



Diurnal and seasonal variations of black carbon and PM_{2.5} over New Delhi, India: Influence of meteorology



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ABSTRACT

Black carbon (BC), which is one of the highly absorbing capacities of solar radiation, reduces albedo of atmospheric aerosol. BC along with fine particulate matters (PM_{2.5}), which play crucial role in climate and health, was monitored online for an entire year of 2011 at an urban megacity of Delhi, situated in the northern part of India. Daily mass concentration of BC varies from 0.9 to 25.5 $\mu\text{g m}^{-3}$, with an annual mean of $6.7 \pm 5.7 \mu\text{g m}^{-3}$ displayed clear monsoon minima and winter maxima; however, PM_{2.5} concentration was ranging from 54.3 to 338.7 $\mu\text{g m}^{-3}$, with an annual mean of $122.3 \pm 90.7 \mu\text{g m}^{-3}$. BC typically peaked between 0800 and 1000 LST and again between 2100 and 2300 LST, corresponding to the morning and evening traffic combined with the ambient meteorological effect. During summer and monsoon, the BC concentrations were found less than 5 $\mu\text{g m}^{-3}$; however, the highest concentrations occurred during winter in segments from <5 to >10 $\mu\text{g m}^{-3}$. In over all study, the BC mass concentration was accounted for ~6% of the total PM_{2.5} mass, with a range from 1.0% to 14.3%. The relationship between meteorological parameters and BC mass concentrations was studied and a clear inverse relationship ($r = -0.53$) between BC and wind speed was observed. Relation between visibility and BC mass concentrations was also significantly negative (-0.81), having relatively higher correlation during post-monsoon (-0.85) and winter (-0.78) periods and lower during summer (-0.45) and monsoon (-0.54) periods. The mixed layer depths (MLDs) were found to be shallower during post monsoon (379 m) and winter (335 m) as compared during summer (1023 m) and monsoon (603 m). The study indicated that during post-monsoon season, the impact of biomass burning is higher as compared to combustion of fossil fuels. Results are well associated with the rapid growth of anthropogenic emissions and ambient meteorological conditions over the station.

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1. Introduction

Black carbon (BC) is an important constituent (90% in PM_{2.5}) of airborne particulate matter (PM) that is often emitted as product of incomplete combustion of fossil fuel and burning of biomass and bio-fuels (Koelmans et al., 2006; Penner et al., 1993; Cooke and Wilson, 1996). Interest in BC has grown due to its strong absorbing nature of short-wave radiation, thereby

degrading visibility (Watson, 2002) and altering the Earth's radiation balance (Penner et al., 1993; Jacobson, 2001). BC can heat the air, alter atmospheric stability, influences large-scale circulations, and affects cloud albedo by changing the hygroscopicity of cloud condensation nuclei (Liousse et al., 1996). BC-containing particles are also associated with adverse health effects (Pope and Dockery, 2006; Badarinarath et al., 2007; Mauderly and Chow, 2008), may be lowering crop yields (Chameides et al., 1999), contaminate building materials (Ghedini et al., 2000), and adversely impact on terrestrial and aquatic ecosystems (Forbes et al., 2006) as well as retreat of

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glaciers (Lau et al., 2010 and references therein). BC has typical lifetime ranging from 1 week to 10 days in the absence of precipitation and hence is amenable for long range transport (Babu and Moorthy, 2001; Reddy and Venkataraman, 1999). Though BC constitutes only a few percent of the total aerosol mass, it can have a significant positive forcing in the atmosphere (Heintzenberg et al., 1997; Satheesh and Ramanathan, 2000; Srivastava et al., 2012a).

A major contribution of fine particle over northern India is due to burning of agricultural residue during the agricultural seasons (Awasthi et al., 2011). Crop residue burning is a process of uncontrolled combustion during which carbon dioxide (CO_2), the principal product of the combustion, is emitted into the atmosphere along with carbon monoxide (CO), un-burnt carbon, nitrogen oxides (NO_x) and sulfur dioxide (SO_2). In Asia, based on the results of a study from Streets et al. (2003), the annual contribution from open burning of biomass is estimated to be 0.37 Tg of SO_2 , 2.8 Tg of NO_x , 1100 Tg of CO_2 , 67 Tg of CO and 3.1 Tg of methane (CH_4). On a global basis, however, approximately 20% of black carbon is emitted from burning of bio-fuels, 40% from fossil fuels, and 40% from open biomass burning (Ramanathan and Carmichael, 2008). This large scale burning of crop residue has been found to aggravate lung and respiratory diseases (Wang and Christopher, 2003).

Although research on BC has been one of the forefront areas of atmospheric research all over the globe, it has special importance for China and India in South Asia, due to its consequential regional climatic effects at these places (Menon et al., 2002; Ramanathan and Carmichael, 2008). Even in megacities, urban air pollution is an important environmental issue (Molina and Molina, 2004), particularly for countries with higher population density; however, there are only limited BC measurements from such large population centers from India (Raju et al., 2011; Safai et al., 2008; Tripathi et al., 2005; Beegum et al., 2009; Tiwari et al., 2009; Bano et al., 2011; Rehman et al., 2011). In order to improve the existing knowledge-base for BC and $\text{PM}_{2.5}$ particles over northern part of India and to understand their regional climatology; a year-long measurement of BC and $\text{PM}_{2.5}$ mass concentrations from an urban-scale site, Delhi, in northern part of India during 2011 was examined for daily, monthly and seasonal temporal scales. Results are also studied in view of potential source emissions and meteorological effects.

2. Description of sampling site, instruments and data

2.1. General climate of Delhi

Delhi, which has more than 17 million inhabitants, is situated ($28^{\circ}35' \text{ N}$; $77^{\circ}12' \text{ E}$) at an altitude of about 300 m above sea level. It is surrounded by the Thar Desert of Rajasthan to the west and the plains of central India to the south and the east. The climate of Delhi is semi-arid and is mainly influenced by its inland position and prevalence of continental air during most of the year. Regarding seasons, according to the classification given by Indian Meteorological Department, Delhi experiences four distinct seasons: winter (December–March), pre-monsoon or summer (April–June), monsoon (July–September) and post-monsoon (October–November) (Perrino et al., 2011). Summer is extremely hot, with maximum temperatures of $45\text{--}48^{\circ}\text{C}$. Dust storms from

nearby Thar Desert are influencing the weather of Delhi during summer (April–June). Humidity is high during monsoon, characterized by heavy rainfall (between 600 and 800 mm); while the air is dry during the rest of the year. Winter is moderately cold, with minimum temperatures becomes around $1\text{--}4^{\circ}\text{C}$. During winter, conditions of atmospheric stability (low wind speed and temperature inversion) lead to the accumulation of atmospheric pollutants in the lower atmospheric layer, causing morning hour foggy conditions. Winds are predominantly westerly and north-westerly during most of the year, except for the monsoon season, when they are easterly and north-easterly.

