ADVANCES IN ATMOSPHERIC SCIENCES, VOL. 34, SEPTEMBER 2017, 1082–1094

• **Original Paper** •

Characterization of Black Carbon in the Ambient Air of Agra, India: Seasonal Variation and Meteorological Influence

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(Received 6 September 2016; revised 12 January 2017; accepted 1 April 2017)

ABSTRACT

This study characterizes the black carbon in Agra, India home to the Taj Mahal—and situated in the Indo-Gangetic basin. The mean black carbon concentration is 9.5 μg m^{-3} and, owing to excessive biomass/fossil fuel combustion and automobile emissions, the concentration varies considerably. Seasonally, the black carbon mass concentration is highest in winter, probably due to the increased fossil fuel consumption for heating and cooking, apart from a low boundary layer. The nocturnal peak rises prominently in winter, when the use of domestic heating is excessive. Meanwhile, the concentration is lowest during the monsoon season because of the turbulent atmospheric conditions and the process of washout by precipitation. The ratio of black carbon to brown carbon is less than unity during the entire study period, except in winter (December). This may be because that biomass combustion and diesel exhaust are major black carbon contributors in this region, while a higher ratio in winter may be due to the increased consumption of fossil fuel and wood for heating purposes. ANOVA reveals significant monthly variation in the concentration of black carbon; plus, it is negatively correlated with wind speed and temperature. A high black carbon mass concentration is observed at moderate (1–2 m s^{-1}) wind speed, as compared to calm or turbulent atmospheric conditions.

Key words: black carbon aerosol, seasonal variation, diurnal variation, meteorological parameter,

Citation: Gupta, P., S. P. Singh, A. Jangid, and R. Kumar, 2017: Characterization of black carbon in the ambient air of Agra, India: Seasonal variation and meteorological influence. *Adv. Atmos. Sci.*, **34**(9), 1082–1094, doi: 10.1007/s00376-017-6234-z.

1. Introduction

Black carbon is one of the most notable aerosol constituents. It causes visibility impairment, radiative forcing and climate change. The concentration of black carbon depends on the strength of the source as well as meteorological parameters, and it varies substantially from region to region and season to season. In general, aerosol particles can affect the earth's radiation balance, leading to a temperature change, i.e., a cooling or heating effect. Black carbon can directly or indirectly affect the atmospheric stability, influence the circulation pattern, and change the cloud albedo by acting as cloud condensation nuclei (Liousse et al., 1996). Aerosol black carbon, a byproduct of an incomplete combustion process, is easily transportable both horizontally and vertically over large regions across the globe (Cooke and Wilson, 1996; Babu and Moorthy, 2002), and hence has attracted worldwide attention from the scientific community.

Black carbon or soot particles (absorbing particles) play a key role in estimating the optical properties in the lower atmosphere from the regional to global scale (Haywood and Shine, 1997). Globally, approximately 8×10^9 kg of black carbon are emitted every year, contributing significantly to global warming after the greenhouse gases, which have a very long lifetime in comparison to aerosols (Bond et al., 2004). Black carbon emissions from India constitute a significant section of the pie in terms of the total global black carbon burden, with its emissions strength being largely due to land use, soil, forestry, transportation, industrial and agricultural practices (Parashar et al., 2005; Ram et al., 2010).The Indo-Gangetic basin is known to be one of the most dominant sources of absorbing aerosols i.e., black carbon (Latha et al., 2017). The Thar Desert of Rajasthan is the major contributor of dust aerosols, while forest areas abundant in the northeast and south of the region are major contributors of organic and black carbon aerosols (Ram and Sarin, 2015). The central region is excessively influenced by various gaseous and anthropogenic sources, such as agricultural waste, biomass burning and industrial emissions (Srivastava et al., 2012; Tiwari et al.,

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2013). The urban landscape of the region is majorly dominated by fossil fuel burning, crop burning/harvesting, automobile and industrial emissions, household cooking practices, etc. (Badarinath et al., 2006; Rehman et al., 2011; Vaishya et al., 2017). These sources are known to be highly absorbing and have strong seasonal and diurnal variability. Atmospheric measurements in the ambient air of rural households and the vehicular emissions from highways in the Indo-Gangetic basin under project "Surya" have revealed cooking with solid biomass fuels to be the major contributor of black carbon in this region (Rehman et al., 2011).

Several recent studies have reported that global climate changes are influenced by black carbon, thus attracting attention for further related research (Highwood and Kinnersley, 2006; IPCC, 2007). Ground-based measurements that reveal the long-term trend for black carbon aerosol over a given location are very rare, and most studies in the Indo-Gangetic basin region have focused on shorter periods, providing limited information in terms of seasonal variability (Ramachandran and Rajesh, 2007; Dumka et al., 2010). Nevertheless, studying black carbon is very important for estimating the emissions contribution in the Indian context, which, as mentioned above, is key in terms of the global burden. Specifically, measuring and studying black carbon at the semi-urban site of Agra, in the Indo-Gangetic basin, could provide significant insights and important progress. Agra is famous worldwide as a tourist attraction because it is home to the Taj Mahal. However, at the same time, it is a highly polluted city. Indeed, recently, the World Health Organization released a report on the 20 most polluted cities in the world, and Agra was among them. Biomass burnt for agricultural and domestic purposes in the Indo-Gangetic basin region has been reported to be the major source of black carbon emissions (Venkataraman et al., 2005). Meanwhile, the topography of the Indo-Gangetic basin region is unique and surrounded by a variety of aerosol emitting sources (Kumar et al., 2007; Srivastava et al., 2011; Kumar and Kumari, 2015), thus making it a hotspot for aerosol research.

This paper describes the measurement and characterization of black carbon and the influence of meteorology over the Agra region. The hope is that this study will play an important role in developing our understanding of the radiative budget of the atmosphere over the Indo-Gangetic basin.

2. Materials and methods

2.1. *Location of study area: site description and meteorology*

Agra (27°10′N, 78°02′E) is located in the central part of northern India in the Indo-Gangetic basin (Fig. 1). It is one of the most famous tourist destinations in the country, and is particularly well known for being home to one of the Seven Wonders of the World—the Taj Mahal. The Indo-Gangetic basin encompasses one fifth of India's land and is densely populated (Nair et al., 2007). The growth of the economy and population in the Indo-Gangetic basin region has resulted in a wide range of anthropogenic activities (Kumar et al., 2003). Vehicular emissions are a major source of black carbon emissions affecting the urban population in Agra. Vehicular pollutants are released at ground level and hence have a considerable impact on the local population. Besides, Mathura refinery is situated on the west side of the city, while Firozabad glass industries are in the east. Agra is spread over an area of 120.57 km^2 and the population, as of 2011, is 1 575 704. The year is typically divided into four seasons: summer (March–June); monsoon (July–September); post-monsoon (October–November); and winter (December–February). The maximum temperature in summer lies between 21.9°C and 40°C, whereas in winter it is between 4.2°C and 31.7°C. The region has a continental sub-tropical climate, featuring along and hot summer from April to September.

