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Variability in atmospheric particulates and meteorological effects on their mass concentrations over Delhi, India

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ABSTRACT

Simultaneous and continuous measurements of $PM_{2.5}$ and PM_{10} along with other co-existent pollutants viz., black carbon (BC), CO, NO and NOx were carried out over Delhi with high resolution (5 min) datasets from 1st Sept. 2010 to 23rd Aug. 2012. Arithmetic mean mass concentrations of $PM_{2.5}$ and PM_{10} were about 130 ± 103 and 222 ± 142 μg m^{-3} respectively during the entire measurement period, which are considerably higher than the annual averages of $PM_{2.5}$ and PM_{10}, stipulated by the National and International standards. It was noticed that the fine mode particles ($PM_{2.5}$) were higher than the coarse mode particles ($PM_{10-2.5}$) during post-monsoon (~89%), winter (~69%) and monsoon (~64%) periods; however, $PM_{10-2.5}$ was higher (~22%) than $PM_{2.5}$ during summer. Arithmetic mean mass concentrations of BC, CO, NO and NOx were about 7 ± 5 μg m^{-3}, 2 ± 1 ppm, 17 ± 17 ppb and 30 ± 24 ppb, respectively. In the present study, highest fraction of BC (~6%) in $PM_{2.5}$ mass was in winter, whereas the lowest fraction (~4%) was in summer. Relationships among PMs (particulate matters) and other pollutants indicated that the fine mode particles are highly correlated with BC (0.74) and CO (0.51). The effects of meteorological parameters on aerosols have been studied and a significant negative relationship (−0.45) between mixing height (MH) and $PM_{2.5}$ has been noticed. Higher correlation was during winter (−0.55), however lower was in summer (−0.16). Relation between visibility (VIS) and $PM_{2.5}$ was higher during post-monsoon (−0.85) and winter (−0.78) when the visibility was around 2 km; however, it was relatively less correlated when VIS was greater than 2 km during summer and monsoon. Relationship between $PM_{2.5}$ and relative humidity (RH) showed a significant negative correlation (−0.56) for the entire study period. A positive correlation (0.32) was observed during the winter period with fine mode particles whereas negative correlation was seen with coarse mode particles during monsoon (−0.70) and summer (−0.51).

1. Introduction

Particulate matters (PMs), including $PM_{2.5}$ (atmospheric dynamics equivalent diameter ≤2.5 μm) and PM_{10} (atmospheric dynamics equivalent diameter ≤10 μm) mass concentrations, have been widely studied in recent years due to their potential impacts on health, environment, visibility and climate (Schwartz et al., 1996; Vedal, 1997; Pillai et al., 2002; IPCC, 2007). Particulate sizes of less than 10 μm are able to absorb toxic substances and can enter the human body by deposition in the lungs through respiration, resulting in various respiratory and cardiovascular diseases (Makkonen et al., 2010), mortality (Lim et al., 2010) and morbidity (Dockery et al., 2005; Katsouyanni et al., 2001). Due to their relatively large size (and mass), these

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particles generally fall to the ground through wet and dry deposition processes whereas fine particles can remain suspended in the atmosphere for several days to weeks and can be transported to the other locations through atmospheric circulations and impact the environment of far locations (Furuta et al., 2005). Furthermore, atmospheric particulates also affect cloud formation and cessation, which regulate heat transfer in the atmosphere, thereby contributing to climate change. These findings have underlined the importance of ambient particles and the need for monitoring PM and its fractions. These data is categorically considered important in order to: i) characterize the physical and chemical properties of particles, ii) identify major particle sources and quantify their contributions, iii) assess the spatial and temporal variability, and iv) investigate the impact of particles on climate and health.

The Indo-Gangetic Basin (IGB) region, encompassing most of the northern part of India, is the world's most populated river basin, where more than 700 million inhabitants are living and exposed to the enormous pollution from various anthropogenic and natural sources (Srivastava et al., 2012a; Tripathi et al., 2006; Dey and Tripathi, 2007). During winter season, the contribution of widespread biomass and agricultural crop residue burning along with the meteorological effect and enhancements in pollution level are common over the IGB region (Awasthi et al., 2011; Badarinath et al., 2006). In addition to this, during summer season, the air over the IGB is strongly affected by mineral dust transport from the Southwest Asian regions including the desert that stretches from Iran through Afghanistan and Pakistan, northwestern India and the Arabian Peninsula (Prospero et al., 2002; Dey et al., 2004). These mineral dusts strongly influence the regional aerosol optical properties over the IGB (Singh et al., 2004; Gautam et al., 2009; A.K. Srivastava et al., 2011; M.K. Srivastava et al., 2011). Over the IGB, however, limited studies have been conducted to characterize the PM concentrations though for a limited time period (Pipal et al., 2011; Kulshrestha et al., 2009; Srivastava et al., 2009; Das et al., 2009; Tiwari et al., 2009, 2012, 2013). In an effort to address some of these particle-related issues, we conducted simultaneous $PM_{2.5}$ and PM_{10} sampling, using USEPA-approved samplers, during the period from 1st Sept. 2010 to 23rd Aug. 2012 in Delhi, India. To the best of our knowledge, this dataset represents the longest ever record of continuous $PM_{2.5}$ and PM_{10} at a fixed site measurement (real time) with high resolution (5 min) in the IGB. In this paper, we used two year datasets for estimating daily levels of $PM_{2.5}$ and PM_{10} mass concentrations and seasonal averages to qualify the data in accordance with standards of NAAQ (National Air Quality), EUAQ (European Union Air Quality) and USEPA (United State Environmental Protection Agency). Furthermore, the study is also done to assess the air quality in the study area in terms of fine ($PM_{2.5}$) and coarse particles ($PM_{10-2.5}$) to inhalable particles (PM_{10}) and to examine the relationships between PM and other co-pollutants such as BC (black carbon), CO (carbon monoxide), and NOx (oxides of nitrogen). The effects of meteorological parameters on PM mass concentrations were also studied.

2. Sampling site and meteorological conditions

Samples of PM and other co-pollutants were collected in the premises of Indian Institute of Tropical Meteorology (Branch), New Delhi (28° 37′N, 77°12′E, ~250 m amsl), located in the heart of the city. Delhi is situated on the banks of the Yamuna River and covers an area of ~1500 km^2. The location is ~1100 km away from the nearer coast of the Arabian Sea and considered as the fourth most polluted city in the world with respect to suspended particulate matters along with high population (~18 million inhabitants) (Goyal and Sidharta, 2002; Tiwari et al., 2012). Apart from this, 4.8 million registered vehicles, three coal-based thermal power plants and small and medium scale industrial units (125,000 nos.) are situated in Delhi (Srivastava et al., 2009). The major sources of pollution over Delhi are thermal power plants, transportation, small-scale industries and domestic cooking/heating. Delhi is located at the border between the rich rain-washed Gangetic plains in the east and semi-arid tracts of Rajasthan to the southwest.