2.2. Local meteorology of the station

Fig. 1 shows variations in the monthly mean meteorological parameters such as temperature, wind speed (WS), related humidity (RH) and mixed layer depths (MLDs). These data were retrieved from the National Oceanic and Atmospheric Administration (NOAA), Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model from the READY Website (<http://ready.arl.noaa.gov>) (Dutkiewicz et al., 2009). The time resolution of the data was one hour interval. Annual mean temperature was found to be about $25(\pm 7)^{\circ}\text{C}$ during the study period. January ($\sim 12^{\circ}\text{C}$) was the colder month which attains maxima during the summer in the month of May ($\sim 34^{\circ}\text{C}$). Temperature increases after January till May and decreases thereafter till December (Fig. 1a). Wind speed indicates maxima in the month of June (3.56 m/s) and minima in December (2.36 m/s) with an annual mean of 3.01 ± 0.4 m/s. The mean annual RH was found to be about $62 (\pm 15)\%$, varying from 35% (April) to 80% (August) as shown in Fig. 1b. On the other hand, MLDs were found to be similar to the temperature variations, which vary from about 220 m (December) to 1085 m (May). Since the impact of ambient aerosols on the radiation budget of the atmospheric changes with source regions and transport pathways, it is important to get detailed meteorological information for aerosol impact assessment. In order to know the possible transport pathways over the station (Delhi), 5-day backward air mass trajectories have been computed using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model of the National Oceanic and Atmospheric Administration, USA (Draxler and Rolph, 2003). Rainfall data was obtained from automatic rain gauge which is installed in the premises of the Institute at New Rajinder Nagar area. Wind direction (WD) data was taken from Indian Meteorological Department, Lodhi Road, New Delhi.

2.3. Instrumentations and data analysis

Sampling of BC and $\text{PM}_{2.5}$ ($\text{PM} \leq 2.5 \mu\text{m}$ in aerodynamic diameter) mass concentrations was simultaneously conducted inside the premises of the Indian Institute of Tropical Meteorology (New Delhi Branch) Pune, located in the central urbanized part of Delhi during 1st January to 31st December, 2011. No major industrial sources are located within 5 km radius around the sampling site. Plenty of vegetation in and around with heavy traffic load in the east is characteristic of the sampling site, which is thus the representative of residential area. The measurements of BC mass concentration were carried out by 7-wavelength Aethalometer (Model AE-31, Magee

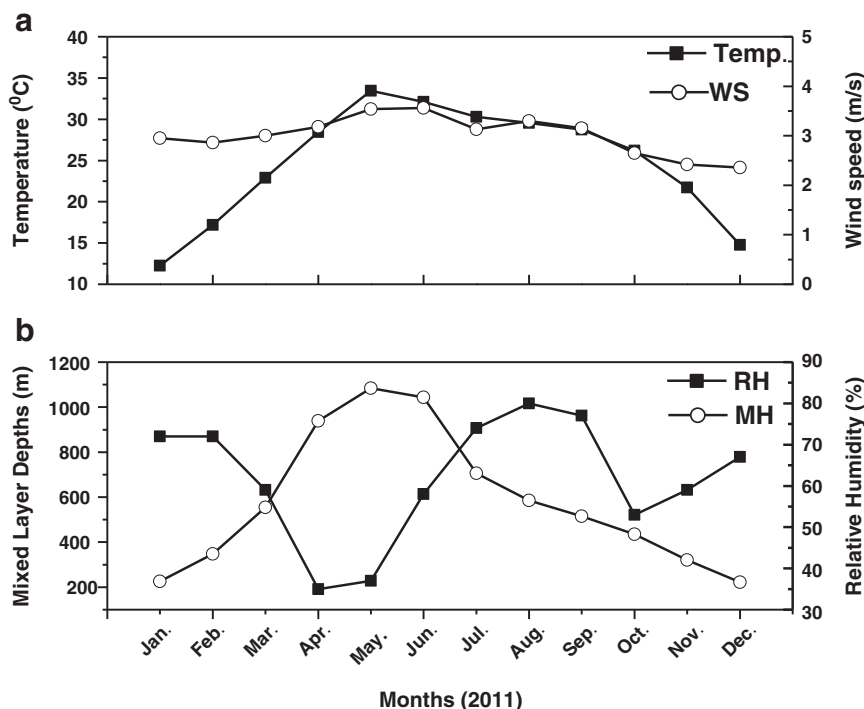


Fig. 1. Monthly variations of meteorological parameters such as (a) temperature (Temp.) and wind speed (WS); (b) relative humidity (RH) and mixed layer depths (MLDs) over Delhi during 2011.

Scientific Company, Berkley, CA, USA) online monitor with temporal resolution of 5 min during study period. It measures the optical attenuation (absorbance) of light from LED lamps emitting at seven wavelengths (370, 470, 520, 590, 660, 880 and 950 nm) with a typical half-width of 20 nm (Hansen, 2005). The black carbon determination assumes that all the light-absorbing materials are composed of BC. The specific mass attenuation efficiency, σ_{ATN} of the Aethalometer used for the determination of the BC concentration was $14,625/\lambda$ [$\text{m}^2 \text{g}^{-1}$], as recommended by the manufacturer. The flow rate was maintained at 4 LPM corresponding to a $2.5 \mu\text{m}$ cut-point of $\text{PM}_{2.5}$ cyclone (BGI Inc., SCC1.829). As shown in Table 1, detection limit of the Aethalometer BC was determined to be $0.16\text{--}0.28 \mu\text{g m}^{-3}$ calculated as three times the standard deviation (3σ) of the dynamic blank. Uncertainty of the Aethalometer reported by the manufacturer is $\pm 5\%$ (Hansen, 2005; Allen et al., 1999; Virkkula et al., 2007); however, further details of the instrument can be found elsewhere (Hansen,

2005; Schmid et al., 2006; Weingartner et al., 2003; Srivastava et al., 2012b). To account for the “shadowing” effect due to filter loading (decrease in Aethalometer sensitivity), $R(\text{ATN})$, the loading correction was done as suggested by Weingartner et al. (2003). Even though humidity effect on the Aethalometer BC measurement is negligible (Schmid et al., 2006), the aerosol was dried to $\text{RH} < 40\%$ by a diffusion dryer prior to particle detection.

Earlier researches found out that certain organic aerosol components of wood combustion have enhanced optical absorption at 370 nm wavelengths relative to 880 nm (Jeong et al., 2004; Park et al., 2006). Hence, a difference between the measured BC at 370 and 880 nm channels can provide useful information on the potential sources of BC, which can be used as a qualitative indicator, particularly for wood smoke particles from the local wood combustion sources (Wang et al., 2011b; Srivastava et al., 2012b). However, for finding the total BC concentration, the 880 nm wavelength

Table 1
Detection limit of BC and its uncertainties at different wavelengths.

Instrument	Parameter	Unit	Dynamic blank	Standard deviation (σ)	Detection limit (3σ)	Uncertainty (%)
Aethalometer (Magee, AE31)	BC (370 nm)	$\mu\text{g m}^{-3}$	0.105	0.052	0.156	10%
	BC (470 nm)	$\mu\text{g m}^{-3}$	0.115	0.054	0.161	
	BC (520 nm)	$\mu\text{g m}^{-3}$	0.106	0.062	0.187	
	BC (590 nm)	$\mu\text{g m}^{-3}$	0.111	0.071	0.213	
	BC (660 nm)	$\mu\text{g m}^{-3}$	0.096	0.076	0.228	
	BC (880 nm)	$\mu\text{g m}^{-3}$	0.090	0.088	0.263	
	BC (950 nm)	$\mu\text{g m}^{-3}$	0.097	0.094	0.281	

has been used in the present study as it was also used in the case of several earlier works (Ramachandran and Rajesh, 2007; Tiwari et al., 2009; Bano et al., 2011). Fine particle aerosol ($PM_{2.5}$) was measured by Thermo Andersen, Inc. Series FH 62 C14 (C14 BETA) at five minute interval, which corresponds to BC measurements. The $PM_{2.5}$ cut-point was achieved through sharp-cut cyclone inlet with the flow rate through the instrument is specified as $1 \text{ m}^3/\text{h}$ (Kenny et al., 2000). The measuring range of instrument is $0\text{--}5000 \text{ }\mu\text{g m}^{-3}$ with minimum detection limit of $1 \text{ }\mu\text{g m}^{-3}$ for 24 h average. More details of the above instruments are given elsewhere (Hyvärinen, et al., 2009).