2.2. *Sampling and analysis*

2.2.1. *Black carbon mass concentration measurement*

Black carbon concentration measurements are carried out from May 2014 to April 2015 at Dayalbagh, Agra, in the Indo-Gangetic basin, using the new seven-channels aethalometer (Model AE-33) produced by Magee Scientific, USA. The aerosol particles are collected on the filter continuously by drawing air containing aerosol through the filter tape. Ambient air is set to a 2 L min^{-1} flow rate through an inlet tube and the black carbon concentration is estimated every minute following an optical attenuation technique. The instrument uses continuous filtration through a quartz filter and estimates optical absorption based on the principle of transmission (Hansen et al., 1984). The aethalometer measures the concentration of optically absorbing suspended particles at seven different wavelengths (370, 470, 520, 590, 660, 880 and 970 nm). The measurement of the absorption of aerosol at 880 nm is interpreted as the true measurement of black carbon, as widely adopted (Pant et al., 2006; Satheesh et al., 2008); whereas, measurement at 370 nm gives information about ultraviolet-absorbing particulate matter (UVPM, i.e., brown carbon). The daily average is calculated during the entire study period.

In the aethalometer, the loading effect is described by the linear function of attenuation. The variation of the loading effect varies (small or zero) from place to place. Under an atmospheric condition that may be highly chemically active, fresher aerosols show a larger loading effect than aged aerosols. Therefore, the linear function of attenuation can be calculated as

$$B_{c,r} = B_0(1 - kA) ,$$

where $B_{c,r}$ represent "reported black carbon", B_0 represent "zero loading black carbon", k is the "loading compensation parameter" and A represent "attenuation" (Virkkula et al., 2007). The loading effect of aerosol is measured using a dual spot technique. These dual spots of filter tape collect the aerosol into two streams that have unequal flow. Therefore, these measurements of attenuation are influenced by the loading effect (Drinovec et al., 2015). To eliminate any loading effect in the overall measurement, extrapolation of the same

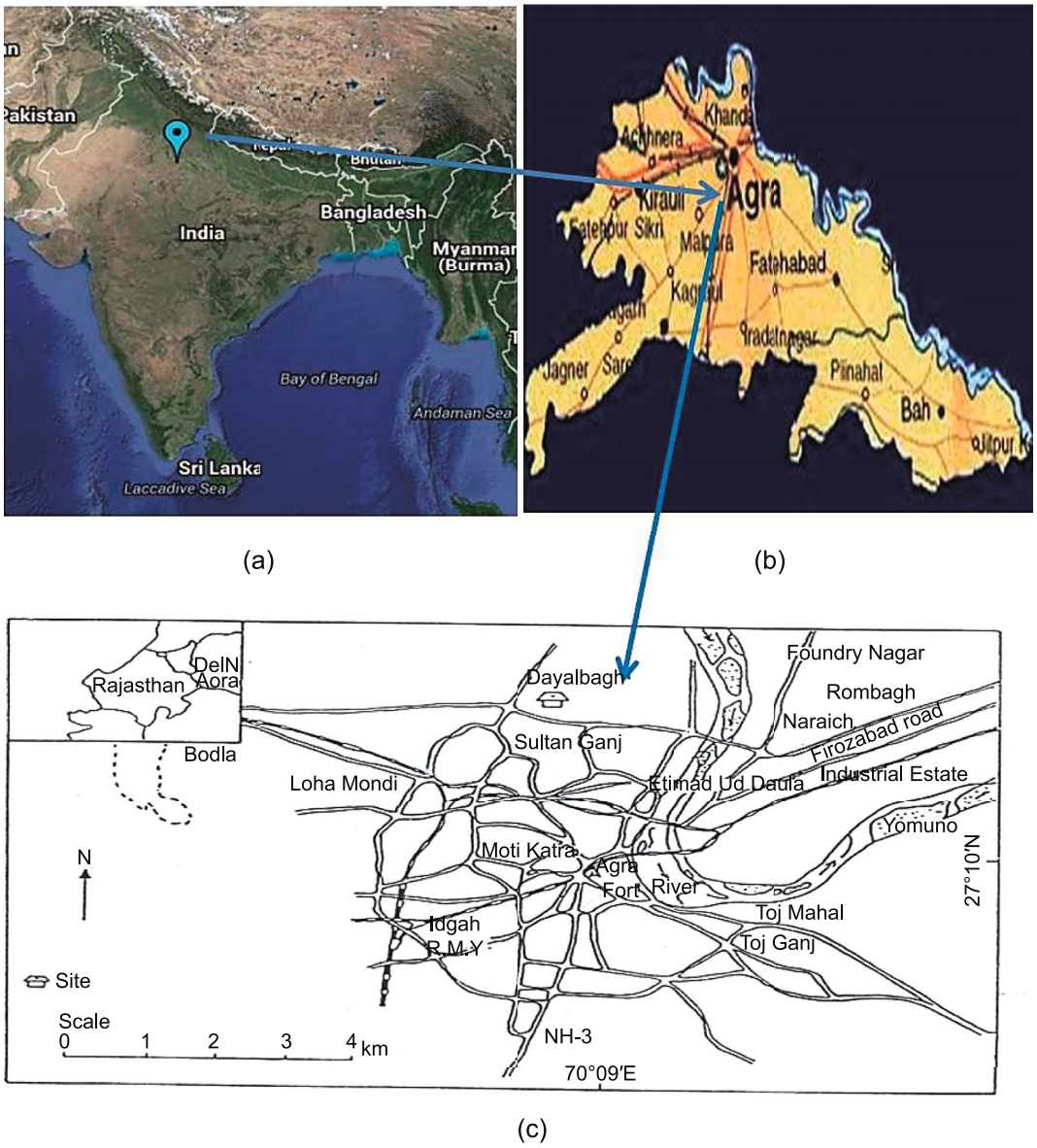

Fig. 1. (a) Map of India showing Agra; (b) map of Agra; and (c) the study site.

