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Research Paper

Measurement of PM_{10}, $PM_{2.5}$ and Black Carbon and Assessment of Their Health Effects in Agra, A Semiarid Region of India

PRATIMA GUPTA[1], ASHOK JANGID[2] and RANJIT KUMAR[1,*]
[1]Department of Chemistry, [2]Department of Physics and Computer Science, Faculty of Science, Dayalbagh Educational Institute (Deemed University), Dayalbagh, Agra 282 005, India

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The present study highlights the annual and seasonal concentration of PM_{10}, $PM_{2.5}$ and Black Carbon (BC) and estimation of health risk assessment in Agra, a semiarid region in India during March 2016-February 2017. The average mass concentrations of PM_{10}, $PM_{2.5}$ and BC are found to be 157 µg m^{-3}, 87 µg m^{-3} and 6 µg m^{-3}, respectively. The mean mass concentrations of PM_{10} is highest in winter (195 µg m^{-3}), followed by post-monsoon (168 µg m^{-3}), summer (186 µg m^{-3}) and monsoon (80 µg m^{-3}) while for the $PM_{2.5}$ and BC the highest mean mass concentrations is in winter (116 µg m^{-3}) followed by post-monsoon (115 µg m^{-3}), summer (103 µg m^{-3}) and monsoon (31 µg m^{-3}). The variation in the concentration may be due to the combined effect of atmospheric conditions, local emissions as well as long-range transport. The mean ratio of $PM_{2.5}/PM_{10}$ was 0.37 and ranged from 0.13 to 0.78. It signifies the fact that the coarse mode particle dominates over fine mode particles in this region. The percentage contribution of BC in $PM_{2.5}$ is 8%. The exceedance factor is found to be more than 1.5 for PM_{10} and $PM_{2.5,}$ which indicates the critical level of the pollution load in the Agra region. The population-weighted concentration of PM_{10}, $PM_{2.5,}$ and BC is 3.2 µg m^{-3}, 1.7 µg m^{-3} and 1.4 µg m^{-3}, respectively. Health risk analysis has been performed using AirQ+ v.1.2 model developed by the World Health Organization (WHO). The relative risk factor analysis representing within the study site indicates the particulate matter from local combustion sources as the cause of mortality. The attributed mortality rate of $PM_{2.5}$ due to Chronic Obstructive Pulmonary Disease (COPD), lung cancer, and stroke in adult (30 years and above) is 1033, 2068, and 3717, respectively. The attributed mortality rate from PM_{10} due to chronic bronchitis in adult and bronchitis in children is 872 and 168, respectively.

Keywords: Particulate Matter; Black Carbon; Population-Weighted Mean Concentration; Health Risk Assessment

Introduction

Ambient air pollution is responsible for serious health problems in developing and developed country both. In today's changing environment, the ability of humankind to intuitively assess and manage health risk has become fundamental for survival. The current technological advancement, rapid industrialization, and modernization are found insufficiently coping with worsening the air quality. Air pollution is linked with various long-term and short-term effects such as respiratory infections and inflammations, cardiovascular dysfunctions, bronchitis, chronic cough, sinusitis, cold, cancer and premature deaths (Brucker *et al.*, 2014). The essential metrics to check the ambient air quality are PM_{10} (particles of size less than 10 µm), $PM_{2.5}$ (particles of size less than 2.5 µm), $PM_{1.0}$ (particles of size less than 1.0 µm) and black carbon (a component of $PM_{2.5}$). The breathing of particulate matter and black carbon poses the most considerable health risks because the particles can find their way deep into lung and bloodstream (Kim *et al.*, 2004; Gauderman *et al.*, 2004).

Various studies on mass concentration of particulate matter and black carbon have been reported from many places in the world. The level of PM_{10} and $PM_{2.5}$ have been measured extensively in ambient air of Shanghai (231 µg m^{-3} and 61.4 µg m^{-3}), Taiwan (128 µg m^{-3} and 88 µg m^{-3}), Hong Kong (406 µg m^{-3} and 64.4 µg m^{-3}), Beijing (142 µg m^{-3} and 96.5 µg m^{-3}), Korea (62 µg m^{-3} and 36 µg m^{-3}), Delhi