3. Results and discussion

3.1. Daily variations of BC and $PM_{2.5}$ aerosols over Delhi

Variations in daily mean mass concentrations of BC and fine particles (i.e. $PM_{2.5}$) from 1st January to 31st December, 2011 are shown in Fig. 2a and b, respectively. The annual averaged concentration of BC was observed to be about $6.7 \pm 5.7 \text{ }\mu\text{g m}^{-3}$ (median: $4.2 \text{ }\mu\text{g m}^{-3}$), varied from 0.9 to $25.5 \text{ }\mu\text{g m}^{-3}$ and displayed clear minima (during monsoon) and maxima (during winter), which follows the order of: winter > post-monsoon > summer > monsoon. The great range of the daily BC variation in Delhi was due to the variation of daily aerosols which is favored by the accumulation of BC aerosol produced from the surroundings and transported from the local, regional, and even the distant locations. BC concentrations in winter ($10.8 \pm 6.4 \text{ }\mu\text{g m}^{-3}$) and post-monsoon ($9.4 \pm 5.4 \text{ }\mu\text{g m}^{-3}$) were approximately three times higher than those during the summer ($3.5 \pm 1.9 \text{ }\mu\text{g m}^{-3}$) and monsoon ($2.8 \pm 1.5 \text{ }\mu\text{g m}^{-3}$). The BC mass concentrations were

found to be exceeded by $\sim 38\%$ of the days to the average during study period. Such high BC mass concentration suggests that the air is mostly polluted by the anthropogenic activities.

The fine particles i.e. $PM_{2.5}$ ($d \leq 2.5 \mu\text{m}$) are responsible for most of the airborne particle threats to human health (Pope et al., 2002; Pope and Dockery, 2006) and visibility degradation. This parameter was also studied simultaneously along with BC (Fig. 2b) because the bulk of BC aerosols ($\sim 90\%$) resides in $PM_{2.5}$ fraction (Viidanoja et al., 2002). The annual mean mass concentration of $PM_{2.5}$ was observed to be about $122.3 \pm 90.7 \text{ }\mu\text{g m}^{-3}$ (median: $90.7 \text{ }\mu\text{g m}^{-3}$), varied from 54.3 to $338.7 \text{ }\mu\text{g m}^{-3}$. Though the magnitude of $PM_{2.5}$ strongly differs with the magnitude of BC, both, however, follow by-and-large similar trends. Similar behavior of mass of $PM_{2.5}$ has been observed in earlier studies over the northern part of India (Hyvärinen et al., 2010; Tiwari et al., 2009; Tiwari et al., 2012a). Awasthi et al. (2011) has reported the mean value of $PM_{2.5} \sim 69 \text{ }\mu\text{g m}^{-3}$ that varied from 44 to $147 \text{ }\mu\text{g m}^{-3}$ at Patiala district of Punjab, India, which is located to the northwest of Delhi. They found that the concentration of fine particulate matter was increased (78%) during rice crop residue burning in the month of October–November with the maxima (100 and $147 \text{ }\mu\text{g m}^{-3}$) in the year 2009. Similarly, Badarinath et al. (2006) through IRS P6 AWiFS satellite study reported exhaustive burning of rice crop residue in Indo-Gangetic Plains in the months of October–November. Therefore, the significantly high concentrations of fine mode particles were observed over Delhi, which may be also attributed to certain anthropogenic activities related to agriculture during the winter season.

Monthly mean variations of BC and $PM_{2.5}$ mass concentrations are shown in Fig. 3 and also given in Table 2. BC mass concentrations started increasing from September ($2.6 \pm 1.0 \text{ }\mu\text{g m}^{-3}$) and peaked during the whole winter period. BC

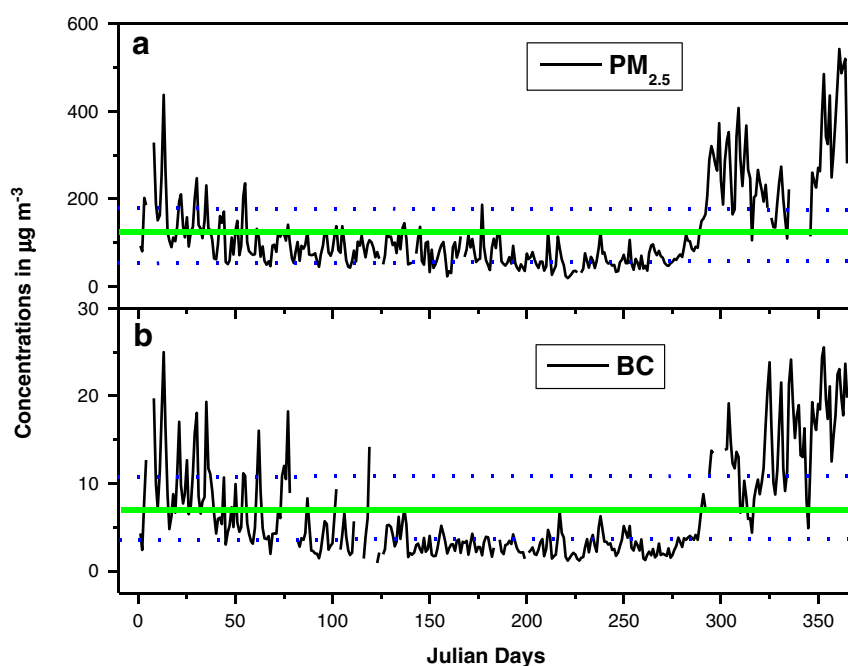


Fig. 2. Daily (Julian days) mass concentrations of $PM_{2.5}$ (a) and BC (b) from 1st January to 31st December 2011 (green line: mean conc. and blue line: mean conc. \pm std.).

concentrations varied by approximately sevenfold, from a low of 2.6 ± 1.0 (September) to $17.9 \pm 4.7 \mu\text{g m}^{-3}$ (December) during the annual cycle. In the month of December, most of the time, wind was in calm condition (mean wind speed: 2.4 m/s), with low mixing height (222 m) along with low temperature (mean: 14.8°C) (Table 2). Also, the monthly relative standard deviations (RSD; standard deviation divided by the mean) were 3–18% higher during post-monsoon and winter (0.58 to 0.59) compared to summer and monsoon (0.56 to 0.42). During the entire study period, the highest peak of BC was observed on 19th December 2011 ($25.5 \mu\text{g m}^{-3}$) when the mixing height depths (168 m), temperature (12.4°C) and wind speed (1.9 m/s) were found to be very low. Coal-burning is a major source of energy in India which contributes 76% of our requirement (SAFAR, 2010) and it is a large emitter of BC into the atmosphere. There are three coal-based thermal power plants located in Delhi, which were estimated to emit about 6000 t of fly ash per day (Srivastava and Jain, 2007). Apart from this, fire crackers, during celebration of festival 'Diwali' also produces huge amount of air pollutants, particularly BC into the atmosphere over Delhi (Tiwari et al., 2012b). During 2011, Diwali was celebrated on 26 October, which is one of the probable causes for the higher BC concentrations in Delhi during this period. Earlier study by Babu and Moorthy (2001) reported that at Thiruvananthapuram the BC concentration increased by a factor of three during the festival. In a recent study, Tiwari et al. (2012a,b) have reported the effect of burning of crackers over Delhi, which leads to the high concentrations of particulate matter and BC. Further, biomass burning is also a large carbon producer in the northern part of India being agriculturally-productive Indo-Gangetic Plains and foothills of the Himalaya (Awasthi et al., 2011). The second largest concentration was observed in the month of March (middle of March), which may be due to the open firing of crops in the northwest part of Delhi (Awasthi et al., 2011; Badarinath et al., 2009). Gadde et al. (2009) have identified the contribution of ~14% (globally) of the open burnt rice straw in the states of Punjab, Haryana and Uttar Pradesh, particularly, during the summer and mid-autumn. In addition to this, Delhi's nighttime temperature sharply decreases during winter ($\sim 2^\circ\text{C}$) and indoor and outdoor heating of bio-fuels are accomplished with

small coal-burning boilers, stoves, open burning of leaves and woods (Ali et al., 2004; Parashar et al., 2005).

