




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 PM₁₀ and PM_{2.5} are 134 ± 48 and 213 ± 80 $\mu\text{g}/\text{m}^3$, respectively, which exceeds both national and international standards. The average value of PM_{2.5}/PM₁₀ 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 SO₄²⁻, Cl⁻, K⁺, NO₃⁻, Na⁺, Ca²⁺, Mg²⁺ 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 PM₁₀ · 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 ($PM_{2.5} \sim 10 \mu\text{g}/\text{m}^3$ and $PM_{10} \sim 20 \mu\text{g}/\text{m}^3$) 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 further-more 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 South-west 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^+ , K^+ , 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 $\text{PM}_{2.5}$ concentrations, ionic (SO_4^{2-} , NO_3^- , 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 sources is 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 $\text{PM}_{2.5}$) 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°N , 82.97°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

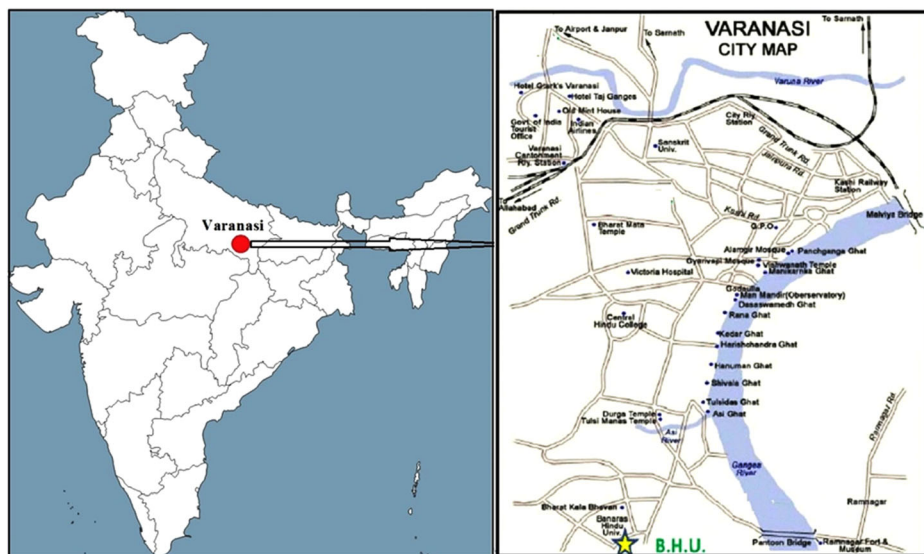


Fig. 1 Map of the sampling location and surrounding area

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 $\sim 10:30$ am (Terra) and $\sim 01:30$ pm (Aqua) local solar time. It measures the radiance at 36 spectral bands in the visible to thermal IR spectral range of $0.41\text{--}14\ \mu\text{m}$ 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; Prjith 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 $2\ ^\circ\text{C}$. Daily variation of meteorological parameters i.e. temperature ($^\circ\text{C}$), 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°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.

