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Research Paper

Assessment of atmospheric aerosols over Varanasi: Physical, optical and chemical properties and meteorological implications

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ABSTRACT

The present paper reports the results derived from $PM_{2.5}$ and PM_{10} concentrations during October 2016–May 2018 using MICROTOPS-II Sunphotometer and high volume samplers at Varanasi. Observed data were categorized and analyzed in order to understand the nature, source of origin and variability with seasons. Observations showed aerosol mass loading during the post-monsoon 2016 and winter 2017 as compared to the pre-monsoon 2018 values, which typically exceed national standard. The close relationships between $PM_{2.5}$ and PM_{10} during post-monsoon ($r = 0.571$) and winter ($r = 0.799$) suggested that both type of particulates might have originated from the same source. Further, the $PM_{2.5}/PM_{10}$ indicated that the fine particles were dominantly present during the post-monsoon and winter season where as coarse particles were found dominant in the pre-monsoon season. The measured high values of aerosol optical depth (AOD) and angstrom exponent (AE) during post-monsoon and winter attributed to the accumulation of aerosols from sources and biomass/crop residue burning in the surrounding region and low dispersal due to shallow boundary layer and lower wind velocity. In contrast during pre-monsoon months the aerosols were accumulated as a mixture of transported from deserts and other far away regions including mineral from earth crust. The effect of meteorological parameters (temperature, wind speed and relative humidity) was also studied. Temperature did not show any relation during the pre monsoon when it was quite high, whereas during the winter months it showed negative trend with concentration. Wind showed negative correlation during the whole observation period. Relative humidity showed weak positive correlation during winter months for $PM_{2.5}$ and PM_{10} where as $PM_{2.5}$ did not show any relation during pre and post monsoon. PM_{10} showed weak negative relation during pre monsoon months. In order to understand elemental and ionic composition Scanning electron microscope (SEM) coupled with energy dispersive X-ray microanalyzer (EDX) analysis were done which showed dominant presence of C, F, O, Si, N, Na, K, Al, Ca and S. The following trend $SO4^{2-} > NO_3^- > Na^+ > Ca^{2+} > K^+ > Cl^- > F^- > Mg^{2+} > Li^+$ was observed from the Ion chromatograph (IC) analysis. The source for these elements may have been different industrial activities, biomass burning and vehicular emissions. The results are useful for further planning of city developments and climate studies.

1. Introduction

Air pollution which defines deterioration in air quality is the most important environmental issue for the human health, climate change as well as for the agricultural production. Aerosols are major contributor to air pollution which also affects global and regional climate changes, directly by scattering and absorption of solar and terrestrial radiations (Satheesh et al., 2006; Srivastava et al., 2014; Tiwari et al., 2018) and nucleation of clouds and modifying their macro and micro-physical properties (Altaratz et al., 2014; Sarangi et al., 2017). In addition aerosols also impact the earth's radiation budget, visibility, health issues and damage of ecosystems (Schwartz et al., 1996; Sharma et al., 2017; Tiwari et al., 2018). Aerosols with diameter ~2.5 μm ($PM_{2.5}$) and ~10 μm (PM_{10}) are quite susceptible in causing heart and lung diseases and aggravating asthma and other respiratory problems (Schwartz et al., 1994, 1996). These social impacts show the need for better

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understanding of physical, optical and chemical characteristics of aerosol particles, and their source of origin.

Heavy aerosol loading has occurred during the last few decades due to large urbanization and continuous increase in population density and related energy demands in the South and East Asian countries mainly India and China (Dey and Di Girolamo, 2010; Lawrence and Lelieveld, 2010; Kang et al., 2016). In India Indo-Gangetic Plain (IGP) faces intense loading of aerosols which varies in density and characteristic features with the seasons (Sayer et al., 2014; Sen et al., 2017; Singh et al., 2017). In this region aerosol emissions from the agricultural residues burning are frequently found to be associated with haze formation (Singh et al., 2018). Srivastava et al. (2014) discussed aerosol-climate relation using surface albedo, optical properties of aerosols and relative position of clouds. In addition to natural and agricultural sources, vehicular emissions (Choudhary et al., 2020), industrial emissions, secondary aerosols (Singh et al., 2017), and biomass burning (Rajput et al., 2014; Wan et al., 2017) are the other sources of aerosols in Asian countries.

The satellite retrieved information using advanced scientific tools are quite useful for the assessment, forecasting and management of air quality. MODIS onboard AQUA and TERRA satellites have been used for predicting air quality (Kumar et al., 2007; Singh et al., 2018). were used for predicting air quality. Aerosols estimation (Dey et al., 2012; Kahn, and Gaitley, 2015), mortality analysis (Corbett et al., 2007; Evans et al., 2013) and climate change (Dey et al., 2004) were studied. Aerosol optical depth (AOD) is used to discuss the air quality, atmospheric conditions and it is also used for the estimation of the aerosol mass concentrations (Dey et al., 2012; Srivastava et al., 2014).

Physical and chemical properties of atmospheric aerosols show a large spatio-temporal variations because of different emission sources, chemical composition and their transportation pathways which results in varying impacts on earth's climate system and air quality (Tiwari et al., 2015, 2018; Sarangi et al., 2017; Sharma et al., 2017). Morphological and chemical compositions of atmospheric aerosols can be used to distinguish their sources and expulsion system. Aerosols morphology, composition of elements and density of particles can be studied using SEM EDX analysis (Conner and Williams, 2004; Srivastava et al., 2009; Pachauri et al., 2013; Singh et al., 2014). The seasonal dependence of aerosol features at a given location is based on the mixture of local and transported from different destinations.

During pre-monsoon season, the coarse-mode aerosol concentrations In IGP region were enhanced because of frequent and intense dust storms which brought huge amount of dust through long-range transport from Arab countries, southwest Asia, and the Thar Desert (Tiwari et al., 2015; Singh et al., 2016). On the other hand higher aerosol concentrations over the central IGP during post-monsoon and winter seasons were attributed to the widespread crop residue burning in west U.P, Punjab and Haryana states (Sharma et al., 2010; Mishra and Shibata, 2012; Sen et al., 2014; Kumar et al., 2015; Tiwari et al., 2016, 2018). The favorable meteorological conditions are responsible to form dense smoke plume that covers the whole IGP. The region is covered by fog and haze due to weak convection, high relative humidity, calm winds, and a shallow atmospheric boundary layer during winter season (Mishra and Shibata, 2012).

Varanasi is a holy city and attracts tourists from all over the world and therefore it is essential to understand different sources of degrading air quality and to scale them, so that using proper management air quality may be improved. The present study is focused on understanding of the aerosols physical, chemical and optical properties in Varanasi during the period October 2016–May 2018. The monthly and seasonal variability are shown in concentrations of $PM_{2.5}$, PM_{10} and $PM_{2.5}/PM_{10}$ as well as AOD and AE observed from MICROTOPS-II and MODIS are presented in section 4. To understand these observations monthly mean temporal variations of meteorological parameters like temperature, wind speed and relative humidity are also presented and used in the present study (section 5). The seasonal dependence of Pearson's coefficient (r) between $PM_{2.5}$ and PM_{10} and between AOD derived from MICROTOPS-II AOD and MODIS data are also presented and discussed. The elemental and ionic composition of aerosols is studied using SEM-EDX and Ion Chromatography (IC) respectively in section in 6 and 7. The present results are extensively compared with previous findings from several aerosol studies carried over the Indian subcontinent. The outcomes of the present study consistent with aerosol optical, physical and chemical characteristics measured by different scientific groups spread over throughout the world. Present results may be quite useful for the climate modelers.

2. Experimental methods

2.1. Experimental site description

The chosen site for the present study is Varanasi (25.2677° N, and 82.9913° E) located in the Eastern part of the state of Uttar Pradesh at an elevation of 80.71 m in the centre of the Ganges valley of North India. The city is a highly polluted city, causing a high aerosol loading throughout the year which shows large seasonal and temporal variability (Tiwari and Singh, 2013; Murari et al., 2017). Varanasi experiences humid sub-tropical climate with extreme summer, intense rainfall during monsoon, and cold weather during winter season. Varanasi is situated in the centre of IGP which is one of the largest, industrialized, densely populated and developing regions of the world (Satheesh et al., 2006; Tiwari et al., 2018). Previous scattered measurements over Varanasi suggested coarse mode aerosol particles (mostly dust) as one of the main contributors to the whole aerosol loading during pre-monsoon months while fine-mode anthropogenic aerosols could be the major contributors of aerosol loading during post-monsoon and winter months (Tiwari and Singh, 2013). In addition increasing population density (>2400 per km^2 as per census 2011) and more than lacs of people daily visitor (dynamic population) and unaccountable vehicles on the road make the situation worst. The sampling topography is of moderate level without having a particular restricted impact of mountains, oceans, or any unequivocal outflow source (Murari et al., 2017).