The mean BC mass concentration measured at Delhi was found to be similar to that observed at Sao Paulo, Brazil ($7.6 \mu\text{g m}^{-3}$; Castanho and Artaxo, 2001) and at Kanpur, India ($6.0\text{--}20.0 \mu\text{g m}^{-3}$; Tripathi et al., 2005) and approximately half of that at Xi'an, China ($14.7 \mu\text{g m}^{-3}$; Cao et al., 2009), Lahore, Pakistan ($21.7 \mu\text{g m}^{-3}$; Husain et al., 2007) and Paris, France ($14 \mu\text{g m}^{-3}$; Ruellan and Cachier, 2001). Other Indian cities such as Pune ($4.1 \mu\text{g m}^{-3}$; Safai et al., 2007) and Bangalore ($4.2 \mu\text{g m}^{-3}$; Babu and Moorthy, 2002) have lower BC concentrations as compared to Delhi. This may also be compared with the measurements from a relatively clean site, Manora Peak, in the Indian Himalayan foothills, where Srivastava et al. (2012a) reported the mean BC of $0.98 \pm 0.68 \mu\text{g m}^{-3}$, ranging from $0.13\text{--}2.12 \mu\text{g m}^{-3}$ during the period from 2006 to 2007. Marinoni et al. (2010) have reported the BC mass concentrations (160.5 ng m^{-3}) at a high-altitude research station in Nepal (Khumbu valley, 5079 m above msl in the Southern Himalayas).

For the annual cycle, annual mean mass concentration of $\text{PM}_{2.5}$ was $122.3 \pm 90.7 \mu\text{g m}^{-3}$ (Fig. 3) which is substantially higher and is far in excess of the 40 and $15 \mu\text{g m}^{-3}$, annual averages stipulated by the Indian National Ambient Air Quality Standards (www.cpcb.nic.in) and the US National Ambient Air Quality Standards (NAAQS, <http://www.epa.gov/air/criteria.html>) respectively. The highest concentrations were observed during post-monsoon ($196.3 \pm 60.1 \mu\text{g m}^{-3}$), which is followed by winter ($164.3 \pm 40.1 \mu\text{g m}^{-3}$), summer ($93.7 \pm 32.8 \mu\text{g m}^{-3}$) and monsoon ($65.9 \pm 18.9 \mu\text{g m}^{-3}$). Higher concentrations of $\text{PM}_{2.5}$ during the post-monsoon period are expected due to burning of fire crackers during the festival (Diwali) and biomass burning; however, minima during monsoon are due to washout effect (Tiwari et al., 2012b). Significant correlation (0.86) was observed between $\text{PM}_{2.5}$ and BC during the entire study period over Delhi indicating the similarity in sources. In general, the atmosphere of Delhi during winter season is characterized by low relative humidity and low solar heating of land accompanied by low ventilation coefficients that result in less dispersion of aerosols, which, in turn, leads to an increase in the concentrations of fine particulate matter.

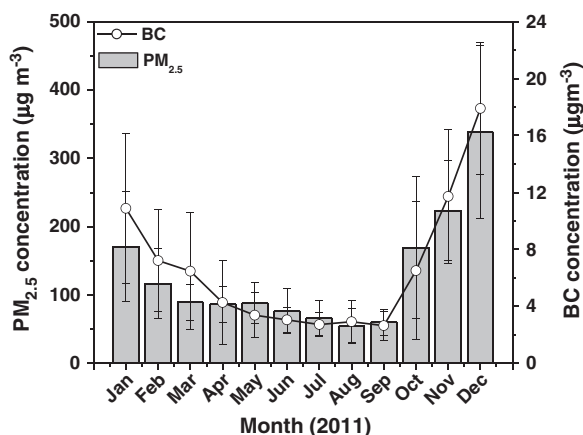


Fig. 3. Monthly mean concentrations of mass BC and $\text{PM}_{2.5}$ along with their standard deviation during 2011 at Delhi.

Table 2Monthly mean mass concentrations of BC and PM_{2.5} in Delhi along with meteorological parameters during 2011.

Sampling period (2011)	BC ($\mu\text{g m}^{-3}$)	PM _{2.5} ($\mu\text{g m}^{-3}$)	MLDs ^a (m)	Temp. ($^{\circ}\text{C}$)	Wind speed (m/s)	Humidity (%)	Visibility (km)
January	10.9	170.5	225.7	12.2	2.95	73	1.84
February	7.2	116.3	347.9	17.2	2.86	72	2.36
March	6.5	89.2	555.9	23.0	3.00	59	2.89
April	4.3	86.1	941.2	28.4	3.18	35	2.99
May	3.4	87.7	1084.4	33.4	3.54	37	3.09
June	3.1	75.9	1043.3	32.1	3.56	58	3.34
July	2.7	65.9	706.2	30.3	3.13	74	3.30
August	2.9	54.3	584.8	29.6	3.30	80	3.07
September	2.6	59.9	514.7	28.7	3.15	77	3.18
October	6.5	169.0	435.4	26.2	2.65	53	2.39
November	11.7	223.5	320.7	21.7	2.42	59	1.71
December	17.9	338.7	222.0	14.8	2.36	67	1.31

^a Mixed layer depths (MLDs).

Also, there is a greater exposure risk as pollutants often get trapped in the lower layers of the atmosphere thereby resulting in high concentrations of PM at surface. In such conditions, the probabilities in the formation of secondary aerosols are higher (Tiwari et al., 2012a). Apart from this, the long range transport of fine particles also plays crucial role during post-monsoon (Tiwari et al., 2009; Hyvarinen et al., 2010).

The annual mean of percentage contribution (i.e. fraction) of BC in PM_{2.5} mass was found to be ~5.3%, ranging from 1.0% to 14.3%, during study period over Delhi indicating that the fine particles have been polluted by the anthropogenic activities and associated BC. Husain et al. (2007) have reported that the BC contributes ~5–15% of PM_{2.5} in urban air. The BC abundance in PM_{2.5} is affected by the mixture of pollution sources and meteorological conditions. Winter had the highest fraction (~6%) of PM_{2.5} mass whereas the lowest fraction (~4%) was found in summer. Tripathi et al. (2005) have reported the fraction of BC of ~7% to 15% to the total suspended particulate (TSP) during the winter period at Kanpur, whereas it was found to be ~4% to 15% to TSP at Delhi (Ganguly et al., 2006), as 7% of TSP at Hyderabad (Latha and Badarinath, 2005), and as 2.3% of TSP for Pune (Safai et al., 2007) in India. In sub-urban regions of Europe and North America, BC contributes about 5% of TSP (Ramanathan and Crutzen, 2003), ~13% and 11% of PM_{2.5} mass at two sites in New York City (Venkatachari et al., 2006). Zhang et al. (2002) have suggested that the decreasing ratio is due non-carbonaceous material (fugitive dust).