**Author for Correspondence: E-mail: rkschem@rediffmail.com*

(219 µg m^{-3} and 97 µg m^{-3}), rural area of Punjab (97 µg m^{-3} and 57 µg m^{-3}), Lucknow (212.5 µg m^{-3} and 129 µg m^{-3}) and Dhaka, Bangladesh (31.6 µg m^{-3} and 29.1 µg m^{-3}) (Ye *et al.,* 2003; Chen *et al.,* 2004; Cao *et al.,* 2006; Chen *et al.,* 2008; Hong *et al.,* 2010; Tiwari *et al.,* 2010; Pandey *et al.,* 2012; Awasthi *et al.,* 2011; Islam *et al.,* 2014). Similarly, the levels of BC mass concentration have also been reported from various places such as Seoul (4.86 µg m^{-3}), Trivandrum (0.5 µg m^{-3}), Mumbai (12.4 µg m^{-3}), Zurich (1.66 µg m^{-3}), Kanpur (6 µg m^{-3}), Asia (1-14 µg m^{-3}), Lahore (21.7 µg m^{-3}), Nepal (0.16 µg m^{-3}) and Europe (3.5 µg m^{-3}) (Kim *et al.,* 1999; Babu and Moorthy, 2002; Venkataraman *et al.,* 2002; Putaud *et al.,* 2003; Tripathi *et al.,* 2005; Koch *et al.,* 2007; Husain *et al.,* 2007; Marinoni *et al.,* 2010; McMeeking *et al.,* 2010). Various studies provide a qualitative assessment of the absorbing nature of particulate matter and black carbon, which helps to understand the relative impact on human health (Li *et al.,* 1996; Salvi *et al.,* 1999; Gong *et al.,* 2003). Various studies in the last decade have excessively red-flagged the contribution of ambient air pollution to increase in morbidity and mortality (Dockery *et al.,* 1993). Exposure to $PM_{2.5}$ caused 4.2 million deaths globally (Cohen *et al.,* 2017). Recent epidemiological studies indicate that associations between adverse health effects and exposure are stronger for BC than for $PM_{2.5}$ (Grahame *et al.,* 2014). Air pollution remained a serious health concern in India (CPCB, 2008; Saud *et al.,* 2011; Dey *et al.,* 2012; Kankaria *et al.,* 2014; Gargava *et al.,* 2016; Cohen *et al.,* 2017). However, limited studies on the health impacts of black carbon have been reported from India. The present investigation aims to characterize air pollution metrics (PM_{10}, $PM_{2.5}$, and BC) and health risk assessment in the Agra region. The population-weighted average concentrations, annual and seasonal variations, and their health risk assessment are reported. The meteorological parameters which affect the particle behavior in the atmosphere were monitored, and their influences on the mass concentration were investigated.

Materials and Method

Site Characteristics

The present investigation has been performed in Agra (27.17 ºN, 78.01 ºE) (Fig. 1). It is 363 km west to the state capital, Lucknow and 200 km south-east from national capital New Delhi. Agra is situated in a semiarid region of India. It has a total area of 87 km^2 and third most populous city in Uttar Pradesh and 24th most populous in India. The population of Agra is 15, 85, 704 as per the census of 2011 and the floating population is about 23 thousand tourists per day. The city of Agra is situated on the western bank of the river Yamuna on National Highway (NH-2). Agra is a famous tourist destination because of the Taj-Mahal, which is one of the seventh wonders in the world. There are many other Mughal-era buildings (Agra Fort, Fatehpur Sikri, Mariyam tomb, etc.) which are known as UNESCO (The United Nations Educational, Scientific and Cultural Organization) World Heritage Sites. The soil in Agra is very fine texture and mix with sand, silt, and clay. The Thar Desert of Rajasthan