2.2. In-situ measurements of particulates

$PM_{2.5}$ and PM_{10} particles were targeted, respectively because these particles affect the human health and constitute the major concern for air quality. Continuous sampling was done from October 2016 to May 2018. Samples were collected twice in a week (24 h) using high volume samplers installed at the building of the Department of Physics, Institute of Science, Banaras Hindu University (BHU), Varanasi. BHU is an educational institution without having impacts from any specific emission source aside occasional burning of garden waste. However, the sampling site could be influenced by local traffic emissions and long-range trans-boundary aerosols from time to time (Kumar et al., 2015). Tiwari et al. (2018) suggested that the local/anthropogenic aerosols could be more prevailing than long range transported aerosols over Varanasi. Particulates of size $\leq$2.5 μm measurements were observed using APM 550, Envirotech on Teflon Micro fibre filter papers (2 μm PTFE). While coarse particulate ($PM_{10,}$ $\leq$10 μm) samples were collected on glass microfiber filters (GF/A, Whatman; 8 × 10 ins) by improved respirable dust sampler (APM-460NL, Envirotech) at a flow rate of 1 m^3/h. The flow rate calculations were made before sample collection to ensure fluctuations within the range. The filter papers were placed in desiccators for the 24 h before and after the sampling for the removal of moisture.

2.3. In-situ and satellite measurements of optical properties

The AOD is directly correlated with optical properties of aerosols in a column-integrated mode (Kaufman et al., 2002). The ground based observations of AOD were carried out using portable multi-band MICROTOPS-II Sunphotometer (Solar Light Company, USA) nearly twice a

week during clear sky condition from 09:00hr to 17:00hr. The field of view is 2.5° with a typical sun targeting accuracy better than 0.1°. The error in the AOD retrieval in the ultraviolet range is ~ ± 0.03, which drops to ~ ± 0.02 in the visible range. MICROTOPS-II Sunphotometer has different filters at 380, 440, 500, 640, and 870 nm wavelengths for data acquisition. For the quality control, sampling were done only on those days which contain proper sunshine having very few/no cloud cover in the sky, particularly at the situation when angles are close to the sun. Triplet MICROTOPS-II Sunphotometer measurements were also taken during every observation to minimize the errors. The observed smaller values are used in analysis because these values correspond to most accurate sun pointing. Higher values are discarded (Porter et al., 2001). MICROTOPS-II Sunphotometer ascertains the corresponding optical depths utilizing the information of solar intensity at the highest point of the atmosphere. Similar calculations were done using Langley method (Schmid and Wehril, 1995). MICROTOPS-II Sunphotometer was calibrated before samples (AOD) collection according to the details provided by Morys et al. (2001). An appropriate log book for every MICROTOPS-II Sunphotometer inspection was prepared and recorded the cloud condition and daylight for every measurement. The MICROTOPS II Sunphotometer AE values at 380, 440, 500, 640, and 870 nm were determined utilizing AOD information of each sample collected on the ground.

The MODIS TERRA (mean_MOD08_D3_6_1_AOD_550_Dark_Target_Deep_ Blue_Combined_Mean) daily AOD satellite data for 1 ° × 1 ° grid during October 2016 to May 2018 were acquired at 550 nm wavelength. MODIS satellite estimates reflected solar radiance and possible outflow which basically relates it with columnar aerosol loading over worldwide at moderate spatial resolutions (Ramachandran et al., 2012). Satellite-based MODIS TERRA data were obtained from MODIS online visualization and analysis system (http://giovanni.gsfc.nasa.gov/giovanni/) by averaging daily AOD at 550 nm and AE (412–470 nm) over Varanasi. In the present investigation a strong spectral and seasonal variability of AOD and AE was observed at Varanasi. Observed good correlation between satellite (MODIS) derived AOD and ground based (MICROTOPS-II) AOD validates the present measurement and analysis system.

3. Methodology

3.1. Methodology for AOD measurement

The spectral AOD and AE are the two optical parameters which describe the columnar aerosol properties and show strong wavelength dependency. The samples collected from MICROTOPS-II Sunphotometer were processed for AOD studies. During each measurement, three continuous scans were taken and processed by the second order polynomial fitting. Only those data were selected which have maximum correlation and minimum error in the second order coefficient (Sharma et al., 2014). The spectral variations of AOD follow the Ångström Power law (Ångström, 1964),

$$\tau_\lambda = \beta\lambda^{-a} \quad (1)$$

where, τ_λ is AOD at the wavelength λ, β is the turbidity coefficient which is equal to AOD at λ = 1 μm and α is the AE.

In the present study the MODIS TERRA AODs are collected at the wavelength of 550 nm whereas MICROTOPS-II AODs are collected at 500 nm wavelength. For comparative studies, the measurements should be at the same wavelength. Therefore, collected AOD values using MICROTOPS-II at 500 nm were interpolated to a common wavelength of 550 nm by Angstrom power law. In the modified form equation (1) could be written as:

$$AOD_{550nm} = AOD_{500nm}(550/500)^{-a} \quad (2)$$

3.2. Methodology for AE measurement

The AE measured at different wavelength bands is a useful tool to distinguish and characterize atmospheric aerosols (Eck et al., 1999). In general, AE is an indicator of aerosol particle size and fraction of fine-to coarse-mode atmospheric aerosols (Schuster et al., 2006; Tiwari et al., 2016). In the present study AE were derived from MODIS satellite observations and were also calculated using the MICROTOPS-II AOD values. The value of AE > 1 values represent fine mode aerosol particles while AE < 1 indicates dominance of coarse mode particles (Eck et al., 1999). Considering measurements at two wavelengths λ1 and λ2 the turbidity coefficient β can be eliminated an AE specified by α can be obtained from the following relation:

$$\alpha = -\ln(\tau_{\lambda 1}/\tau_{\lambda 2})/\ln(\lambda_2/\lambda_1) \quad (3)$$

Using AOD ($\tau_{\lambda 1}$, $\tau_{\lambda 2}$) values observed by MICROTOPS-II Sunphotometer instrument for the pair of wavelength in range 440–870 nm AE values are evaluated using the above relation. While MODIS AE values were used directly provided by NASA Giovanni in the wavelength ranges (412–470 nm) (http://giovanni.gsfc.nasa.gov/giovanni/).

3.3. Particulate chemical analysis using SEM-EDX and IC instruments

Using IC and SEM-EDX measurements chemical composition and ionic content are the characterized (Singh et al., 2014; Pratap et al., 2017). IC provides information about ionic content of the particulates while SEM-EDX provides elemental composition and surface morphology of particulate matters. SEM is a 'Supra' type made by Carl Zeiss Microscopy, Germany, and EDX is a 'Genesis' type with Si–Li-Detector made by EDAX, Germany. For the elemental composition and surface morphology analysis the sample paper was randomly cut in size of about 1 mm^2 out of the main filter paper. A very thin film of carbon was deposited on the surface of the prepared samples to make electrically conductive using vacuum coating unit. These prepared samples were mounted on electron microprobe stubs for the further analysis (Singh et al., 2014). The dominance of Carbon (C), Fluorine (F), Oxygen (O), Silicon (Si), Sodium (Na), Potassium (K), Aluminium (Al), Nitrogen (N), Calcium (Ca), and Sulphur (S) was observed for all the days of observation. Some other elements such as, Iron (Fe), Zink (Zn), Magnesium (Mg), Chlorine (Cl) and Barium (Ba) were also found in small amount in these samples.

Ionic contents of the particulates were obtained from the analysis of anion and water-soluble cations using IC. One - fourth of the sample filters was extracted into 20 ml of de-ionized water (18.2 MΩ) and then ultrasonicated for half an hour and there after filtered using Whatman filters. 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 sample. IC analysis indicated the dominance of Sulphate ($SO4^{2-}$), Nitrate (NO_3^-), Chloride (Cl^-), Sodium (Na^+), Calcium (Ca^{2+}), Potassium (K^+) and Fluoride (F^-) ions. However, some cations Magnesium (Mg^{2+}) and Lithium (Li^+) were also present. The source of SO_4^{2-} concentration can be attributed to the oxidation of Sulphur dioxide originated from the burning of fossil fuels and vehicular emissions (Sharma et al., 2010). NO_3^- concentration is attributed to the vehicular emissions and fertilizer used in agricultural farms. The presence of Mg^{2+} concentration could be cement dust from the infrastructure development being carried out in Varanasi across the year.

3.4. Meteorological observations

The daily basis meteorological data (temperature, relative humidity, wind speed and wind direction) were taken from the India Meteorological Department (IMD) local station functional in the nearby building inside the BHU campus. Temperature record showed increasing trend from February to June in the pre-monsoon months, the lowest value was

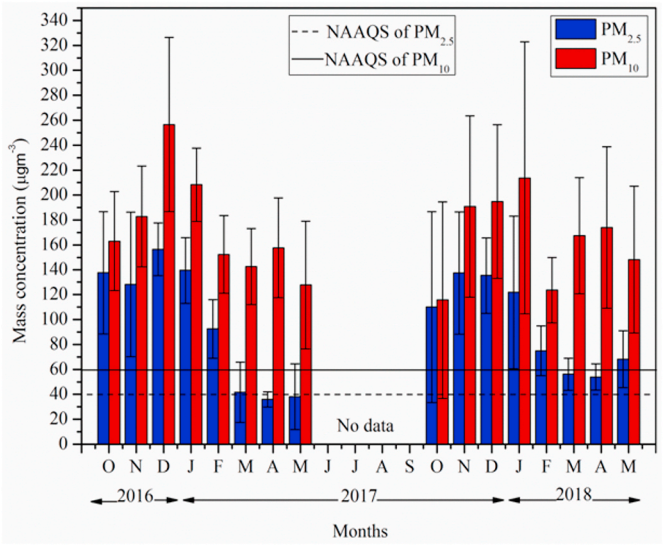

Fig. 1. Monthly mean variability of mass concentrations of $PM_{2.5}$ and PM_{10} particulates and its standard deviation in Varanasi, India.

in the January month of winter season. Relative humidity showed opposite behavior to the temperature. Higher wind speeds were observed during pre-monsoon and monsoon months. The lower wind velocity was recorded in the post-monsoon and winter months. Relative humidity was recorded higher during monsoon, post-monsoon and winter months with lower values in the pre-monsoon months.