Fig. 4 shows the frequency distribution of BC during different seasons which was divided into six different categories starting from <5 to >30 $\mu\text{g m}^{-3}$. BC was found to be skewed toward higher to lower BC mass concentration in respect of corresponding spectrum except at 15–20 $\mu\text{g m}^{-3}$, which was 56% (<5 $\mu\text{g m}^{-3}$); 20% (5–10 $\mu\text{g m}^{-3}$); 11% (10–15 $\mu\text{g m}^{-3}$); 2% (15–20 $\mu\text{g m}^{-3}$); 8% (20–25 $\mu\text{g m}^{-3}$) and 3% (25–>30 $\mu\text{g m}^{-3}$). Season-wise, the trend was similar during summer and monsoon; however, during winter and post-monsoon, the highest contributions of hourly BC concentrations in higher ranges were observed. During summer and monsoon, approximately 80% and 95% of concentrations of BC level were observed to be less than 5 $\mu\text{g m}^{-3}$ whereas only 28% and 23% were during post-monsoon and winter seasons, respectively. During winter, BC concentrations were highest (32%) in the segment between <5 and >10 $\mu\text{g m}^{-3}$; however,

fewer BC mass concentrations (3%) were in the segment between <15 and >20 $\mu\text{g m}^{-3}$. The distinct feature during post-monsoon and winter was observed due to the dominance of hourly BC mass concentrations by approximately 46% higher for >10 $\mu\text{g m}^{-3}$ spectrum compared to summer (1%). During monsoon period, most of the BC concentrations were found to be <10 $\mu\text{g m}^{-3}$. This feature is caused due to meteorological conditions which are discussed in Section 3.4 and by the mixture of pollution sources.

3.2. Diurnal variations of mass of BC and PM_{2.5}

Besides the daily variations, BC and PM_{2.5} mass concentrations also exhibit a pronounced diurnal variability, 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; Srivastava et al., 2012b). Fig. 5a and b shows a diurnal cycle of BC and PM_{2.5} mass concentrations during different seasons in 2011 over Delhi. Significant diurnal variations in the concentrations of BC and PM_{2.5} were observed over the study period with relatively lower values during the monsoon as compared to other seasons with discernible minima during afternoon. The observed diurnal variation of BC is mainly attributed to the dynamics of the local atmospheric boundary layer, though the urban and local human activities might also be contributing to the nocturnal peak.

The diurnal variations of BC mass concentrations along with mixed layer depths were plotted for different seasons and depicted in Fig. 6. The diurnal profile for BC observed in this study is very similar to that observed by others at different locations (Husain et al., 2007; Nair et al., 2007; Tripathi et al., 2005; Safai et al., 2007; Babu and Moorthy, 2002; Bhugwant et al., 2000; Latha and Badrinath, 2004) with a gradual build-up starting at 0600 LST (Local standard time). The decreasing trend in BC between midnight and 0600 LST appears to be primarily caused by decreased emissions due to reduced vehicular traffic as the mixing height and wind speed remained relatively constant and very low during this period. In between 0800 to 1000 LST and 2100 and 2300 LST, higher concentration of BC is consistent with the morning and evening rush-hour traffic pattern, respectively. However, an usual decrease in BC concentration slowly after midnight, presumably due to

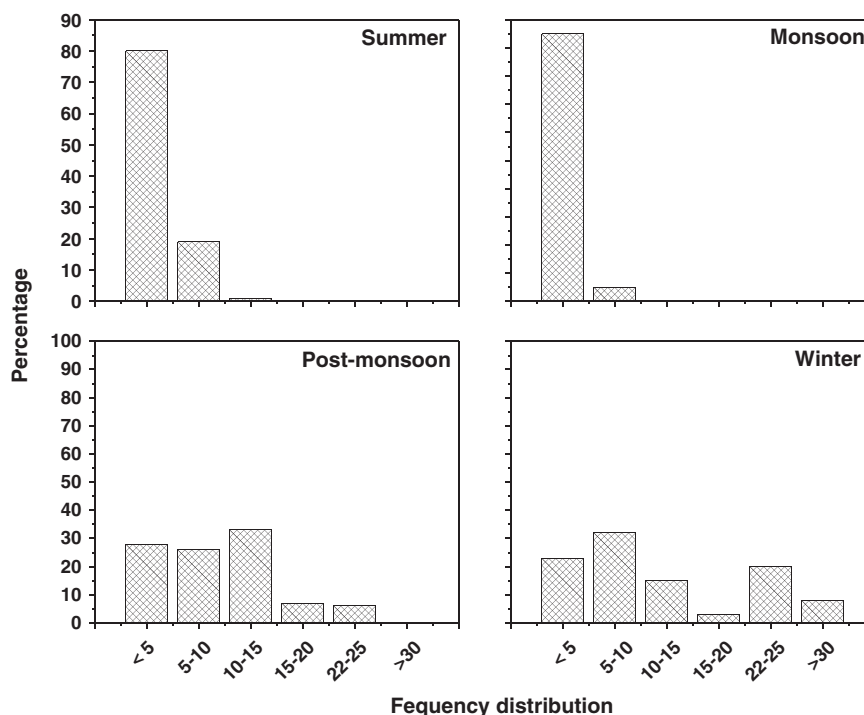


Fig. 4. Frequency distribution of BC mass concentrations at Delhi during 2011.

decreased vehicular emissions and meteorological effect. The morning peak is followed by a gradual decrease in BC concentrations through the afternoon. This feature might be explained in part by the growth of the mixed layer depths (Fig. 6) and stronger atmospheric ventilation during the afternoon. Peak BC concentrations during the evening and morning were about 3.6, 2.8, 11.3 and 12.9 $\mu\text{g m}^{-3}$ and 4.8, 3.5, 12.1 and 14.7 $\mu\text{g m}^{-3}$ for summer, monsoon, post-monsoon and winter, respectively. These levels are more than thrice to those measured during the afternoon. These patterns can be explained by a combination of emissions and meteorology. The morning peak might 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 increase turbulent effects and a deeper boundary layer, leading to faster dispersion of aerosols and hence a dilution of BC concentration occurs near to the surface during the late-afternoon (1600 LST). After 1600 LST, the evening rush hour commences as does cooking and residential heating during the winter. Soon after sunset, the surface inversion begins to form, trapping more primary pollutants related to BC. The earlier onset of the morning buildup and the later onset of the evening accumulation in summer than in winter correspond with the earlier sunrises and later sunsets in summer. Seasonal diurnal variations (Fig. 5b) in mass $\text{PM}_{2.5}$ were similar to BC except post monsoon due to long range transport of pollutant and festival period along with meteorological effects.