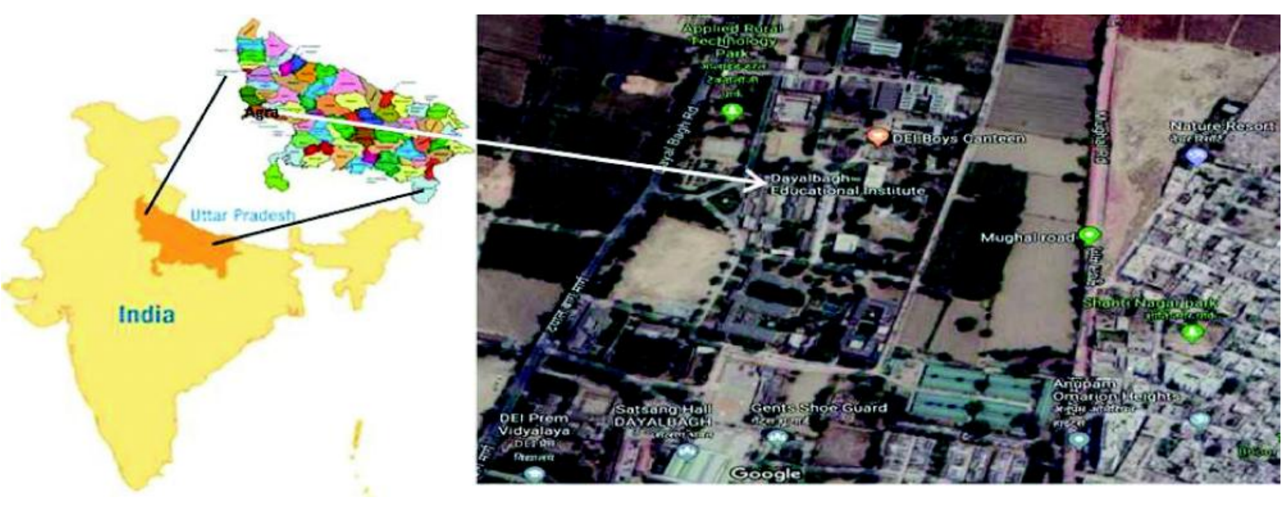

Fig. 1: Location of study site

surrounds two-third of its borderlines (SE, W, and NW). In the North, Agra is bounded by the Mathura district, in the South, it is bounded by Dhaulpur, in the East, Firozabad bounds it, and in the West, it is bounded by Bharatpur. Agra, situated on the Indo-Gangetic plain is about 169 m above mean sea level (MSL). It has a semiarid climate with average atmospheric temperature in summer lies between 21.9 ^{o}C to 40 ^{o}C, and in winter temperature is between 4.2 ^{o}C to 31.7 ^{o}C. The climate of Agra is generally divided into four seasons, summer (March to June), monsoon (July to August), post-monsoon (October-November) and winter (December to February). The wind pattern in Agra projects the prevalence of the calm condition, which is higher in winter than summer. The calm condition represents the wind velocity < 1 m s^{-1}. The dominant wind direction is SW, NW, W, and SE during summer; SE, E, W and NW during monsoon; NW during post-monsoon and NW, W during winter. The primary source of air pollution in Agra is the soil dust, urban population, open barren land, fossil fuel, biomass burning, poor traffic system and vehicular emission. The commercial and personal vehicles in Agra have increased from 4 to 6.4 Lakhs during 2003-2011 (SPA, 2018). The main contributor to particulate matter in Agra is truck, auto, bus, and two-wheeler. There are about 180 glass based industries in Firozabad, and a large number of small scale household industries in Agra contribute emission of pollutants (SPA, 2018). These small scale domestically operated industries significantly use coal, cow dung, wood, and agro-waste as a primary fuel. The major industrial works are rubber processing, engineering, brick-kiln and chemical works. Besides, several petha industries are also operating in the city. They generally use coal as fuel. The urban population of Agra is severely affected by vehicular emission. Vehicular pollutants are released at ground level, and hence the impact on the recipient population will be more. Besides, Mathura refineries and Firozabad glass industries are situated at a distance of about 40 km NW and NE, respectively, from the study site.

Sampling and Analysis

Measurement of Particulate Mass Concentration of PM_{10} and $PM_{2.5}$: PM_{10} and $PM_{2.5}$ sample were collected using the Polltech fine dust sampler at a flow rate of 16.7 LPM (liter per minute) for 24 hrs using glass fiber filter and PTFE filter, respectively. All the filter papers were carefully weighed and kept in desiccators before and after the sampling for 24 hours. The difference in the mass of filter paper after and before the sampling gives PM_{10} and $PM_{2.5}$ load.

Exceedance Factor : The exceedance factor is a ratio of the annual mean concentration of a pollutant with that of a respective standard (NAAQMS, 2014). Particulate matter concentration levels in ambient air are characterized as mean mass concentration and acceptance of daily limit concentration. It is a useful method to analyze the deterioration of air quality due to rising levels of air pollution. The exceedance Factor (EF) is determined using the following formula:

$$\text{EF (Exceedance Factor)} = \frac{\text{Observed concentration of criteria pollutant}}{\text{Annual standard for the respective pollutant and area class}}$$

Air quality has been categorized into four categories based on the concept of exceedance factor. The four categories of air quality are critical pollution (C) when EF is more than 1.5, high pollution (H) when EF is between 1.0 to 1.5, moderate pollution (M) when EF is between 0.5 to 1.0 and low pollution (L) when EF is less than 0.5.

Measurement of Black Carbon Mass Concentration : The black carbon mass concentration has been measured by using real-time AE-33 seven channels Aethalometer (Magee Scientific, USA). It is a self-contained optical transmission based automatic instrument. It was run at a flow rate of 2 LPM, and data were collected at an interval of one minute. The mass concentration of BC is calculated by the change in optical attenuation at 880 nm at any selected time duration (Drinovec *et al.*, 2015). The measured concentration is attributed to BC. The following equation has been used for fractionation of sources:

$$b_{abs}(\lambda) = b_{abs}FF(\lambda) + b_{abs}WB(\lambda)$$

where, $b_{abs}(\lambda)$ is the spectrally dependent mass absorption efficiency at 880 nm and $b_{abs}WB(\lambda)$ is the spectrally dependent mass absorption efficiency at 370-950 nm. The first assumption implies that the aerosol absorption coefficient $b_{abs}(\lambda)$ at a given wavelength can be expressed as the sum of the light absorption of aerosols emitted by b_{abs} FF(λ) and b_{abs} WB(λ) from fossil fuel and wood burning emissions, respectively.

Exposure Contributions and Health Impact Calculations : The population-weighted mean concentration is determined to understand the risk of pollutants on exposed populations of that particular area, and it is calculated by using the following equation:

$$\sum_{i=1}^{n} \frac{p_i \times C_i}{P_{tot}}$$

where, i designates each computational cell in the domain, p_i is the population at a given cell location, C_i is the particulate concentration in the same cell location, and p_{tot} is the total population in the domain of interest (i.e., air basin wide total population) (Mahmud *et al.*, 2012). In the present study, the total population for the computational cell was taken to be 15, 85, 704, and the population at the given cell location was considered to be 2830 for Dayalbagh region.

Health Risk Assessment : The estimation of health risk assessment due to pollution has been performed using AirQ+ v.1.2 model (World Health Organization, 2016). AirQ+v.1.2 software adopts the risk of mortality of $PM_{2.5}$ and PM_{10} with the impacts of long term and short term exposure on human health in a specific period (Table 1). It works by logarithmic functions, matrix calculations, and statistical functions. AirQ+ v.1.2 provides the associated relative risks (RR) of exposure. The relative risk of exposure is defined as the incidence rate in the population exposed (number of new cases per 100, 000 population/year) with the incidence rate in the counterfactual population. The AirQ+ v.1.2 software uses WHO specified input of relative risk (RR) values (per 10 $\mu g\ m^{-3}$) and their incidences for the different air pollutants as well as types of diseases (e.g., cardiovascular, respiratory, COPD, hospital admissions respiratory disease, etc.) (WHO, 2006; WHO, 2014). The health risk assessment is based on the population attributable fraction (PAF) which is defined as the fraction of health consequences in public exposed to a specific air pollutant (Maji *et al.*, 2017; Khaniabadi *et al.*, 2018). The PAF was calculated by the following equation (WHO, 2003):

$$PAF = RR - 1/RR \quad (1)$$

(in case of total population exposed)

$$PAF = p\,(RR-1)\,/p\,(RR-1)+1, 0 < p \leq 1 \quad (2)$$

(in case of number population exposed)

where, RR is the changed relative risk for the health outcome, and *p* is the proportion of the exposed population.