The climate of Delhi is semi-arid and has large variations in meteorological parameters such as mixing height (MH), wind speed (WS), temperature (Temp.), wind direction (WD), relative humidity (RH) and visibility (VIS) during the different seasons. However, in an earlier study, Tiwari et al. (2009) have reported the maximum temperature of ~45–48 °C during summer and a minimum of ~1–2 °C during winter over Delhi. To look into the seasonal trend of atmospheric aerosols, it was divided into four distinct seasons i.e., pre-monsoon or summer (April–May–June), monsoon (July–August–September), post-monsoon (October–November) and winter (December–January–February–March) (Perrino et al., 2011). Generally, the pre-monsoon period is impacted by dust episodes transported through northwesterly and westerly winds from northwest India and Southwest Asia. However, monsoon period is dominated by high humidity. During the post-monsoon and winter seasons, the wind mostly reaches from northwest azimuth which brings dry, cool air (called western disturbances) and winds spill across the northwest Himalayas and flow to the central part of IGB and southeast across the country (Mishra and Shibata, 2012). During the study period, meteorological parameters except MH were taken from the India Meteorological Department observatory (automatic weather station) which is about 500 m away from the center in the premises of Indian Agricultural Research Centre. The boundary layer mixing height was obtained from the reanalysis data available on 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://www.arl.noaa.gov/ready.html; Draxler and Rolph, 2003). These data were collected on hourly intervals and analyzed for daily, monthly, and seasonal intervals to obtain the behavior of the meteorological parameters during the observational period.

3. Instrumentations

On-line beta-attenuation analyzers of Thermo Andersen, Inc. USA (Series FH 62 C14) were used for the measurement of ambient mass aerosols such as $PM_{2.5}$ and PM_{10} with five minute intervals at a height of 3 m from the ground. The $PM_{2.5}$ cut-point was achieved through sharp-cut cyclone inlet with the flow rate through the instrument specified as 1 m^3/h (Kenny et al., 2000). The measuring range of the instrument is 0–5000 μg m^{-3} with a minimum detection limit of 1 μg m^{-3} for 24 h average. More details of the above instruments are

given elsewhere (Hyvarinen et al., 2009). The measurements of BC mass concentrations were carried out by 7-Wavelength Aethalometer (Model AE-31, Magee Scientific Company, Berkley, CA, USA) with a temporal resolution of 5 min. The mass concentration of BC was estimated from the light attenuation at 880 nm using a specific mass absorption cross-section (MAC) of 16.6 $m^2\ g^{-1}$, as recommended by the manufacturer (Hansen, 2005). The detailed measurement uncertainties were described elsewhere (Weingartner et al., 2003; Collaud et al., 2010).

NOx measurement was performed continuously using a NOx analyzer (Model 42i, Thermo Scientific, USA). The instrument analyzes ambient air by the chemiluminescence technique, at a time base of 1 min. Basically, the reaction of NO on ozone produces NO_2 in the excited state, which subsequently returns to stable state, while emitting a chemiluminescent radiation. NOx concentration is measured by sampling air through a molybdenum catalytic converter. CO was also simultaneously measured directly by CO analyzer (Model 49i, Thermo Scientific, USA). The Model 49i is based on the principle that carbon monoxide (CO) absorbs infrared radiation at a wavelength of 4.6 μm. The data, used for interpretation, were taken by daily arithmetic average of the observed data of $PM_{2.5}$, PM_{10}, BC, CO and NO_X.

4. Results and discussion

4.1. Mass concentrations of atmospheric particulate matters (PMs)

Simultaneous measurement of $PM_{2.5}$ and PM_{10} mass concentrations was done for two years from 1st Sept. 2010 to 23rd Aug. 2012 at Delhi and its day to day variability is shown in Fig. 1. Log-normal distribution of mass concentrations of PM_{10}, $PM_{2.5}$ and $PM_{10-2.5}$ was also plotted and are depicted in Fig. 2. The arithmetic mean mass concentrations of $PM_{2.5}$ and PM_{10} for the entire two year period were 129.8 ± 103.4 and 222.0 ± 142.0 μg m^{-3}, respectively. These concentrations are considerably higher than the annual average standards stipulated by the National Ambient

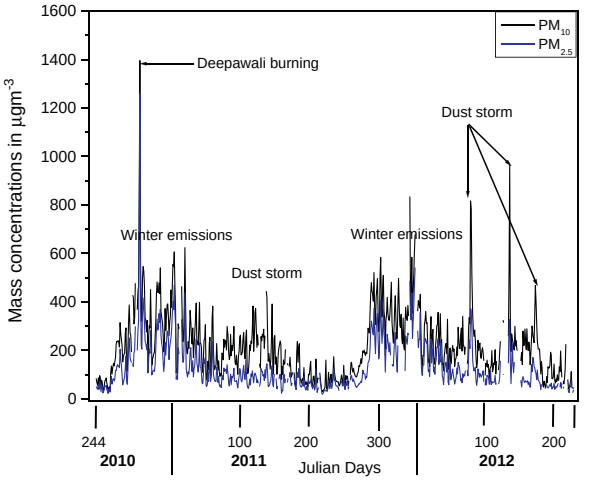

Fig. 1. Day to day variations of $PM_{2.5}$ and PM_{10} from 2010 (September) to 2012 (August) over Delhi.

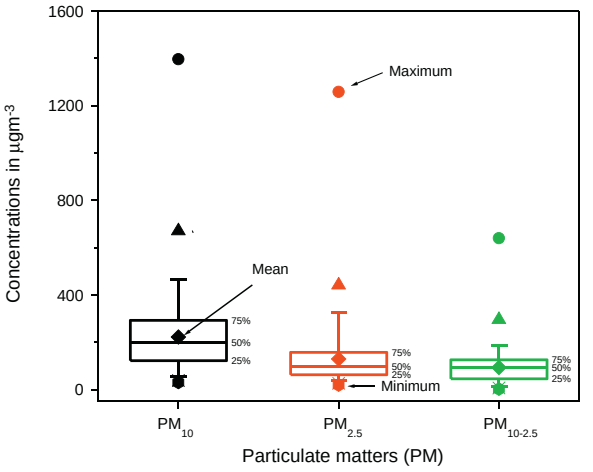

Fig. 2. The boundaries of the box indicate the 25th and 75th percentiles; a line within the box marks the median; whiskers indicate the 5th and 95th percentiles of mass PM_{10}, $PM_{2.5}$ and $PM_{10-2.5}$.

