Chemical characteristics of particulate matters and their emission sources over Varanasi during winter season

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Abstract

The chemical composition of particulate matter impacts both human health and climate. In this study, the chemical characteristics of particulate matter was measured for four months (November 2016–February 2017) at Varanasi, which is located in the middle of the Indo-Gangetic Basin (IGB). The daily observed mean values of PM_{10} and $PM_{2.5}$ are 134 ± 48 and 213 ± 80 μg/m³, respectively, which exceeds both national and international standards. The average value of $PM_{2.5}/PM_{10}$ ratio is 0.64 ± 0.16 which indicates a relatively higher fraction of fine particles that are attributed to anthropogenic emission sources (biomass/post-harvest burning) as corroborated by MODIS fire counts and back trajectory analysis. Ion chromatographic measurements showed that SO_4^{2-}, Cl^-, K^+, NO_3^-, Na^+, Ca^{2+}, Mg^{2+} are the major ionic species present in the aerosol. Scanning Electron Microscopy with Energy Dispersive X-Ray (SEM–EDX) analysis shows the prevalence of carbon-rich particles at Varanasi which is likely due to biomass burning and other anthropogenic sources.

Keywords PM_{10} · $PM_{2.5}$ · Ion chromatography · SEM–EDX · Chemical analysis

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1 Introduction

In last few decades, the study of particulate matter, one of the major components of air pollutants, has become a matter of research interest world wide due to its detrimental effects on regional air quality, atmospheric visibility, human health, earth's ecosystem and climate system (Singh et al. 2016a; Seinfeld et al. 2016; Chowdhury and Dey 2016; Ghude et al. 2016; Penner et al. 2011; Lau and Kim 2006). They are emitted from both natural as well as anthropogenic sources resulting to a complex, multi-component mixture containing water-soluble inorganic species (ionic species), organic as well as metals also (Chernyshev et al. 2019; Shah et al. 2018; Tiwari et al. 2016; Onat et al. 2012). They can travel a large distance from one continent to another continent under suitable meteorological conditions (Tiwari et al. 2020; 2018; Ancelet et al. 2015). The rapid increase in industrialization/urbanization and associated energy demands led to an increasing trend of air pollution over the south and southeast Asia (mainly China and India) (Tiwari et al. 2020; Liu et al. 2019; Pani et al. 2019; Ghude et al. 2016) which have a substantial impact on the earth's atmosphere and human health (Sweerts et al. 2019; Cohen et al. 2017; Burnett et al. 2014; WHO 2014a, 2014b). Recently, World Health Organization (WHO 2018) reported that most of the metropolitan Indian cities have exceeded the WHO's annual particulate exposure guideline ($PM_{2.5} \sim 10$ μg/m^3 and $PM_{10} \sim 20$ μg/m^3) resulting to more than 90% population exposed by polluted air. Recent studies also reported more than half-million pre-mature death over India occurred due to air pollution (Ghude et al. 2016; Chowdhury and Dey 2016). Besides, PMs have also ability to perturb the earth's radiation budget through their direct effect (by effecting scattering and absorption of solar radiation), indirect effect (by serving as Cloud Condensation Nuclei), semi-direct effect (evaporation of cloud) and associated with a large uncertainty because of their spatial and temporal variability (Tiwari et al. 2018; Tiwari et al. 2015; Seinfeld et al. 2016; Fan et al. 2016). Although several comprehensive studies have been performed worldwide to understand the characteristics of these particulates and their impacts on different aspects (Guttikunda 2013; Zhang et al. 2014) yet their scientific understanding is not up to satisfactory level. These uncertainties are mainly due to their different emission sources, chemical compositions and interaction with complex atmospheric processes (IPCC 2013). Thus, a furthermore in-depth study of particulate matter along with their chemical composition and emission sources is highly needed, especially at a regional scale.

Indo – Gangetic Basin (IGB), one of the largest river basins in the world, experience heavy aerosol loading throughout the year which shows a strong spatio-temporal heterogeneity in various aerosol characteristics, their inter/intra seasonal variability and associated with significant heterogeneity in their types, emission sources, and composition (Tiwari et al. 2018, 2016, 2015; Gautam et al. 2011). During pre-monsoon season, IGB experiences prevailing westerly/southwesterly winds in association with the dry weather condition which is favorable for the frequent and intense dust storm. These dust storms enhance the concentration of coarse mode aerosol thorough the long-range transportation from the Middle East, Thar Desert and Southwest Asia (Tiwari et al. 2019, 2013; Singh et al. 2016b; Kumar et al. 2015a; Srivastava et al. 2014; Singh and Naseema Beegum 2013). The enhanced concentration of coarse mode particle causes degradation of air quality and thus atmospheric visibility (Alkheder and AlKandari 2020; Taneja et al. 2020; Smith et al. 2019). However, crop residue/biomass burning is a common phenomenon over the northwestern IGB (especially Haryana and Punjab state) during the post-monsoon season and causes a dense smoke plume under suitable meteorological conditions (Ojha et al. 2020; Sarkar et al. 2018; Kumar et al. 2016, 2015b;

Kaskaoutis et al. 2014). On the other hand, during the winter season, IGB is mostly covered by dense haze/fog on a time scale of few hours to week due to weak convection, high relative humidity and shallow atmospheric boundary layer (Ali et al. 2019; Kulkarni et al. 2012; Mishra and Shibata 2012). The well admixing of minerals or desert dust with anthropogenic pollutants (like carbonaceous aerosols) from various emission sources results in the formation of a thick aerosol layer, popularly referred to as atmospheric brown clouds (Bonasoni et al. 2010). Water-soluble inorganic ionic species (WSIIS: like Na^+, Ka^+, F^-, Cl^- etc.) are the major components of the particulate matter and their contribution is highly influenced by the meteorological conditions, emissions sources and their chemical transformation (Pani et al. 2019; Singh et al. 2016a; Satsangi et al. 2013; Tsai et al. 2012; Pipal et al. 2011). Furthermore, SO_4^{2-}, NO_3^- and NH_4^+ are another major component of particulate matter which are mainly emitted from the anthropogenic sources (gas to particle conversion) (Tiwari et al. 2016; Satsangi et al. 2016; China et al. 2013; Tsai et al. 2011). Several researchers have attempted to understand the chemical composition and mass size distribution of particulate matter at different locations in IGB like at Patiala (Singh et al. 2016a), at Delhi (Ali et al. 2019; Goel et al. 2018), Kanpur (Rajput et al. 2014), Agra (Satsangi et al. 2016, 2013), Allahabad, Hisar (Ram et al. 2012), Kolkata (Roy et al. 2016) and Kharagpur (Verma et al. 2013). In addition to this, Rastogi et al. (2014) found a pronounced variability in $PM_{2.5}$ concentrations, ionic ($SO4^{2-}$, $NO3^-$, NH_4^+, K^+, etc.) at Patiala during October 2011 to March 2012. To the best of my knowledge, very limited studies have been performed to evaluate the chemical composition of particulate matter and their emission sources over Varanasi (Tiwari et al. 2016; Murari et al. 2016; Singh et al. 2014). Thus, a detailed analysis of particulate matter concentration, chemical composition, and emission sourcesis highly needed at study region which will be helpful for policymakers to take legislative action for environmental protection especially on a regional scale. In this context, the present study represents the mass concentration of particulate matter (both PM_{10} and $PM_{2.5}$) and its chemical composition along with the emission sources during winter season at Varanasi, located in central Ganges Valley.