4. Results and discussions

4.1. Variation and correlation analysis between $PM_{2.5}$ and PM_{10}

The mass concentrations of fine and coarse mode aerosols were continuously measured and analyzed during October 2016–May 2018 and monthly mean variations in $PM_{2.5}$ and PM_{10} are shown in Fig. 1 along with Indian National Ambient Air Quality Standard (NAAQS) levels. Particulate mass anomalies clearly distinguish the higher level wintertime and post-monsoon peaks before being reduced into pre-monsoon months. The winter season recorded pattern and mean mass concentrations of $PM_{2.5}$ (129.808 ± 36.348 μgm^{-3} for 2016; 110.732 ± 42.581 μgm^{-3} for 2017) and PM_{10} (207.992 ± 66.861 μgm^{-3} for 2016; 177.344 ± 68.971 μgm^{-3} for 2017) typically resemble to that of Kumar et al. (2015) for the same region. The wintertime higher aerosol concentrations may be understood in terms of prevailing meteorological conditions of slow wind and low temperature and burning of biomass/biofuel burning including increased vehicular circulation (Banerjee et al., 2015). The mean mass concentrations of $PM_{2.5}$ (132.434 ± 51.752 μgm^{-3} for 2016; 121.46 ± 65.374 for 2017) and PM_{10} (173.784 ± 37.671 μgm^{-3} for 2016; 153.577 ± 82.595 μgm^{-3} for 2017) were recorded during post monsoon season. The high concentrations of aerosols loading exceeded the 24-h average national standards ($PM_{2.5}$ = 60 μgm^{-3}; PM_{10} = 100 μgm^{-3}; cpcb. nic.in) and USEPA standards ($PM_{2.5}$ = 35 μgm^{-3}; PM_{10} = 150 μgm^{-3}) (http://www.epa.gov/air/particlepollution/) which becomes a threat to human health and wealth. During the post-monsoon the $PM_{2.5}$ mean mass concentrations were observed to be higher in comparison to the winter season. Whereas the PM_{10} mean mass concentrations were higher during winter season than to the post-monsoon season. Further the widespread crop residue burning in Punjab and Haryana states leads to aerosols loading in the fine mode during post-monsoon as well as winter season in Varanasi region. In the pre-monsoon season the recorded mean mass concentrations of $PM_{2.5}$ (39.211 ± 22.872 μgm^{-3} for 2017; 59.413 ± 17.933 μgm^{-3} for 2018) and PM_{10} (142.618 ± 40.034 μgm^{-3} for 2017; 160.54 ± 54.866 μgm^{-3} for 2018) showed lesser mean mass concentrations of $PM_{2.5}$ than PM_{10}. This reveals the domination of coarse particles. During pre-monsoon dust storm and higher wind velocity carry coarse particles in plenty and dominate Asian aerosols from Middle-East countries and Thar Desert, Rajasthan, India (Murari et al., 2015). Both types of aerosols exceeded the annual Indian NAAQS ($PM_{2.5}$ = 40 μgm^{-3}; PM_{10} = 60 μgm^{-3}; cpcb. nic.in) and WHO ($PM_{2.5}$ = 10 μgm^{-3}; PM_{10} = 20 μgm^{-3}) standards. For the entire monitoring period, the higher variability from 4.33 to 231.405 μgm^{-3} for $PM_{2.5}$ and 36.679 to 372.102 μgm^{-3} for PM_{10} were recorded. Sampling was not done during the monsoon (June–September) months due slight to higher rainfall in the region which wash out the aerosol particles (see Fig. 2).

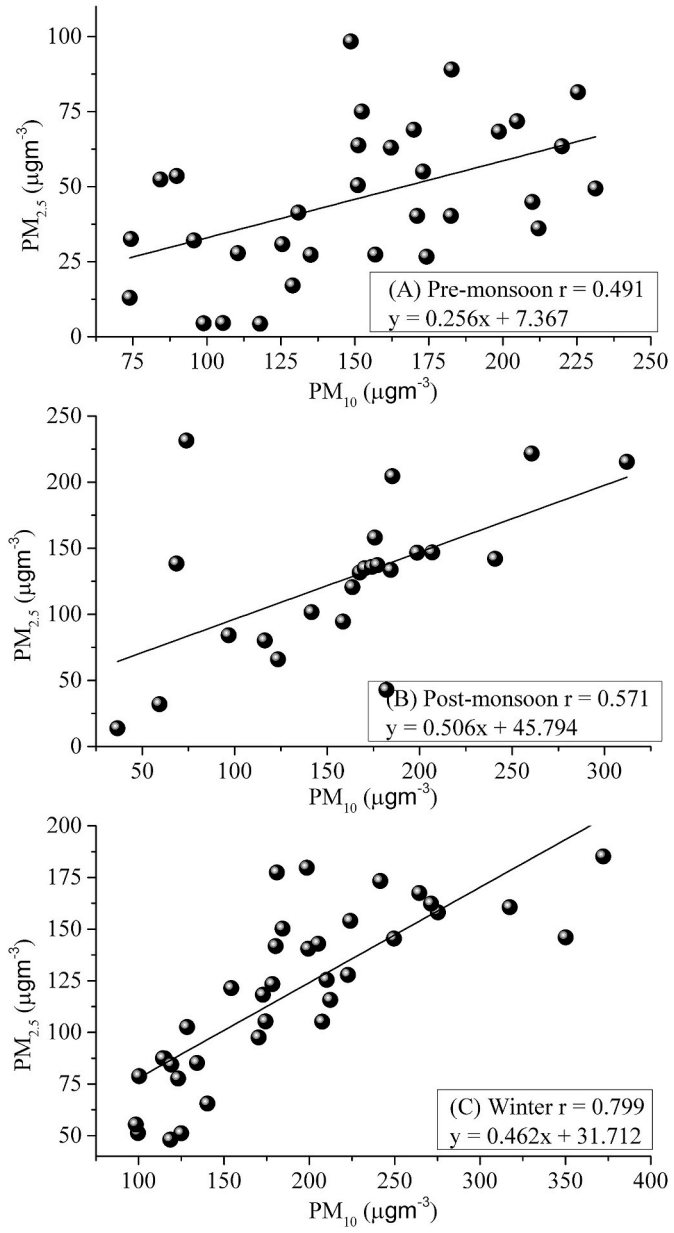

Fig. 2. Correlation between mass concentrations of $PM_{2.5}$ and PM_{10} particles during (A) Pre-monsoon (B) Post-monsoon and (C) Winter seasons in Varanasi, India.

The statistical relation between $PM_{2.5}$ and PM_{10} during pre-monsoon (r = 0.491), post-monsoon (r = 0.571) and winter (r = 0.799) revealed close association between the fine and coarse particulates (figures) which may also be inferred from their similar origin. Incidentally, a high degree of statistical association was observed between $PM_{2.5}$ and PM_{10}

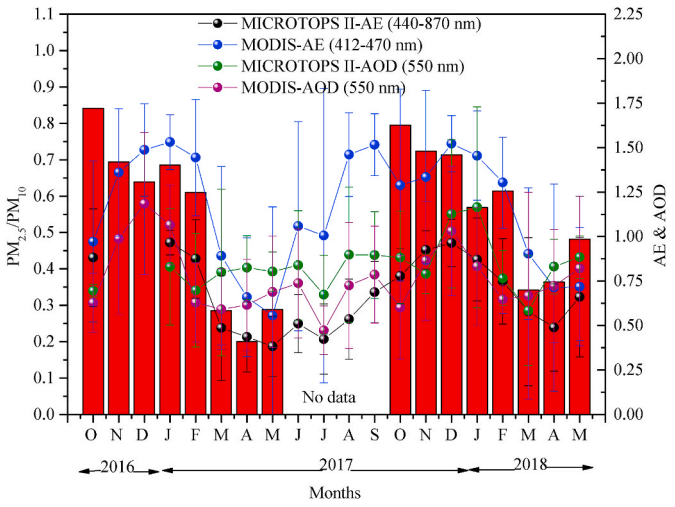

Fig. 3. Monthly variations of the particulate ratios ($PM_{2.5}$ to PM_{10}) with respect to MICROPTOPS II and MODIS AOD and AE.

(r = 0.799) during winter season in the present study as compared to the previous years findings (r = 0.65) reported by Kumar et al. (2015).