Air Quality Standards (NAAQS: http://cpcb.nic.in/National-Ambient-Air-Quality-Standards.php) ($PM_{2.5}$ = 40 μg m^{-3} and PM_{10} = 100 μg m^{-3}), as well as by the United States Environmental Protection Agency (USEPA) ($PM_{2.5}$ = 15 μg m^{-3} and PM_{10} = 50 μg m^{-3}) standards (http://www.epa.gov/air/criteria.html) and European Union Air Quality Standards (EUAQS) ($PM_{2.5}$ = 25 μg m^{-3} and PM_{10} = 40 μg m^{-3}) (http://ec.europa.eu/environment/air/quality/standards.htm). However, one should keep in mind that the United States compliance with the air quality $PM_{2.5}$ standard is based on measurements from multiple monitoring sites within the city over a period of three years. This is to ensure that the $PM_{2.5}$ compliance measurements are representative of the average population exposure (population based standard). In our study, the average concentration of $PM_{2.5}$ (129.8 ± 103.4 μg m^{-3}) was based on measurements conducted at a single site for a two year sampling, located next to a highly trafficked and frequently congested street in the heart of Delhi. $PM_{2.5}$ and PM_{10} mass concentrations showed considerable daily variability ($PM_{2.5}$ = 19.4–1257.9 μg m^{-3}; PM_{10} = 29.9–1395.5 μg m^{-3}) in agreement with the previous works conducted in the northern part of India (Tiwari et al., 2009 and reference therein). During precipitation, which efficiently removes mostly coarse particles from the atmosphere, very low concentrations of $PM_{2.5}$ and PM_{10} were observed. To assess the association between different sizes of PM, linear regression analysis between $PM_{2.5}$ and PM_{10} was carried out. A significant positive relationship between PM_{10} and $PM_{2.5}$ ($y = 0.7088x - 26.946$: $r^2 = 0.88$) was observed which indicated the possibility of similar sources implying that variation in PM_{10} mass is governed by the variation in $PM_{2.5}$ mass. During the study periods, approximately 70% and 87% of the daily $PM_{2.5}$ concentrations exceeded the NAAQS and USEPA limit respectively. The highest mass concentrations of PM_{10} (408 μg m^{-3}) and $PM_{2.5}$ (318 μg m^{-3}) were observed in the month of December 2011, whereas the low concentrations of PM_{10} (75 μg m^{-3}) and $PM_{2.5}$ (47 μg m^{-3}) were observed in the month of August, 2011 and 2012, respectively. Pipal et al. (2011) have reported a similar level of $PM_{2.5}$ (90.16 ± 7.21 μg m^{-3}) and PM_{10} (278.67 ±

106.58 μg m^{-3}) over Agra, which is another urban location ~200 km away from Delhi. Daily mean mass concentrations of coarse mode particles ($PM_{10-2.5}$) were calculated and varied between 2.4 and 639 μg m^{-3} (with mean of 93.02 ± 62.6) (Fig. 2). To date, neither the European Union nor the United States have established an air quality standard for $PM_{10-2.5}$. Currently, the USEPA is in the process of developing a coarse particle standard, $PM_{10-2.5}$, in lieu of the existing PM_{10} standard. Frequency distribution of fine ($PM_{2.5}$) and coarse ($PM_{10-2.5}$) mode particles was separated in 11 segments ranging between <25 and >275 μg m^{-3} with a 25 μm interval (Fig. 3). $PM_{10-2.5}$ was found to be higher (~20%) in the spectrum of 75 to 100 μg m^{-3} as compared to $PM_{2.5}$, whereas lower (~50%) was in the spectrum of 200 to 225 μg m^{-3}. In the case of fine particle ($PM_{2.5}$), it was higher (~23%) in the segment of 50–75 μg m^{-3} and lower (~2%) in the spectrum of 175 to 200 μg m^{-3}. Stringent feature was seen in $PM_{2.5}$, which is ~11% higher in the segment of >250–275 μg m^{-3} whereas in the case of coarse mode particle, it was only ~2%. This distinct feature in the case of fine particle during the study period was mainly due to anthropogenic activities and meteorological effects.

On the basis of seasonal analysis, the variations of $PM_{2.5}$ (fine) and $PM_{10-2.5}$ (coarse) mass concentrations were calculated. The mass loading was found to be in the order of: post-monsoon (205.51 μg m^{-3}) > winter (169.42 μg m^{-3}) > summer (91.00 μg m^{-3}) > monsoon (56.30 μg m^{-3}) for $PM_{2.5}$ however, for $PM_{10-2.5}$, it was summer (116.23 μg m^{-3}) > post-monsoon (108.88 μg m^{-3}) > winter (100.08 μg m^{-3}) > monsoon (34.38 μg m^{-3}). During the study period, it was noticed that the $PM_{2.5}$ was ~89%, 69% and 64% higher than the $PM_{10-2.5}$ during post-monsoon, winter and monsoon, respectively; however, during summer, the $PM_{10-2.5}$ was higher (~22%), due to dust storm activities (Kulshrestha et al., 2009). In addition to this, large variations in mass concentrations of both $PM_{2.5}$ and PM_{10} aerosols during different seasons over Delhi could be due to the thermodynamic conditions in the planetary boundary layer. In the case of $PM_{2.5}$, it was observed that the concentrations were higher during post-monsoon and winter, which is due to very frequent and persistent thermal inversion and foggy conditions at ground level causing a considerable amount of aerosols to accumulate in the lower layers of the atmosphere; however, during monsoon, it was minimum due to the wash-out effect. Apart from the meteorological effect, the impact of fire crackers during festivals such as Diwali and massive biomass burning of crop residue over the western part of India, especially in the states of Haryana and Punjab, plays a crucial role in the enhancement of fine mode aerosol mass concentrations during post-monsoon and winter (Awasthi et al., 2011; Tiwari et al., 2009; Hyvarinen et al., 2010). In a recent study, Guttikunda and Gurjar (2012) have reported two to three times higher concentrations of PM during winter as compared to the summer season over Delhi, which was due to the effect of local meteorology and pollution. In another study, Tiwari et al. (2009, 2013) have found that the atmosphere of Delhi during winter is characterized by low MH, WS, and RH and 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. 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 the surface. In general, $PM_{2.5}$ and PM_{10} show lower concentrations (order of magnitude) during rainy events since precipitation removes the particles from the atmosphere. PM_{10} indicates higher concentrations in summer, which were often associated with intense sporadic peaks of mineral dust (Kubilay et al., 2000; Kocak et al., 2004).

Mass concentrations of PM ($PM_{2.5}$ and PM_{10}) in the present study over Delhi were compared with earlier reported mass concentrations of PM in different parts of northern India and other global locations and were depicted in Table 2. The level of PM mass concentrations over Delhi in the present study is largely similar to that of the other Indian sites, especially in the northern part of India such as at Agra (Pipal et al., 2011), Delhi (Dey et al., 2012; Tiwari et al., 2009; Guttikunda, and Calori, 2013), Lucknow (Pandey et al., 2012), Kanpur (Sharma and Maloo, 2005) and Patiala (Awasthi et al., 2011). Kulshrestha et al. (2009) have also observed higher mass concentrations of PM_{10} (170 μg m^{-3}) and $PM_{2.5}$ (136 μg m^{-3}) at Agra and reported much higher concentrations in summer during dust

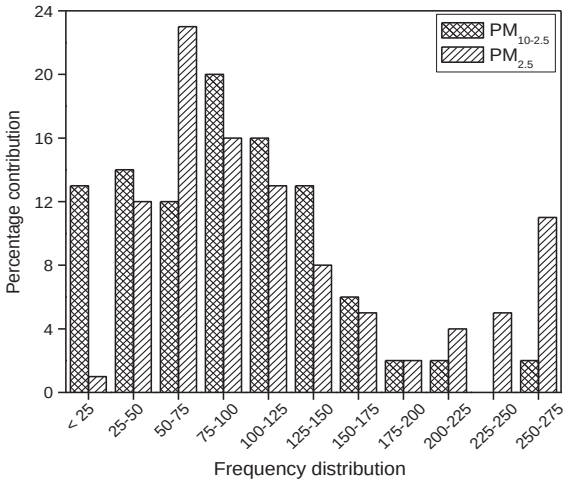

Fig. 3. Absolute frequency distribution of $PM_{2.5}$ and $PM_{10-2.5}$ over Delhi.