2 Experimental observations and methodology

2.1 Site description and meteorological condition

The particulate matter sampling is performed at the terrace of the Department of Physics; Banaras Hindu University (BHU), Varanasi during the winter season (November 2016 – February 2017). Varanasi (25.20^0 N, 82.97^0 E, ~ 83 m above msl), a semi-urban city, located in central IGB covers an area of around 112 km^2 with high population density i.e. 2399 residents per square kilometer (Census India 2011). Figure 1 shows the map of Varanasi and the detailed descriptions about the surroundings of the sampling site. Varanasi is heavily polluted and experiences high aerosol loading with significant heterogeneity in their types, emission sources on seasonal as well as an annual basis (Tiwari et al. 2018; Kumar et al. 2015b; Tiwari and Singh 2013). The dominance of coarse mode aerosol is found during pre-monsoon season. The abundance of fine mode aerosols over Varanasi during post-monsoon and winter season is mainly attributed to the biomass/crop residue and fossil fuel burning (Tiwari et al. 2018; Kumar et al. 2015a; Kaskaoutis et al. 2014). However, in monsoon season, on a certain occasion, it is also influenced by the marine aerosol coming from the Bay of Bengal through long-range transportation (Tiwari and Singh 2013). The spatial distribution of
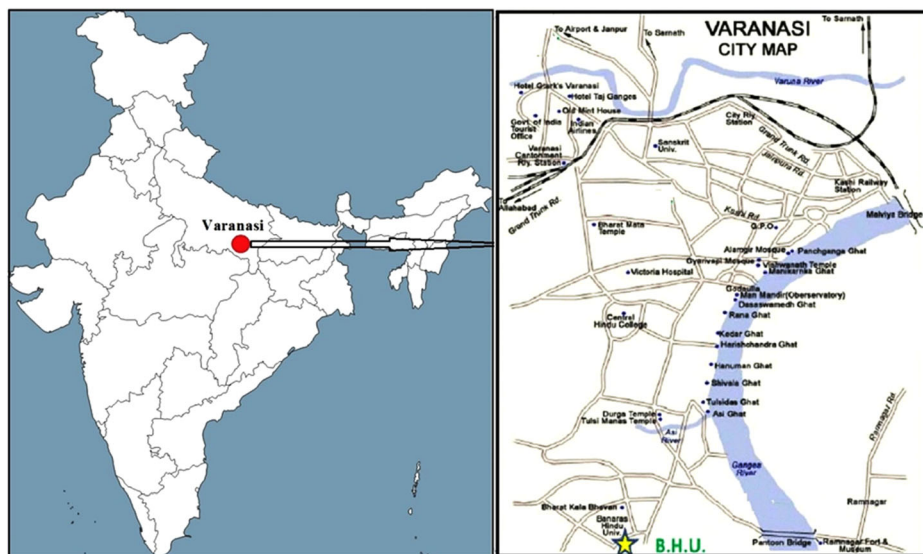
Fig. 1 Map of the sampling location and surrounding area

aerosol optical depth (AOD; at 550 nm) over the Indian subcontinent, obtained from the Moderate Resolution Imaging Spectroradiometer (MODIS: level 3 version 6.1) onboard of Aqua satellite during the study period (November 2016 – February 2017) is shown in Fig. 2. It has a wide swath of 2330 km and passes over the Indian region at ~10:30 am (Terra) and ~ 01:30 pm (Aqua) local solar time. It measures the radiance at 36 spectral bands in the visible to thermal IR spectral range of 0.41– 14 μm with a spatial resolution of $1^0 \times 1^0$ (Kaufman et al. 1997). The uncertainty in AOD measurement over oceanic regions is relatively higher i.e. $\pm (0.03 \pm 0.05 \tau)$ than over the land i.e. $\pm (0.05 \pm 0.15 \tau)$ (Tiwari et al. 2019; 2016a; Prijith et al. 2013; Levy et al. 2010). Further details about the MODIS, its data retrieval algorithm and associated uncertainties are discussed elsewhere (Levy et al. 2010; Remer et al. 2005; Kaufman et al. 1997). Figure reflects relatively higher value AOD over the entire IGB than the rest part of the Indian subcontinent. A negative gradient from western to eastern IGB resulting in the higher value of AOD which may be mainly due to wind down flow during this time (Kumar et al. 2015a). The mean value of AOD over Varanasi and surrounding (represented by a blue star in Fig. 2) is significantly higher (> 0.8) resulting in an interesting region for aerosol study. Earlier studies also reported higher AOD value over Varanasi during the winter season (Tiwari et al. 2018; Murari et al. 2016; Kumar et al. 2015b; Tiwari and Singh 2013).

During the winter season, Varanasi experienced cold and stable atmospheric conditions with relatively lower ambient temperature which sometimes reached up to nearly 2 °C. Daily variation of meteorological parameters i.e. temperature (°C), relative humidity (RH %), wind speed (m/s) and wind direction (degree), obtained from the Central Pollution Control Board (CPCB), during the study period (November 2016–February 2017) over Varanasi is shown in Fig. 3. The relative humidity is found in the wide range from 31 to 97% while the ambient temperature varied in a range of nearly 2 to 28^0 C. The weather condition is calm and cold during the study period and winds are found mostly north easterly with the relatively lower speed in the range of 0.27 to 2.4 m/s. Further details about the meteorological conditions at Varanasi are given in Table 1.
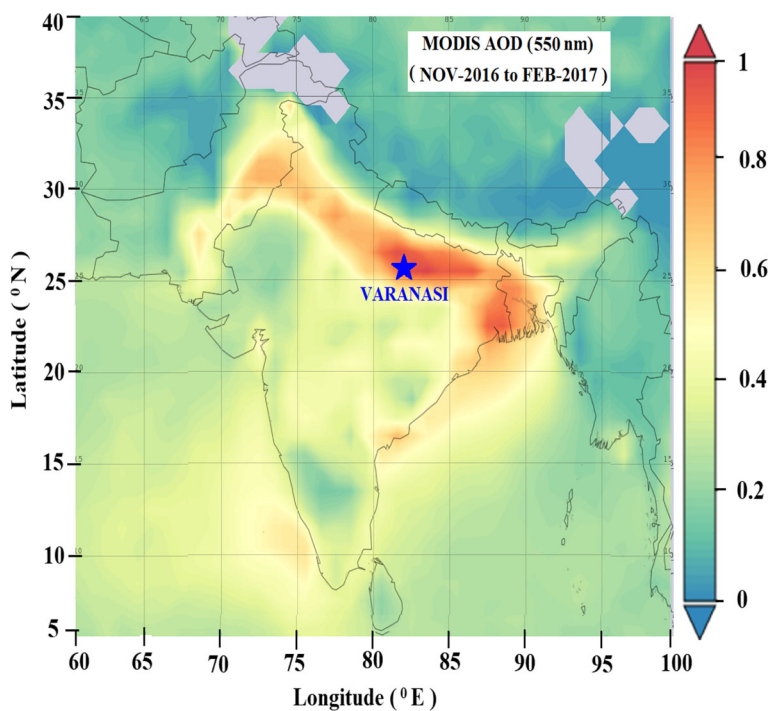
Fig. 2 Map of the study area showing the spatial variability of MODIS derived $AOD_{550\ nm}$ during November 2016 – February 2017. The blue star represents the location of Varanasi city

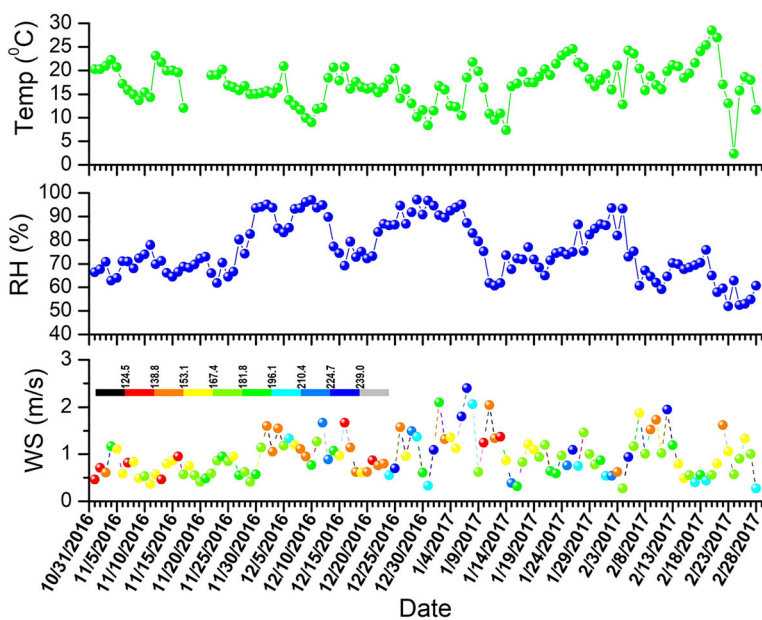**Fig. 3** Diurnal variation of meteorological parameters like temperature (°C), relative humidity (RH %), wind speed (m/s) and wind direction (degree) over Varanasi during the study period. The color scale represents the wind direction

Table 1 Monthly mean and ranges for meteorological parameters at Varanasi during the study period