to zero loading is carried out. Drinovec et al. (2015) incorporated this measurement technique into this next-generation aethalometer. Dual spot technology is highly advantageous in that it eliminates data artifacts due to filter loading. The aethalometer is statistically and technically a very advanced instrument, used for real-time measurement and analysis of the optical absorption of black carbon. This newly developed advanced version of this aethalometer, i.e., AE-33, inherits all the qualities of previous versions, i.e., AE-10 and AE-30, following the same basic principles. It is a self-sufficient and automatic instrument, reproducing the data spontaneously. It collects samples on a quartz fiber filter and performs a continuous optical analysis. The tape does not move forwards during this process. It moves only when the spot has reached a certain density. The re-initialization process takes a certain amount of time, during which the output of valid data is suspended. Each spot advance requires one minute, and the tape drive motors move extremely slowly in order to not tear or stretch the tape. In addition to this, the algorithm allows for a period of stabilization before valid data are generated for analysis of a fresh spot. A tape advance results in the loss of data; hence, the time base is reduced to one minute in AE-33 for urban locations, where tape advances may be more frequent, in order to not lose any more data than necessary. The sampling of aerosol particles goes through continuous optical attenuation, being analyzed at a high temporal resolution of every minute. Therefore, the determination of black carbon at different wavelengths (370 nm and 880 nm) is important. This can also be effectively used as an indicator of excessive local biomass combustion (Wang et al., 2011; Srivastava et al., 2012). The method used for the measurement of attenuation is whether a light beam is allowed to transmit through the sample collected over the fibrous filter. This quantity is linearly proportional to the amount of black car-

bon deposited on the filter indicating 880 nm of black carbon measurement, whereas UVPM at 370 nm indicates the result of brown carbon. The difference between these quantities can have a range of values from zero (if the aerosol content is mostly from biomass burning) to a larger value (if the content of aerosol is from fossil fuel combustion). Sometimes, a negative value may appear, which is due to the subtraction of two large and slightly noisy signals negating each other. This advanced seven-channel aethalometer is programmed in such a way that it needs no correction and can be taken directly into consideration (Drinovec et al., 2015; Vaishya et al., 2017).

2.2.2. *Measurements of meteorological parameters*

Meteorological parameters (temperature, relative humidity, wind speed and wind direction) are monitored using a WDL 1002 Data Logger system, Dyna Lab, Pune, India. A summary of the synoptic meteorology (e.g., wind pattern, air temperature and relative humidity) over Agra is shown in Table 1. The dominant wind direction over the Agra region is northwesterly and westerly, showing its influence on the concentration of black carbon at this site (Fig. 2). The weather in Agra during the summer season is very hot, with a maximum average temperature reaching around 39.4°C. The minimum wind speed tends to occur in winter, and varies between 0.45 and 0.9 m s^{-1}; whereas, the maximum wind speed is usually in summer and varies within 3–8 m s^{-1}. Wind speeds of less than 1.0 m s^{-1} are considered as calm conditions. High relative humidity is observed during the monsoon and winter seasons; however, dry conditions (relative humidity less than 65%) (Saha and Despiau, 2009) prevail during the sampling period.

2.2.3. *Air-mass back trajectories*

Air-mass back trajectories are calculated using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Ralph, 2003). The air-mass back trajectories at 500 m above ground level (AGL) are investigated to explore the role of black carbon long-range transport. The study site is also influenced by local emissions sources at 500

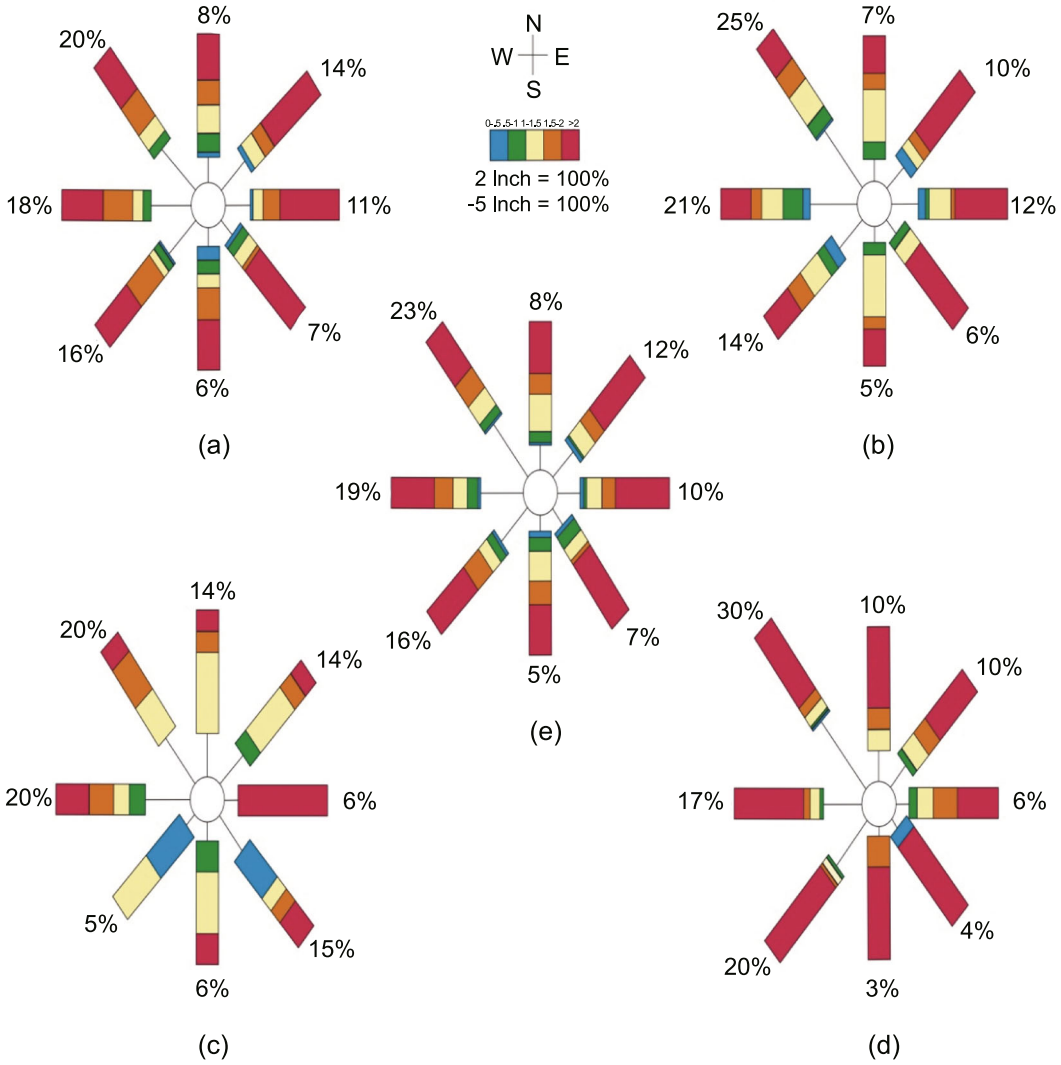

Fig. 2. Wind roses during the study period: (a) summer; (b) monsoon season; (c) post-monsoon season; (d) winter; and (e) whole year.

Table 1. Meteorological conditions during the study period.