$$RR = 1 + (Ca\text{-}Cw) \times (RR - 1)/10$$

where, Ca is the ambient air pollutant concentration, and Cw is the WHO recommended threshold level of that pollutant. In a specific population, the number of cases or the fraction of all cases attributable to a specific pollutant and health outcome is of interest to calculate the attributable burden (AB):

$$AB = BoD \cdot PAF \quad (3)$$

where, BoD is the total burden of a specific health outcome. The total attributable burden for health outcome of specific pollutant is the sum over all age groups:

$$AB_{pollutant} = \Sigma_{\text{all age group}}\, AB_{pollutant} \text{ (health outcome, age group)}$$

Table 1: WHO specified default values of Relative risk (RR) (per 10 $\mu g\ m^{-3}$) with 95% confidence intervals (CI) implemented in AirQ+ v.1. 2 software

	Mortality (95% CI)	Relative risk	Baseline incidence per 100,000
PM_{10}	Total mortality	1.0174 (1.0062-1.0086)	1013
	Cardiovascular mortality	1.008 (1.005-1.018)	497
	Respiratory mortality	1.012 (1.008-1.037)	66
$PM_{2.5}$	Total mortality	1.0123 (1.0045-1.0201)	-
	Hospital cardiovascular admission	1.0091 (1.0017-1.0166)	-
	Hospital respiratory admission	1.0190 (0.9982-1.0402)	-

This means repeating the same calculation for all the age groups, considering the respective values of mortality and RRs.

Meteorological Condition : The Meteorological parameters viz., temperature, relative humidity and wind speed were monitored using automatic weather station (AWS 10.0, M/s. Rave Innovations) during the study period (Fig. 2A). The daily average temperature in summer lies between 21.9 ^{o}C to 40.9 ^{o}C and in winter temperature varies between 4.2 ^{o}C to 23.2 ^{o}C. The average relative humidity is 60%, which varies between 7.5% and 98%. The high relative humidity was observed in monsoon and lowest in the summer season. The wind speed tends to occur less than 1 m s^{-1} (calm wind) throughout the year. The wind comes from N, NE during monsoon while from NW during rest of the seasons.

Results and Discussion

Mean Mass Concentration of PM_{10}, $PM_{2.5}$ and Black Carbon

Table 2 presents the mean mass concentration of PM_{10}, $PM_{2.5}$ and BC. The concentration of PM_{10}, $PM_{2.5}$ and BC is 157 µg m^{-3}, 87µg m^{-3} and 6 µg m^{-3}, respectively. The high mass concentration of PM_{10}, $PM_{2.5}$, and BC may be due to the high source strength (automobile emission, diesel exhaust, crop harvesting, waste burning, and the road paved dust) and variation in the meteorological conditions (temperature, wind speed, and wind direction). The majority of the population in northern rural areas uses wood, crop residue, and cow dung cakes as a fuel (Badarinath *et al.*, 2006; Safai *et al.*, 2007; Gupta *et al.*, 2017). The concentration of PM_{10}, $PM_{2.5}$, and BC is compared with various studies reported from other places of the world (Table 3). The mass concentration of PM_{10} and $PM_{2.5}$ at the present location is higher than sites in the United States, Spain, Singapore, Taiwan, and Seoul (Burton *et al.*, 2000; Qurol *et al.*, 2001; Balasubramniam *et al.*, 2003; Chen *et al.*, 2004; Hong *et al.*, 2010) while lower than the Hong Kong, Delhi, Lucknow, Beijing, and Faisalabad (Cao *et al.*, 2006; Tiwari *et al.*, 2010; Pandey *et al.*, 2012; Wang *et al.*, 2014; Niaz *et al.*, 2016). The poor road and traffic system, use of biomass as a domestic fuel, the practice of crop burning, upliftment of soil and dust (re-suspended) may be responsible for high load at the present site. The BC concentration in Agra is higher than cities of U.S., Taiwan, Singapore, Hong Kong, Spain, and Seoul (Koch *et al.*, 2007; Chaw *et al.*, 2010; Zhang *et al.*, 2015; Deng *et al.*, 2016; Vallie *et al.*, 2017; Kim *et al.*, 2017). The level of BC in Agra is lower than Beijing, and Delhi (He *et al.*, 2010; Tiwari *et al.*, 2015) which may be due to considerably lower local sources strength.

Seasonal variations in the concentration of PM_{10}, $PM_{2.5}$, and BC have been observed amongst the seasons (Table 2). The mean mass concentrations of PM_{10} is highest in winter (195 µg m^{-3}), followed by post-monsoon (168 µg m^{-3}), summer (186 µg m^{-3}) and monsoon (80 µg m^{-3}) while for the $PM_{2.5}$ and BC the highest mean mass concentrations is in winter (116 µg m^{-3}) followed by post-monsoon (115 µg m^{-3}), summer (103 µg m^{-3}) and monsoon (31 µg m^{-3}). The seasonal variation on the particulate matter mass concentrations in Agra can also be explained as the combined impact of weather conditions, local emissions as well as long-range transport. The higher mass concentrations of the PM_{10} during summer season may be due to fact that the present study location is fed by particulate matter emissions from Delhi, agricultural waste combustion of Punjab and Haryana in addition to the dust storms of Rajasthan under the influence of downwind. The present site lies between Mathura and Firozabad. Wind rose (Fig. 2B) analysis