4.2. Particulate ratio ($PM_{2.5}/PM_{10}$) analysis

Average of PM concentrations ratio is an indicator of the relative presence of fine and coarse particulates in the air. In the current analysis, seasonal mean of $PM_{2.5}/PM_{10}$ (0.768 for 2016; 0.759 for 2017) during the post-monsoon season indicates dominance of very fine particles. The lower seasonal mean of $PM_{2.5}/PM_{10}$ (0.258 for 2017; 0.396 for 2018) may possibly be due to accumulation of coarser particulates through trans-boundary movement during pre-monsoon season. The winter seasonal mean $PM_{2.5}/PM_{10}$ (0.645 for 2016; 0.632 for 2017) strengthens the fact that the aerosol characteristics over Varanasi are mostly dominated by finer particulates. This is supported by the ranges of the daily mean $PM_{2.5}/PM_{10}$ for post-monsoon (0.235–1.104), winter (0.405–0.98) and pre-monsoon (0.132–0.661) seasons. The ambient aerosol concentrations contain both PM2.5 and PM10 particulates in mixed form. Therefore the ratio of PM2.5 to Pm10 may be taken as the mixing ratio. These ranges for different seasons are an indication of degree of domination of finer particulates. The particulate ratios appeared well above 0.50 during post-monsoon (91%), winter (86%) and pre-monsoon season (15%) clearly indicates domination of finer particulates during post-monsoon and winter months where as coarse particles dominate during pre-monsoon months in the ambient aerosol concentration. Based on the present study it may be concluded that lower ratios during pre-monsoon (increase in non-spherical crustal dust) and higher during post-monsoon and winter (dominance of biomass/waste/wild fire) months are consistent with previous studies (Murari et al., 2015). In addition to $PM_{2.5}/PM_{10}$ variation, AOD and AE variations observed from MICROTOPS-II and MODIS are also presented in Fig. 3.

Similar trend between MICROTOPS-II and MODIS AOD at 550 nm was observed during almost all the months. The higher monthly mean AOD values were observed almost in all the months except in the months of February, March and July. Fig. 3 clearly show differences in AE derived from MICROTOPS-II and MODIS. The observed differences are due to different wavelength ranges. However trend of the both plots were similar. The higher AE values observed in the monsoon, post-monsoon and winter seasons are indicative of dominance of fine particulates. Increased AE values during pre-monsoon months indicate dominance of coarser particulates.

4.3. AOD and AE for different types of aerosols

The optical properties of aerosols depend on their types, size and their concentration in the atmosphere (El-Metwally et al., 2008; Moorthy et al., 2009). In order to study impact of aerosols in different regions surrounding Varanasi AOD at 500 nm and AE at 380–870 nm are summarized in Table 1. Fig. 3 and Table 1 show monthly and seasonal variations of AOD and AE. AOD was high in the months of October, November, December and January and low in February and thereafter an increasing trend was found increasing from March to June month. Enhancement in AOD during pre-monsoon months was attributed to aerosol loading due to dust-storm events from far away source region such as Arabia peninsula and African countries (Tiwari and Singh, 2013). Sudden decrease of AOD in July was attributed to wash out of the aerosols from the rain. In fact concentration of $PM_{2.5}$ and PM_{10} clearly exhibit its effect in AOD. Further, AE in the months of October, November, December, January and February was very high which indicated the dominance of fine mode particles mainly from biomass/biofuel burning and vehicular emissions. While lower AE values during March, April, May, June and July indicated the presence of coarse mode particles in the region. The AE values started increasing from August and September months which indicated the presence of mixture of local and transported aerosols over the region. The difference in the Monthly mean AE values derived from MICROTOPS-II and MODIS measurements was attributed to different wavelength ranges used in the measurements. However trend of variation remained similar. The mean AOD values in the pre monsoon season were (0.766 ± 0.255 in 2017; 0.748 ± 0.274 in 2018 as observed from MICROTOPS-II and 0.551 ± 0.198 in 2017; 0.649 ± 0.311 in 2018 from MODIS. While during

Table 1
Seasonal AOD_{500nm} and $AE_{380–870nm}$ at Varanasi and its surroundings.

Locations	Study period	AOD				AE				References
		Prm	M	Pom	W	Prm	M	Pom	W	
Varanasi	2016–2018	0.79	0.78	0.75	0.86	0.63	0.72	1.03	1.12	Present study
Varanasi	2011–2014	0.73	0.73	0.95	0.87	0.76	0.86	1.16	1.09	Tiwari et al. (2018)
Delhi	2011–2013	0.82	0.86	1.00	0.95	0.51	0.89	1.03	1.02	Tiwari et al. (2016)
G Noida	2010–2012	0.78	0.73	0.98	0.87	0.68	1.02	1.19	1.13	Sharma et al. (2014)
Delhi	2001–2012	0.78	0.74	0.91	0.77	0.49	0.66	0.93	0.97	Lodhi et al. (2013)
Jodhpur	2004–2012	0.75	0.71	0.59	0.48	0.54	0.75	0.86	0.81	Bhaskar et al. (2015)
Varanasi	2011	0.67	0.68	0.78	0.90	0.70	0.72	1.20	1.11	Tiwari and Singh (2013)
Jaipur	2011	0.50–0.80				0.38–0.60				Tiwari et al. (2013)
G College	2011	0.60–0.85				0.85–1.05				Tiwari et al. (2013)
Kanpur	2005–2010	0.59	0.60	0.76	0.63	0.66	0.77	1.27	1.24	Kaskaoutis et al. (2012)
G College	2009	0.51–0.77				0.65–0.91				Srivastava et al. (2011)
Gual Pahari	2009	0.64				0.52				Gautam et al. (2011)
Nainital	2005–2008	0.30	0.15	0.09	0.11					Ram et al. (2010)
Kanpur	2001–2003	0.54	0.66	0.63	0.57	0.60	0.66	1.12	1.26	Singh et al. (2004)

Prm = Pre-monsoon, *M* = Monsoon, *Pom* = Post-monsoon, *W* = Winter.

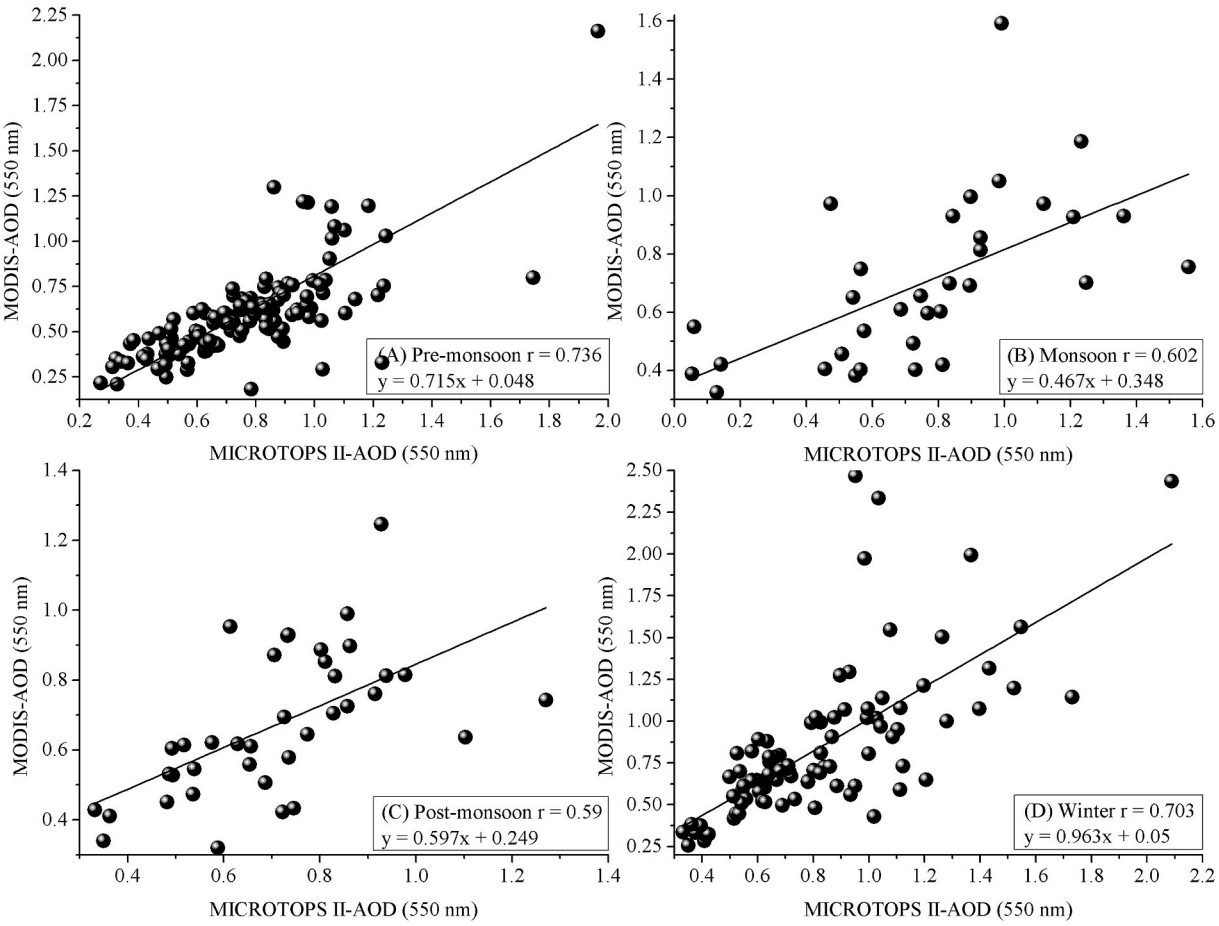

Fig. 4. Correlation between MICROTOPS II-AOD and MODIS TERRA-AOD during (A) Pre-monsoon, (B) Monsoon, (C) Post-monsoon and (B) Winter seasons in Varanasi India.