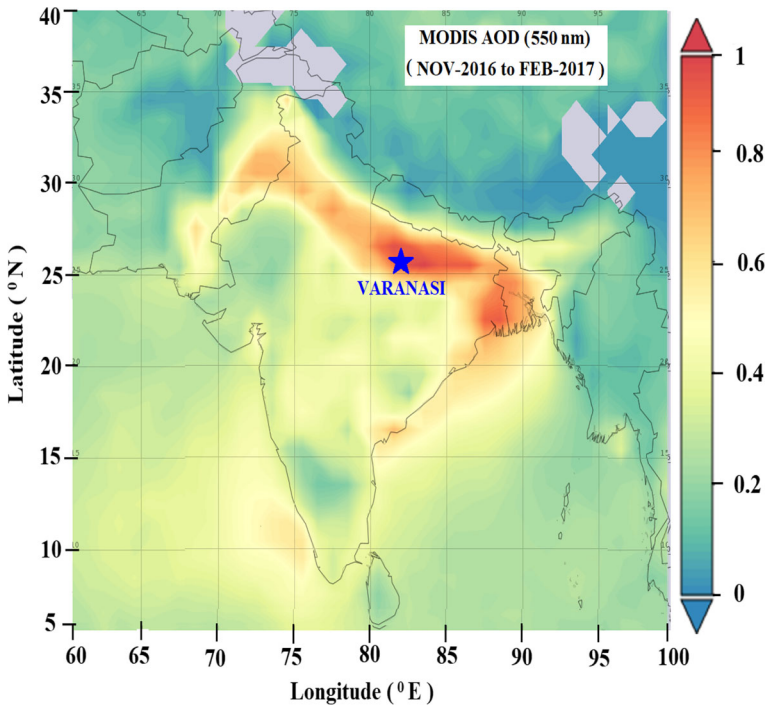


Fig. 2 Map of the study area showing the spatial variability of MODIS derived $AOD_{550\text{ nm}}$ during November 2016 – February 2017. The blue star represents the location of Varanasi city

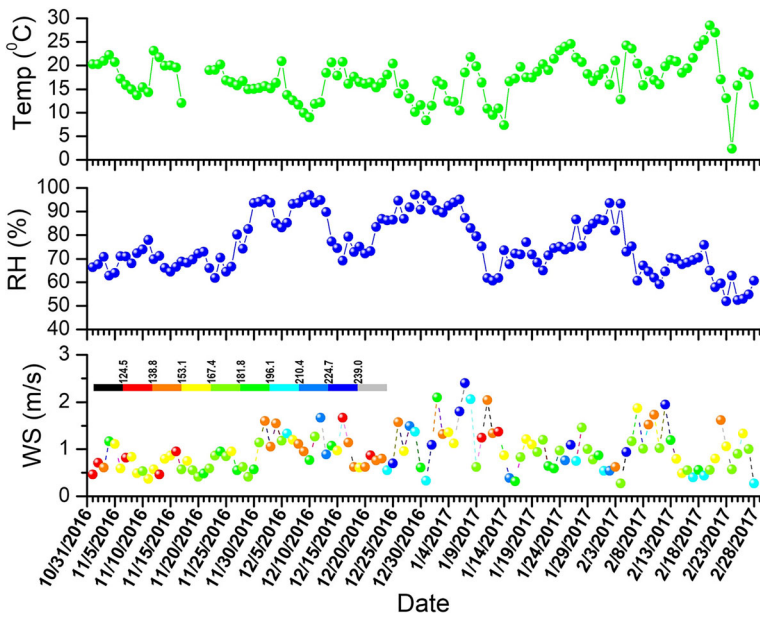


Fig. 3 Diurnal variation of meteorological parameters like temperature ($^{\circ}\text{C}$), relative humidity (RH %), wind speed (m/s) and wind direction (degree) over Varanasi during the study period. The color scale represents the wind direction