Table 2: Annual and seasonal mean mass concentration of particulate matter and black carbon

	PM_{10}			$PM_{2.5}$			BC		
	Mean±SD	Min	Max	Mean±SD	Min	Max	Mean±SD	Min	Max
Annual	157±84	29	375	87±46	14	388	6±2	0.21	68
Summer	186±80	50	375	103±58	17	222	6±2	0.5	38
Monsoon	80±35	29	136	31±17	14	58	2±0.6	0.17	26
Post-monsoon	168±54	89	267	115±76	46	283	8±2	0.21	68
Winter	196±65	54	245	116±88	25	388	11±6	0.75	58

Table 3: Comparison of PM_{10}, $PM_{2.5}$ and BC with different sites of the world

Location	PM_{10} ($\mu g\ m^{-3}$)	$PM_{2.5}$ ($\mu g\ m^{-3}$)	BC ($\mu g\ m^{-3}$)	References
Agra	157	87	6	Present study
Beijing	175	99	8.7	Wang *et al.*, 2014; He *et al.*, 2010
US	23.1	14.1	0.1-0.5	Burton *et al.*, 2000; Koch *et al.*, 2007
Singapore	-	27.5	3.3	Balasubramniam *et al.*, 2003; Zhang *et al.*, 2015
Spain	49.8	35	3.7	Qurol *et al.*, 2001; Vallie *et al.*, 2017
Taiwan	128	88	5.3	Chen *et al.*, 2004; Chaw *et al.*, 2010
Faisalabad, Pakistan	109-164.3	22-49	-	Niaz *et al.*, 2016
Hong Kong	406	64.4	2.4	Cao *et al.*, 2006; Deng *et al.*, 2016
Seoul	62	36	2.6	Hong *et al.*, 2010; Kim *et al.*, 2017
Zonguldak, Turkey	66.7-116.7	32.4-83.3	-	Akyuz *et al.*, 2009
Delhi	219	97	12.1	Tiwari *et al.*, 2010; Tiwari *et al.*, 2015
Lucknow	212	129	-	Pandey *et al.*, 2012

reveals the dominance of wind from NW, W, E and SW. The higher mass concentrations of the PM_{10} during the summer in comparison to post-monsoon may be due to the turbulent wind and the upliftment of soil dust. The highest mass concentrations of $PM_{2.5}$ and BC during the winter and post-monsoon season may be due to calm wind speed, low mixing height, low atmospheric boundary layer, increased biomass and coal burning (Tiwari *et al.,* 2012; Misra *et al.,* 2012; Pipal *et al.*, 2014; Villalobos *et al.*, 2015; Gogikar and Tyagi, 2016). The lowest mass concentration of particulate matter and BC in monsoon season may be due to high precipitation (Kumar *et al.,* 2007; Kumar and Sarin, 2009; Deshmukh *et al.*, 2011). Meteorological parameters influence the concentration of PM_{10}, $PM_{2.5,}$ and BC. The BC mass concentration was observed to be the highest in the moderate wind condition (Gupta *et al.*, 2017).

The monthly mean concentrations of $PM_{2.5}$, PM_{10} and BC have been shown in (Fig. 3). The highest monthly mean mass concentration of PM_{10} (249 $\mu g\ m^{-3}$), $PM_{2.5}$ (177 $\mu g\ m^{-3}$) and BC (21 $\mu g\ m^{-3}$) has been observed in December while the lowest concentration of PM_{10} (57 $\mu g\ m^{-3}$) $PM_{2.5}$ (26 $\mu g\ m^{-3}$) and BC (1.6 $\mu g\ m^{-3}$) has been seen in August or September. The month of December-January is generally under the influence of local biomass burning and moderate or calm wind condition ($< 1\ m\ s^{-1}$). The highly stable nature of atmosphere during October to February led to lower surface convection and, trapping of the enormously increased aerosol particle within the lower atmosphere. Various studies have also shown the higher concentration of particulate matter and BC in the month of November-December which has been attributed to the poor dilution of air pollutants (Taneja *et al.*, 2008; Kulshrestha *et al.*, 2009; Awasthi *et al.*, 2011; Tiwari *et al.*, 2012; Gupta *et al.*, 2017).

Source Attribution of PM_{10}, $PM_{2.5}$ and Black Carbon

The ratio of $PM_{2.5}$ and PM_{10} has been analyzed to understand the mode and nature of the particulate matter. The $PM_{2.5}/PM_{10}$ monthly mean mass concentration ratios at the present study site have been presented in Fig. 4A. The mean ratio of $PM_{2.5}/PM_{10}$ was 0.37 and ranged from 0.13-0.78. It signifies the fact that the coarse mode particle dominates over this region (Kumar *et al.*, 2007; Kumar and Kumari, 2015). It also suggests the contribution of natural sources (soil, dust, the road paved dust as well as grinding of marble and red stone) (Kumar *et al.*, 2007; Satsangi *et al.*, 2014). The similar study has also been reported from Kanpur, which intended that the coarse and the fine particle are associated with local sources and meteorological phenomenon (Sharma and Maloo, 2005). The Thar Desert of Rajasthan and open barren land around the site might be the reason for the dominance of the coarse particles over this region.