Months	Temp. (°C) Mean ± Std (Min – Max)	RH (%) Mean ± Std (Min – Max)	WS(m/s) Mean ± Std (Min – Max)	WD (Degree) Mean ± Std (Min – Max)
Nov. 2016	18 ± 3 (12–23)	70 ± 6 (61–93)	0.7 ± 0.2 (0.37 ± 1.17)	162 ± 17 (124–187)
Dec. 2016	15 ± 3 (8–20)	86 ± 8 (69–97)	1.04 ± 0.4 (0.33–1.67)	170 ± 27 (136–233)
Jan. 2017	17 ± 4 (7–24)	78 ± 10 (31–95)	1.14 ± 0.50 (0.32–2.4)	181 ± 27 (135–237)
Feb. 2017	19 ± 5 (2–28)	67 ± 11 (52–93)	0.92 ± 0.48 (0.27–1.95)	180 ± 24 (149–239)

2.2 Sampling and chemical analysis

Particulate matter samples (both PM_{10} and $PM_{2.5}$) were collected for 24 h during the study period, using two Respirable Dust Sampler (APM 460 NL and APM 550 with flow rate 1.0 m³/min) manufactured by Envirotech Pvt. Ltd. which is well accepted and widely used in air pollution sampling (Sah et al. 2019; Ambade 2018; Bansal et al. 2019). The samples were collected twice in a week on Whatmann filter papers for the entire study period and after sampling, these were kept in a refrigerator at 4^0 C for further analysis. The concentration of particulate matter is calculated using the gravimetric method. For the analysis of anion and water-soluble cations, one - fourth of the sample filter was extracted into 20 ml of de-ionized water (18.2 MΩ) and then ultrasonicated for half an hour and then filtered using Whatman filters. After that, the filtered solution was again filtered using 0.22 μm syringe filters. Then this filtrate was executed in Ion Chromatograph (Metrohm, 930 Compact IC Flex, Switzerland) to examine anions and cations present in the solution.

The elemental composition and surface morphology of the particulate matter was analyzed using Scanning Electron Microscopy with Energy Dispersive X-Ray SEM-EDX) analysis. The sample was randomly cut in the size of about 1 mm² out of the main filter (Singh et al. 2014). A very thin film of carbon was deposited on the surface of the samples to make electrically conductive using a vacuum coating unit. These samples were mounted on electron microprobe stubs. The SEM was a 'Supra' type, manufactured by Carl Zeiss Microscopy, Germany. The EDX was a Genesis type with Si-Li-Detector manufactured by EDAX, Germany.

2.3 Fire count and Back trajectory analysis

The fire count over any specific region provides valuable information about the biomass/crop residue burning which is capable to inject plenty amount of fine mode aerosol particles as well other environmental pollutants into the atmosphere. The MODIS derived biomass fire hotspots data with a threshold confidence level ($\geq 80\%$) are obtained from the National Aeronautics and Space Administration (NASA) Land-Atmosphere near-Real-Time Capability for Earth Observing System (EOS) Fire Information for Resource Management System (NASA LANCE FIRMS) fire archive covering an area from 5^0 N to 40^0 N and 60^0 E to 95^0 E. However, the five days air mass back trajectory is computed from the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single-Particle Lagrangian Integrated Trajectory Model (HYSPLIT 4.9) using National Center for Environmental Prediction (NCEP) reanalysis wind data as input (Draxler and Rolph 2003). It provides a three-dimensional (latitude, longitude, and altitude) information about the air mass pathways as a function of time and directly associated with PMs concentration as well as composition also.

3 Result and discussions

3.1 Mass concentration of particulate matter

The mass concentration of particulate matter observed in the range of 123–464 μg/m³ and 42–257 μg/m³ with the mean value 213 ± 80 μg/m³ and 134 ± 48 μg/m³ for PM_{10} and $PM_{2.5}$ respectively. The daily observed concentration of PMs (both PM_{10} and $PM_{2.5}$) is manifold higher than the standard daily mean concentration of PMs referred by National Ambient Air Quality Standard (NAAQS: $PM_{10} = 100$ μg/m³ and $PM_{2.5} = 60$ μg/m³), US Environmental Protection Agency (USEPA: $PM_{10} = 150$ μg/m³ and $PM_{2.5} = 35$ μg/m³) and World Health Organization (WHO: $PM_{10} = 50$ μg/m³ and $PM_{2.5} = 25$ μg/m³) which indicated towards a serious threat to the environment, as well as health also. The observed mean value of PMs concentration is quite comparable to the mean values reported at different locations over IGB (Table 2). For example, the mean concentration of PM_{10} is reported ~285 ± 87 μg/m³ during the winter season at Varanasi (Murari et al. 2016; Kumar et al. 2015a), 203 ± 40 μg/m³ at Kanpur (Tare et al. 2006), 268 ± 74 μg/m³ at Kharagpur (Thurston et al. 2011), 232 ± 131 μg/m³ at Delhi (Tiwari et al. 2009). Similarly, Guttikunda (2013) also reported the concentration of $PM_{2.5}$ and PM_{10} to be 123 ± 87 and 208 ± 14 μg/m³ over Delhi. In addition, Pipal et al. (2014) also observed that the PMs concentration exceeded two to three times higher than the standard limit value ($PM_{2.5}$: 121 μg/m³ and PM_{10}: 230 μg/m³) over Agra city during 2010–2011. They have also studied elemental composition and morphology of these particulates and have reported C, O and Si-rich particles in both types of particulates. Sen et al. (2017) reported a large temporal as well as spatial variation in particulate matter concentration over IGB and Indo-Himalayan range for summer and winter season. They found that the PMs concentrations exceed NAAQS limit over IGB with value at Varanasi (PM_{10}: 238 ± 81 μg/m³), Kolkata ($PM_{2.5}$: 131 ± 58 μg/m³), Lucknow ($PM_{2.5}$: 130 ± 73 μg/m³ and PM_{10}: 196 ± 90 μg/m³), Agra ($PM_{2.5}$: 144 ± 79 μg/m³) and Delhi ($PM_{2.5}$: 117 ± 79 μg/m³ and PM_{10}: 191 ± 127 μg/m³) during the year 2014 which may be due to the rapid industrialization and other anthropogenic activities. However, the PMs concentration (both PM_{10}, and $PM_{2.5}$) in the Indo-Himalayan region found within the NAAQS limit with value at Kashmir ($PM_{2.5}$: 20 ± 13 μg/m³; PM_{10}: 32 ± 28 μg/m³), Kullu ($PM_{2.5}$: 31 ± 17 μg/m³, PM_{10}: 49 ± 16 μg/m³), Nainital (PM_{10}: 42 ± 24 μg/m³), Dehradun ($PM_{2.5}$: 53 ± 38 μg/m³) and Darjeeling ($PM_{2.5}$: 24 ± 14 μg/m³, PM_{10}: 49 ± 18 μg/m³). In addition, they have also reported a decrease in $PM_{2.5}$ particles mass concentration (μg/m³) from 2014 to 2015 over Patiala ($PM_{2.5}$: 93 ± 35), Varanasi, Kolkata, and Delhi while enhancement in particulate over Lucknow and Agra.

The temporal variation in particulate matter concentration along with their ratio ($PM_{2.5}/PM_{10}$) shows a large variability (Fig. 4) during the study period, suggesting the significant role of meteorological condition and heterogeneity in emission sources in particulate loadings. Earlier studies also reported significant day-to-day variability in particulate matter loading during winter season over Varanasi (Murari et al. 2016; Mukherjee and Toohey 2016; Kumar et al. 2015b). Figure also reflects nearly similar day to day variation of PM_{10}, $PM_{2.5}$, and $PM_{10-2.5}$ concentration, suggesting the similarity in their emission sources which is further confirmed by their significant correlation coefficient ($r = 0.75$ for PM_{10}, $r = 0.81$ for $PM_{2.5}$). On few days, peak in particulate matter loading is associated with a relatively lower value of $PM_{2.5}/PM_{10}$ ratio suggesting the existence of polluted dust over the region. The evidence of polluted dust is also corroborated from the spaceborne Lidar i.e. Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation: CALIPSO,