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

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

2.2 Sampling and chemical analysis

Particulate matter samples (both PM₁₀ 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^o 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 M Ω) and then ultrasonicated for half an hour and then filtered using Whatman filters. After that, the filtered solution was again filtered using 0.22 μ m 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^o N to 40^o N and 60^o E to 95^o 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 (HYSPPLIT 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 $\mu\text{g}/\text{m}^3$ and 42–257 $\mu\text{g}/\text{m}^3$ with the mean value 213 ± 80 $\mu\text{g}/\text{m}^3$ and 134 ± 48 $\mu\text{g}/\text{m}^3$ for PM_{10} and $\text{PM}_{2.5}$ respectively. The daily observed concentration of PMs (both PM_{10} and $\text{PM}_{2.5}$) is manifold higher than the standard daily mean concentration of PMs referred by National Ambient Air Quality Standard (NAAQS: $\text{PM}_{10} = 100$ $\mu\text{g}/\text{m}^3$ and $\text{PM}_{2.5} = 60$ $\mu\text{g}/\text{m}^3$), US Environmental Protection Agency (USEPA: $\text{PM}_{10} = 150$ $\mu\text{g}/\text{m}^3$ and $\text{PM}_{2.5} = 35$ $\mu\text{g}/\text{m}^3$) and World Health Organization (WHO: $\text{PM}_{10} = 50$ $\mu\text{g}/\text{m}^3$ and $\text{PM}_{2.5} = 25$ $\mu\text{g}/\text{m}^3$) 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 $\sim 285 \pm 87$ $\mu\text{g}/\text{m}^3$ during the winter season at Varanasi (Murari et al. 2016; Kumar et al. 2015a), 203 ± 40 $\mu\text{g}/\text{m}^3$ at Kanpur (Tare et al. 2006), 268 ± 74 $\mu\text{g}/\text{m}^3$ at Kharagpur (Thurston et al. 2011), 232 ± 131 $\mu\text{g}/\text{m}^3$ at Delhi (Tiwari et al. 2009). Similarly, Guttikunda (2013) also reported the concentration of $\text{PM}_{2.5}$ and PM_{10} to be 123 ± 87 and 208 ± 14 $\mu\text{g}/\text{m}^3$ 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 ($\text{PM}_{2.5}$: 121 $\mu\text{g}/\text{m}^3$ and PM_{10} : 230 $\mu\text{g}/\text{m}^3$) 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 $\mu\text{g}/\text{m}^3$), Kolkata ($\text{PM}_{2.5}$: 131 ± 58 $\mu\text{g}/\text{m}^3$), Lucknow ($\text{PM}_{2.5}$: 130 ± 73 $\mu\text{g}/\text{m}^3$ and PM_{10} : 196 ± 90 $\mu\text{g}/\text{m}^3$), Agra ($\text{PM}_{2.5}$: 144 ± 79 $\mu\text{g}/\text{m}^3$) and Delhi ($\text{PM}_{2.5}$: 117 ± 79 $\mu\text{g}/\text{m}^3$ and PM_{10} : 191 ± 127 $\mu\text{g}/\text{m}^3$) during the year 2014 which may be due to the rapid industrialization and other anthropogenic activities. However, the PMs concentration (both PM_{10} , and $\text{PM}_{2.5}$) in the Indo-Himalayan region found within the NAAQS limit with value at Kashmir ($\text{PM}_{2.5}$: 20 ± 13 $\mu\text{g}/\text{m}^3$; PM_{10} : 32 ± 28 $\mu\text{g}/\text{m}^3$), Kullu ($\text{PM}_{2.5}$: 31 ± 17 $\mu\text{g}/\text{m}^3$, PM_{10} : 49 ± 16 $\mu\text{g}/\text{m}^3$), Nainital (PM_{10} : 42 ± 24 $\mu\text{g}/\text{m}^3$), Dehradun ($\text{PM}_{2.5}$: 53 ± 38 $\mu\text{g}/\text{m}^3$) and Darjeeling ($\text{PM}_{2.5}$: 24 ± 14 $\mu\text{g}/\text{m}^3$, PM_{10} : 49 ± 18 $\mu\text{g}/\text{m}^3$). In addition, they have also reported a decrease in $\text{PM}_{2.5}$ particles mass concentration ($\mu\text{g}/\text{m}^3$) from 2014 to 2015 over Patiala ($\text{PM}_{2.5}$: 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 ($\text{PM}_{2.5}/\text{PM}_{10}$) 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} , $\text{PM}_{2.5}$, and $\text{PM}_{10-2.5}$ 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 $\text{PM}_{2.5}$). On few days, peak in particulate matter loading is associated with a relatively lower value of $\text{PM}_{2.5}/\text{PM}_{10}$ 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 (µg/m ³)	PM _{2.5} (µg/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)