The mean ratios of $BC/PM_{2.5}$ and the percentage

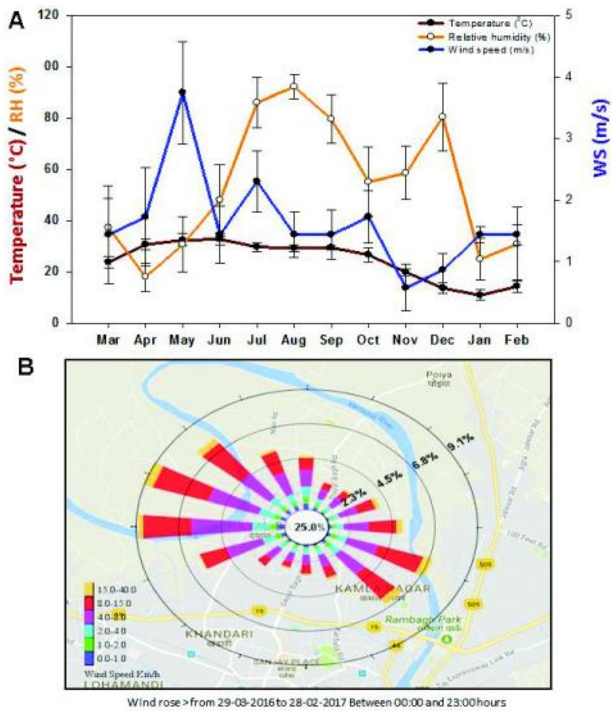

Fig. 2: (A) Meteorological parameter (temperature, relative humidity and wind speed) during the study period and (B) Wind rose during the study period

contribution of BC have been shown in Fig. 4B. The $BC/PM_{2.5}$ mass ratio varied from (0.01-0.17) with a mean value of (0.08). The average percentage contribution of BC in $PM_{2.5}$ is 8%. The lower ratio of $BC/PM_{2.5}$ is due to aeolian and other fugitive dust, which increases $PM_{2.5}$ with non-carbonaceous material, thereby, decreasing the $BC/PM_{2.5}$ ratio (Zhang *et al.*, 2008). The abundance of BC in $PM_{2.5}$ is affected by the mixture of pollution sources and meteorological conditions. Tripathi *et al*. (2005) reported BC as 7 to 15% of total suspended particulate matter in Kanpur. The contribution of BC in $PM_{2.5}$ has been reported to be 7% in Hyderabad (Latha and Badrinath, 2005), 2.3% in Pune (Safai *et al.*, 2007), 1.6 to 15.6% in Xi'an, China (Cao *et al.*, 2009) and about 5% in suburban regions of Europe and North America (Ramanathan and Crutzen, 2002). The percentage fractionation of sources has been made using spectrally dependent mass absorption efficiency and presented in Fig 5. The natural sources contribute about 63%, while anthropogenic sources contribute 37%. Anthropogenic sources include biomass burning, use of fossil fuel, solid waste incineration, transportation, and industrial emissions. The percentage of contributions of fossil fuel, biomass combustion, and other sources are 28%, 7%, and 65%, respectively.

The particulate matter (PM_{10} and $PM_{2.5}$) and black carbon can easily get long-range transported under the influence of wind. The air mass back trajectory analysis was employed with seasonal data. The seven days air mass back trajectories of air pollutants have been obtained by using the HYSLPIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) model from NOAA Air Resources Laboratory (http://www.arl.noaa.gov/ready/hysplit4.8 html) (Fig. 6A). The transport pathways of pollutants are from various sources viz., local as well as via long-range transport. Wind from the west and northwest were dominant over this region in the post-monsoon and winter season. Crop burning is in common practice in Punjab and Haryana, which may be possible reason for high pollution load in Agra. MODIS (Moderate Resolution Imaging Spectroradiometer) image derived from the Aqua satellite NASA, USA also shows influences of crop burning over this region in the year of 2016 and 2017 (Fig. 6B). The calm and stable atmospheric conditions and the low boundary layer also contribute to an increase in the concentration of pollutants. These conditions retain the high loading of particulate matter (PM_{10} and $PM_{2.5}$) and BC in the winter season. The southwest prevails during the monsoon season, but pollutants get settled under the influence of precipitation. Several studies have reported similar trends (Srivastava *et al.*, 2012; Tiwari *et al.*, 2013; Kumar and Kumari, 2015; Gupta *et al.*, 2017).

Health Risk Assessment

The health risk assessment has been carried out by using exceedance factor analysis, population weighted mean analysis, and by using AirQ+ model. The exceedance factor has been analyzed for the level of particulate matter mass concentration (PM_{10} and $PM_{2.5}$) at Agra (Fig. 7). The monthly analysis of exceedance factor of particulate mass concentration (PM_{10} and $PM_{2.5}$) shows significant variation from moderate to the critical level. The exceedance factor is found to be more than 1.5 for PM_{10} and $PM_{2.5,}$ which indicates the critical level of pollution load. There is an alarming rise in the pollutant level as the maximum number of days has crossed double the standard values. The month of November and December shows the highest critical exceedance