	Temperature (°C)				Wind speed (m s^{-1})				Relative humidity (%)			
	Mean	SD	Min	Max	Mean	SD	Min	Max	Mean	SD	Min	Max
Whole year	21.7	3.8	4.4	39.4	1.5	1.7	0.4	6.2	53.6	16.3	8	100
Summer	28.4	3.0	20.6	39.4	2.1	0.9	0.4	5.4	30.7	13.5	8	65
Monsoon	14.6	5.2	21.6	24.4	1.8	0.9	0.4	6.2	72.4	18.9	29	100
Post-monsoon	24.1	3.5	17.8	31.0	1.2	4.3	0.9	8.3	43.5	14.8	19	74
Winter	19.9	3.9	4.4	29.4	1.0	0.9	0.4	2.3	67.8	18.3	27	98

Notes: SD, standard deviation; Min, minimum; Max, maximum.

m AGL, known as shorter trajectories, and these tell us about the origin of sources at the site. The air-mass back trajectories on a daily basis for the whole of each season are used to evaluate the role of the long-range transport of black carbon over the study region.

2.2.4. *Statistical analysis*

All the datasets are subjected to statistical analysis (correlation analysis, regression analysis, ANOVA) to explore the contributions of different sources and to examine their significance in the variations of black carbon. ANOVA is a technique that can facilitate the investigation of any number of factors, and gives information on whether differences in mean values are significant. The calculated value of F is compared with tabular values of the F distribution (critical values) and, if the calculated F value is higher than the critical value, the mean values are not equal. A two-way ANOVA test is applied to the entire dataset to identify whether the seasonal and monthly variation in the concentration of black carbon is significant.

3. Results and discussion

3.1. *Annual concentration of black carbon*

Table 2, which presents the statistics on the daily average concentration of black carbon over Agra, shows that the annual daily average concentration of black carbon is 9.5 $\mu g\ m^{-3}$. The black carbon mass concentration is high, but its variation is large. The wide range of average black carbon concentration values in Agra may be due to the variation in daily aerosol, which is determined by biomass burning, crop harvesting (Kanokkanjana et al., 2011) and anthropogenic activities (automobile exhaust, emissions from the glass industry and Mathura refinery), both locally and regionally, i.e., over the broader Indo-Gangetic basin (Badarinath et al., 2006; Sharma et al., 2002; Rehman et al., 2011; Latha et al., 2017). The mean black carbon mass concentration measured at Agra is similar to the metropolitan city of Delhi, attributed to the high accumulation of black carbon aerosol from local, regional and even distant locations (Tiwari et al., 2013). Similar variation has also been observed in many other studies carried out in India, dominated by the local boundary conditions (Latha and Badarinath, 2005).

Table 2 compares the black carbon concentration measured at the present site with those reported at other sites in the Indo-Gangetic basin. The scope of the comparison may not be very wide, but it nonetheless depicts the nature of the variation in the black carbon mass concentration in different environments across this region. The concentration of black carbon over different parts of India shows differences, especially in the months of October–December, which may be due to increased biomass burning from agricultural practices, the use of fossil fuels for heating and cooking, transportation, and industrial emissions over the Indo-Gangetic basin region (Venkataraman et al., 2005; Rehman et al., 2011; Srivastava et al., 2012; Vadrevu et al., 2012). It can be seen from Table 2 that the black carbon concentration is higher at Agra than at several other cities in the Indo-Gangetic basin, such as Dibrugarh (Pathak et al., 2010), Chandigarh (Chowdhury et al., 2007), Kolkata (Chatterjee et al., 2012), Ranchi (Kumar et al., 2011), and Nainital (Dumka et al., 2010). This

Table 2. Comparison of black carbon at different locations over the Indo-Gangetic basin.

Location	Region	Altitude (m)	Average black carbon concentration ($\mu g\ m^{-3}$)	Period	Reference
Agra	Northern India	169	9.5	May 2014 to April 2015	This study
Nainital	Northwestern Himalaya	2084	1.0	November 2004 to December 2007	Dumka et al. (2010)
New Delhi	Northern India	214	12.1	December 2011 to March 2012	Tiwari et al. (2015)
Agartala, Tripura	Northeastern India	13	9.35	September 2010 to September 2012	Guha et al. (2015)
Dibrugarh	Northeastern India	108	3.4	June 2008 to May 2009	Pathak et al. (2010)
Varanasi	Northern India	80.7	12.7	March–December 2013	Murari et al. (2016)
Chandigarh	Northern India	350	3.7	March 2001 to January 2002	Chowdhury et al. (2007)
Ranchi	Northeastern India	652.27	1–1.8	July 2010 to March2011	Kumar et al. (2011)
Kolkata	Eastern India	9	5.8	January–December 2006	Chatterjee et al. (2012)

may be due to relatively high source strength at the present site. In Agra, the use of biomass burning and cow dung cake is very common in practice. In addition, Agra is a famous tourist destination, where the floating population is approximately 35 000 per day, largely due to the attraction of the Taj Mahal. Nevertheless, the black carbon concentration at Agra is lower than that at New Delhi (Tiwari et al., 2015) and Varanasi (Murari et al., 2016), possibly because—in the case of Delhi—it being the most polluted city and one that is badly affected by biomass burning over Punjab and Haryana. Agra is downwind from Delhi. So, the source strength at Delhi is also much higher than at Agra. With respect to Varanasi, the lower black carbon concentration at Agra may be due to the high source strength at Varanasi. Ritualistic practices such as burning of wood, grains and ghee to mark births, marriages, and funeral enhance the concentration of black carbon (Chakrabarty et al., 2014).

3.2. *Seasonal variation of the black carbon concentration*

As shown in Fig. 3, high black carbon concentration is observed in winter (15.4 ± 5.9 μg m^{-3}), and the minimum (5 ± 2.2 μg m^{-3}) in the monsoon season. The highest concentration being in winter and the post-monsoon season may be due to the increased consumption of biomass and fossil fuels for heating and cooking purposes, as well as the decrease in the thickness of the boundary layer; whereas, the lowest black carbon concentration being in the monsoon season may be due to the low source strength and strong winds, as well as the process of washout by precipitation (Tiwari et al., 2013). The long-range transportation of black carbon may also contribute during the post-monsoon and winter seasons, as the dominant wind is from the northwest, to which Agra is downstream. The black carbon concentration in winter and the post-monsoon season is about 62% and 35% higher than the annual concentration, respectively, while in the summer and monsoon seasons the mean black carbon concentration is about 27% and 47% lower than the annual mean, respectively. The black carbon concentration in winter is approximately three times higher than during the monsoon season. The monthly mean variation of black carbon, shown in Fig. 4, indicates that the trend of the black carbon concentration features an increase from September to December (the peak of the annual cycle). Agra is the second largest tourist destination in the world, and so the floating population of about 35 000 per day due largely to the attraction of the Taj Mahal—may also be a contributing factor over this region. The geographical situation of Agra is such that wind coming from European countries and the U.S. also contributes towards its

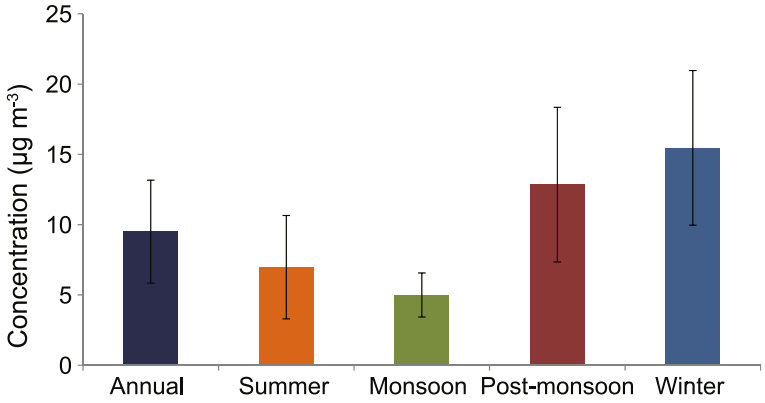

Fig. 3. Seasonal variation in mean concentration of black carbon along with standard deviation.