Table 1
Correlation analysis among PM and gaseous pollutants during the different seasons.

	BC	CO	NO	NO_X
PM_{10}				
Over all study	0.69	0.43	0.41	0.47
Winter	0.75	0.40	0.48	0.43
Summer	0.07	0.06	−0.02	0.09
Monsoon	0.18	−0.16	−0.07	−0.06
Post-monsoon	0.62	0.46	0.39	0.43
$PM_{10-2.5}$				
Over all study	0.34	0.14	0.38	0.30
Winter	0.38	0.04	0.36	0.33
Summer	−0.04	0.09	−0.21	−0.03
Monsoon	0.07	−0.13	−0.27	−0.28
Post-monsoon	0.53	0.22	0.21	0.26
$PM_{2.5}$				
Over all study	0.74	0.51	0.32	0.46
Winter	0.80	0.53	0.45	0.39
Summer	0.23	−0.03	0.27	0.23
Monsoon	0.23	−0.18	0.17	0.19
Post-monsoon	0.55	0.46	0.38	0.42

Table 2
Comparison of $PM_{2.5}$ and PM_{10} mass concentrations in the present study over Delhi with earlier reported mass concentrations of PMs in different parts of northern India and other global locations.

Places	$PM_{2.5}$ (μg m^{-3})	PM_{10} (μg m^{-3})	References
Delhi	129.8 ± 103.4	222.0 ± 142.0	Present study
Agra	90.16 ± 7.2	278.67 ± 106.6	Pipal et al. (2011)
Delhi	148.4 ± 67	ND	Dey et al. (2012)
Delhi	97 ± 56	219 ± 84	Tiwari et al. (2009)
Delhi	123 ± 87	208 ± 14	Guttikunda and Calori (2013)
Lucknow	101.05 + 22.5	204.0 + 26.7	Pandey et al. (2012)
Kanpur	95	281	Sharma and Maloo (2005)
Patiala	57 ± 2	97 ± 2	Awasthi et al. (2011)
Agra	136	170	Kulshrestha et al. (2009)
Lahore, Pakistan	91	489	Colbeck et al. (2011)
Iran	69 ± 583.2	319 ± 407.1	Shahsavani et al. (2012)
Beijing, China	86.6	ND	Zhao et al. (2009)
Taipei, Taiwan	21.82 ± 7.5	39.45 ± 11.6	Gugamsetty et al. (2012)
Haarlemmerweg, Germany	17.8	27.5	Boogaard et al. (2011)
USA	2.9 ± 1	9.4 ± 7	Wagner et al. (2012)
Helsinki, Finland	9.6	18.7	Laakso et al. (2003)
Athens, Greece	40.2	75.5	Chaloulakou et al. (2003)
Ordos, China	51.8	89.1	Wang et al. (2012)
Fuzhou city, China	44.33 ± 16.3	ND	Xu et al. (2012)
Dhaka, Bangladesh	36.7	97.7	Begum et al. (2013)

ND = No data.

storm whereas lower concentrations was observed during monsoon due to the washout effect. In other studies, similar concentrations of PM in Delhi were also observed at Lahore, Pakistan (Colbeck et al., 2011), Iran (Shahsavani et al., 2012), and Beijing, China (Zhao et al., 2009). However, Taipei, Taiwan (Gugamsetty et al., 2012), Haarlemmerweg, Germany (Boogaard et al., 2011), USA (Wagner et al., 2012), Helsinki, Finland (Laakso et al., 2003), Athens, Greece (Chaloulakou et al., 2003), Ordos, China (Wang et al., 2012), Fuzhou City, China (Xu et al., 2012) and Dhaka, Bangladesh (Begum et al., 2013) were observed with relatively lower concentrations of PM in comparison to the present study. The present study indicates that the Delhi is one of the most polluted cities in Asia, as well as in the world, with respect to fine particulate matter, which is more responsible for respiratory problems and human health (Pope et al., 2009).

4.2. Relationships between $PM_{2.5}$, PM_{10} and $PM_{10-2.5}$

The ratios between $PM_{2.5}$ and PM_{10} were calculated on a monthly basis and was found to have a large variability, ranging from 0.41 (June) to 0.74 (December) with a mean value of 0.57 ± 0.10 over the station during the study period, however, the ratios between $PM_{10-2.5}$ and PM_{10} were 0.43 ± 0.10, ranging from 0.32 (December) to 0.58 (June). Season wise ratios between $PM_{2.5}$ and PM_{10} were also analyzed and were found to be highest during post-monsoon (0.63) followed by winter (0.62), monsoon (0.61) and summer (0.45) which clearly indicated that the maximum contribution of fine particles ($PM_{2.5}$) was during post-monsoon and winter and decreases during the summer (0.44). The ratio between $PM_{2.5}$ and PM_{10} was higher during the monsoon period due to the removal of coarse mode particles. During post-monsoon and winter, the boundary layer heights, which are generally low, play a crucial role in the trapping of fine particles near the surface emitted by man-made activities such as vehicular exhaust, power plants and industries as compared to other seasons (Stull, 1988; Ramachandran and Rajesh, 2007; Tiwari et al., 2013). In the summer, the ratio was lower due to the enhanced concentration of coarse mode particle, mostly from long range transport of dust from the western part of India where Thar Desert is located (Pandithurai et al., 2008; A.K. Srivastava et al., 2011; M.K. Srivastava et al., 2011). Singh et al. (2005) reported that the high suspended particulate matter (SPM) concentration was due to desert aerosols during the pre-monsoon period (April to June) in 2003 over Delhi. They have also reported low Angstrom exponent (AE: 0.328) with a drastic decrease up to 0.06 during severe dusty event. The seasonal ratios between coarse mode ($PM_{10-2.5}$) and PM_{10} were also calculated and were found to be different than fine particle behavior. It was observed to be higher during summer (0.55) followed by monsoon (0.39), winter (0.38) and post-monsoon (0.37). This large variability in fine and coarse mode particles in PM_{10} may be due to the variability in aerosol chemical composition and its sizes, which is affected by various sources (both natural and man-made) as well as meteorological conditions.

We have compared the ratio ($PM_{2.5}/PM_{10}$) with other studies conducted in India and abroad. The ratios were reported on the basis of a large number of urban and semi-rural US areas and were varied between 0.3 and 0.7 (EPA, 2001). Higher ratios were observed in the Northeastern US, especially during summer, when sulfate compounds represent a large fraction of PM_{10}. In contrast, low $PM_{2.5}/PM_{10}$ ratios were measured in the semi-arid Western US, where a large fraction of PM_{10} consists of soil particles. Harrison et al. (1997) found that $PM_{2.5}$ comprises about 80% of PM_{10} during the winter months at Birmingham, UK. Also, a lower fraction was reported for the summer months with $PM_{2.5}$ accounting for approximately 50% of PM_{10} concentrations. Sharma and Maloo (2005) have reported similar ratio (~0.58) at three different sampling locations in a highly polluted city in the

northern part of India, at Kanpur during the winter season of 2002–03. Kocak et al. (2007) have reported a great variability in the ratios of $PM_{2.5}/PM_{10}$ in Turkey that ranged from 0.25 to 0.90. Rajsic et al. (2004) have reported a higher ratio (0.78) of $PM_{2.5}/PM_{10}$ at an urban city, Belgrade, with a lower value during summer (0.62) and higher during winter (0.85). They have also reported that during the intensive photochemical episode, the ratio reached 0.96; and in severe-pollution episode during winter, the ratio was 0.88. Pipal et al. (2011) have reported a very high $PM_{2.5}/PM_{10}$ ratio in a rural site (0.70) in Agra and lower at a road site location (0.32). Wang et al. (2013) reported a very high ratio (0.70) between $PM_{2.5}$ and PM_{10} in Shanghai, China due to an industrial effect. Tiwari et al. (2009) have reported the ratio ~0.48 ± 0.2 over Delhi which varied between 0.18 (June) and 0.86 (February), suggesting the dominance of coarse mode particles in summer and fine mode particles in winter.