monsoon season mean AOD values in 2017 were 0.755 ± 0.363 0.701 ± 0.281 as observed from MICROTOPS-II and MODIS respectively. The identical values during pre monsoon and monsoon season causes concern because during monsoon season aerosol concentration is expected to decrease because of removal from the atmosphere during precipitation and reduction in aerosol injection in to the atmosphere due to wet cloud condition. However, increased developmental work and enhanced vehicular circulation due to developmental and other activities may compensate. During the post-monsoon season higher values of mean AOD (0.805 ± 0.193 MICROTOPS-II and 0.739 ± 0.233 from MODIS) were recorded in 2017. In 2016 lower values of AOD (0.655 ± 0.194 from MICROTOPS-II and 0.636 ± 0.187 from MODIS) were observed. This may be due to availability of only few AOD data in the month of November 2016. The highest seasonal mean AOD was recorded during the winter of 2017, which was 0.867 ± 0.332 from MICROTOPS-II, 0.923 ± 0.473 from MODIS. Similarly lower mean value for winter season in 2016 from MICROTOPS-II - 0.737 ± 0.304 and MODIS - 0.698 ± 0.366 may be due to unavailability of data in December 2016 and partly in January 2017.

The highest monthly mean AE from MICROTOPS-II was 0.966 ± 0.069 in January 2017; and 0.964 ± 0.134 in December 2017. The high AE values recorded during post-monsoon and winter seasons were attributed to the accumulation of aerosols from urban sources and biomass/crop residue burning emissions within the shallow boundary layer. However, the lowest monthly mean AE was recorded 0.383 ± 0.169 in May 2017 along with other low values during pre-monsoon season. In pre-monsoon season AE values were generally smaller and less than 1.0 which indicated the presence of larger size of particulates.

Table 2
Seasonal variations of correlation coefficients (r) between PM, its ratio with meteorological parameters over Varanasi.

Seasons	Temperature	Equations	RH	Equations	WS	Equations
Pre-monsoon						
$PM_{2.5}$	0.097	Y = 0.528x+30.261	−0.083	Y = -0.142x+54.186	−0.295	Y = -2.538x+53.097
PM_{10}	0.104	Y = 1.083x+118.047	−0.451	Y = -1.481x+238.330	−0.040	Y = -0.667x+151.625
$PM_{2.5}/PM_{10}$	0.075	Y = 2.026x+28.586	0.101	Y = 8.661x+57.184	−0.429	Y = -7.303x+5.190
Post-monsoon						
$PM_{2.5}$	0.012	Y = 0.210x+121.337	0.052	Y = 0.319x+99.762	−0.229	Y = -6.720x+141.317
PM_{10}	−0.367	Y = -7.519x+352.461	−0.161	Y = -1.112x+253.752	−0.318	Y = -10.502x+182.613
$PM_{2.5}/PM_{10}$	0.370	Y = 2.008x+23.891	0.057	Y = 0.916x+83.730	0.125	Y = 0.420x+1.811
Winter						
$PM_{2.5}$	−0.429	Y = -7.256x+253.200	0.541	Y = 1.872x-40.669	−0.258	Y = -4.711x+128.131
PM_{10}	−0.470	Y = -13.766x+443.697	0.497	Y = 2.976x-64.652	−0.276	Y = -8.732x+206.022
$PM_{2.5}/PM_{10}$	−0.024	Y = -0.414x+18.706	0.145	Y = 12.330x+77.618	−0.013	Y = -0.211x+1.988

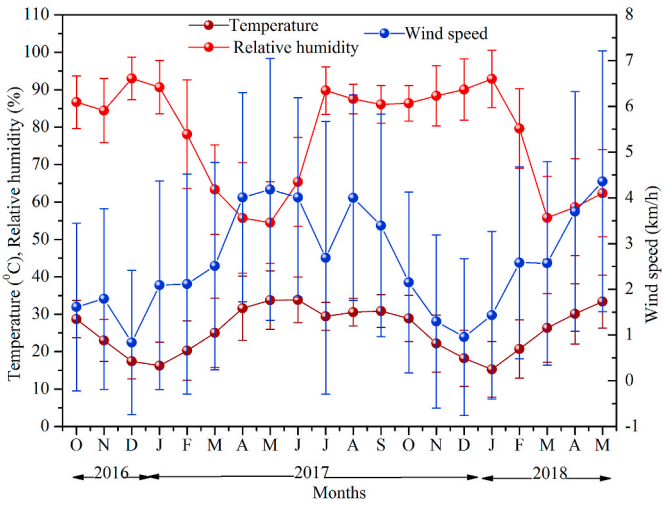

Fig. 5. Monthly mean variation of the meteorological parameters, i.e. temperature, relative humidity, wind direction, and wind speed over Varanasi for October 2016–May 2018.

These may be due to burning of agricultural field products and dust aerosols from Thar Desert and Sahara Desert (Singh et al., 2004).

The high variability in AOD and AE may be due to aerosol particles were from different emission sources (Kaskaoutis et al., 2013). Figure (4) showed A significant positive correlation between MICROTOPS-II and MODIS AOD during pre-monsoon (r = 0.736), monsoon (r = 0.602), post-monsoon (r = 0.59) and winter (r = 0.703) seasons which revealed accuracy and validation of satellite data. The regression line slope away from unity indicated the probability of some inconsistency between microphysical and optical retrieval model used which could not represent the existing situation (Zhao et al., 2002).

5. Influence of regional meteorology on particulate mass

Meteorological conditions are strong regulators of particulate loading in both local and regional scales. Correlation of particulate concentrations and meteorological factors could provide a quantitative extent of such impact. Ideally, individual meteorological variable influences particulate loading in different ways while the impact of ambient environment is cumulative effect and the result may become significantly different. Correlation coefficients (r) between PM, its ratio and meteorological parameters are given in Table 2. Table 2 showed that temperature and wind speed were inversely correlated with particulate loading while relative humidity during winter season was found to play a significant association with $PM_{2.5}$ (r = 0.541) and with PM_{10} (r = 0.497). Similar relationships between meteorological parameters and particulate ratio were observed during pre-monsoon, post-monsoon and winter seasons. To decipher the meteorological implications aerosol mass concentration was monitored twice a week and average for each individual month and was obtained and compared with the monthly average meteorological data.

5.1. Influence of temperature on aerosols

During pre-monsoon months May and June temperature usually remain high. For example in 2017, monthly mean temperature in May was 33.76 ± 7.82 °C and in June 33.83 ± 6.06 °C, where during May 2018 it was 33.34 ± 7.06 °C. An increase in ambient temperature is usually associated with the possibly of enhanced wind circulation which increases atmospheric dispersion (Murari et al., 2017). This may cause decrease in aerosol mass concentration. But increased temperature associated with reduced humidity increases the potential of generating windblown crustal materials to the environment as a result aerosol increases (Kumar et al., 2015). The resultant impact will depend on the efficiency of these two processes. The monthly mean temperatures were observed very low during the months of December 2017 (17.421 ± 4.709 °C), 2018 (18.229 ± 7.471 °C) and January 2017 (16.247 ± 6.335 °C), 2018 (15.258 ± 7.424 °C). Decreasing temperature and higher relative humidity level during winter months possibly assisted in aerosol accumulation and hence higher AODs. Further, the convection of winds because of varying temperature and fluctuations in the boundary layer could also influence the aerosols generation and transportation may affect aerosol concentration. Fig. 5 show monthly mean variation of meteorological parameters, i.e., temperature, relative humidity, and wind speed, for October 2016–May 2018. The minimum temperature occurs in the January months and there after increases continuously until June and then slightly decreases during monsoon and post-monsoon months with a minimum in the winter. Thus a significant temperature variation observed over Varanasi. The present analysis showed almost no significant correlation during pre – monsoon period. During post monsoon $PM_{2.5}$ did not any correlation, however PM_{10} showed negative (r = - 0.37) correlation. During winter both $PM_{2.5}$ and PM_{10} showed negative (r = - 0.43 and – 0.47) correlation. The temperature dependence needs further investigation because of opposite impact due to dispersal and injection impacts as indicated earlier.