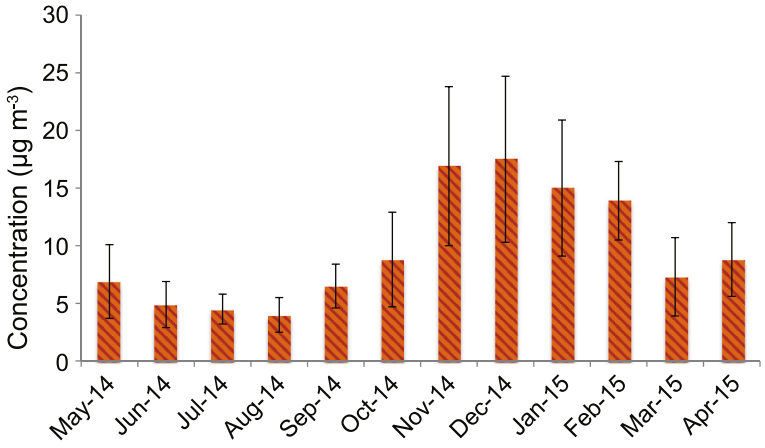

Fig. 4. Monthly variation in mean concentration of black carbon along with standard deviation.

pollution (Kumar et al., 2007, 2015). In the month of December, usually, the wind is characterized by calm conditions, with a low mixing height, while the consumption of fossil fuels/biomass burning are at their peak because of domestic heating and cooking. Two-way ANOVA is employed to examine the significance in the variation of the level of black carbon in different seasons and months, and significant variation in mean concentrations of black carbon is found among months.

3.3. *Diurnal variability of the black carbon concentration*

As shown in Fig. 5, the concentration of black carbon is around twice as high at night than during the day. The nighttime peak shows higher values from October (8.8 μg m^{-3}) to February (13.9 μg m^{-3}), while much lower concentrations are seen in July–September, and are relatively high value in April (8.1 μg m^{-3}). The diurnal variation in black carbon shows higher concentrations at nighttime than during daytime, in spite of the fact that the density of vehicles is higher during the day, and biomass burning for cooking purposes occurs during daytime as well as nighttime. This is because, during daytime, relatively unstable atmospheric conditions and solar insolation lead to dispersion; whereas, at nighttime, calm conditions and a low boundary layer lead to the trapping of pollution near the surface. Moreover, during daytime, convective mixing tends to be homogenous, leading to dispersion and dilution due to solar insolation within the atmospheric boundary level. This results in the distribution of surface black carbon in a greater atmospheric volume and, hence, reduction in the nearby surface concentration (Nair et al., 2007; Aruna et al., 2013). On the other hand, at night, mixing is generally inhibited and, after sunset, as the land cools faster, the black carbon concentration increases near the boundary layer. The black carbon concentration during nighttime may be attributable to the increase in bio-fuel/biomass burning, especially for cooking but also for heating to provide warmth against the cold weather (Safai et al., 2007).

As we can see from Fig. 6, there is a gradual buildup of

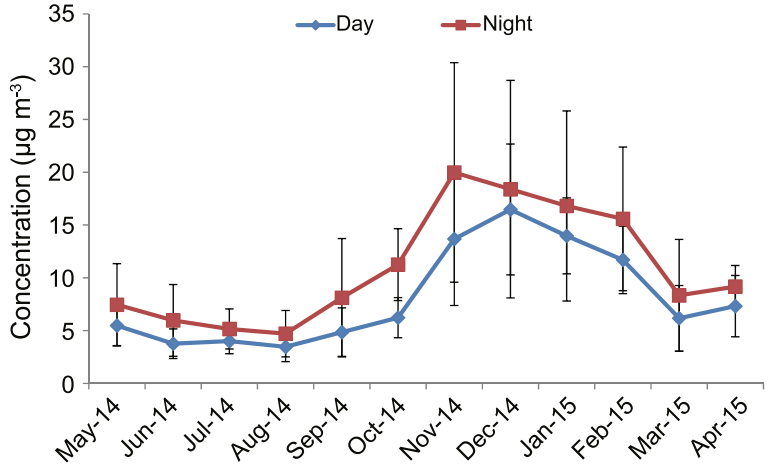

Fig. 5. Monthly day–night variation in the mean concentration of black carbon along with standard deviation.

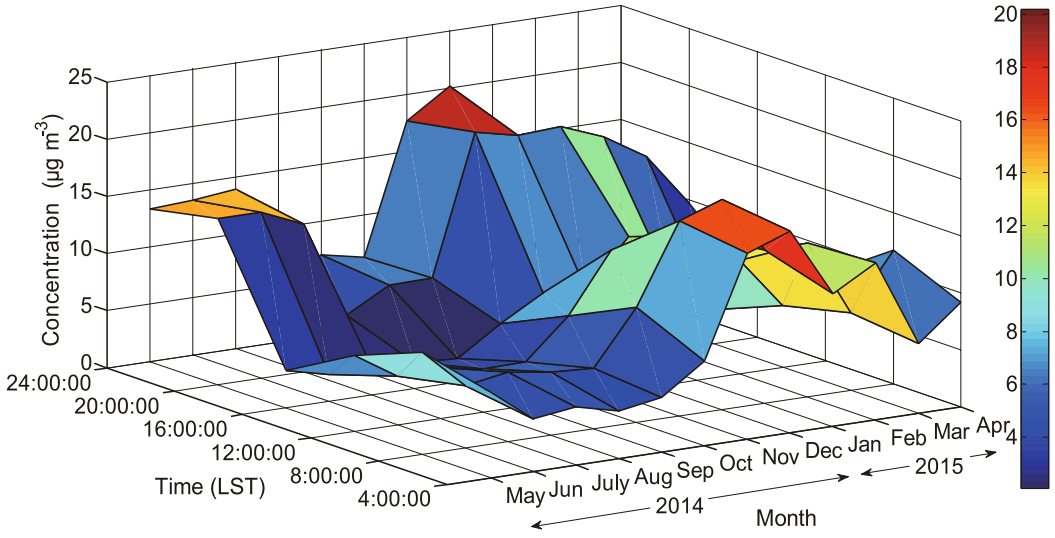

Fig. 6. Monthly diurnal variation in the concentration of black carbon.