4.3. Relationships among PM and other pollutants

Black carbon (BC) and other co-pollutants such as CO, NO, and NOx along with PM were also studied simultaneously during the same period and their arithmetic means (two years) were found to be 7.01 ± 5.3 μg m^{-3}, 2.0 ± 1.4 ppm, 17.2 ± 17.0 ppb and 29.6 ± 24.1 ppb, respectively. The BC abundance in $PM_{2.5}$ is affected by the mixture of pollution sources and the meteorological conditions. In the present study, the highest fraction of BC (~6%) in $PM_{2.5}$ mass was in winter, whereas the lowest fraction (~4%) was in summer. Husain et al. (2007) have reported that the BC contributes ~5–15% of $PM_{2.5}$ in the urban air (Lahore, Pakistan). Tripathi et al. (2005) have reported the fraction of BC to be ~7% to 15% to the total suspended particulate (TSP) during the winter period at Kanpur, whereas it was ~4% to 15% to the TSP at Delhi (Ganguly et al., 2006), 7% of the TSP at Hyderabad (Latha et al., 2005), and 2.3% of the TSP in Pune (Safai et al., 2007). In the sub-urban regions of Europe and North America, the BC contributed about 5% of TSP (Ramanathan and Crutzen, 2003) and ~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 contribution is due to non-carbonaceous material (fugitive dust). Badarinath et al. (2009) have found that the fraction of mass BC to the total mass concentration of aerosols was ~6% at Anantapur, a rural environment in Andhra Pradesh. On the other hand, BC was found to contribute <3% to the total aerosol mass at Manora Peak, situated in the Himalayan foothills (Srivastava et al., 2012b). Overall, the concentrations of $PM_{2.5}$ and PM_{10} were observed to be higher, as discussed earlier, due to anthropogenic activities, which are responsible in generating the aerosols from incredible establishment of small industries, vehicle density, house cooking, thermal power plants and unplanned road and constructional activities. Moreover, the pollutants are also transported from the nearest polluted areas of IGB (Mishra and Shibata, 2012) as well as from western azimuth (M.K. Srivastava et al., 2011). Correlation analysis among fractions of PM (PM_{10}, $PM_{2.5}$ and $PM_{10-2.5}$) mass concentrations and BC, CO, NO, and NOx was calculated (Table 1). The correlation coefficient (r) between mass of PM_{10} and BC concentrations was (0.69) for the entire study period. When the data were analyzed for various seasons, the correlation between PM_{10} and BC was 0.75 and 0.62 for winter and post monsoon, respectively; however, during summer and monsoon, no significant correlations was seen. The correlation coefficient between PM_{10} and CO was 0.43 for the entire period, whereas it was about 0.40 during winter and 0.46 during the post-monsoon period. Similar strong associations between $PM_{2.5}$ and other pollutant concentrations were also found in the winter and post-monsoon periods with a lesser extent. For fine mode aerosols, significant correlation (0.74) was observed between $PM_{2.5}$ and BC during the entire study period over Delhi indicating the similarity in sources, which was relatively high (0.80) during winter and low during monsoon and summer (~0.23). Furthermore, the correlation for NO and NOx was observed to be 0.41 and 0.43 with PM_{10} whereas it was 0.32 and 0.46 with $PM_{2.5}$ during the entire study period. In general, the atmosphere of Delhi during the winter season is characterized by low relative humidity and 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. 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 the surface. In such conditions, the probabilities in the formation of secondary aerosols are higher (Tiwari et al., 2012). Strong associations between particle mass concentrations and primary gaseous pollutants had been previously reported for Birmingham, UK air quality study, suggesting the importance of road traffic related emissions (Harrison et al., 1997).

4.4. Diurnal variations of PM

The data of PM_{10} and $PM_{2.5}$ were diurnally analyzed and the seasonal diurnal variability in PM_{10} and $PM_{2.5}$ concentrations was shown in Fig. 4a and b, respectively. Two peaks were observed in both PM_{10} and $PM_{2.5}$ concentrations with gradual build-up around 0800 h (Local Standard Time, LST), peaking around 0900 to 1100 h LST, and low values in the afternoon and secondary maxima between 2100 h LST till midnight. Diurnal mean lowest and highest of $PM_{2.5}$ and PM_{10} varied from 81.2 (1700 h LST) to 167.7 (0100 h LST) μg m^{-3} and 155.5 (1800 h LST) to 257.8 (0900 h LST) μg m^{-3}. The higher concentrations of $PM_{2.5}$ were observed during mid-night, whereas PM_{10} was higher during evening time; it may be due to the impact of boundary layer and re-suspension of inhalable particle. In day and night comparison, ~20% of $PM_{2.5}$ mass was found to be higher during nighttime (1900 to 0600 h LST) as compared to daytime (700 to 1800 h LST); however, in the case of PM_{10}, it was opposite with higher (~3%) during daytime as compared to nighttime. This indicates that the source of PM_{10} was from re-suspension of soil dust particles due to traffic movement. On the basis of diurnal seasonal analysis, it was observed that the nighttime $PM_{2.5}$ and PM_{10} were higher during post-monsoon as compared to other seasons. We reached to the conclusion that both meteorological parameters especially boundary layer condition and WS as well as man-made emissions from various sources play a crucial role in the dispersal and accumulation of the aerosols over Delhi (detailed discussions in Section 4.5). In an urban environment, the amount of PM variation of the exposure of aerosols from anthropogenic activities is also observed in India (Badarinath et al., 2009; Ramachandran and Kedia, 2010; Pathak et al., 2010).