Table 2 Comparison of PM mass concentration recorded in this study with other previous studies over IGB and other locations in India

| Study Locations	PM_{10} ($\mu g/m^3$)	$PM_{2.5}$ ($\mu g/m^3$)	$PM_{2.5}/PM_{10}$	Site Description	Sampling Period (24 h)	Reference
Varanasi	213±80	134±48	0.62	Semi– Urban	Nov 2016 – Feb. 2017	Present Study
Kashmir	32.0±27.6	20.3±13.1	0.63	Hilly area	Winter 2015	Sen et al. (2017)
Kullu	48.5±16.2	30.8±17.2	0.63	Hilly area	Winter 2015	Sen et al. (2017)
Patiala	140	NA	NA	Urban	17–21 October 2017	Bansal et al. (2019)
Delhi	208±14	123±87	0.59	Urban	2008 to 2011	Guttikonda and Calori (2013)
Delhi	191.6±58.1	125.7±56.6	0.65	Urban	Winter 2015	Sen et al. (2017)
Agra	230.5	121.2	0.52	Semi-Urban	2010 to 2011	Pipal et al. (2014)
Agra	214.6	NA	NA	Urban	March 2016 to March 2017	Sah et al. (2019)
Lucknow	195.9±90.3	130.5±73.0	0.66	Urban	Winter 2015	Sen et al. (2017)
Varanasi	161.3±110.4	81.78±66.4	0.50	Semi – Urban	Jan. 2014 –Dec. 2014	Murari et al. (2017)
Jamshedpur	500	NA	NA	Urban	21–26 October 2014	Ambade (2018)
Raipur	NA	150.9±75.6	NA	Semi -Urban	July 2009–June 2010	Deshmukh et al. (2010)
Darjeeling	47.8±18.1	24.3±13.5	0.50	Hilly area	Winter 2015	Sen et al. (2017)
Chennai	135	73	0.54	Urban	Jan 2008- Feb 2008	Bathmanabhan et al. (2010)
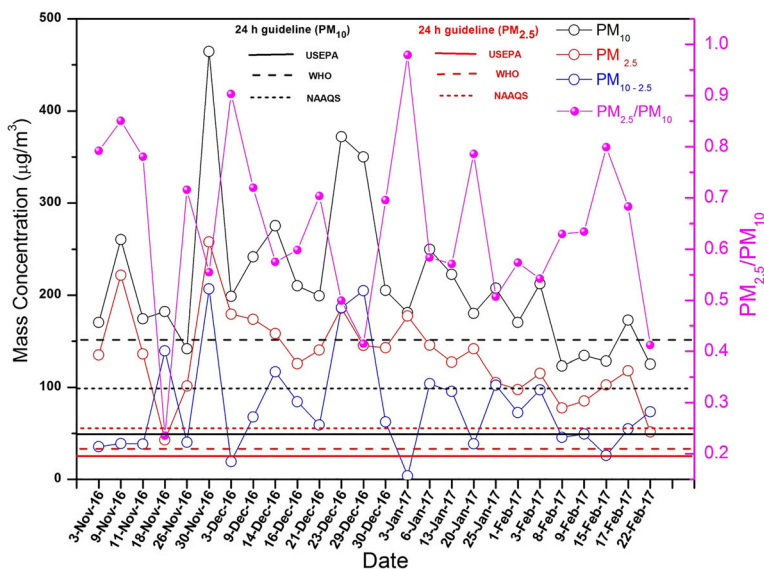
Fig. 4 Temporal variation of Particulate Matter (PM_{10}, $PM_{2.5}$ and $PM_{10-2.5}$) and their ratio ($PM_{2.5}/PM_{10}$) during the study period

which shows an elevated dense layer (up to 1–3 km) of polluted dust over Varanasi and other nearby places of IGB region (Fig. S1). Recently, Kumar et al. (2015b) also reported the long-range transportation of polluted dust at higher altitudes over Varanasi during the winter season. Similar to the particulate variation, their ratio also shows a large day-to-day variation in the range of 0.23–0.98 with a mean value 0.65 during the study period, suggesting the dominance of fine mode aerosol particles which are mainly attributed from the anthropogenic emission. Higher concentration of fine mode aerosol particles during the winter season is also reported at different locations in IGB (Satsangi et al. 2016; Kumar et al. 2015a; Tiwari et al. 2015; Pipal et al. 2014) and other places of the world (Kang et al. 2016; Rupakheti et al. 2019).

3.2 Characteristics of ionic species in the particulate matter

The chemical composition of particulate matter is characterized by the major anion (Cl^-, NO_3^-, SO_4^{--}) and cations (Na^+, K^+, Ca^{2+}, Mg^{2+}). The mass concentration of major ions are found in the order of $SO_4^{2-} > Cl^- > NO_3^- > Na^+ > Mg^{2+} > K^+ > Ca^{2+}$ and $SO_4^{2-} > NO_3^- > Cl^- > Na^+ > K^+ > Mg^{2+} > Ca^{2+}$ for $PM_{2.5}$ and PM_{10} respectively suggesting the anions as the major components with nearly 62% and 77% contribution of total ionic species mass concentration in $PM_{2.5}$ and PM_{10} samples respectively. The average mass concentrations of major ionic species in PM_{10} and $PM_{2.5}$ samples are summarized in Table 3 and their percentage contribution is shown in Fig. 5. The ratio Cl^-/Na^+ are found in the range of 0.49–4.63 and 0.55–3.90 with average value 1.62 and 1.73 for PM_{10} and $PM_{2.5}$ samples respectively which are often different than the sea water ratio (1.8) during the study period, suggesting a relatively lesser influence of sea salt in particulate loading during the study period. A poor correlation (~ 0.04) between Cl^- and Na^+ indicate towards their dissimilarities in their emission sources or depletion of Cl^- in the presence of specific acidic species like SO_4^{--}, NO_3^{--} (Wang et al. 2015; Thurston et al.

Table 3 Statistical analysis of major ionic species concentration in PM_{10} and $PM_{2.5}$ samples over Varanasi

Species	PM_{10} ($\mu g/m^3$)				$PM_{2.5}$ ($\mu g/m^3$)			
	Min	Max	Mean	STD	Min	Max	Mean	STD
SO_4^{2-}	8.9	55.7	27.5	13.5	4.6	33.4	16.9	8.6
Cl^-	10.68	20.6	16.2	3.2	4.8	21.5	10.5	5.0
NO_3^{2-}	4.4	42.4	20	12.1	4.4	15.9	6.5	4.4
Na^+	3.6	14.7	6.8	9.4	2.4	13.1	6.7	2.4
Mg^{2+}	2.1	9.2	5	2.5	0.1	0.9	0.3	0.19
K^+	3.2	14.9	8.2	3.6	1.3	12.9	3.5	2.6
Ca^{2+}	1.1	10.7	4.1	3.2	1.1	5.3	2.4	0.9

2011). Earlier studies also reported negligible contribution of sea salt aerosol during winter season over Varanasi (Tiwari et al. 2016; Murari et al. 2016; Kumar et al. 2015b) as well as at different locations in IGB (Goel et al. 2018; Satsangi et al. 2016; Pipal et al. 2014; Thurston et al. 2011). However, the ratio of Ca^{2+}/Na^+, Mg^{2+}/Na^+ is found lower than seawater ratio and SO_4^{--}/Na^+ is found higher while Cl^-/Mg^{2+} and Na^+/Mg^{2+} are found to very close to corresponding local soil (Satsangi et al. 2016), indicating that these species are mainly from the soil-derived, anthropogenic and local terrestrial emission. The non-sea salt (nss) components of water-soluble ions are calculated using Na^+ reference element of sea salt concentration (Lee and Hieu 2013; Duce et al. 1983). The mass concentration of nss-component SO_4^{--}, K^+ and Ca^+ are found in the range of 75% - 96% of their total mass which further confirmed the insignificant marine influence. The major sources of nss-SO_4^{--} are coal combustion, fossil fuel, biomass burning, vehicular emission and oxidation of SO_2 (Sharma et al. 2010; Ram et al. 2012; Ram **et al**. 2010; Pratap et al. 2017). One of the other possible sources of anions may be the emission from the surroundings 'Saree" and Carpet industries which emit a significant amount of chemicals into the environment. K^+ is considered as biomass tracer (Rajput et al. 2016, 2014; Ram et al. 2012) and a relatively higher concentration of K^+ in November is mainly attributed because of heavy fireworks during the Diwali festival and massive biomass/crop residue burning. Recently, Sarkar et al. (2018) also reported an extensive crop residue burning over the entire IGB during the post-monsoon season (October – November) which enhances the concentration of fine mode particles in a manifold.
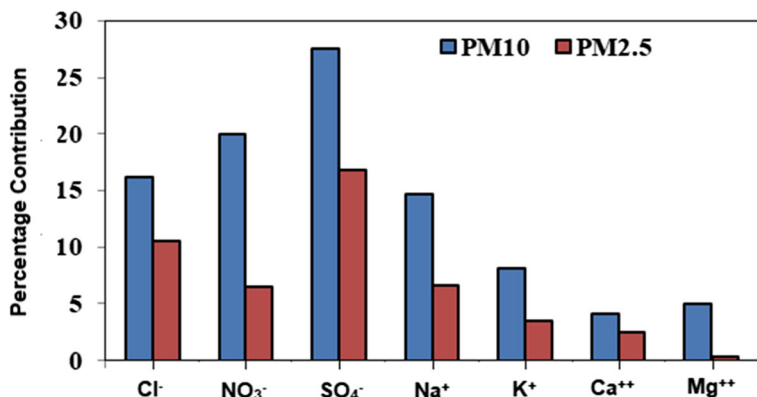