5.2. Influence of relative humidity on aerosols

The relative humidity usually remains high during monsoon, post-monsoon and winter months. However, comparatively lower value was observed during pre-monsoon months. The monthly mean relative humidity during December 2016 and January 2017 was respectively 93 ± 5.698% and 90.677 ± 7.12%. The highly humid conditions (relative humidity > 85%) is mostly credited to the hygroscopic escalation of fine particulates over the region (Altaratz et al., 2014). The monthly mean relative humidity values are given in Fig. 5, which show almost similar values in March, April and May of 2017 and 2018 with slight variation in magnitude. The monthly mean relative humidity was found to increase from June 2017 (65.4 ± 11.883%) up to January 2018 (92.871 ± 7.676%). During the monsoon season, tropospheric ozone reached its lowest value over middle IGP due to the influence of decreased solar radiation associated with high humidity. Monsoon months having high relative humidity usually associates with the amount of rain which essentially reduce the implications of humidity in particulate loading (Murari et al., 2017). However, during winter season relative humidity exhibited strong association with $PM_{2.5}$ (r = 0.541) and PM_{10} (r = 0.497) because of lower probability of rain. During wet winter, high relative humidity in lower atmosphere helps to induce hygroscopic growth of particulates and thereby increases PM loadings (Kumar et al., 2015). During pre and post – monsoon season $PM_{2.5}$ almost did not exhibit any perceptible relation (Table 2). Although PM_{10} showed negative relation with r = - 0.45 (pre – monsoon) and r = - 0.16 in the post – monsoon. Pre-monsoons over IGP are relatively dry while winters are presumably wet.

5.3. Influence of wind speed and direction on aerosols

Wind is the carrier and the influential component of aerosol dispersal from one place to the other. Fig. 6 shows the wind vector distribution at Varanasi for the study period. During pre-monsoon months wind vector were relatively high with westerly and south westerly dominance. The calm wind conditions have only 26.09%. The air mass carries dry dust particles from the western Thar Desert and Middle East countries to the study region. In these months frequent dust storms from western region also occur which enhances dry aerosols in the region and makes the dry weather condition. Winds during monsoon months are easterly and south westerly with about 27.05% calm conditions and carry moisture causing drastically increased humidity. During post – monsoon easterly and south westerly winds occur with relatively calm condition about

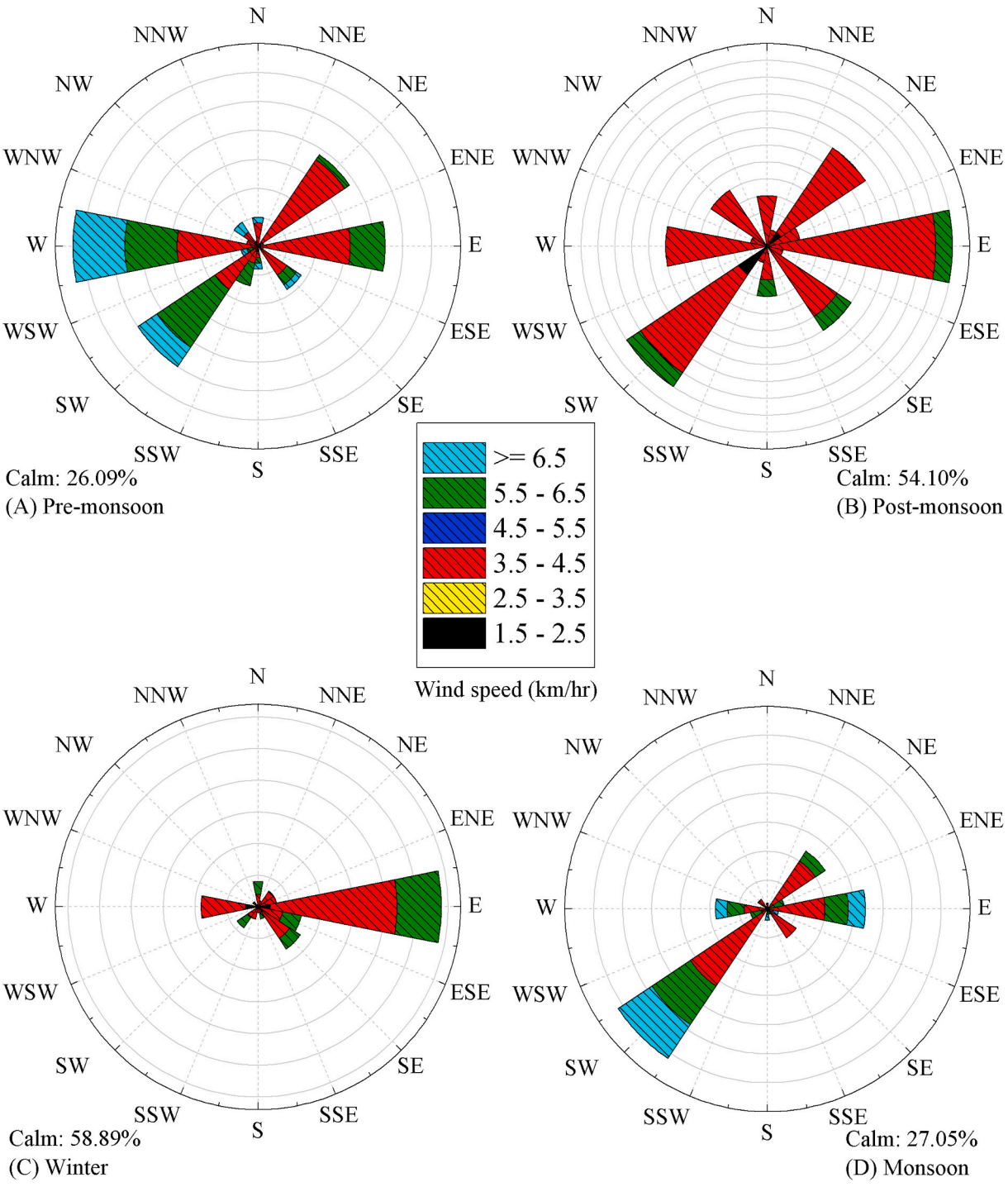

Fig. 6. Variations of wind profiles during (A) Pre-monsoon, (B) Post-monsoon, (C) Winter and (D) Monsoon seasons over Varanasi, India.

54.10%. During winter months winds are usually easterly with calm condition 58.89%. The monthly mean wind speed was relatively high monthly mean speed during pre-monsoon month May 2017 (4.182 ± 2.868 km/h) and 2018 (4.361 ± 2.851 km/h) compared to the winter months of December 2016 (0.836 ± 1.574 km/h) and December 2017 (0.956 ± 1.716 km/h). The atmospheric conditions over IGP during winter months are dominated by high relative humidity, calm winds, and shallow atmospheric boundary layer which led to intense fog and haze (Mishra and Shibata, 2012), which resulted in low dispersion of aerosols. Calm and weak winds from south-western and eastern directions during the post-monsoon and winter seasons are unfavorable conditions for aerosol dispersion and thus help the temperature inversions formation and accumulation of aerosols and air pollutants. Dry pre-monsoon with relatively high wind speed induces coarser crustal materials which increases PM_{10} concentrations. The relation between wind and aerosol particulates (both $PM_{2.5}$ and PM_{10}) is negative and varies with season (Table 2). A negative association between wind speed and particulates signifies the implications of surface loading from local to regional sources. The effect of local sources including regional acts of biomass-waste burning, vehicular emissions and resuspension of crustal components are associated with negative correlation between wind speed and aerosols (Kumar et al., 2015).

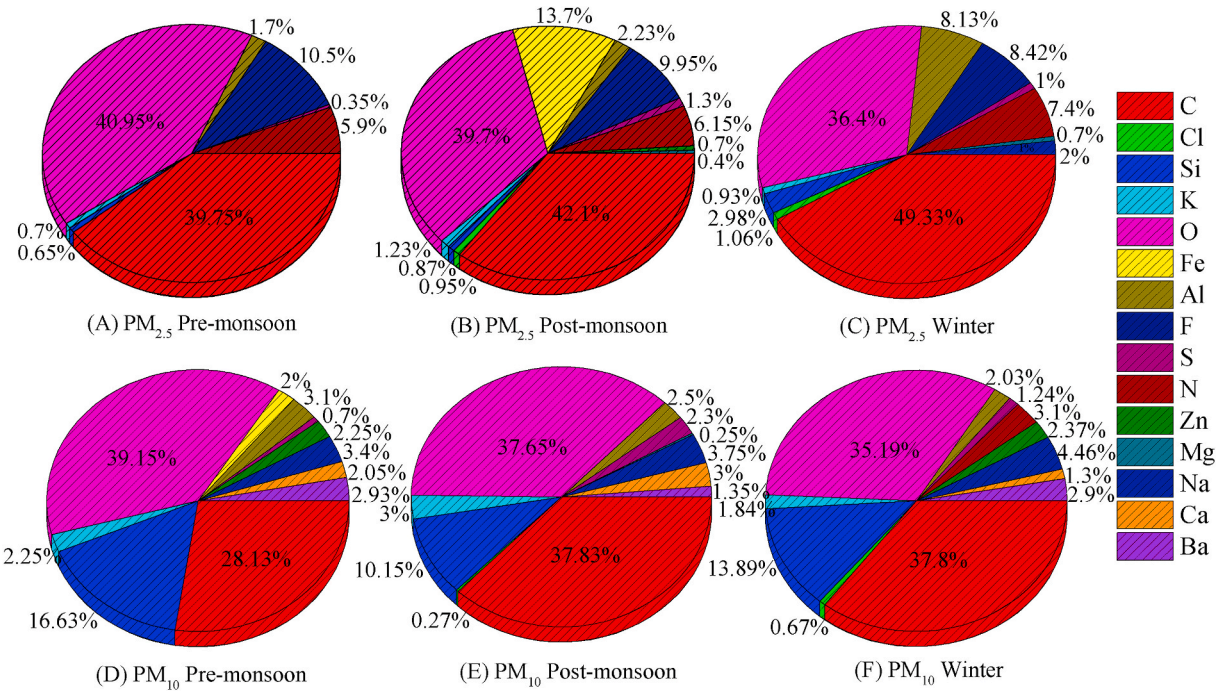

Fig. 7. Seasonal variability in particulate elemental compositions using SEM-EDX analysis over Varanasi.