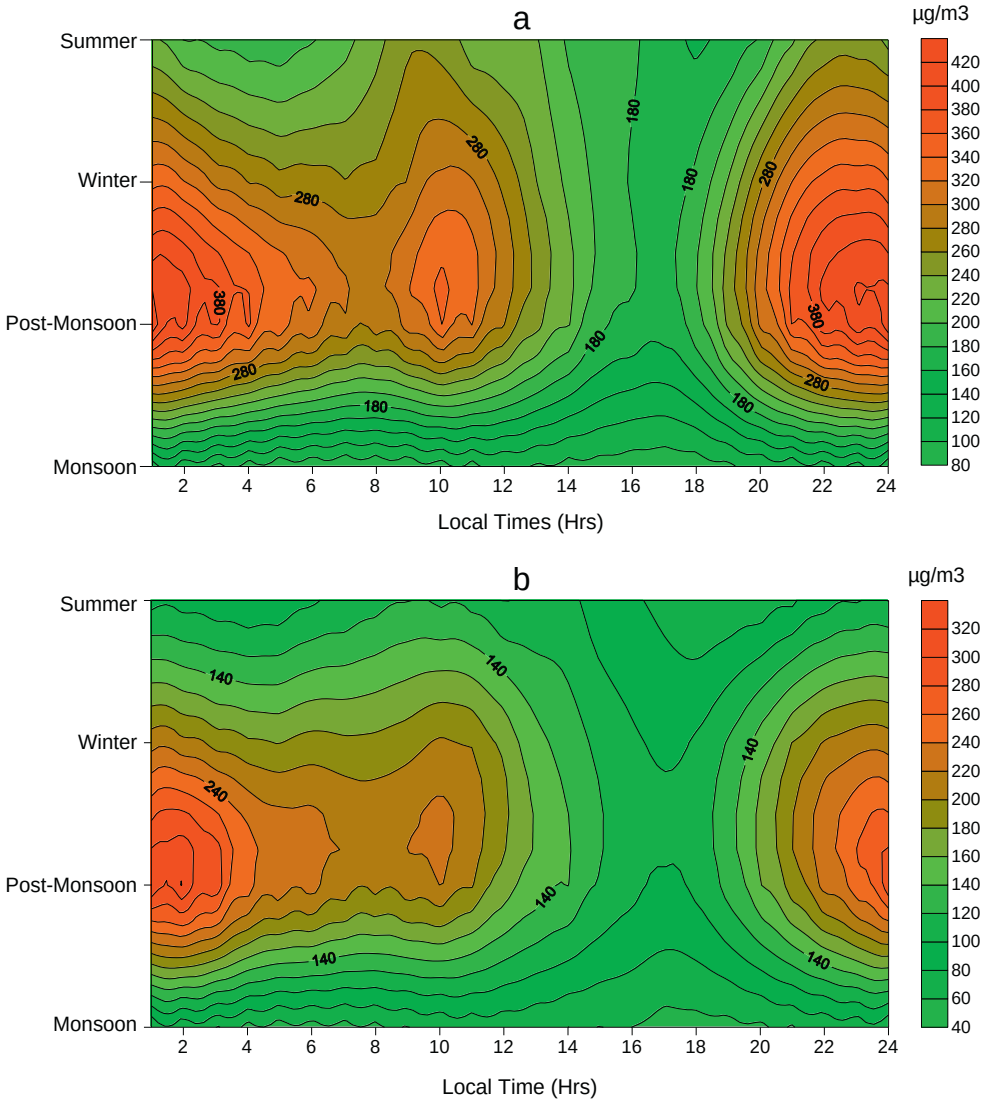

Fig. 4. a: Seasonal diurnal variation of mass PM_{10} concentrations (µg m^{-3}) from September 2010 to August 2012. b: Same as panel a for $PM_{2.5}$ mass concentrations (µg m^{-3}).

This fact was also observed by DeGaetano and Doherty (2004) in New York City and Zhao et al. (2009) in Beijing, China. The results indicate the seriousness of fine particulate pollution in Delhi and its influence in the surrounding areas.

4.5. Relationships among PM and meteorological parameters

Relationships between particle mass ($PM_{2.5}$ and PM_{10}) concentrations and meteorological parameters, such as mixing height (MH), wind speed (WS), temperature (Temp.), relative humidity (RH), rainfall (RF) and visibility (VIS), were studied to understand the impact of meteorological parameters on aerosols. During the study period, the daily average value of two years of meteorological parameters with their variation was recorded as MH, varied from 98.9 to 1752.3 m (mean: 603 ± 388.1 m); Temp., varied from 7.5 to 39.5 °C (mean: 24.9 ± 7.4 °C); WS, varied from 5.48 to 19.54 km/h (mean: 10.81 ± 2.85 km/h); RH, varied from 14.6 to 98.0% (mean: 60.2 ± 16.53%) and VIS, varied from 0.4 to 4.8 km (mean: 2.7 ± 0.9 km). The maximum ambient temperature, MH and WS were obtained during summer; however, minimum MH and WS occurred during the winter and post-monsoon seasons. Higher RH was observed during monsoon and lower in the summer season. These parameters can change the atmospheric conditions, which resulted in variations of PM mass concentration levels in distinct seasons in the northern part of India. $PM_{2.5}$ was negatively correlated with MH (−0.42), WS (−0.45), Temp. (−0.52) and VIS (−0.75); however, coarse mode particles were highly correlated with RH (−0.56) and VIS (−0.28). Fig. 5 shows the relationship between $PM_{2.5}$ and PM_{10} concentrations and WS, respectively. The negative correlation (−0.45) between WS and $PM_{2.5}$ concentrations indicates the dominance of local sources. Strong winds 'flush' the pollution out of the system, and low winds allow pollution levels to rise within the system. Significant

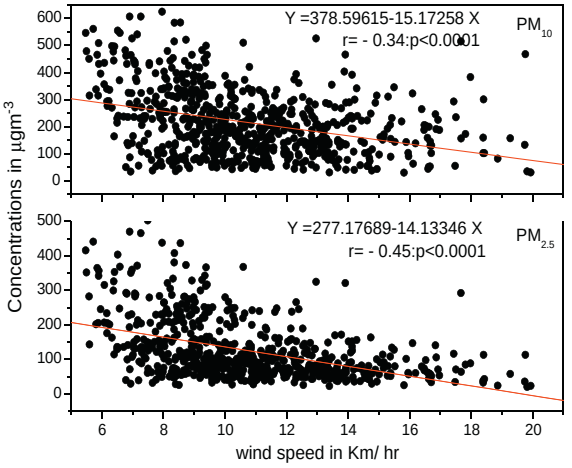

Fig. 5. Relationship between PM_{10} and $PM_{2.5}$ concentrations with wind speed over Delhi.