The ratio between nighttime (1900 to 0600 LST) and daytime (0700 to 1800 LST) of BC mass concentrations was estimated and found to be larger than the unity (1.58), which clearly indicated that the nocturnal peak is the evident by

changes in emission sources and variability in meteorological conditions. Season-wise, its ratio was found to be 1.62, 1.60, 1.75 and 1.53 during the summer, monsoon, post-monsoon and winter seasons, respectively suggesting relatively large meteorological effect in the nighttime as compared to daytime. In most of the months, the ratios were within 1.38 (February) to 2.11 (May), which verified that there was a remarkable accumulation of BC at nighttime mainly attributed due to the prevailing meteorological conditions such as low mixed layer depths. During winter, the ratio was low because there was no large variation observed in mixing height (Fig. 6) during day and night as compared to summer and monsoon. Juan et al. (2010) have reported similar ratio values at Taklimakan Desert, which was in between 1.17 and 1.50. Babu and Moorthy (2002) and Fochesatto et al. (2001) have also suggested that the ABL evolves after sunrise, in which the strengthened thermal lift eventually breaks the nighttime inversion, causing the aerosols in the residual layer to mix with those near the surface, leading to a sharp increase in the near-surface concentrations. The deepening of the ABL during daytime and the associated convective turbulence thoroughly mixes and redistributes aerosols. On the other hand, at evening, the solar heating decreases and after the sunset, the land cools rapidly leading to inhibition of the thermal-inversion. The mixed layer deforms to a shallow stable boundary layer near the surface and a residual layer aloft separated by a layer of inversion aerosols. Apart from the dynamics of the local ABL, the local traffic and burning of fossil fuels from industrial and urban activities play a crucial role in modifying the diurnal pattern of BC (Husain et al., 2007; Nair et al., 2007). Since the timing of the morning peak varied from day to day and season to season, the morning rush-hour peak in the average profile was broad.

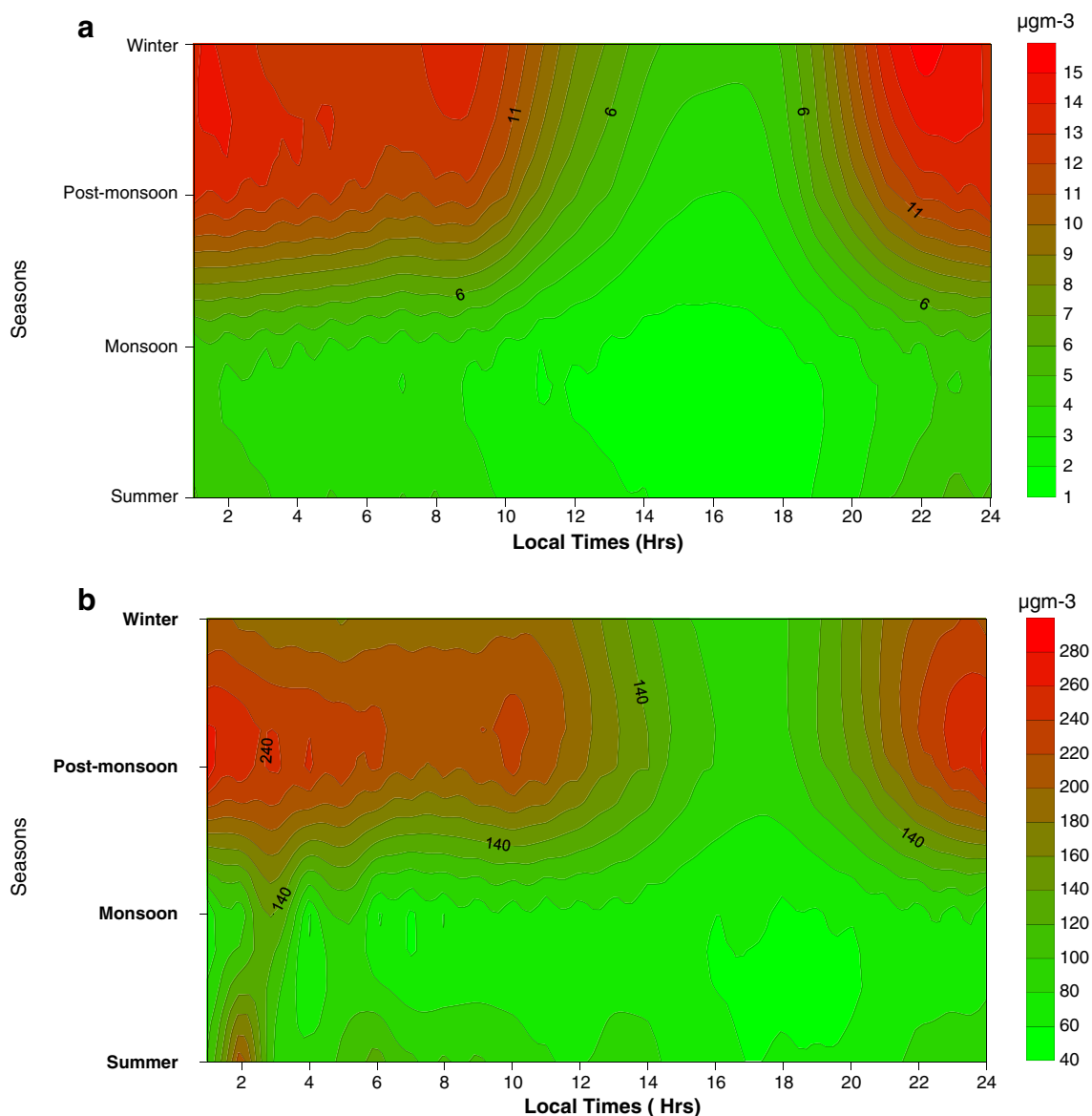


Fig. 5. a. Diurnal variation of mass BC concentrations ($\mu\text{g m}^{-3}$) during different seasons in 2011 at Delhi. b. Diurnal variation of mass $\text{PM}_{2.5}$ concentrations ($\mu\text{g m}^{-3}$) during different seasons in 2011 at Delhi.

3.3. Source identification of BC

As described previously, the AE-31 Aethalometer has seven wavelengths from 370 to 950 nm. A comparative analysis of BC measured at UV (370 nm) and near-IR (880 nm) wavelengths was made which is useful for the source identification of BC (Srivastava et al., 2012b and references therein). The 370 nm is used for identifying the presence of UV absorbing organic compounds (BC_{UV}) even if the values of components measured using this second wavelength cannot be quantified as real BC mass concentration. The levels of BC_{UV} were typically less than or similar to Aethalometer BC (i.e. BC_A at 880 nm) concentrations during the normal measurement period. However, the ratio between BC_{UV} (370 nm) and BC_A (880 nm) of hourly data

during study period was calculated (Fig. 7a) and was found to more than unity during post-monsoon (>1.0) and winter periods (~ 1.0) as compared to summer (0.90) and monsoon (0.80) except in few cases. The study indicates that the UV absorbing organic compound concentrations increased significantly during the colder period than BC_A concentrations. It can be anticipated that some of the most abundant UV absorbing organic species such as PAHs since biomass burning emits various hydrocarbons (McMahon and Tsoukalas, 1978; Jenkins et al., 1996). Thus, the instrument does respond to UV absorbing species and provides information that the mixture of sources of BC has been changed.