Table 3
The elemental percentage composition in $PM_{2.5}$ and PM_{10} over Varanasi from SEM-EDX analysis.

Elements	$PM_{2.5}$			PM_{10}		
	Pre - monsoon	Post - Monsoon	Winter	Pre - monsoon	Post - Monsoon	Winter
C	39.75%	42.1%	49.33%	28.13%	37.83%	37.8%
O	40.95%	39.7%	36.4%	39.15%	37.65%	35.19%
F	10.5%	9.95%	8.42%	–	–	–
N	5.9%	6.15%	7.4%	–	–	3.1%
Fe	–	13.7%	–	2%	–	–
Al	1.7%	2.23%	8.13%	3.1%	2.5%	2.03%
Si	0.65%	0.87%	2.98%	16.63%	10.15%	13.89%
Na	–		2%	3.4%	3.75%	4.46%
Ba	–			2.93%	1.35%	2.9%
Ca	–	–	–	2.05%	3%	1.3%
K	0.7%	1.23%	0.93%	2.25%	3%	1.84%
Cl		0.95%	1.06%		0.27%	0.67%
S	0.35%	1.3%	1%	0.7%	2.3%	1.24%
Zn		0.7%		2.25%		2.37%
Mg		0.4%	0.7%		0.25%	

6. Elemental characteristics by SEM-EDX analysis

For the better understanding of the elemental composition, sources and transport of pollutant, SEM-EDX analysis was done. Fig. 7 A–F shows the seasonal variability in particulate elemental compositions using SEM-EDX analysis. The elemental percentage composition in $PM_{2.5}$ and PM_{10} are given in Table 3. The high C content during almost all the seasons may be due to automobile fuels being exhausted from automobile vehicles during highly choked traffic level. Increasing vehicular traffic (Choudhary et al., 2019) and anthropogenic activities may be the source of C, F, Si, Fe and Al aerosol particles in the air over the region. Presence of Ca and Na in the aerosol samples may affect hygroscopicity of the air. However, K element present with C and O in aerosol samples may be due to biomass burning, a common practice of local residents in Varanasi region (Murari et al., 2015).

Varanasi being one of the oldest and densely populated cities of India contains variety of small scale industries which are the sources of various types of pollutants. In addition, coal burning industries, coal fired fixed chimneys, open refuge burning, home furnaces, kitchen stoves etc widely utilized for common purposes. The presence of F elements within finer particulates may be associated with industrial emissions (Pipal et al., 2014). The presence of Fe, C and Si in fine particulates may be mixture of smaller size Si and ferrous oxide with absorbed C soot particles. Fly ash produced by industrial burning may contain Si (Tegen and Kohfeld, 2006). Association of S possibly signifies additional contribution from biomass/fuel combustion (Paoletti et al., 2003). The trace metals such as Ca, Fe, Zn, Na, K, Al, S, Cl, K, N, Zn, Mg, Na and Ba are mainly accumulated from the soil/earth crust, road dust and re-suspended dust particles over the site. Small presence of Cl having carbonaceous aerosols with traces of K and Si indicated about the contribution by regional livelihoods from farming and burning practices (Murari et al., 2016). Fig. (8A) and figure (8B) show the example of morphological representation of aerosol particles in $PM_{2.5}$ and PM_{10} samples respectively by SEM-EDX analysis.

7. Chemical characteristics using IC analysis

The presence of ionic contents in the collected samples of aerosol

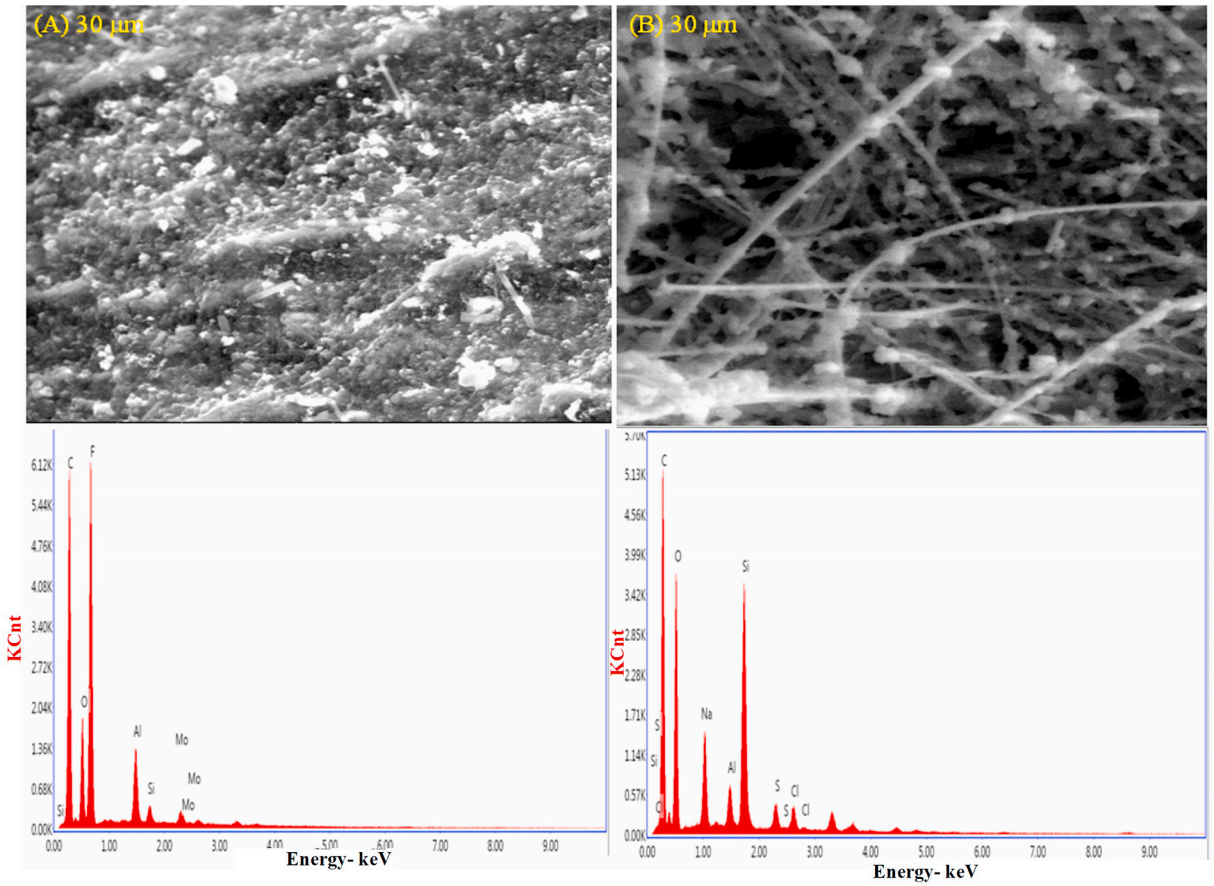

Fig. 8. (A) Morphological representation of aerosol particles in $PM_{2.5}$ samples by SEM-EDX analysis. (B) Morphological representation of aerosol particles in PM_{10} samples by SEM-EDX analysis.

Table 4
The presence of ionic contents in $PM_{2.5}$ and PM_{10} over Varanasi from IC analysis.

Elements	$PM_{2.5}$			PM_{10}		
	Pre - monsoon	Post - Monsoon	Winter	Pre - monsoon	Post - Monsoon	Winter
SO_4^{-2}	20.41%	18.61%	20.10%	21.26%	19.29%	20.55%
NO_3^-	18.06%	16.66%	19.72%	17.18%	16.36%	18.46%
Cl^-	10.14%	9.62%	8.45%	9.66%	9.43%	9.99%
Ca^{2+}	13.04%	13.03%	11.48%	14.14%	13.31%	11.21%
Na^+	11.28%	13.81%	14.42%	13.05%	12.91%	14.76%
K^+	9.18%	13.01%	11.34%	9.63%	12.32%	12.35%
F^-	11.31%	9.11%	8.31%	9.23%	8.26%	6.18%
Li^+	2.04%	2.22%	1.11%	1.31%	2.10%	1.03%
Mg^{+2}	4.53%	3.94%	5.05%	4.53%	3.98%	5.44%

from IC analysis showed the following trend: $SO4^{2-} > NO_3^- > Cl^- > F^-$ in both $PM_{2.5}$ and PM_{10} samples during all the seasons (Table 4). The cations showed the following trend: $Na^+ > Ca^{2+} > K^+ > Mg^{2+} > Li^+$ in $PM_{2.5}$ and PM_{10} samples. These harmful and problematic chemical species may have different sources. One of the major sources could be carpet and Saree industries abundantly operative in and around Varanasi which emit significant amount of chemicals into the environment and may be one of the possible sources of anions. In the urban zone of Varanasi, the ionic substance of the aerosols has demonstrated the following pattern: $Na^+ > SO4^{2-} > Ca^{2+} > Cl^- > Mg^{2+} > NO_3^- > K^+ > HCO_3^+ > F^-$. In the suburban region Singh et al. (2014) reported $Na^+ > SO4^{2-} > NO_3^- > Ca^{2+} > Cl^- > Mg^{2+} > K^+ > HCO_3^+ > F^-$. They found lesser amount of F^-, although in the present study enhanced amount of F^- was observed.