Fig. 5 The percentage contribution of major ions in PM_{10} and $PM_{2.5}$ samples

Table 4 Weight % (± standard deviation) of the elements quantified in $PM_{2.5}$ and PM_{10} sample during study period at receptor site

Elements	PM_{10} (weight %)	$PM_{2.5}$ (weight %)
C	38.30 ± 6.57	38.35 ± 21.81
O	34.72 ± 6.00	12.26 ± 9.83
Fe	0.06 ± 0.18	NA
Al	2.28 ± 0.74	2.55 ± 6.21
Si	11.97 ± 4.15	0.98 ± 2.79
S	1.51 ± 1.12	0.62 ± 0.71
Cl	0.46 ± 0.25	0.52 ± 0.51
K	2.1 ± 0.80	0.45 ± 0.69
F	1.96 ± 7.8	23.66 ± 16.20
N	0.38 ± 1.05	1.59 ± 3.40
Zn	0.58 ± 1.06	NA
Mg	0.06 ± 0.12	0.04 ± 0.18
Na	4.08 ± 1.25	0.46 ± 1.16
Ca	0.79 ± 1.11	NA
Ba	0.90 ± 1.25	NA

3.3 Elemental composition of particulate matter

The elemental composition of the particulate matter is also other important parameters that affect the scattering properties of the particulates, heterogeneous chemical reactions and rate of water uptake (Li et al. 2016). The weight percentage of different elements measured in the PM_{10} and $PM_{2.5}$ samples during the study period are summarized in Table 4. The average concentration of crustal elements in PM_{10} and $PM_{2.5}$ samples are found in order of $C > O > Si > Na > Al > K > S > F > Ba > Ca > Zn > Cl > N > Fe > Mg$ and $C > F > O > Al > N > Si > S > Cl > Na > K > Mg$ respectively according to their weight percentage. Results suggest the highest contribution (in weight %) of carbon (Fig. 6) in both PM_{10} and $PM_{2.5}$ (38%) attributed to the dominance of anthropogenic sources like biomass/crop residue burning and traffic derived emission. Fluorine (F) is found to be the second highest contributor in $PM_{2.5}$ particles suggesting the significant impact of the atmospheric pollution from continental crust and soil (Teotia and Teotia 1994). The contribution of S, Al, and Na are also found in a comparatively significant amount which is mainly emitted from anthropogenic activities like industrial activities and vehicular emission (Liu et al. 2019). The presence of Si is found mostly due
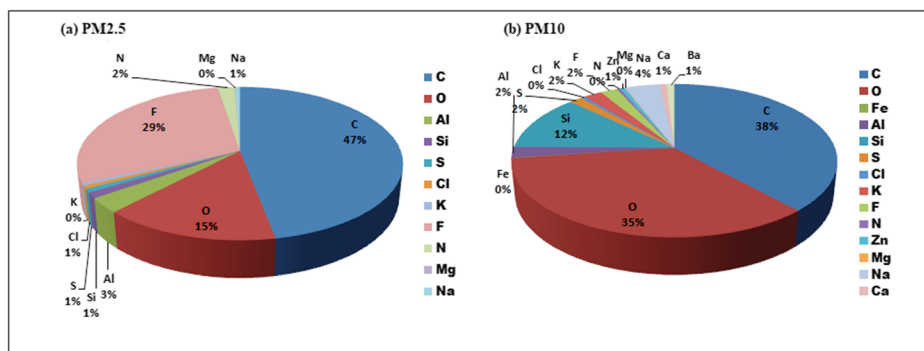
Fig. 6 Percentage (%) elemental composition particulate samples for (a) $PM_{2.5}$ and (b) PM_{10}

to transported dust and has a relatively higher contribution in PM_{10} samples, however, the presence of K is mostly attributed to biomass burning (Pachon et al. 2013; Pipal et al. 2014).

3.4 Source identification of particulate matter and their transportation

In order to identify the potential emission sources and transported pathways of the particulate matter arriving at the sampling site, five days air mass backward trajectories on sampling days are computed at three different altitudes i.e. 500 m 1000 m and 1500 m using NOAA HYSPLIT model. The computed trajectories are superimposed over MODIS derived fire hot spot during study period (Fig. 7). Figure 7 reveals that most of the pollutants are mainly coming either from Northern India (which have a relatively higher density of fire hot spots) through the transportation or from local areas around Varanasi (mostly at lower altitude i.e. 500 m), indicating the dominance of anthropogenic emission sources. Earlier studies also reported the dominance of fine mode particles from the anthropogenic emission sources at Allahabad (Ram et al. 2012) and at Kanpur (Rajput et al. 2014) which are nearly 120 and 250 km towards west from the sampling site. On the other hand, on some epoch, the air masses at higher altitudes are reached at receptor site from the desert region passing through Northern India suggesting the mixing of dust with smoke resulting to polluted dust. The evidence of
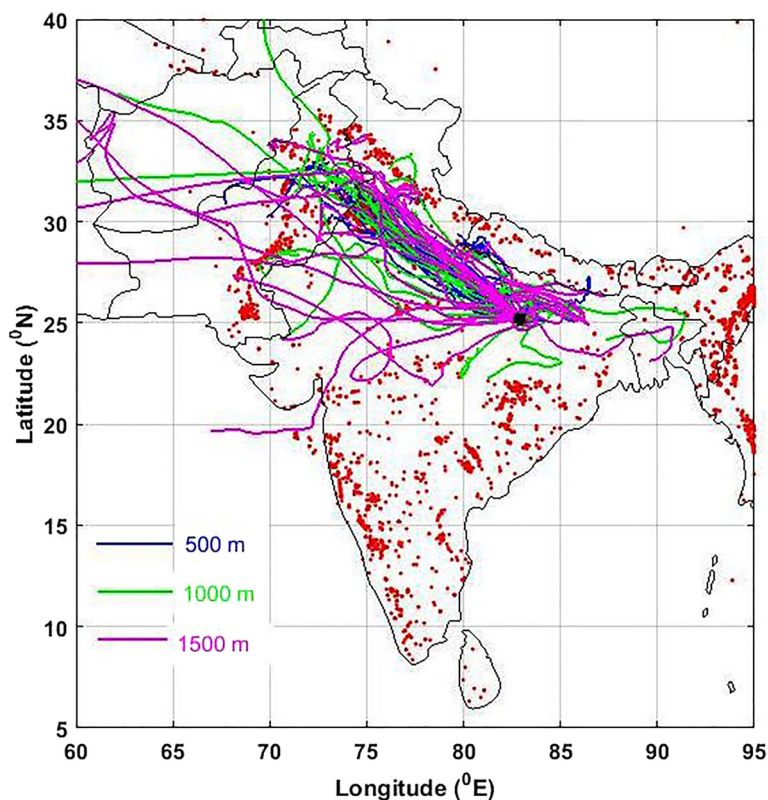

Fig. 7 Spatial variability of biomass fire hot spot during November 2016 to February 2017 superimposed with five days airmass back trajectories on sampling dates at three different altitudes i.e. 500 m (blue), 1000 m (green) and 1500 m (magenta). The black dot represents the receptor site

polluted dust over the receptor site is also corroborated with the lower value of $PM_{2.5}/PM_{10}$ ratio (Fig. 4) and further confirmed by the CALIPSO images (Fig. S1). Recently, Kumar et al. (2015a) also found the presence of long-range transported dust over Varanasi during the winter season.