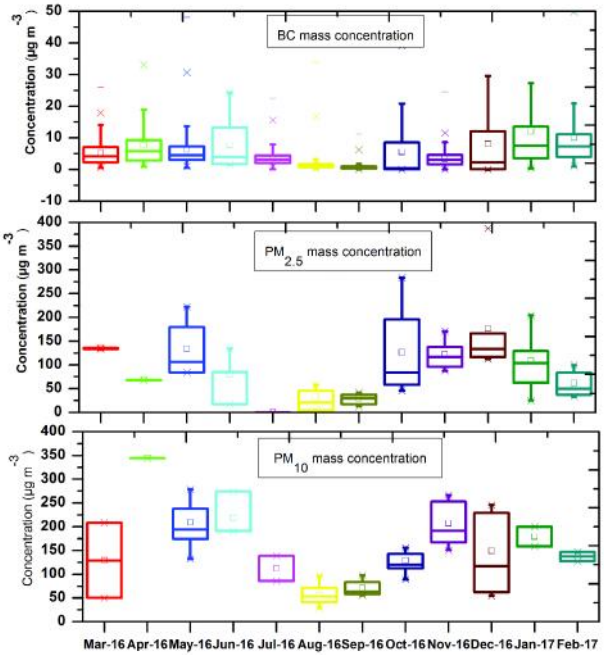

Fig. 3: The monthly mean mass concentration of PM_{10}, $PM_{2.5}$ and BC

which may be due to the peak winter season and excessive use of fossil fuel for cooking and heating purposes. The population-weighted mean concentration of PM_{10}, $PM_{2.5,}$ and BC have been determined to understand the risk of pollutants on exposed populations of that particular area and presented in Table 4. $PM_{2.5}$ has been considered as an indicator and has been attributed for most deaths, while BC also put weight on health risk. Various short term studies on health effects have shown the associations with BC are more robust than $PM_{2.5}$ and PM_{10} (Schwartz *et al.*, 1992; Gotschi *et al.*, 2002; Johanneson *et al.*, 2007; Schaap *et al.*, 2007; WHO, 2012; Segersson *et al.*, 2017), which causes the cardiovascular mortality, and cardiopulmonary hospital admissions (Atkinson *et al.*, 2001; Anderson *et al.*, 2001; Analitis *et al.*, 2006; Zanobetti and Schwartz, 2006; Brauer *et al.*, 2007).

The health risk assessment and relative risk factors of $PM_{2.5}$ and PM_{10} has been calculated using AirQ+ model (Table 5). The emission sources of these pollutants are local, biomass combustion, waste burning, vehicular emission, as well as long-range transport, which cause more premature deaths (Kim *et al.*, 2014). The attributed deaths to all natural causes in adult (above 30 years) were 1651 and 1372 for PM_{10} and $PM_{2.5}$, respectively. The attributed

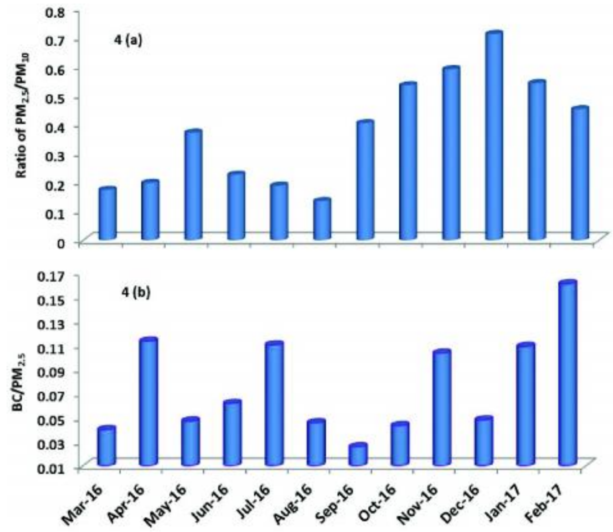

Fig. 4: (A) Fraction of $PM_{2.5}/PM_{10}$ (B) the ratio of $BC/PM_{2.5}$

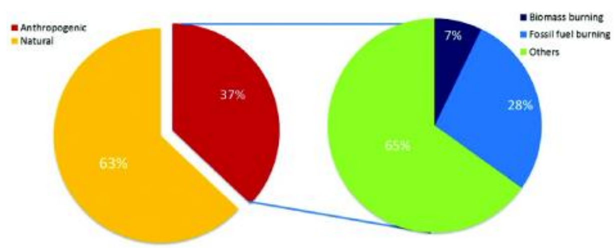

Fig. 5: Fractionation of different type of sources

mortality rate of $PM_{2.5}$ due to Chronic Obstructive Pulmonary Disease (COPD), lung cancer, and stroke in adult (30 years and above) is 1033, 2068 and 3717, respectively. The attributed mortality rate from PM_{10} due to chronic bronchitis in adult and bronchitis in children is 872 and 168, respectively. The excess number of death has been reported from chronic bronchitis in adults due to PM_{10}. Adults are more suffering from the PM_{10} as adults are more exposed in the ambient environment. Various studies have also reported the excess number of mortality due to PM_{10} (Gurjar *et al.*, 2010; Nagpure *et al.*, 2014; Maji *et al.*, 2017). The premature deaths in India from PM_{10} and $PM_{2.5}$ have been reported to be a total of 4, 38, 000 and 4, 86, 100, respectively (IHME, 2014; Chowdhury and Dey, 2016). Similar studies have been reported in Cape Town, California, Shanghai, Delhi, Kolkata, Pune for mortality rate due to cardiovascular mortality, respiratory mortality and hospital admission of COPD (Carincross *et al.*, 2007; Mahmud *et al.*, 2012; Zhao *et al.*, 2013; Lelieveld *et al.*, 2015;