From Table 4 it was observed that, $SO4^{2-}$ concentration was maximum in both types of aerosol samples, whereas concentration of Li^+ ion was observed minimum. Srimuruganandam and Nagendra (2011) reported $SO4^{2-}$, NO_3^- and Mg^{2+} as dominant species in the urban areas of India. In the current investigation $SO4^{2-}$ and NO_3^- were observed to be higher while the amount of Mg^{2+} was obtained in lesser amount.

Tiwari et al. (2009) reported SO_4^{-2} to be the highest concentration over Delhi among all the chemical species present in both finer and corser aerosol samples, whereas the concentration of F^- was the lowest. The increasing amount of F^- in the atmosphere of the Varanasi should be further studied. SO_4^{-2} could be originated from fuel combustion, industries and transportation (De, 2006). Murari et al. (2016) reported negligible contribution of sea salt aerosol over Varanasi whereas in the present study sea salt was found to have significant contribution. Pipal et al. (2014) reported lesser amount of sea salt contribution over Agra as compared to the Varanasi. Fig. 9 shows the seasonal variation in aerosol chemical constituents along with their standard deviation values in $PM_{2.5}$ and PM_{10} during (A) Pre-monsoon, (B) Post-monsoon and (C)

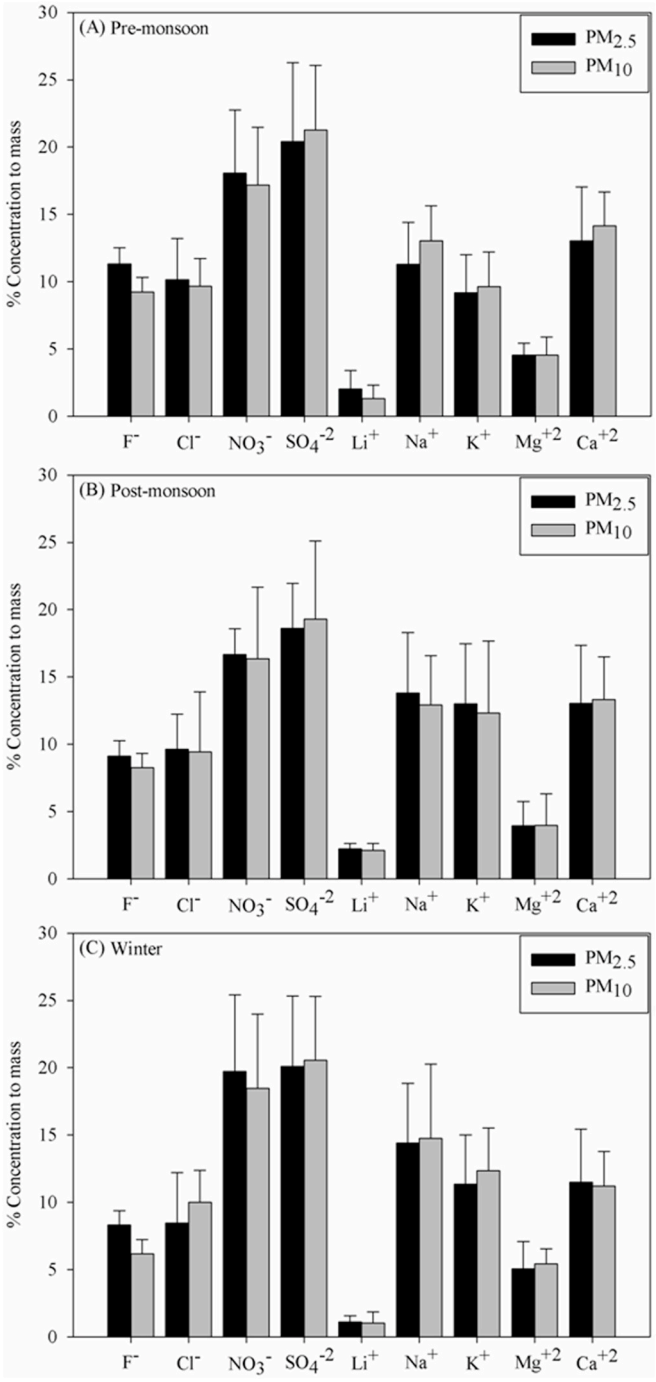

Fig. 9. Seasonal variation in aerosol constituents with standard deviation during (A) Pre-monsoon, (B) Post-monsoon and (C) Winter seasons by IC analysis over Varanasi.

Winter seasons.

In the present study, SO_4^{2-}, NO_3^-, and F^- may be from the source of fuel burning. It is also known that Ca^{2+}, Mg^{2+}, Cl^- and Na^+ generally originated from soil, while SO_4^{2-}, NO_3^-, F^- and K^+ may have originated from anthropogenic activities. Higher concentrations of $SO_4^{2-}$are also attributed to the burning of coal which is widely used in cooking and heating processes in and around the city. NO_3^- is generally emitted from vehicles exhausts and Diesel generators. These are frequently used in Varanasi and aerosols and thus NO_3^- concentration. Elevated fractions of NO_3^- and K^+ in the samples of winter also signified the contribution of biomass burning (Murari et al., 2016). Major sources of F^- might be due to coal burning (Safai et al., 2005), fertilizers and pesticide uses. The high concentrations of Cl^- may be attributed to resuspension of road dust including shows some anthropogenic emissions emitted from wood burning and tobacco smoking processes. The higher fractions of K^+ ions might be emissions from biomass/wood burning (Niemi et al., 2005). Mg^+ and Ca^{2+} elements could originate from the crustal and road-dust re-suspension (Banerjee et al., 2015). The presence of higher fractions of Na^+ and Ca^{+2} in the sample may be accounted from the long range transport of minerals dust and sea salts (Murari et al., 2015). The elemental analysis of the aerosols establishes the presence of some elemental components in large amount which clearly shows significant decline of regional air quality leading to negative health impacts.

8. Conclusions

Measurements of aerosol concentrations using MICROTOPS–II are carried out during October 2016–May 2018. Observed data were categorized and analyzed in order to understand the nature, source of origin and variability with seasons. Based on the study following conclusions may be derived:

(i) Higher concentration of aerosols (both $PM_{2.5}$ and PM_{10}) were observed during the winter months (November, December, January) as compared to the pre – monsoon months (March, April, May) when higher temperature, lower humidity and relatively higher wind velocity were observed. In the pre monsoon period dust storms and emission from the Earth's surface due to higher temperature and wind velocity dump more aerosol particles into the atmosphere. However, it seems source during winter months were found to be dominants. This point should be further exposed with larger amount of data sets.

(ii) Concentration of $PM_{2.5}$ was found to be higher than that of PM_{10} during the whole period of observation. A linear relation was obtained between them. The coefficients and constants of linear relation varied from season to season. This result could be useful for modelers using aerosols in their work. The ratio $PM_{2.5}/PM_{10}$ more than 0.5 during the post-monsoon and winter months showed fine mode anthropogenic particles dominance while during pre-monsoon months (less than 0.5) represented the presence of coarse particles. The burning of agricultural residue, solid waste and biomass around Varanasi and regulated by prevailing meteorological factors were found to be the cause of fine mode particles domination whereas the polluted continental and mixed types of aerosols were more frequent in the pre-monsoon months.

(iii) The effect of local meteorological parameters (temperature, humidity and wind) on $PM_{2.5}$ and PM_{10} distribution were studied. Temperature effect could not be sorted out because of its association with wind and humidity which have rival impacts. Moreover data sets were limited. Controlled field experiments are needed in which one parameter is varied keeping the other two parameter constants. Wind showed a negative correlation in all the season which indicated dispersal impact along with the impact of boundary layer. This also suggested the importance of local sources than long range transportation from far away region.

(iv) The percent content of various elements and ionic charges were studied to understand the sources of aerosols in the atmosphere of Varanasi during different seasons. Enhanced concentration of observed elements and ionic charges suggested the sources to be small and large scale industries, biomass burning, coal burning and vehicular activities.

In order to improve quality of life (air quality), it is suggested to reduce emissions from industries, stop burning of agricultural products and coal and reduce vehicle flux on the road. Further studies are

required both with large scale data set over extended period and with controlled temperature/humidity/wind speed.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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