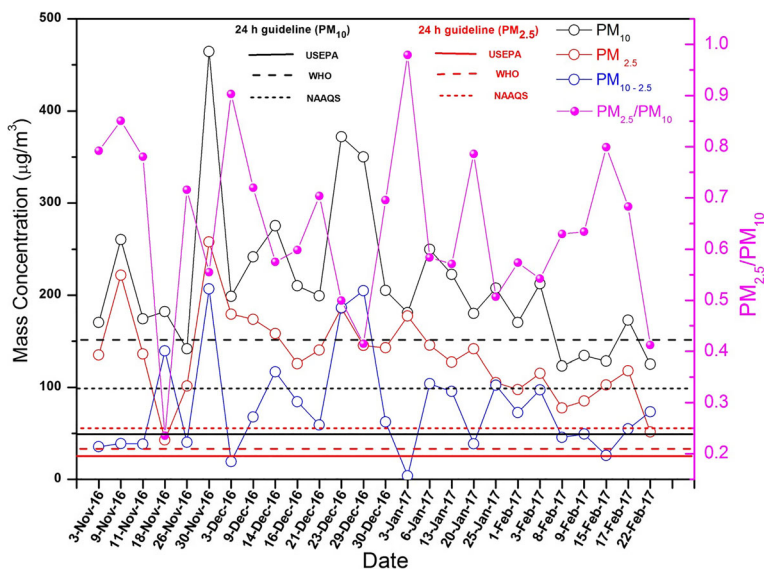


Fig. 4 Temporal variation of Particulate Matter (PM_{10} , $PM_{2.5}$ and $PM_{10-2.5}$) and their ratio ($PM_{2.5}/PM_{10}$) during the study period

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^{2-}) 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^{2-} , NO_3^- (Wang et al. 2015; Thurston et al.

Table 3 Statistical analysis of major ionic species concentration in PM₁₀ and PM_{2.5} samples over Varanasi

Species	PM ₁₀ (µg/m ³)				PM _{2.5} (µg/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 Ca²⁺/Na⁺, Mg²⁺/Na⁺ is found lower than seawater ratio and SO₄⁻/Na⁺ is found higher while Cl⁻/Mg²⁺ and Na⁺/Mg²⁺ 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 SO₄⁻, 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-SO₄⁻ are coal combustion, fossil fuel, biomass burning, vehicular emission and oxidation of SO₂ (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.

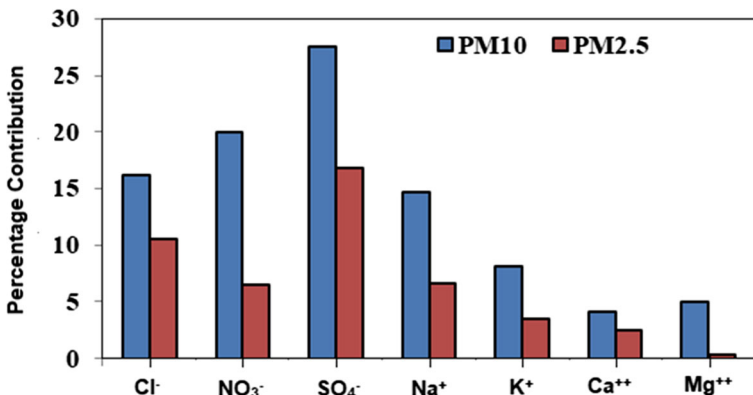
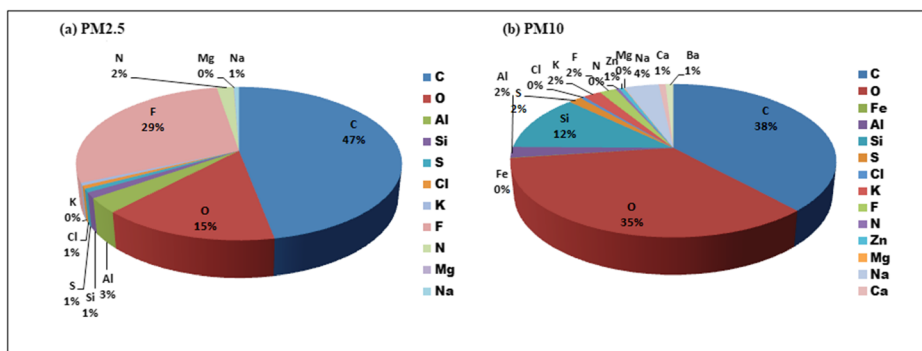
**Fig. 5** The percentage contribution of major ions in PM₁₀ and PM_{2.5} samples

