to know the exact contribution of different primary sources without performing a comprehensive source analysis. Castro et al. (1999) suggested that the primary OC/EC ratio could be replaced by the minimum OC/EC ratio observed in the atmospheric samples which contain exclusively primary carbonaceous compounds. Thus, SOC can be calculated as

$$OC_{sec} = OC_{tot} - EC\left(\frac{OC}{EC}\right)_{Min} \quad (2)$$

An estimation of SOC in Delhi is made according to Eq. (2). It must be noted that this method can give only semi-quantitative information because of the high uncertainties associated with them. Table 3 also shows monthly mean concentrations of $(OC/EC)_{Primary}$, SOC and SOC/OC at Delhi during the study period. SOC concentrations varied significantly from month to month. Calculated mean SOC concentration was 25.5 μg m^{-3} that varied between 14.6 (February 2011) to 30.8 μg m^{-3} (December 2010); accounting for 47 % of the total OC. In the month of December, the OC concentration was ~48 % higher than that of February; it is due to the low temperature (14 °C) and photochemical activities.

The high concentration of OC during winter can be attributed due to the two reasons: (1) stable atmospheric condition and its prolonged residence time that strengthens atmospheric oxidation and (2) low temperature in winter, which would enhance the condensation of volatile secondary organic compounds on pre-exist aerosols. Thus, the amount of SOC is significantly higher during the periods with meteorological conditions favorable to the occurrence of photochemical activity. These results suggest that the secondary organic aerosols may be significant contributor to fine organic particles like OC.

4.7 Diurnal variations of carbonaceous aerosol

Diurnal variations of EC and OC along with mixing height during the study period at Delhi are shown in Fig. 7. As expected, EC and OC showed two peaks during the whole study period where the maximum occurred during the morning at 0800–1000 h LT and in the late evening at 2000–2300 h LT. The minimum OC and EC occurred at about 1600 h LT in the afternoon that rapidly increased after the sunset. The evening highest peak was found at 2200 h LT, which is to be dominating to the morning peak (at 0900 h LT). Very high OC and EC concentration generally prevailed throughout the evening and decreased slowly after midnight, presumably due to decreased vehicular activities. Higher OC and EC concentrations during night time are attributed to the increase in bio-fuel burning such as burning of grass, dry leaves, wood, etc. for getting warmth against cold weather during the winter in northern part of India (Safai et al. 2009). The nocturnal peak is also due to the dynamics of local boundary layer. The morning broad peak between about 0800–1000 h LT is consistent with the morning rush-hour traffic. This peak can also be attributed to the local mobile sources combined with the low mixing heights and consequent poor dispersion conditions in the morning hours. The daily variations in the concentrations of OC and EC were strongly affected by the diurnal variations in the mixing height. OC and EC concentrations were low during the day time when the mixing height was higher (>1,100 m). However, the concentrations were high during the night when the mixing height was below 100 m. Babu et al. (2002) have also suggested that the ABL evolves after sunrise, the strengthened thermals lift and eventually break the nighttime inversion causing the aerosols in the residual layer to mix

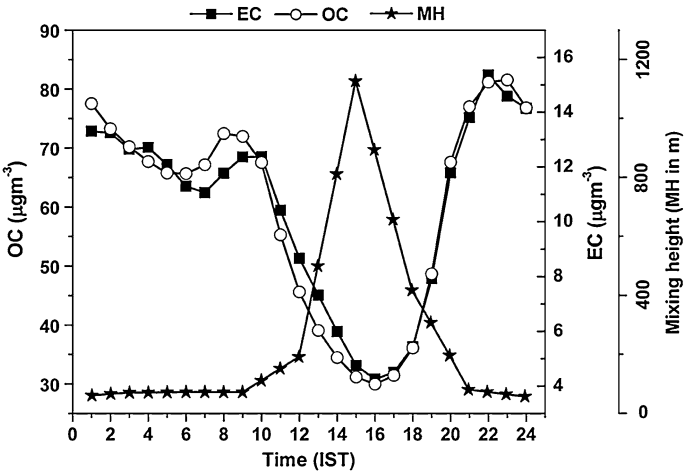

Fig. 7 Diurnal variations of EC and OC along with mixing height (MH) during the study period

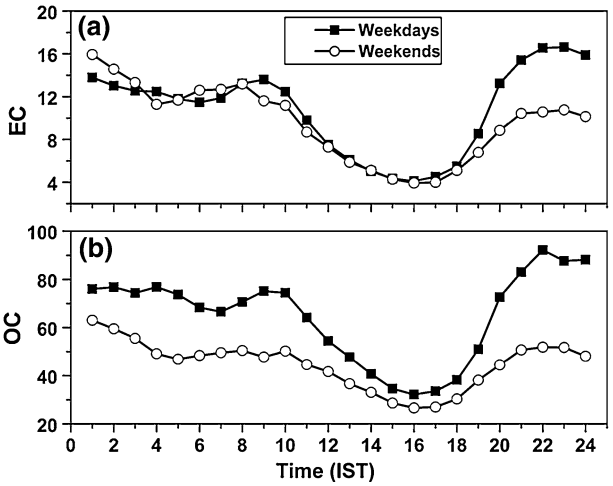

Fig. 8 Diurnal variations of **a** EC and **b** OC in weekdays and weekends during the study period

with those near to the surface, leading to a sharp increase in the near-surface concentrations during morning hours; an effect known as fumigation (Stull 1999). Apart from the dynamics of the local boundary layer variations, the effect of traffic intensity, burning of fossil fuels from industrial and urban activities also play crucial role in modifying the diurnal pattern of EC. Safai et al. (2004) have reported the bimodal behavior of aerosols concentration at an urban site, Pune with higher concentrations during morning and evening hours when the traffic density was more inside the city.