the black carbon mass concentration in the morning hours, and there is a peak between 0700 and 1000 LST (Local Standard Time, LST=UTC+5:30) at about an hour after sunrise. The peak in the evening between 1700 and 2100 LST, is attributable to the combined effect of the boundary layer and the volume of traffic. As the day progresses, the atmospheric boundary layer rises with increased solar heating; plus, turbulence increases, which comprehensively mixes and redistributes aerosols at higher altitude. This then causes faster dispersion and, hence, dilution of the aerosol concentration from afternoon to evening (1200–1600 LST). The observed low concentration of black carbon in the afternoon may be influenced by the diurnal variation of wind speed, which is calculated to be several notches higher in the afternoon than in the morning and nighttime hours (Srivastava et al., 2012). Babu and Moorthy (2002) also stated that the increased solar heating as the day advances produces a deeper and more turbulent boundary layer, which leads to faster dispersion and dilution of black carbon near the surface. The daily anthropogenic activities that take place during the morning hours may also be a reason for the morning peak in diurnal variation. As the observational site is neither highly industrialized nor highly populated, being a distance from the main city, the anthropogenic causes may be confined to the morning hours. The national highway is about 2 km away, but the traffic density during office hours (from 0900 LST) is not significantly higher compared to other big cities in India. Local traffic activity reaches a peak between 1700 and 1900 LST, after office hours, complemented by the burning of fuels (fossil fuels and bio-fuels); heating, cooking and other household activities in the adjacent residential areas in the evening also add to the rise in the black carbon concentration. The surface aerosol loading in urban areas typically decreases from late at night to early in the morning (0300–0600 LST) and increases after sunrise (0700–1000 LST). It then drops in the afternoon (1500 LST) until the occurrence of the second peak at 1600–2000 LST (Jin et al., 2005).The trend across the seasons is the same, but they do differ in values, suggesting a relatively large meteorological effect during the night as compared to the day. In winter, the excessive amount of biomass burning by local communities and villagers contribute to the black carbon concentration at night. Of note is that the diurnal variation of the black carbon concentration over Agra is much higher than that over other urban locations. Moreover, it has been observed that the boundary layer is shallower at night than during the day (Kunhikrishnan et al., 1993; Tiwari et al., 2015). In general, wind speeds are lower at night, resulting in the confinement of aerosols during this part of the day. These factors result in an increase in the black carbon mass concentration during the early part of the night (Babu and Moorthy, 2002).

3.4. *Identification of the sources of black carbon at the UVPM and IR wavelengths*

Figure 7 presents the monthly variation in the levels of black carbon at the infrared and ultraviolet wavelengths, respectively referred to as BC_{880} and $UVPM_{370}$, and the ratio of BC_{880} to $UVPM_{370}$. The $UVPM_{370}$ level is higher than that of BC_{880} throughout the year, except in November and December. The high $UVPM_{370}$ level is probably due to the dominance of crop burning and vehicular emissions over this region. The monthly variation shows that $UVPM_{370}$ is lowest in the month of August and highest in the month of November. The low concentration in August (in the monsoon season) may be due to the washing out of pollutants by precipitation, while the high concentration in November (in the post-monsoon season) may be due to long-range transportation from Punjab and Haryana, where crop (paddy) burning takes place in this month (Badarinath et al., 2006; Srivastava et al., 2012). The sharp rise in $UVPM_{370}$ from August to November supports this suggestion, as also reported by Tiwari et al. (2013). The increasing trend in the concentration of $UVPM_{370}$ from March to May (in summer) may be due to the crop harvesting that takes place at this time of year. The northwesterly wind might also be a contributing factor to this increasing trend, as in Punjab and Haryana crop har-

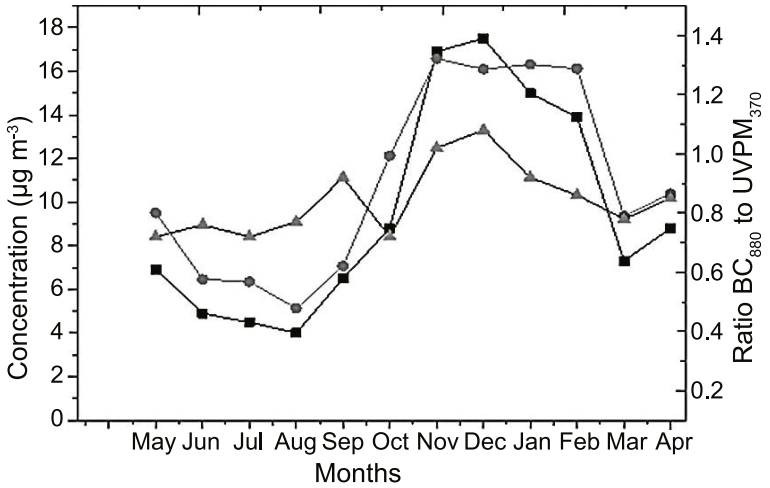

Fig. 7. Levels of black carbon (line with squares), UVPM (line with circles), and the ratio of BC_{880} to $UVPM_{370}$ (line with triangles).

vesting and burning takes place in this season, to which Agra is downstream. The level of BC_{880} is lowest in August and highest in December. The high concentration in December may be due to the increase in the consumption of fossil fuels for heating and cooking purposes (Safai et al., 2007). It may also be due to the fact that, in the months of November and December, the temperature falls and the consumption of fossil fuels enhances, leading to an increase in BC_{880}. Several studies, using various methods, have attempted to identify sources of black carbon (Kirchstetter et al., 2004; Herich et al., 2011; Tiwari et al., 2013; Crilley et al., 2015; Li et al., 2016). Here, source identification is performed on the basis of the ratio of the concentration of black carbon at 880 nm to that at 370 nm, as well as on the basis of the absorption coefficient.

The ratio of $UVPM_{370}$ to BC_{880} is calculated (Fig. 7) to reveal the influences of biomass and fossil fuel emissions. Black carbon that absorbs radiation and has a warming influence on the atmosphere, depending on the brightness of the underlying surface, is considered as organic carbon, sometimes known as brown carbon. So, the ratio of black carbon to brown carbon can be used as an emissions indicator for different types of black carbon sources (Srivastava et al., 2012). Organic aerosol material, such as aromatic organic species of wood smoke or biomass burning, enhance UV absorption at 370 nm, relative to 880 nm (Kumar et al., 2011; Wang et al., 2011). The mean ratio of BC_{880} to $UVPM_{370}$ is less than 1.0 for the entire study period, except in November and December. The highest ratio (> 1.0) of BC_{880} to $UVPM_{370}$ is observed in winter, indicating the dominance of the contribution from fossil fuel and coal consumption for heating and cooking purposes. In summer and in the post-monsoon season, the ratio is less than 1.0, indicating the dominance of crop burning and vehicular emissions (INCCA, 2011). Similar ratio trends have been reported from Switzerland, New Delhi, London and the Himalayan Tibetan Plateau (Herich et al., 2011; Tiwari et al., 2013; Crilley et al., 2015; Li et al., 2016). Biomass emissions are identified as a major cause of $UVPM_{370}$. There are consistent trends of biomass combustion and fossil fuel burning over the study region. Li et al. (2016) also reported similar trends in the southern Langtag valley, which is influenced by the urban effects of Kathmandu over the Indo-Gangetic basin. Kirchstetter et al. (2004) stated that an absorption coefficient of black carbon that is close to 1 represents vehicular emissions, while more that 2 or 2.5 represents emissions form biomass burning at a lower wavelength. The mean value of the absorption coefficient at Agra is 1.5, over the entire study period. Hence, vehicular and biomass emissions are the dominant source over this region, throughout the year, except in November and December, when the absorption coefficient is more than 2.5, attributable to fossil fuel combustion.