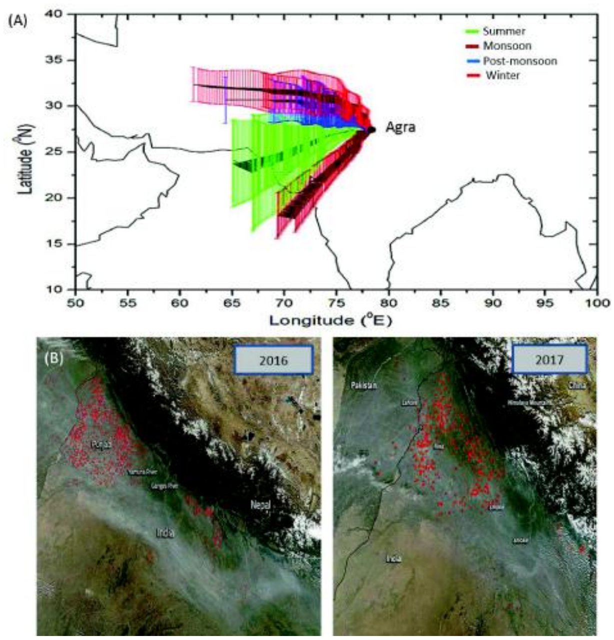

Fig. 6: (A) Back air trajectories on daily basis for Agra region (500 AGL) and (B) Source: Moderate Resolution Imaging Spectroradiometer (MODIS) image from the Aqua satellite, NASA, USA. Smoke from fires in the states of Punjab and Haryana was blowing in a northwesterly direction. Actively burning fires are marked with red dots

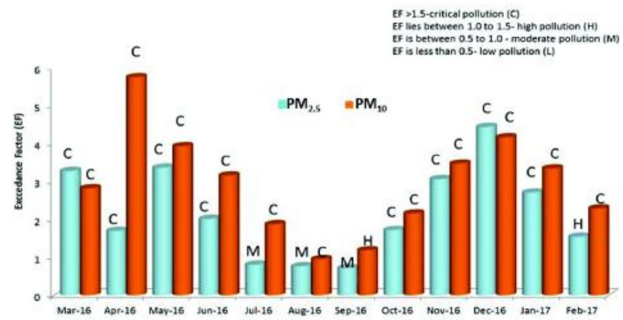

Fig. 7: Exceedance factor of PM_{10} and $PM_{2.5}$

Nagpure *et al.*, 2014; Maji *et al.*, 2016). Many epidemiological studies have found conclusive relationships between the increase in suspended particles to increased mortality, besides, to increase in hospitalization, due to respiratory or cardiovascular diseases (Venners *et al.*, 2003; Kan *et al.*, 2008; Dockery, 2009; Pascal *et al.*, 2013). Combating with air pollution needs a multipronged approach. It may be monitored and planned at the city level or regional level to develop methods to overcome the problem. Agra like megacities viz., Mumbai, and Delhi are under acute risk with its inhabitants prone to severe diseases. Consolidated steps at various levels need to be taken to frame environmental policies and plan health risk management.

Table 4: Population-weighted annual average concentrations of the different fractions of pollutants from different source categories

Fraction of pollutants	Sources of pollutant	Populated mean concentration ($\mu g\ m^{-3}$)
PM_{10}	Natural and anthropogenic	0.13
$PM_{2.5}$	Automobile emission	0.15
BC	Biomass and automobile emission	0.011

Table 5: Attributable mortality rate due to $PM_{2.5}$ and PM_{10}

		Attributable factor (%)	Attributed deaths
$PM_{2.5}$	All natural causes mortality	8.9	1372
	COPD for adults (30+ above)	6.7	1033
	Lung cancer adults (30+ above)	13.3	2068
	Stroke for adults	24.1	3717
PM_{10}	All natural causes mortality	10.3	1631
	Chronic bronchitis in adults	11.1	872
	Bronchitis in children	16.1	168

Conclusion

The mass concentration PM_{10}, $PM_{2.5}$ and Black Carbon and their health effect were measured over Agra in the Indo-Gangetic basin during March 2016-February 2017. The average mass concen-trations of PM_{10}, $PM_{2.5}$ and BC were found to be 157 µg m^{-3}, 87 µg m^{-3} and for 6 µg m^{-3}, respectively. The BC mass concentration during December-January is generally under the influence of local biomass burning and moderate and calm wind condition. The percentage contribution of Black Carbon (BC) in $PM_{2.5}$ is 8%. The mean ratio of $PM_{2.5}/PM_{10}$ was 0.37 and ranged from 0.13-0.78. It signifies the fact that the coarse mode particle dominates over this region. Natural sources contribute 63%, and anthropogenic contributes 37% during the study period. The contribution of fossil fuel, biomass burning, and other sources towards anthropogenic sources are 28%, 7%, 65%, respectively. The exceedance factor is found to be more than 1.5 for PM_{10} and $PM_{2.5,}$ which indicates the critical level of pollution load. The population-weighted mean concentration of PM_{10} (3.2 µg m^{-3}), $PM_{2.5}$ (1.7 µg m^{-3}) and BC (1.4 µg m^{-3}) have been determined to understand the risk of pollutant on the exposed population. The attributed mortality rate of $PM_{2.5}$ due to Chronic Obstructive Pulmonary Disease (COPD), lung cancer and stroke in adult (30 years and above year) are 1033, 2068 and 3717, respectively, while attributed mortality rate from PM_{10} due to chronic bronchitis in adult and bronchitis in children are 872 and 168, respectively. The health risk on the common public in Agra is high like megacities of India viz., Mumbai and Delhi.

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