differences were observed in seasonal regression analysis between WS and $PM_{2.5}$ e.g., strong correlation during winter (−0.55) and post-monsoon (−0.32) when WS was lower. On the other hand, relatively less, but negative correlation, was observed during the monsoon (−0.26) for the considered period, when the WS was relatively higher. Results are found to be similar to those observed over a sub-urban and urban site in Canada (Sharma et al., 2002) and an urban site in China (Cao et al., 2009). In another study, Cheng and Lam (1998) have investigated the impact of WS on TSP concentrations in Hong Kong and found a similar relationship. $PM_{2.5}$ concentrations measured near a highly trafficked road in Paris were inversely proportional to WS (Ruellan and Cachier, 2001). The relationship between PM mass concentrations and temperature is also explored, which shows no significant correlation between $PM_{2.5}$ and Temp., but in the case of coarse mode, a good correlation was observed between $PM_{10-2.5}$ and temperature during summer (0.46) and monsoon (0.58). Stull (1988) has suggested that mixing height (MH) determines the volume through which surface-emitted pollutants can be diluted and reflected boundary layer turbulence. The surface boundary layer is shallow over Delhi during winter, which results in pollutant trapping near the surface. With an increase in surface temperature and convective activities during summer, pollutants are dispersed as the boundary layer deepens, thus lowering the PM concentrations. Negative relationship (−0.42) between MH and $PM_{2.5}$ was seen during the study period with a higher correlation in winter (−0.48) and poor in monsoon (0.06). Seasonal mean magnitude of MH and $PM_{2.5}$ was about 1109 m and 210.1 μg m^{-3} in summer; 628 m and 56.3 μg m^{-3} in monsoon; 348 m and 230.7 μg m^{-3} in post-monsoon and 327 m and 169.7 μg m^{-3} in winter. Pollutant dispersion is weaker during the lower MH, which is in agreement with the observations made in a coastal area of south China (Cheng et al., 2006). However, coarse mode particles are significantly and positively correlated (0.54) with MH (628 m) during monsoon, as compared to other seasons due to lower concentrations of coarse mode particle (35.9 μg m^{-3}) with bigger size ranges. 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). The relation between VIS and $PM_{2.5}$ was studied on seasonal basis that show higher correlation for post-monsoon (−0.85) and winter (−0.78), when the VIS was around 2 km; however, relatively less correlation was observed during summer and monsoon, when visibility was greater than 2 km. It is due to the rapid growth of anthropogenic emissions especially fossil fuel consumption during post-monsoon and winter. Tiwari et al. (2013) have found that during post-monsoon and winter, the majority of wind was calm and the region experiences a thick foggy weather during winter with low boundary layer heights. During such conditions, pollutants could not be dispersed and mix 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). Relationship between PM and RH was also calculated, and no significant correlation was observed with $PM_{2.5}$ and PM_{10}; however, a significant relationship (−0.56) was seen with coarse ($PM_{10-2.5}$) mode particles. Seasonal analysis indicated a positive correlation (0.32) during the winter period with fine mode particles, whereas negative correlation with coarse mode particle during monsoon (−0.70) and summer (−0.51) and very low negative correlation during post-monsoon. Most of the RF occurred in July to September which is called the southwest summer monsoon period. Data of RF and PM during the monsoon period were separated and correlation analysis was carried out. A poor negative relationship (−0.14) was found between them. It may be because most of the PM is scavenged out by RF and may also be due to longer dry periods with less RF over the station as compared to other parts of India. In another study, Ramachandran and Rajesh (2007) have done a correlation analysis between BC (highly absorbing aerosols) and RF over Ahmedabad, in western India and observed a significant negative correlation of about −0.35. Further, Babu and Moorthy (2002) have also reported a significant negative correlation between BC and RF at Trivandrum (−0.74) a coastal station in south India.

The influences of WD on atmospheric aerosols were also studied over Delhi. Seasonal wind rose along with mass concentrations of $PM_{2.5}$ and PM_{10} and was plotted and shown in Fig. 6a–d: during (a) monsoon, (b) post-monsoon, (c) winter and (d) summer seasons, respectively. We noticed that both $PM_{2.5}$ and PM_{10} concentrations showed substantial variations during the measurement period with WD. During the post-monsoon and winter periods, the occurrence of elevated concentration episodes were accompanied with a wind from the NNE–NE–ENE wind sectors. When the wind blows on this direction, the concentrations were found to be higher in both cases. The open burning of crop residues and biomass is a common practice in this region during post-monsoon, which results in an abundance of biomass aerosols and their transport towards the study region. In the overall study period, the higher concentrations of PM were observed during calm condition. In the case of PM_{10}, it was highest during winter (427.2 μg m^{-3}), followed by post-monsoon (399.7 μg m^{-3}), summer (241.0 μg m^{-3}) and monsoon (122.5 μg m^{-3}); however, in the case of $PM_{2.5}$, it was 287.5 μg m^{-3} (winter), followed by 260.5 μg m^{-3} (post-monsoon), 107.0 μg m^{-3} (summer) and 76.1 μg m^{-3} (monsoon). We have also

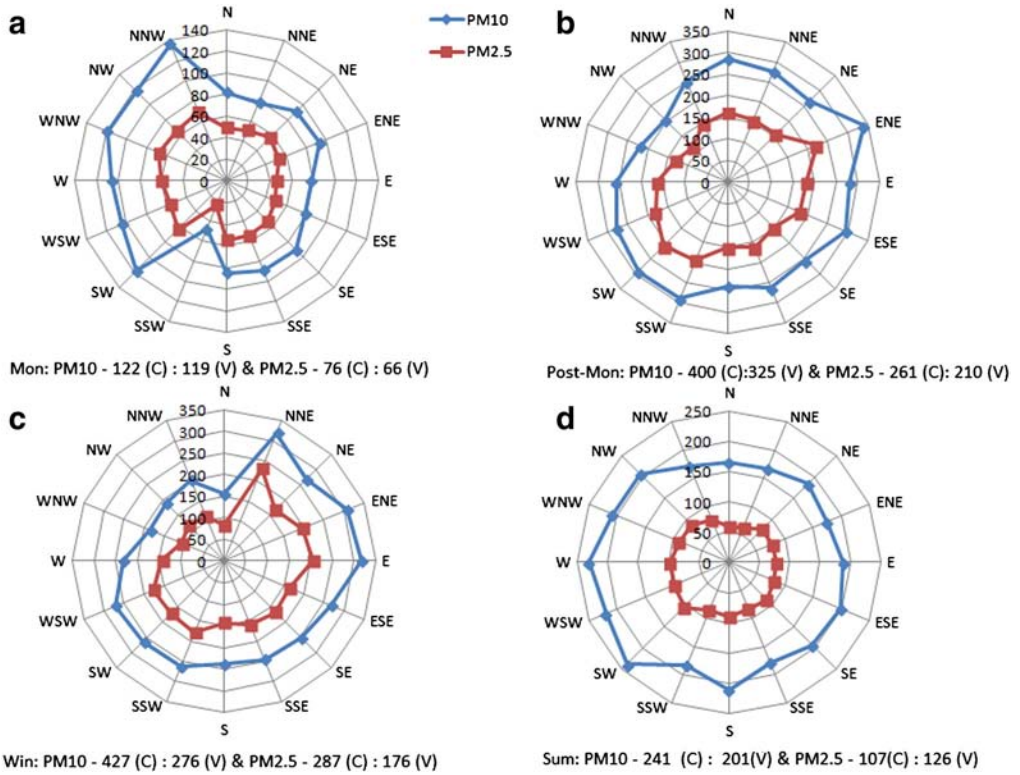

Fig. 6. Mean mass concentrations of $PM_{2.5}$ and PM_{10} with respect to wind directions during (a) monsoon, (b) post-monsoon, (c) winter and (d) summer seasons for calm condition (C) and variable condition (V) during the study period over Delhi.

analyzed these particulate matters during variable conditions, which were different during calm conditions. It was highest during post-monsoon (325.0 μg m^{-3}), followed by winter (276.2 μg m^{-3}), summer (201.0 μg m^{-3}) and monsoon (119.8 μg m^{-3}) for PM_{10}. A similar trend was also observed for $PM_{2.5}$ e.g., post-monsoon (209.0 μg m^{-3}), winter (176.1 μg m^{-3}), summer (125.8 μg m^{-3}) and monsoon (65.9 μg m^{-3}). This analysis suggest possible source regions for PM concentration over Delhi. There are large-scale industrial complexes with densely populated area, including the power plants and chemical industrial plants in the eastern sector over Delhi City. Thus, the easterly wind brought in a large amount of pollutants from the industrial source regions. However, this technique (wind rose) does not disassociate local from distant emission sources.