4 Conclusion

The insitu measurement of PMs samples has been carried out from November 2016 to February 2017 over Varanasi, a semi-urban site and analyzed to investigate their chemical composition. The daily mean mass concentration of PMs ($PM_{2.5}$: 134 ± 48 μg/m^3 and PM_{10}: 213 ± 80 μg/m^3) is found two to three times higher compared to established safe limits from the different national/international agencies. The observed mean concentration is associated with a relatively higher value of $PM_{2.5}/PM_{10}$ ratio (0.65) with a wide range (0.23–0.98) suggesting the mixing of fine mode aerosol with smoke. On a few days, an elevated layer (up to 1–3 km) of polluted dust is observed because of the mixing of long-range transported dust and smoke from the biomass/crop residue burning. The different WSIIS (i.e. SO_4^{2-}, Cl^-, K^+, NO_3^-, Na^+, Ca^{2+}, Mg^{2+}) is detected in PMs samples with the dominance of SO_4^{2-}, NO_3^- and Cl^- ions in both types of samples which contribute more than 50% of the total concentration of particulates. However, elemental analysis suggests the dominance of carbonaceous aerosols as well as trace and transitions elements that are mainly emitted from the biomass/crop residue burning and other anthropogenic emission and reached at receptor site through the long-range transportation.

References

Ali, M.U., Liu, G., Yousaf, B., Ullah, H., Irshad, S., Ahmed, R., Hussain, M., Rashid, A.: Evaluation of floor-wise pollution status and deposition behavior of potentially toxic elements and nanoparticles in air conditioner dust during urbanistic development. J. Hazard. Mater. **365**, 186–195 (2019)

AlKheder, S., AlKandari, A.: The impact of dust on Kuwait international airport operations: a case study. Int. J. Environ. Sci. Technol., https://doi.org/10.1007/s13762-020-02710-3 (2020), 2020

Ambade, B.: The air pollution during Diwali festival by the burning of fireworks in Jamshedpur city, India. Urban Clim. **26**, 149–160 (2018)

Ancelet, T., Davy, P.K., Trompetter, W.J.: Hourly variations in particulate matter source contributions in New Zealand's southernmost city. Air Quality and Climate Change. **49**(3), 26 (2015)

Bansal, O., Singh, A., Singh, D.: Short-term perturbation in aerosol characteristics over northwestern India: a case study during Diwali festival. Journal of Earth System Science. **128**(7), 193 (2019)

Bonasoni, P., Laj, P., Marinoni, A., Sprenger, M., Angelini, F., Arduini, J., Bonafè, U., Calzolari, F., Colombo, T., Decesari, S., di Biagio, C., Disarra, A.G., Evangelisti, F., Duchi, R., Facchini, M.C., Fuzzi, S., GobbiGP, M.M., Panday, A., Roccato, F., Sellegri, K., Venzac, H., VerzaGP, V.P., Vuillermoz, E., Cristofanelli, P.: Atmosphericbrown clouds in the Himalayas: first two years of continuous observations at the Nepal Climate Observatory-Pyramid (5079 m). Atmos Chem Phys. **10**, 7515–7531 (2010)

Burnett, R.T., Pope III, C.A., Ezzati, M., Olives, C., Lim, S.S., Mehta, S., Shin, H.H., Singh, G., Hubbell, B., Brauer, M., Anderson, H.R.: An integrated risk function for estimating the global burden of disease attributable to ambient fine particulate matter exposure. Environ. Health Perspect. **122**(4), 397–403 (2014)

Census India (2011) http://censusindia.gov.in/2011-Common/CensusData2011.html

Chernyshev, V.V., Zakharenko, A.M., Ugay, S.M., Hien, T.T., Hai, L.H., Olesik, S.M., Kholodov, A.S., Zubko, E., Kokkinakis, M., Burykina, T.I., Stratidakis, A.K.: Morphological and chemical composition of particulate matter in buses exhaust. Toxicol. Rep. **6**, 120–125 (2019)

China, S., Mazzoleni, C., Gorkowski, K., Aiken, A.C., Dubey, M.K.: Morphology and mixing state of individual freshly emitted wildfire carbonaceous particles. Nat. Commun. **4**, 2122 (2013)

Chowdhury, S., Dey, S.: Cause-specific premature death from ambient PM2.5 exposure in India: estimate adjusted for baseline mortality. Environ. Int. **91**, 283–290 (2016)

Cohen, A.J., Brauer, M., Burnett, R., Anderson, H.R., Frostad, J., Estep, K., Balakrishnan, K., Brunekreef, B., Dandona, L., Dandona, R., Feigin, V., Freedman, G., Hubbell, B., Jobling, A., Kan, H., Knibbs, L., Liu, Y., Martin, R., Morawska, L., Pope III, C.A., Shin, H., Straif, K., Shaddick, G., Thomas, M., van Dingenen, R., van Donkelaar, A., Vos, T., Murray, C.J.L., Forouzanfar, M.H.: Estimates and 25-year trends of the global burden of disease attributable to ambient air pollution: an analysis of data from the global burden of diseases study 2015. Lancet. **389**, 1907–1918 (2017)

Draxler, R.R., Rolph, G.D.: HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory). NOAA Air Resources Laboratory, College Park, MD (2003)

Duce, R.A.,Arimoto, R., Ray, B.J.,Unni, C.K., and Harder, P.J.: Atmospheric trace elements at Enewetak Atoll: 1. Concentrations, sources, and temporal variability: Journal of Geophysical Research. Oceans, 88(C9), 5321–5342 (1983)

Fan, J., Wang, Y., Rosenfeld, D., Liu, X.: Review of aerosol–cloud interactions: mechanisms, significance, and challenges. J. Atmos. Sci. **73**(11), 4221–4252 (2016)

Gautam, R., Hsu, N.C., Tsay, S.C., Lau, K.M., Holben, B., Bell, S., Smirnov, A., Li, C., Hansell, R., Ji, Q., Payra, S., Aryal, D., Kayastha, R., Kim, K.M.: Accumulation of aerosols over the indo-Gangetic Plains and southern slopes of the Himalayas: distribution, properties and radiative effects during the 2009 pre-monsoon season. Atmos. Chem. Phys. **11**, 12841–12863 (2011)

Ghude, S.D.,Chate, D.M., Jena, C.Beig, G. Kumar, R. Barth, M.C. Pfister, G.G.Fadnavis, S. and Pithani, P.: Premature mortality in India due to PM2.5 and ozone exposure: Geophysical Research Letters, 43(9), 4650–4658 (2016)

Goel, V., Mishra, S.K., Ahlawat, A., Sharma, C., Vijayan, N., Radhakrishnan, S.R., Dimri, A.P., Kotnala, R.K.: Effect of reduced traffic density on characteristics of particulate matter over Delhi. Curr. Sci. **115**(2), 315 (2018)

Guttikunda, S.K.: And Calori, G.:a GIS based emissions inventory at 1 km× 1 km spatial resolution for air pollution analysis in Delhi, India. Atmos. Environ. **67**, 101–111 (2013)

IPCC, 2013. IPCC Fifth Assessment Report, Climatic Change: The Physical Science Basis. The Intergovernmental Panel on Climate Change (2013). https://doi.org/10.1017/CBO9781107415324

Kang, N., Kumar, K.R., Yu, X., Yin, Y.: Column-integrated aerosol optical properties and direct radiative forcing over the urban-industrial megacity Nanjing in the Yangtze River Delta, China. Environmental Science and Pollution Research. **23**(17), 17532–17552 (2016)

Kaskaoutis, D.G., Kumar, S., Sharma, D., Singh, R.P., Kharol, S.K., Sharma, M., Singh, A.K., Singh, S., Singh, A., Singh, D.: Effects of crop residue burning on aerosol properties, plume characteristics, and long-range transport over northern India. Journal of Geophysical Research: Atmospheres. **119**(9), 5424–5444 (2014)

Kaufman, Y.J., Tanré, D., Remer, L.A., Vermote, E.F., Chu, A., Holben, B.N.: Operational remote sensing of tropospheric aerosol over land from EOS moderate resolution imaging spectroradiometer. J. Geophys. Res. **102**(D14), 17051–17067 (1997)

Kulkarni, J.R., Maheskumar, R.S., Morwal, S.B., Padmakumari, B., Konwar, M., Deshpande, C.G., Joshi, R.R., Bhalwankar, R.V., Pandithurai, G., Safai, P.D., Narkhedkar, S.G.: The cloud aerosol interactions and precipitation enhancement experiment (CAIPEEX): overview and preliminary results. Curr. Sci. **102**(3), 413–425 (2012)

Kumar, M., Singh, R.K., Murari, V., Singh, A.K., Singh, R.S., Banerjee, T.: Fireworks induced particle pollution: a spatio-temporal analysis. Atmos. Res. **180**, 78–91 (2016)

Kumar, M., Tiwari, S., Murari, V., Singh, A.K., Banerjee, T.: Wintertime characteristics of aerosols at middle indo-Gangetic plain: impacts of regional meteorology and long-range transport. Atmos. Environ. **104**, 162–175 (2015a)