3.5. *Effect of meteorological parameters and long-range transport on the black carbon concentration*

Several meteorological parameters are responsible for high variability in the concentration of black carbon. Analysis of the relationship between the black carbon mass concentration and meteorological parameters (wind speed, temperature and relative humidity) (Fig. 8) reveals results that are similar to many other studies reported in Delhi (Srivastava et al., 2012; Tiwari et al., 2013) and Ahmadabad (Ramachandran and Rajesh, 2007). Table 3 shows that, irrespective of wind direction, the concentration of black carbon is highest when the wind speed is moderate (1–2 m s^{-1}). Northwesterly, westerly, southwesterly and northeasterly winds are dominant in all seasons, but the speed varies (Fig. 2). The highest wind speed (> 2 m s^{-1}) causes more dispersion and dilution, which in turn leads to low concentrations of black carbon. The relationship between black carbon and wind speed is not always straight forward. The black carbon concentration is high when atmospheric conditions are calm and the wind direction is southeasterly, southwesterly, westerly or northwesterly. High black carbon concentrations are also observed when the wind is from the north, northeast or east. This may be due to the fact that Agra is downwind from the glass industry at Firozabad, which is northeast of Agra. This industry is based on coal combustion and, hence, may contribute towards high levels of black carbon when the wind is from the northeastern side. In the post-monsoon and winter seasons, however, northwesterly, westerly and southwesterly winds are dominant, but the wind speed is moderate or calm, resulting in higher concentrations of pollutants. Wind speed during the monsoon season is lower in comparison to summer. The concentration of black carbon must be higher, but due to precipitation it is washed out in the monsoon. The major contributor to the black carbon concentration in Agra is local anthropogenic sources, which accumulate under low wind speed in the post-monsoon and winter seasons. Similar results have also been found for suburban and an urban site in Canada (Sharma et al., 2002), as well as at an urban site in China (Cao et al., 2009). The daily mean black carbon mass concentration shows an inverse relationship with temperature. A correlation of $r = 0.3$ and above is considered as significant (Kothari, 2004). Black carbon is also found to be inversely related to wind speed during the measurement periods, which is relatively more pronounced during the winter and monsoon seasons.

Table 3. The concentration of black carbon with respect to wind speed (WS) and wind direction.

	Black carbon concentration (μg m^{-3})		
Wind direction	WS < 1 m s^{-1}	WS =1–2 m s^{-1}	WS > 2 m s^{-1}
Northerly	4	7	4.3
Northeasterly	3.3	5.2	3.4
Easterly	3.1	5.8	3.7
Southeasterly	5.6	8.8	4.2
Southerly	4.9	6.3	2.2
Southwesterly	5.7	7.5	2.5
Westerly	6.8	8.5	3.4
Northwesterly	4.5	9	1

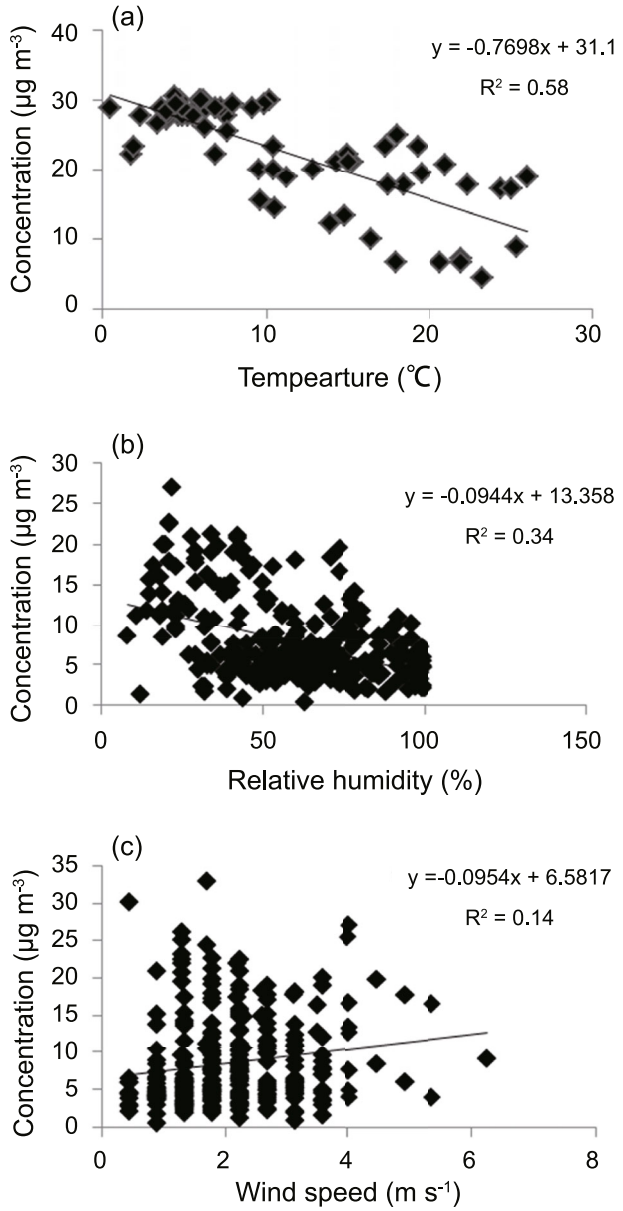

Fig. 8. Relationship of black carbon with temperature (a), relative humidity (b), and wind speed (c).