During post-monsoon period, 5 min interval data at both wavelengths (370 and 880nm) during 16th – 20th November

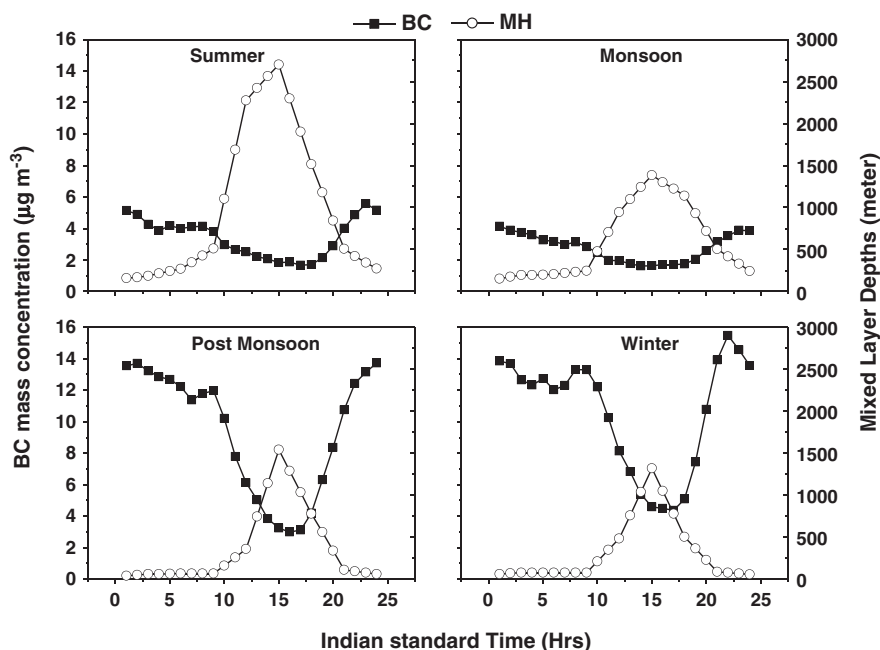


Fig. 6. The diurnally seasonal variations of BC mass concentrations along with mixed layer depths (MLDs) at Delhi during 2011.

2011 were analyzed (Fig. 7b). A positive magnitude of the difference between 370 and 880 nm wavelength was observed on November 16th (1.86), 17th (1.46) and 18th (0.91), which indicates the presence of biomass burning smoke into the atmosphere whereas a negative number on November 19th (−1.63) and 20th (−2.80) indicates no impact from biomass burning. Negative values can also occur due to desorption of UV absorbing semi-volatile organic species from the filter between consecutive samples (Wang et al., 2011a,b). Fine particle ($PM_{2.5}$) was also separated during the same period and found higher concentrations on November 16th ($266.1 \mu g m^{-3}$) followed by 17th ($241.2 \mu g m^{-3}$), 18th ($219.1 \mu g m^{-3}$) and 19th ($193.9 \mu g m^{-3}$). In order to know the effect of long range transport of pollutants during these particular days, 5-day backward air mass trajectories have been computed using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model of the National Oceanic and Atmospheric Administration, USA and shown in Fig. 8. The trajectory analysis indicated that the source of fine particles and BC during these days is basically from the west and northwest parts of India. Ultimately the study indicated that during post monsoon season, the impact of biomass burning was higher as compared to combustion of fossil fuels.

3.4. Meteorological effects on BC

In order to understand the effect of meteorological parameters on BC mass concentrations, a correlation analysis between BC mass concentrations and the corresponding measured meteorological parameters such as wind speed, temperature, relative humidity (RH), visibility and rainfall was studied over Delhi during 2011. BC was found to be negatively correlated with the wind speed ($r = -0.53$) during the entire measurement periods, which is relatively more pronounced during

post-monsoon ($r = -0.64$), winter ($r = -0.56$) and monsoon ($r = -0.54$) periods when the corresponding mean wind speed was 2.53, 2.73 and 3.12 m/s respectively. On the other hand, relatively less, but negative correlation, was observed during the summer ($r = -0.29$) periods when the corresponding wind speed was relatively higher (3.40 m/s). Results are consistent with earlier studies reported by Ramachandran and Rajesh (2007) in Ahmedabad and Srivastava et al. (2012b) in Delhi. It suggests that the major contribution of BC over Delhi is from the local sources, which can accumulate under lower wind speed (observed mostly during the winter and post-monsoon seasons). On the other hand, no or less significant correlation, as observed during the summer suggests that the minor contribution of BC originates from distant sources (Ramachandran and Rajesh, 2007). The open burning of crop residues is a common practice in the north-west part of India during the summer, which results an abundance of biomass aerosols and their transport towards the study region (Srivastava et al., 2012b). Results are found to be similar to that observed over a sub-urban and an urban site in Canada (Sharma et al., 2002) and at an urban site in China (Cao et al., 2009).

In northern part of India, most of the precipitation occurred during southwest summer monsoon period (July to September). The total precipitation during monsoon period of 2011 was 616 mm with highest rainfall recorded in the month of September (150 mm). Correlation analysis between BC and rainfall was also carried out and non-significant correlation (-0.01) was found between these two parameters. However, a significant negative correlation between BC and rainfall was reported in Ahmedabad ($r = -0.35$), in western India (Ramachandran and Rajesh, 2007) and at Trivandrum ($r = -0.74$) in southern India (Babu and Moorthy, 2002). This distinct feature may be due to the long interval of consistent rainfall over northern part of India. Hourly average

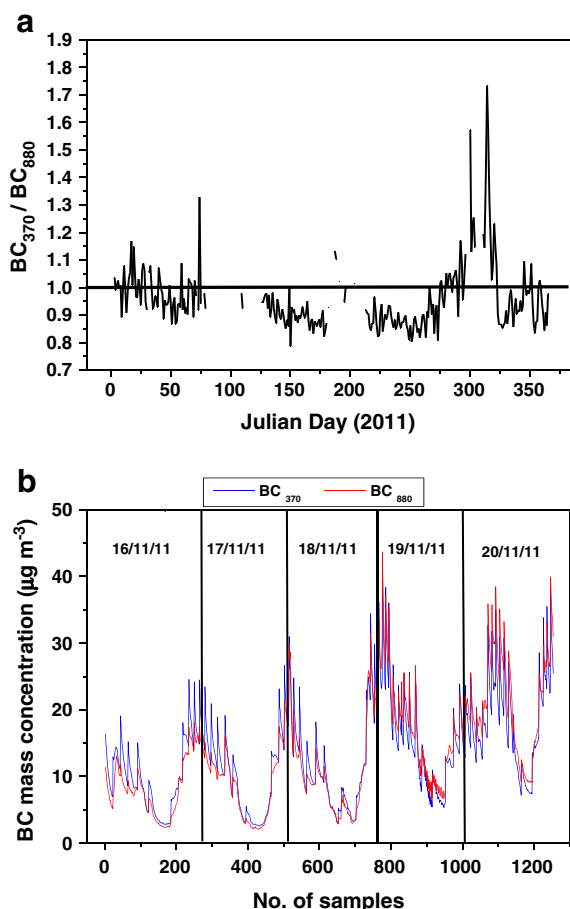


Fig. 7. a. Ratio between BC_{370} and BC_{880} of hourly data during 2011 over Delhi. b. Comparison of Aethelometer BC measured at 880 nm and 370 nm wavelength during 2011 over Delhi.

temperatures ranged from 4 to 44 °C, and an inverse relationship ($r = -0.64$) between temperature and BC can be observed on the basis of daily mean. This is consistent with the expected increase in residential coal burning during low temperatures, as well as with the more stable atmosphere. Cao et al. (2005) showed that residential coal-combustion contributed ~44% of the total carbon in China at Xi'an during the winter period. Stull (1988) has suggested that mixed layer depths (MLDs) determines the volume through which surface-emitted pollutants can be diluted and reflect boundary layer turbulence. The surface boundary layer is shallow over Delhi during winter, which results in pollutant trapping near to the surface. With increases in surface temperatures and convective activity during summer, pollutants are dispersed as the boundary layer deepens, thus lowering the BC concentrations. Negative relationship ($r = -0.54$) between MLDs and BC was seen with relatively higher values in winter and lower in summer. Maximum MLDs was in the month of May (1084 m) when BC was $3.5 \mu\text{g m}^{-3}$; however, lowered MLDs in the month of November (222 m) caused higher BC ($17.9 \mu\text{g m}^{-3}$). MLDs were shallower during post monsoon (379 m) and winter (335 m) as compared to summer (1023 m) and monsoon

(603 m). Seasonal mean magnitude of MLDs and BC was about 1022 m and $3.5 \mu\text{g m}^{-3}$ in summer; 602 m and $2.8 \mu\text{g m}^{-3}$ in monsoon; 379 m and $9.3 \mu\text{g m}^{-3}$ in post-monsoon and 325 m and $10.8 \mu\text{g m}^{-3}$ in winter, respectively. Pollutant dispersion is weaker during the lower MLDs, which is in agreement with the observations made in a coastal area of south China (Cheng et al., 2006).