Table 4 Weight % (\pm standard deviation) of the elements quantified in PM_{2.5} and PM₁₀ sample during study period at receptor site

Elements	PM ₁₀ (weight %)	PM _{2.5} (weight %)
C	38.30 \pm 6.57	38.35 \pm 21.81
O	34.72 \pm 6.00	12.26 \pm 9.83
Fe	0.06 \pm 0.18	NA
Al	2.28 \pm 0.74	2.55 \pm 6.21
Si	11.97 \pm 4.15	0.98 \pm 2.79
S	1.51 \pm 1.12	0.62 \pm 0.71
Cl	0.46 \pm 0.25	0.52 \pm 0.51
K	2.1 \pm 0.80	0.45 \pm 0.69
F	1.96 \pm 7.8	23.66 \pm 16.20
N	0.38 \pm 1.05	1.59 \pm 3.40
Zn	0.58 \pm 1.06	NA
Mg	0.06 \pm 0.12	0.04 \pm 0.18
Na	4.08 \pm 1.25	0.46 \pm 1.16
Ca	0.79 \pm 1.11	NA
Ba	0.90 \pm 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 PM₁₀ and PM_{2.5} samples during the study period are summarized in Table 4. The average concentration of crustal elements in PM₁₀ 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 PM₁₀ 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) PM₁₀

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

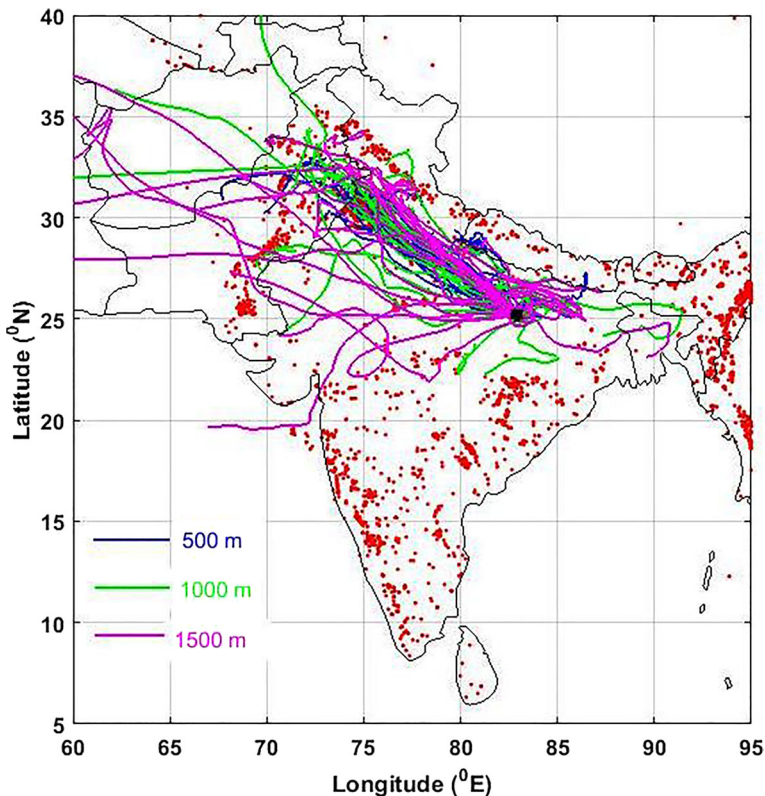


Fig. 7 Spatial variability of biomass fire hot spot during November 2016 to February 2017 superimposed with five days air mass back trajectories on sampling dates at three different altitudes i.e. 500 m (blue), 1000 m (green) and 1500 m (magenta). The black dot represents the receptor site

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 ($PM_{2.5}$: $134 \pm 48 \mu\text{g}/\text{m}^3$ and PM_{10} : $213 \pm 80 \mu\text{g}/\text{m}^3$) 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 WSIS (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 transition 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.

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