Kumar, S., Kumar, S., Kaskaoutis, D.G., Singh, R.P., Singh, R.K., Mishra, A.K., Srivastava, M.K., Singh, A.K.: Meteorological, atmospheric and climatic perturbations during major dust storms over indo-Gangetic Basin. Aeolian Res. **17**, 15–31 (2015b)

Lau, K.M., and Kim, K.M.: Observational relationships between aerosol and Asian monsoon rainfall, and circulation. Geophysical Research Letters, 33(21) (2006)

Lee, B.K., Hieu, N.T.: Seasonal ion characteristics of fine and coarse particles from an urban residential area in a typical industrial city. Atmos. Res. **122**, 362–377 (2013)

Levy, R.C., Remer, L.A., Kleidman, R.G., Mattoo, S., Ichoku, C., Kahn, R., Eck, T.F.: Global evaluation of the Collection 5 MODIS dark-target aerosol products over land. Atmos. Chem. Phys. **10**, 10399–10420 (2010)

Li, G., Su, H., Li, X., Kuhn, U., Meusel, H., Hoffmann, T., Ammann, M., Pöschl, U., Shao, M., Cheng, Y.: Uptake of gaseous formaldehyde by soil surfaces: a combination of adsorption/desorption equilibrium and chemical reactions. Atmos. Chem. Phys. **16**(15), 10299–10311 (2016)

Liu, K., Wang, F., Li, J., Tiwari, S., and Chen, B. Assessment of trends and emission sources of heavy metals from the soil sediments near the Bohai Bay. Environmental Science and Pollution Research, 1–15 (2019)

Mishra, A.K., Shibata, T.: Climatological aspects of seasonal variation of aerosol vertical distribution over central indo-Gangetic belt (IGB) inferred by the space borne lidar CALIOP. Atmos. Environ. **46**, 365–375 (2012)

Mukherjee, A., Toohey, D.W.: A study of aerosol properties based on observations of particulate matter from the US Embassy in Beijing, China. Earth's Future. **4**(8), 381–395 (2016)

Murari, V., Kumar, M., Singh, N., Singh, R.S., Banerjee, T.: Particulate morphology and elemental characteristics: variability at middle Indo-Gangetic plain. J. Atmos. Chem. **73**(2), 165–179 (2016)

Ojha, N., Sharma, A., Kumar, M., Girach, I., Ansari, T. U., Sharma, S. K., Singh, N., Pozzer, A., Gunthe, S. S.: On the widespread enhancement in fine particulate matter across theIndo-Gangetic Plain towards winter. Scientific Reports, 10:5862 https://doi.org/10.1038/s41598-020-62710-8 (2020)

Onat, B., Şahin, Ü.A., Bayat, C.: Assessment of particulate matter in the urban atmosphere: size distribution, metal composition and source characterization using principal component analysis. J. Environ. Monit. **14**(5), 1400–1409 (2012)

Pachon, J.E., Weber, R.J., Zhang, X., Mulholland, J.A., Russell, A.G.: Revising the use of potassium (K) in the source apportionment of PM2. 5. Atmospheric Pollution Research. **4**(1), 14–21 (2013)

Pani, S.K., Chantara, S., Khamkaew, C., Lee, C.T., Lin, N.H.: Biomass burning in the northern peninsular Southeast Asia: aerosol chemical profile and potential exposure. Atmos. Res. **224**, 180–195 (2019)

Penner, J.E., Xu, L., Wang, M.: Satellite methods underestimate indirect climate forcing by aerosols. Proc. Natl. Acad. Sci. **108**(33), 13404–13408 (2011)

Pipal, A.S., Kulshrestha, A., Taneja, A.: Characterization and morphological analysis of airborne PM2. 5 and PM10 in Agra located in north central India. Atmospheric environment. **45**(21), 3621–3630 (2011)

Pipal, A.S., Jan, R., Satsangi, P.G., Tiwari, S,. and Taneja, A .: Study of surface morphology, elemental composition and origin of atmospheric aerosols (PM2.5 and PM10) over Agra, India. Aerosol and Air Quality Research, 14(6), 1685–1700 (2014)

Pratap V., Kumar A., Singh, A.K .: Short Term Air Quality Degradation by Firecrackers Used during Diwali Festival in Varanasi, India: 18158–18165 (2017). https://doi.org/10.15680/IJIRSET.2017.0609079

Prijith, S.S., Rajeev, K., Thampi, B.V., Nair, S.K., Mohan, M.: Multi-year observations of the spatial and vertical distribution of aerosols and the genesis of abnormal variations in aerosol loading over the Arabian Sea during Asian summer monsoon season. J. Atmos. Sol. Terr. Phys. **105**, 142–151 (2013)

Rajput, P., Gupta, T., Kumar, A.: The diurnal variability of sulfate and nitrate aerosols during wintertime in the indo-Gangetic plain: implications for heterogeneous phase chemistry. RSC Adv. **6**(92), 89879–89887 (2016)

Rajput, P., Sarin, M.M., Sharma, D., Singh, D.: Atmospheric polycyclic aromatic hydrocarbons and isomer ratios as tracers of biomass burning emissions in northern India. Environ. Sci. Pollut. Res. **21**(8), 5724–5729 (2014)

Ram, K., Sarin, M.M., and Tripathi, S.N.: A 1 year record of carbonaceous aerosols from an urban site in the Indo Gangetic Plain: Characterization, sources, and temporal variability. Journal of Geophysical Research: Atmospheres, 115 (D24) (2010)

Ram, K., Sarin, M.M., and Tripathi, S.N.: Temporal trends in atmospheric PM2.5, PM10, elemental carbon, organic carbon, water-soluble organic carbon, and optical properties: impact of biomass burning emissions in the Indo-Gangetic Plain. Environmental science & technology, 46(2), 686–695 (2012)

Rastogi, N., Singh, A., Singh, D., Sarin, M.M.: Chemical characteristics of PM2.5 at a source region of biomass burning emissions: evidence for secondary aerosol formation. Environ. Pollut. **184**, 563–569 (2014)

Remer, L.A., Kaufman, Y.J., Tanré, D., Mattoo, S., Chu, D.A., Martins, J.V., Li, R.R., Ichoku, C., Levy, R.C., Kleidman, R.G., Eck, T.F.: The MODIS aerosol algorithm, products, and validation. J. Atmos. Sci. **62**(4), 947–973 (2005)

Roy, A., Chatterjee, A., Tiwari, S., Sarkar, C., Das, S.K., Ghosh, S.K., Raha, S.: Precipitation chemistry over urban, rural and high altitude Himalayan stations in eastern India. Atmos. Res. **181**, 44–53 (2016)

Rupakheti, D., Kang, S., Rupakheti, M., Cong, Z., Panday, A.K., Holben, B.N.: Identification of absorbing aerosol types at a site in the northern edge of indo-Gangetic plain and a polluted valley in the foothills of the Central Himalayas. Atmos. Res. **223**, 15–23 (2019)

Sah, D., Verma, P.K., Kandikonda, M.K., Lakhani, A.: Pollution characteristics, human health risk through multiple exposure pathways, and source apportionment of heavy metals in PM_{10} at indo-Gangetic site. Urban Clim. 27, 149–162 (2019)

Sarkar, S., Singh, R.P., Chauhan, A.: Crop residue burning in northern India: increasing threat to greater India. Journal of Geophysical Research: Atmospheres. 123(13), 6920–6934 (2018)

Satsangi, A., Pachauri, T., Singla, V., Lakhani, A., Kumari, K.M.: Water soluble ionic species in atmospheric aerosols: concentrations and sources at Agra in the indo-Gangetic plain (IGP). Aerosol Air Qual. Res. 13(6), 1877–1889 (2013)

Satsangi, P.G., Pipal, A.S., Budhavant, K.B., Rao, P.S.P., Taneja, A.: Study of chemical species associated with fine particles and their secondary particle formation at semi-arid region of India. Atmospheric Pollution Research. 7(6), 1110–1118 (2016)

Seinfeld, J.H., Bretherton, C., Carslaw, K.S., Coe, H., DeMott, P.J., Dunlea, E.J., Feingold, G., Ghan, S., Guenther, A.B., Kahn, R., Kraucunas, I.: Improving our fundamental understanding of the role of aerosol−cloud interactions in the climate system. Proc. Natl. Acad. Sci. 113(21), 5781–5790 (2016)