Sloane and White (1986) have suggested that the loss of visibility is an easily measured manifestation of air pollution, arising from a loss of contrast between the object and the background and attenuation of the light signal from the object due to scattering and absorption of light by fine particulates and gaseous pollutants. In urban areas, it is regarded as a primary index of ambient air quality (Watson, 2002). Relation between visibility and BC was studied and significant inverse correlation (-0.81) was observed over Delhi having higher during post-monsoon (-0.85) and winter (-0.78) when the visibility was generally less than 3 km; however, relatively less correlation was observed during summer (-0.45) and monsoon (-0.54) when visibility was greater than 3 km. It is due to rapid growth of anthropogenic emissions especially fossil fuel consumption and meteorological effects mainly degradation in temperature, MLDs and visibility during winter (Tiwari et al., 2011). A significant proportion (43%) of winds are in calm conditions whereas about 52% are below 2.1 m/s and experiences a thick foggy weather during the winter and show low boundary layer height in the entire northern part of India. During such conditions, pollutants could not be dispersed and mixed with free troposphere. The impact of such conditions is discernible as poor visibility and high levels of pollutants in this region (Mohan and Bhati, 2009). In a recent study, Xu et al. (2012) have also observed negative impact of BC on visibility, having a significant correlation of -0.79 over Shanghai in China. To see the relationship between BC mass concentrations and surface wind direction (WD), a frequency distribution in five categories [East (NE, E, NNE), west (W, WSW, SSW, SW), North (N, NNW, NW, WNW), south (S, SE, ESE, SSE) and Calm] was performed during the study period. The percentage frequency of wind direction was in order of North (56%) > Calm (23%) > East

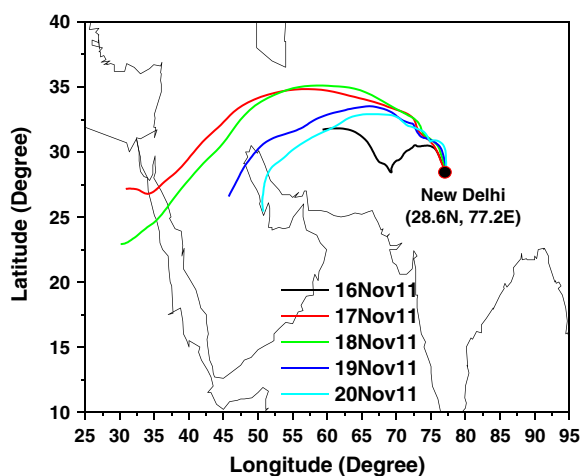


Fig. 8. 5-day back-trajectory analysis of the air masses in Delhi region during 2011.

(18%)>West (14%)>South (13%) during 2011, which corresponds to the average BC concentrations of 7.6, 9.6, 7.6, 10.5 and $3.5 \mu\text{g m}^{-3}$, respectively. In the case of winter, the BC mass concentrations were observed to be higher ($10.8 \pm 6.4 \mu\text{g m}^{-3}$) when the wind was in mostly calm ($\sim 29\%$) condition. In the month of November, we observed that the wind blows mostly from the northwest direction ($\sim 61\%$). At that time, the mean concentration of BC was $\sim 11 \mu\text{g m}^{-3}$, which confirms that the major source of BC during winter period was both localized and with long-range transport of pollutants.

4. Conclusions

Year-long real-time data sets of BC and $\text{PM}_{2.5}$ at 5-min intervals at an urban megacity of “Delhi” were examined for daily, monthly and seasonal temporal scales as well as the influence of meteorology. We observed that the annual mean mass concentrations of BC and $\text{PM}_{2.5}$ were observed to be $6.7 \pm 5.7 \mu\text{g m}^{-3}$ (ranging from 0.9 to $25.5 \mu\text{g m}^{-3}$) and $122.3 \pm 90.7 \mu\text{g m}^{-3}$ (ranging from 54.3 to $338.7 \mu\text{g m}^{-3}$), respectively. Monsoon minima ($2.8 \mu\text{g m}^{-3}$) and winter maxima ($10.8 \mu\text{g m}^{-3}$) of BC can be explained by changes in emission sources and variability in ambient meteorological conditions, especially low wind speeds and shallow mixed layer depths in winter. However, the concentration of $\text{PM}_{2.5}$ was higher during post-monsoon and lower during monsoon. Diurnal distributions showed BC concentration to peak between 0800 and 1000 LST and between 2100 and 2300 LST. The first peak corresponds to morning traffic, while the second peak results from evening traffic combined with meteorological conditions and the nighttime residential cooking and heating (during winter). On an average, the BC aerosol was accounted for $\sim 6\%$ to the $\text{PM}_{2.5}$ mass, with a range of 1.0% and 14.3%. Source identification of BC during post-monsoon in a specified period was performed on the basis of absorption of particles in two different wavelengths at 370 and 880 nm from 16th to 20th November 2011. It clearly indicated that the impact of biomass burning (agricultural residue) was a major source of BC on 16th (1.86), 17th (1.46) and 18th (0.91) November, 2011 while no such impact was observed on 19th (-1.63) and 20th (-2.80) November, 2011. The percentage frequency of WD which was in order of North (56%)>Calm (23%)>East (18%)>West (14%)>South (13%) during the study period corresponds to the BC concentrations of 7.6, 9.6, 7.6, 10.5 and $3.5 \mu\text{g m}^{-3}$, respectively. Correlation analysis between BC and meteorological parameters such as WS, rainfall, and visibility was performed and a large variation was observed. BC was found to be negatively correlated with the WS ($r = -0.53$) during the entire measurement periods, which is relatively more pronounced during post-monsoon ($r = -0.64$), winter ($r = -0.56$) and monsoon (-0.54) periods when the corresponding mean wind speed was 2.53, 2.73 and 3.12 m/s, respectively. On the other hand, relatively less, but negative correlation, was observed during the summer (-0.29) periods when the corresponding wind speed was relatively higher, i.e. 3.40 m/s. Significant inverse relationship was observed between BC and visibility (-0.81) during the entire study period, with higher correlation during post-monsoon (-0.85) and winter (-0.78) season; however, poor correlation was observed during summer (-0.45) and monsoon (-0.54). It is due to rapid growth of anthropogenic

emissions and meteorological effects. Such a systematic and continuous dataset of BC with diurnal variability during different seasons would be helpful in developing emission inventories and validating the chemical transport models.

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