In terms of the long-range atmospheric transportation of air pollutants, black carbon aerosol, being relatively small in size (< 1 μm), can be transported for long distances at high altitudes by the prevailing winds (Wolff, 1981). To explore the role of long-range transport in the level of black carbon, air-mass back trajectories at an altitude of 500 m AGL are computed on a seasonal daily basis over the Agra region (Fig. 9) using the HYSPLIT model. The transportation of black carbon on the global as well as regional scale potentially affects visibility over a wide range of regions. Studies have shown that aerosol concentrations are closely related to wind speed and direction, and trajectory analysis can help in identifying the sources and paths taken aerosols in reaching a particular site, as well as improve our understanding of their properties and types (Bian et al., 2011). The trajectory analysis is run using seasonal data, for which the main atmospheric circulation paths influencing the black carbon concentration are identified. Figure 9 illustrates that, in each season, the sources or source strength are different. It is evident that the pathways of transported black carbon from different sources vary with season. In summer, there is a large error bar, which indicates the transportation of black carbon from various directions and, possibly, from various sources. It also hints at a gusty nature of the wind and an unstable atmosphere during summer. Winds from the northwest, west and southwest are dominant during summer. Dust from Thar Desert, Rajasthan, might also be a contributing factor during summer. Wind from the southwest prevails during the monsoon season, but pollutants get settled under the influence of precipitation. Recently, several studies have reported similar scenarios over the Indo-Gangetic basin (Srivastava et al., 2011, 2012; Tiwari et al., 2013). Westerly and northwesterly winds dominate during the post-monsoon and winter seasons over this region. Crop burning in Punjab and Haryana takes place in the post-monsoon period, having a substantial effect. Indeed, this may be the reason for the high concentration of black carbon, as well as UVPM, in the months of November and December over this region. The calm and stable atmospheric conditions also contribute to an increasing concentration of black carbon during the winter season. These conditions retain the high loading of black carbon in the ambient air for a longer period. Crop burning over the Indo-Gangetic basin is reported to be higher in the post-monsoon and winter seasons (Kanokkanjana et al., 2011; Paliwal et al., 2016). The trajectory analysis also reveals that this region is affected by winds blowing from European countries. Agra supports the transportation of dust over long ranges in the North Indian region. During summer, air masses appear to be transported from the central northwest and southwest at low altitudes, and then enter Agra (northern India) and the Indo-Gangetic basin (Tiwari and Singh, 2013). Meanwhile, during the monsoon season, they enter India from the Arabian Sea, covering the whole of the Deccan peninsula and central India before entering into Agra.

4. Conclusion

In this study, the black carbon concentration is measured over the Agra region for a period of one year (2014–15). The observational findings highlight the high loading of black carbon in Agra. The annual mean mass concentration of black carbon is 9.5 μg m^{-3} over Agra, and exhibits seasonal variation with a maximum concentration during winter, followed by the post-monsoon, summer and monsoon seasons. ANOVA reveals significant monthly variation in the concentration of black carbon. The black carbon concentration is higher at night compared to daytime, because of the burning of biomass and the calm atmospheric conditions. The diurnal variation of the black carbon concentration is attributable to local sources and changes in the local boundary layer, whereas monthly black carbon variations are

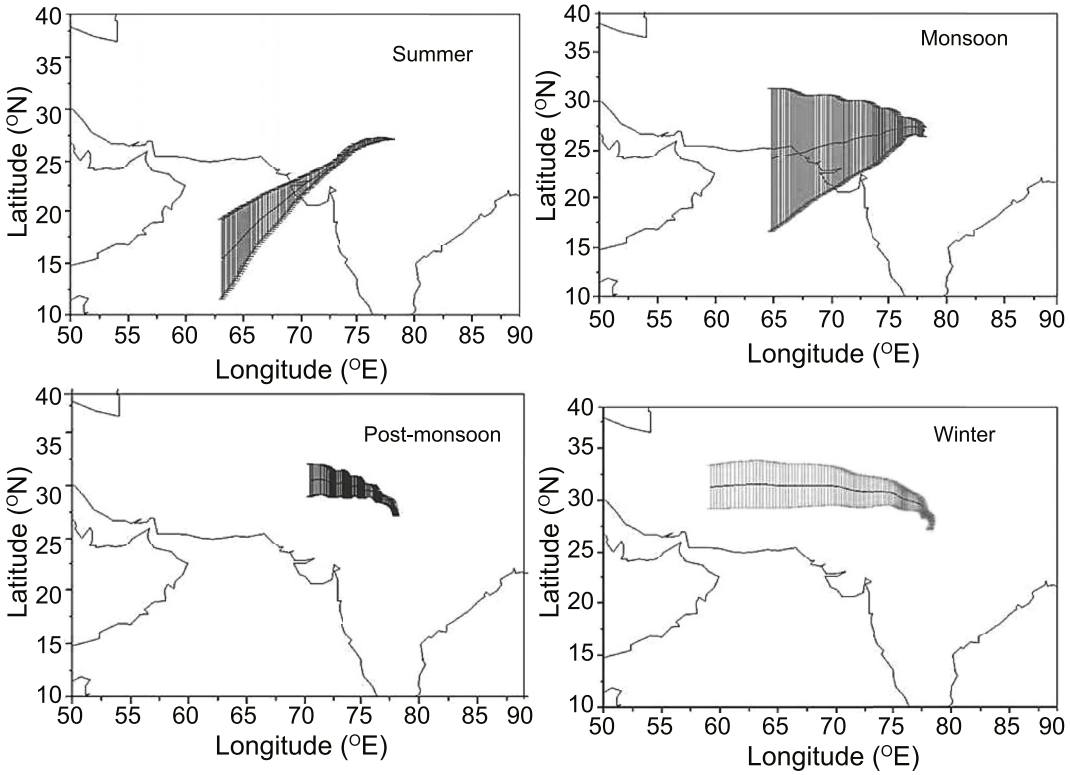

Fig. 9. Air-mass back trajectories, analyzed on a daily basis, for each season in the Agra region (500 m AGL).

due to changes in wind patterns. The peaks during daytime (0700–1000 LST) and in the evening (1700–2100 LST) are attributable to vehicle emissions arising from rush-hour traffic, and boundary-layer dynamics. The major source of black carbon is biomass combustion and diesel exhaust, throughout the year, with an increase in local fossil fuel combustion during the winter season. This is supported by the ratio of BC_{880} to $UVPM_{370}$, which is less than 1 during the entire study period, except in winter (December). This may be due to the fact that biomass combustion and diesel exhaust are greater contributors over this region, whereas the higher ratio in winter may be due to the increase in consumption of fossil fuel and wood for heating purposes. Black carbon is found to be inversely related to wind speed and temperature during the measurement periods, which is relatively more pronounced during the winter and monsoon seasons. Emission sources and wind patterns influence the concentration of black carbon over the Agra region. Air-mass back trajectories, analyzed on a daily basis using the HYSPLIT model, indicate large seasonal variation in sources and the effects of long-range transported pollutants.

Acknowledgements. This work was carried out as a part of the Aerosol Radiative Forcing over India project of the Indian Space Research Organization's Geosphere Biosphere Programme. We are grateful to Dr. B. B. RAO, Principal, Technical College, and Prof. Sahab DAS, Head, Department of Chemistry, Deemed University, Dayalbagh, for their kind support and encouragement. Sampling assistance from Mr. Hazur SARAN is highly appreciated. Prof. C.P. NIGAM is gratefully acknowledged for reading throughout the manuscript. Finally, we wish to acknowledge Google Maps.

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