Sen, A., Abdelmaksoud, A.S., Ahammed, Y.N., Banerjee, T., Bhat, M.A., Chatterjee, A., Choudhuri, A.K., Das, T., Dhir, A., Dhyani, P.P., Gadi, R.: Variations in particulate matter over indo-Gangetic Plains and indo-Himalayan range during four field campaigns in winter monsoon and summer monsoon: role of pollution pathways. Atmos. Environ. 154, 200–224 (2017)

Shah, V., Jaeglé, L., Thornton, J.A., Lopez-Hilfiker, F.D., Lee, B.H., Schroder, J.C., Campuzano-Jost, P., Jimenez, J.L., Guo, H., Sullivan, A.P., Weber, R.J.: Chemical feedbacks weaken the wintertime response of particulate sulfate and nitrate to emissions reductions over the eastern United States. Proc. Natl. Acad. Sci. 115(32), 8110–8115 (2018)

Sharma, S.K., Datta, A., Saud, T., Saxena, M., Mandal, T.K., Ahammed, Y.N., Arya, B.C.: Seasonal variability of ambient NH3, NO, NO2 and SO2 over Delhi. J. Environ. Sci. 22(7), 1023–1028 (2010)

Singh, A., Rastogi, N., Patel, A., Satish, R.V., Singh, D.: Size-segregated characteristics of carbonaceous aerosols over the northwestern indo-gangetic plain: year-round temporal behavior. Aerosol Air Qual. Res. 16(7), 1615–1624 (2016a)

Singh, A., Tiwari, S., Sharma, D., Singh, D., Tiwari, S., Srivastava, A.K., Rastogi, N., Singh, A.K.: Characterization and radiative impact of dust aerosols over northwestern part of India: a case study during a severe dust storm. Meteorog. Atmos. Phys. 128, 779–792 (2016b)

Singh, A.K., Srivastava, M.K., Singh, M., Srivastava, A.K., Sravan, K., Tiwari, S., Singh, B.P., Bisht, D.S.: Characterization of atmospheric aerosol by SEM-EDX and ion-chromatography techniques for eastern indo-Gangetic plain location, Varanasi. India. International Journal of Advances in Earth Sciences. 3, 41–51 (2014)

Singh, S., Naseema Beegum, S.: Direct radiative effects of an unseasonal dust storm at a western indo Gangetic plain station Delhi in ultraviolet, shortwave, and longwave regions. Geophys. Res. Lett. 40(10), 2444–2449 (2013)

Smith, C.L., Moores, J.E., Lemmon, M., Guzewich, S.D., Moore, C.A., Ellison, D., Khayat, A.S.J.: Visibility and line-of-sight extinction estimates in Gale crater during the 2018/MY34 global dust storm. Geophys. Res. Lett. (2019). https://doi.org/10.1029/2019GL083788

Srivastava, A.K., Bisht, D.S., Ram, K., Tiwari, S., Srivastava, M.K.: Characterization of carbonaceous aerosols over Delhi in ganga basin: seasonal variability and possible sources. Environ. Sci. Pollut. Res. 21(14), 8610–8619 (2014)

Sweerts, B., Pfenninger, S., Yang, S., Folini, D., van der Zwaan, B., Wild, M.: Estimation of losses in solar energy production from air pollution in China since 1960 using surface radiation data. Nat. Energy. 4, 657–663 (2019)

Taneja, K., Ahmad, S., Ahmed, K., Attri, S.D.: Impact assessment of a severe dust storm on atmospheric aerosols over an urban site in India. Curr. Sci. 118(5), 737–749 (2020)

Tare, V., Tripathi, S.N., Chinnam, N., Srivastava, A.K., Dey, S., Manar, M., Kanawade, V.P., Agarwal, A., Kishore, S., Lal, R.B., and Sharma, M. : Measurements of atmospheric parameters during Indian Space Research Organization Geosphere Biosphere Program Land Campaign II at a typical location in the Ganga Basin: 2: chemical properties. Journal of Geophysical Research: Atmospheres, 111(D23) (2006)

Teotia, S.P.S., Teotia, M.: Dental caries: a disorder of high fluoride and low dietary calcium interactions (30 years of personal research). Fluoride. 27, 59–59 (1994)

Thurston, G.D., Ito, K., Lall, R.: A source apportionment of US fine particulate matter air pollution. Atmos. Environ. 45(24), 3924–3936 (2011)

Tiwari, S., Kun, L., Chen, B.: Spatial Variability of Carbonaceous Aerosol over the South Yellow Sea, China: Impact of anthropogenic emission and long-range transportation. Environmental Science and Pollution Research. (2020). https://doi.org/10.1007/s11356-020-08686-4

Tiwari, S., Kumar, A., Pratap, V., Singh, A.K.: Assessment of two intense dust storm characteristics over indo–Gangetic basin and their radiative impacts: a case study. Atmos. Res. **228**, 23–40 (2019)

Tiwari, S., Kaskaoutis, D., Soni, V.K., Attri, S.D., Singh, A.K.: Aerosol columnar characteristics and their heterogeneous nature over Varanasi, in the Central Ganges valley. Environ. Sci. Pollut. Res. **25**(25), 24726–24745 (2018)

Tiwari, S., Dumka, U.C., Kaskaoutis, D.G., Ram, K., Panicker, A.S., Srivastava, M.K., Tiwari, S., Attri, S.D., Soni, V.K., Pandey, A.K.: Aerosol chemical characterization and role of carbonaceous aerosol on radiative effect over Varanasi in central indo-Gangetic plain. Atmos. Environ. **125**, 437–449 (2016)

Tiwari, S., Hopke, P.K., Pipal, A.S., Srivastava, A.K., Bisht, D.S., Tiwari, S., Singh, A.K., Soni, V.K., Attri, S.D.: Intra-urban variability of particulate matter ($PM_{2.5}$ and PM_{10}) and its relationship with optical properties of aerosols over Delhi, India. Atmos. Res. **166**, 223–232 (2015)

Tiwari, S., Singh, A.K.: Variability of aerosol parameters derived from ground and satellite measurements over Varanasi located in the indo-Gangetic Basin. Aerosol Air Qual. Res. **13**, 627–638 (2013)

Tiwari, S., Srivastava, A.K., Bisht, D.S., Bano, T., Singh, S., Behura, S., Srivastava, M.K., Chate, D.M., Padmanabhamurty, B.: Black carbon and chemical characteristics of PM 10 and PM 2.5 at an urban site of North India. Journal of Atmospheric Chemistry. **62**(3), 193–209 (2009)

Tsai, D.H., Amyai, N., Marques-Vidal, P., Wang, J.L., Riediker, M., Mooser, V., Paccaud, F., Waeber, G., Vollenweider, P., Bochud, M.: Effects of particulate matter on inflammatory markers in the general adult population. Particle and fibre toxicology. **9**(1), 24 (2012)

Tsai, T.C., Jeng, Y.J., Chu, D.A., Chen, J.P., Chang, S.C.: Analysis of the relationship between MODIS aerosol optical depth and particulate matter from 2006 to 2008. Atmos. Environ. **45**(27), 4777–4788 (2011)

Verma, S., Pani, S.K., Bhanja, S.N.: Sources and radiative effects of wintertime black carbon aerosols in an urban atmosphere in East India. Chemosphere. **90**(2), 260–269 (2013)

Wang, Y.H., Liu, Z.R., Zhang, J.K., Hu, B., Ji, D.S., Yu, Y.C., Wang, Y.S.: Aerosol physicochemical properties and implications for visibility during an intense haze episode during winter in Beijing. Atmos. Chem. Phys. **15**(6), 3205–3215 (2015)

WHO, 2014a Burden of Disease from Ambient Air Pollution for 2012, Description of Method, Version 1.3. WHO, Geneva. The Global Health Observatory Retrieved 01.03.2014 (2014)

WHO, 2014b. WHO's Ambient Air Pollution Database e Update 2014.Data Summary ofthe AAP Database.WHO,Geneva. http://www.who.int/phe/health_topics/outdoorair/databases/AAP_database_results_2014.pdf?ua¼1

WHO, 2018 .https://apps.who.int/iris/bitstream/handle/10665/272596/9789241565585-eng.pdf

Zhang, J.K., Sun, Y., Liu, Z.R., Ji, D.S., Hu, B., Liu, Q., Wang, Y.S.: Characterization of submicron aerosols during a month of serious pollution in Beijing, 2013. Atmos. Chem. Phys. **14**(6), 2887–2903 (2014)