4.6. A case study during dust storm episode

During the pre-monsoon (summer) season (April to June), dust storm conditions were seen over the northern part of India with a thick layer of dust that enveloped the area and reduced the visibility levels (A.K. Srivastava et al., 2011; M.K. Srivastava et al., 2011). Such dust storm affects the day-to-day life and environment of the affected region. Dust storms generally called Andhi are generated by strong winds lifting particles of dust or sand into the air from regions that are mainly deserts, dry lakebeds and semi-arid. It is very dynamic, and as a result, the particle size and the concentrations of aerosols vary significantly (MacKinnon and Chavez, 1993; Singh et al., 2005). IGB that experiences dust storm, during the pre-monsoon period, have a major threat to agricultural resources and also produce large scattering of incoming solar radiation (Pandithurai et al., 2008; El-Askary et al., 2004). Concentrations of aerosols over the Indian region are found to be increasing, and studies report that the aerosol optical depth (AOD) over the northern part of India is higher as compared to the southern part (Singh et al., 2004; Sarkar et al., 2006; Gautam et al., 2007). The AOD in the northern part of India shows an annual variability with higher aerosol loading during the dry season due to episodes of dust events. During the study period, an influence of unusual dust storm was observed over the station on 20–21 March 2012 (Singh and Beegum, 2013). To understand the impact of dust storm on PM concentrations, we have analyzed eight day PM data over the station from 18 to 25 March 2012. Hourly mean mass concentrations of $PM_{2.5}$ and PM_{10} were plotted for this episode (Fig. 7a), which clearly indicated the impact of aerosols on regional climate over the northern part of India. The figure clearly shows that the concentrations of PM started increasing around 10 am on 20 March 2012 (PM_{10} > 1600 μg m^{-3}), which continued to exist till next day. Daily mean mass concentrations of $PM_{2.5}$ and PM_{10} were 207.9 and 310.2 μg m^{-3} on 19th March 2012 and a sudden increase was observed on 20 March 2012 which reaches up to 373.6 and 816.8 μg m^{-3}, respectively. Further, it was observed to decrease and reached to its normal condition on 25 March 2012 when $PM_{2.5}$ and PM_{10} were 114.9 μg m^{-3} and 257.9 μg m^{-3} respectively. There were no

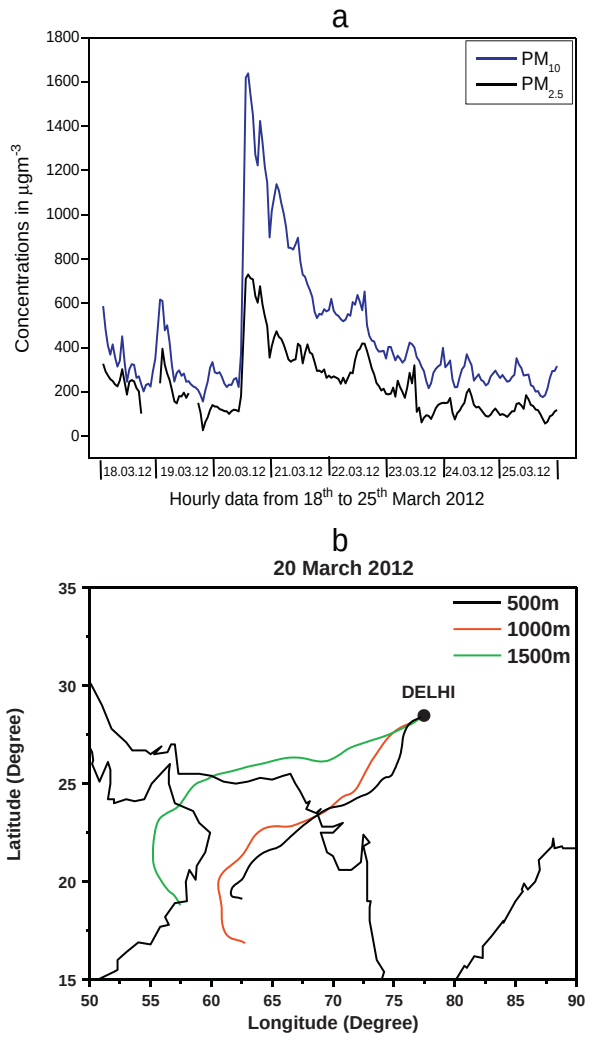

Fig. 7. a: Hourly interval mass concentrations of $PM_{2.5}$ and PM_{10} during a dust storm over Delhi. b: 5-day air mass back trajectory at different altitudes over Delhi on 20 March 2012.

large variations seen in the case of fine mode particle during the dust event as compared to coarse mode particles. Since the impact of ambient aerosols on radiation budget of the atmosphere changed with source regions and transport pathways, it is important to know the details of their sources. In order to know the transport pathways over the station (Delhi), 5-day air mass backward trajectories have been computed (Fig. 7b) for 20 March 2012, using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model of the National Oceanic and Atmospheric Administration (NOAA), USA (Draxler and Rolph, 2003). The back-trajectories were calculated at 7:00 GMT (i.e. 12:30 h local time) at three different altitudes at 500 m (representing within boundary layer), 1000 m (representing the boundary layer) and 1500 m (representing above the boundary layer). The transport of air masses is a path to bring aerosols from the major source regions. It is clearly evident from the figure that the back-trajectories over the station are coming from the Thar Desert region at all the altitude levels, which is one of the major dust sources in the northern part of India and one of the major sources of enhanced PM mass concentrations over the present station in Delhi.

5. Conclusions

Simultaneous measurements of mass concentrations of $PM_{2.5}$ and PM_{10} along with other co-existence pollutants viz., BC, CO, NO and NOx were studied with high resolution (five minute) datasets from 1st Sept. 2010 to 23rd Aug. 2012 at a mega city 'Delhi' in the northern part of India. The arithmetic mean mass concentrations of $PM_{2.5}$ (fine particle) and PM_{10} (inhalable particle) were 129.8 ± 103.4 and 222.0 ±142.0 μg m^{-3}, respectively during the entire study period. During the period, fine particles were found to be higher than the coarse mode particle during post-monsoon (~89%), winter (~69%) and monsoon (~64%); however, it was opposite (~22% lower) during summer. Relationships among PM and other pollutants indicated that the $PM_{2.5}$ was highly correlated with BC (0.74) and CO (0.51) especially during winter; however, $PM_{10-2.5}$ particles were less correlated with other gaseous pollutants. To see the impact of meteorological parameters on aerosols, regression analysis was performed among PM and MH, WS, Temp., RH, RF and VIS. A negative correlation (−0.45) between WS and $PM_{2.5}$ was found, indicating the predominance of local sources of fine particle. Significant differences were observed in seasonal regression analysis between WS and $PM_{2.5}$ e.g., strong correlation during winter (−0.55) and post-monsoon (−0.32) when WS was lower. On the other hand, relatively less, but a negative correlation, was observed during the monsoon (−0.26) for the considered period, when the WS was relatively higher. Relation between visibility and $PM_{2.5}$ was higher during post-monsoon (−0.85) and winter (−0.78) when the visibility was around 2 km; however, the correlation was relatively less when the visibility was greater than 2 km, mainly during summer and monsoon. The trajectory analysis indicated that the major source of PM, especially coarse mode particles ($PM_{10-2.5}$), is from the Thar Desert as well as from the Southwest Asian regions.

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