Diurnal variations of OC and EC during weekdays and weekends were also shown in Fig. 8a and b, respectively. The concentration of OC and EC was substantially higher ($\sim$45 and 13 %, respectively) during weekdays than weekends. There are no large variations observed in the morning and day time in EC concentrations except in the late evening, during weekdays and weekends. During the late evening (2000–2300 h LT), the concentration of EC was observed to be $\sim$53 % higher during weekdays than weekends. These results strongly suggest that the source of EC was closely related to the emissions from motor vehicles and biomass burning. In contrast, large variations in OC concentrations were observed during weekdays and weekends. Also, very high concentration of OC was observed in the late evening time (2000–2300 h LT) contributing $\sim$67 % during weekdays. The concentrations of EC and OC were $\sim$36 and 28 % higher during night time (1900–0600 h LT) as compared to day time (0700–1800 h LT), respectively. This feature is mainly due to the low mixing heights during night hours. The ratio of night time to day time concentrations was estimated for both EC and OC and was observed as larger than unity (1.87 for EC and 1.51 for OC), indicating accumulation of EC and OC near the surface at night.

5 Conclusions

Carbonaceous aerosols are the major component in fine particulate matter (i.e., $PM_{2.5}$), which play important role in the regional atmospheric chemistry and radiation balance that leads to climate change. The paper presents first time measurements of OC, EC and BC

along with $PM_{2.5}$ concentrations from an urban location at Delhi, in the northern part of India over the IGB region during the winter period of 2010–11. The salient results are:

- The city of Delhi is heavily polluted with fine particulate matters ($PM_{2.5}$) having mean mass concentration of about 210 ± 146 μg m^{-3}, ranging from 51 to 1,258 μg m^{-3}, which is substantially higher and far excess from the annual mean standards stipulated by the Indian and US-EPA NAAQS, which are 40 and 15 μg m^{-3}, respectively.
- The average mass concentration of OC and EC was about 54 ± 39 and 10 ± 5 μg m^{-3}, respectively during the study period. Contribution of TCA to $PM_{2.5}$ mass was found to be ~46 %. Secondary organic carbonaceous aerosol concentration over the station was found to be 25 μg m^{-3}, which varied between 14.6 (February 2011) and 37.0 μg m^{-3} (December 2010) and accounting for 47 % of the total OC.
- Mean BC mass concentration over the station was about 12 ± 5 μg m^{-3}, varied from 2.4 to 25.0 μg m^{-3}. Monthly mean BC values were found to be varied twofold, from 7.2 (February 2011) to 15.2 μg m^{-3} (December 2010). Approximately 39 % frequency level of BC showed its concentration between 5 and 10 μg m^{-3}; however, only 4 % showed the BC concentration >21 μg m^{-3} (except Diwali).
- Concentrations of OC and EC were strongly affected by the diurnal variations in the mixing height and wind speed. The concentrations were found to be low during the day time when the mixing height was higher (>1,100 m). However, the concentration was found to be high during the night when the mixing height was <100 m. Regression analysis among BC versus mixing height and wind speed suggested dispersion of BC aerosols into the atmosphere, associated with high wind speed and large mixing height. Diurnal distributions showed OC-EC concentrations peak occurring from 0800 to 1000 h LT and 2000 to 2300 h LT.
- The concentrations of OC and EC were ~45 and 13 % higher during weekdays than the weekends due to large emissions from domestic biomass burning and traffic exhaust emissions. About 67 and 53 % higher concentrations of OC and EC were observed in the late evening (2000–2300 h LT) during weekdays than the weekends indicating their similar types of sources. However, concentrations of EC and OC were found to be ~36 and 28 % higher during night as compared to day, which could be due to the prevailing meteorological conditions of the station. The ratio of EC and OC between night time and day time was observed to be larger than the unity (1.87 for EC and 1.51 for OC), indicating accumulation of EC and OC near the surface at night.

The present study revealed significantly high magnitude of OC, EC, BC and $PM_{2.5}$ mass concentrations in an urban region (Delhi) during the winter period, which suggests an urgent need to focus on air quality management in such regions. Also, an assessment of the impact of these aerosol particles on climate and health perspectives should be done.

Acknowledgments Authors express their sincere gratitude to Prof. B. N. Goswami, Director, IITM, Pune and Dr. P. C. S. Devara, Head, PM&A, IITM for their encouragement and support during the study period.

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