Source Apportionment of PM_{10} Over Three Tropical Urban Atmospheres at Indo-Gangetic Plain of India: An Approach Using Different Receptor Models

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Abstract

The present work is the ensuing part of the study on spatial and temporal variations in chemical characteristics of PM_{10} (particulate matter with aerodynamic diameter $\leq 10\ \mu m$) over Indo Gangetic Plain (IGP) of India. It focuses on the apportionment of PM_{10} sources with the application of different receptor models, i.e., principal component analysis with absolute principal component scores (PCA-APCS), UNMIX, and positive matrix factorization (PMF) on the same chemical species of PM_{10}. The main objective of this study is to perform the comparative analysis of the models, obtained mutually validated outputs and more robust results. The average PM_{10} concentration during January 2011 to December 2011 at Delhi, Varanasi, and Kolkata were 202.3 ± 74.3, 206.2 ± 77.4, and $171.5 \pm 38.5\ \mu g\ m^{-3}$, respectively. The results provided by the three models revealed quite similar source profile for all the sampling regions, with some disaccords in number of sources as well as their percent contributions. The PMF analysis resolved seven individual sources in Delhi [soil dust (SD), vehicular emissions (VE), secondary aerosols (SA), biomass burning (BB), sodium and magnesium salt (SMS), fossil fuel combustion, and industrial emissions (IE)], Varanasi [SD, VE, SA, BB, SMS, coal combustion, and IE], and Kolkata [secondary sulfate (Ssulf), secondary nitrate, SD, VE, BB, SMS, IE]. However, PCA-APCS and UNMIX models identified less number of sources (besides mixed type sources) than PMF for all the sampling sites. All models identified that VE, SA, BB, and SD were the dominant contributors of PM_{10} mass concentration over the IGP region of India.

Worsening air quality has become the foremost perturb in the world, causing approximately 7 million global deaths as revealed by World Health Organization (WHO) (2014)

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and 3.2 million annual deaths solely due to airborne fine particulates (WHO 2014). Particulate matter (PM) is a multicomponent mixture of solid and liquid particles suspended in air (Ramgolam et al. 2009). They originate from a wide range of sources and transform eventually through numerous microphysical processes, such as coagulation, nucleation, and condensation, which scavenge off through wet and dry deposition (Perrino et al. 2011). The role of atmospheric PM is being increasingly recognized in Earth's climate system due to its potential to exert climate forcing by affecting the Earth's radiative balance directly by the extinction of the incoming solar radiation and indirectly by acting as cloud condensation nuclei (CCN) (IPCC 2013). In the past two decades, health-related issues are statistically associated with worsening air quality due to the presence of aerosols in the atmosphere (Pope et al. 2009; Li et al. 2004). Recently, several studies about the increasing risks of myocardial ischemia, systemic inflammation, and blood pressure due to traffic and other combustion related exposures have unveiled the significant role of PMs size, composition, and sources in

health-related concerns (Dachs and Eisenreich 2000; Sharma et al. 2014a; Brauer et al. 2015; Karagulian et al. 2015).

To assess aerosols effects on air quality and climate, it is crucial to understand their primary and secondary sources along with their characteristics in time and space. The exigency to comprehend the potential source categories and their contributions has become imperative to reduce the PM pollution. The US EPA receptor models and other mathematical or statistical procedures essentially quantify the contribution of individual sources to particulate loading at a particular location, based on source and receptor characteristics and in certain cases, with the nature of pollutants (Hopke 2016; Hopke et al. 2006; Kong et al. 2010). Fundamentally, the three selected receptor models used for the present study, i.e., principal component analysis (PCA) with absolute principal component score (APCS), UNMIX, and positive matrix factorization (PMF), adopt similar chemical mass balance procedures. The assumptions include that the chemical species remain unchanged from their respective sources to receptor location, and their time dependence will be same at the receptor sites coming from the same sources (Belis et al. 2013). These receptor models are based on the following equation:

$$X = GF + E \qquad (1)$$

All of the variables denote a matrix: X for concentration, G for source contribution, F for source profile, and E for unfit elemental concentration (Paatero and Tapper 1994; Hopke et al. 2006). These receptor models based on factor analysis require quantitative data of the chemical characteristics of PM along with little qualitative knowledge about different sources at the receptor site (Pant and Harrison 2012). The theoretical and statistical approaches that define these models have already been explicated in detail in a number of research articles (Thurston and Spengler 1985; Paatero 1999; Henry 2003; Hopke 2016; Song et al. 2006; Chen et al. 2007; Zheng et al. 2007; Viana et al. 2008; Begum et al. 2010; Harrison et al. 2011; Gugamsetty et al. 2012; Pant and Harrison 2012; Shi et al. 2014; Sharma et al. 2014a, b, 2015; 2018a, b; Cesari et al. 2018; Jain et al. 2017a, b). In respect of source apportionment studies of PM_{10} in India, the majority of studies were performed using PCA and classic factor analysis, which has drastically been shifted to PMF model recently, accredited to its idiosyncrasies (Banerjee et al. 2015). PMF is efficient in handling missing or below detection level data as well as includes individual level data uncertainties. PCA investigates the structure and variation in a dataset and accordingly fetches the pattern in the data (Viana et al. 2008). UNMIX is the least studied model in India, i.e., only 3% studies have been reported through the year 2014 (Banerjee et al. 2015). UNMIX exploits the single-value decomposition method for procuring the number of contributing sources to PM. Further explications on different aspects of these models are available in the next section of this paper.

The present work, which is an ensuing part of the previous study published by our group (Sharma et al. 2016b) on spatial and temporal variations in chemical characteristics of PM_{10} over IGP of India, deals in the application of different receptor models (i.e., PCA-APCS, UNMIX, and PMF) on the same data set to obtain the mutually validated outputs and more robust results. The study adopts a synergistic approach to monitor the different parameters, coupling different models known to achieve in a united manner what could not have been achieved alone. The overall goal of this study is to narrow the gaps in the understanding of region-specific potential sources of PM_{10} over urban cities of IGP of India. The study accounts the detailed analysis of aerosol particles' chemical properties and quantifying the contribution of each and every source to PM_{10} pollution. IGP, known to be the major food basket of India, has ranked among the world's most densely populated and intensely farmed areas. IGP is one of the most expeditiously growing regions in terms of industrialization and urbanization. The region is treacherously congested, overpopulated, and majorly culpable of contributing PM pollution throughout India and Indian subcontinents, which is burgeoning from the sources, such as fossil fuel combustion, biomass burning, and agricultural activities. This study aids in deciphering the intricacies involved in source apportionment of PM_{10} over the IGP region using receptor models, such as PCA-APCS, UNMIX, and PMF.

Materials and Methods

Experimental Sites

The IGP, popularly called the north Indian river plain, is an extensive alluvial crescent, stretching from the Indus river system in Pakistan, encompassing most of northern and eastern India, to the Ganges delta in Bangladesh. The IGP is a 255-million hectare (ha) fertile plain, comprising homogenous topography and has ranked among the world's most densely populated and intensely farmed areas (Sharma et al. 2016b). The sampling sites selected for the present study were chosen to represent the vast stretch of IGP in the Indian subcontinent, having exceedingly polluted atmospheric conditions, i.e., Delhi, Varanasi, and Kolkata, which epitomizes the upper, middle, and lower regions of IGP, respectively (Fig. 1). The chosen sites represent the characteristic urban environments and exemplify the concoction of local sources and transboundary features of atmospheric pollutants.
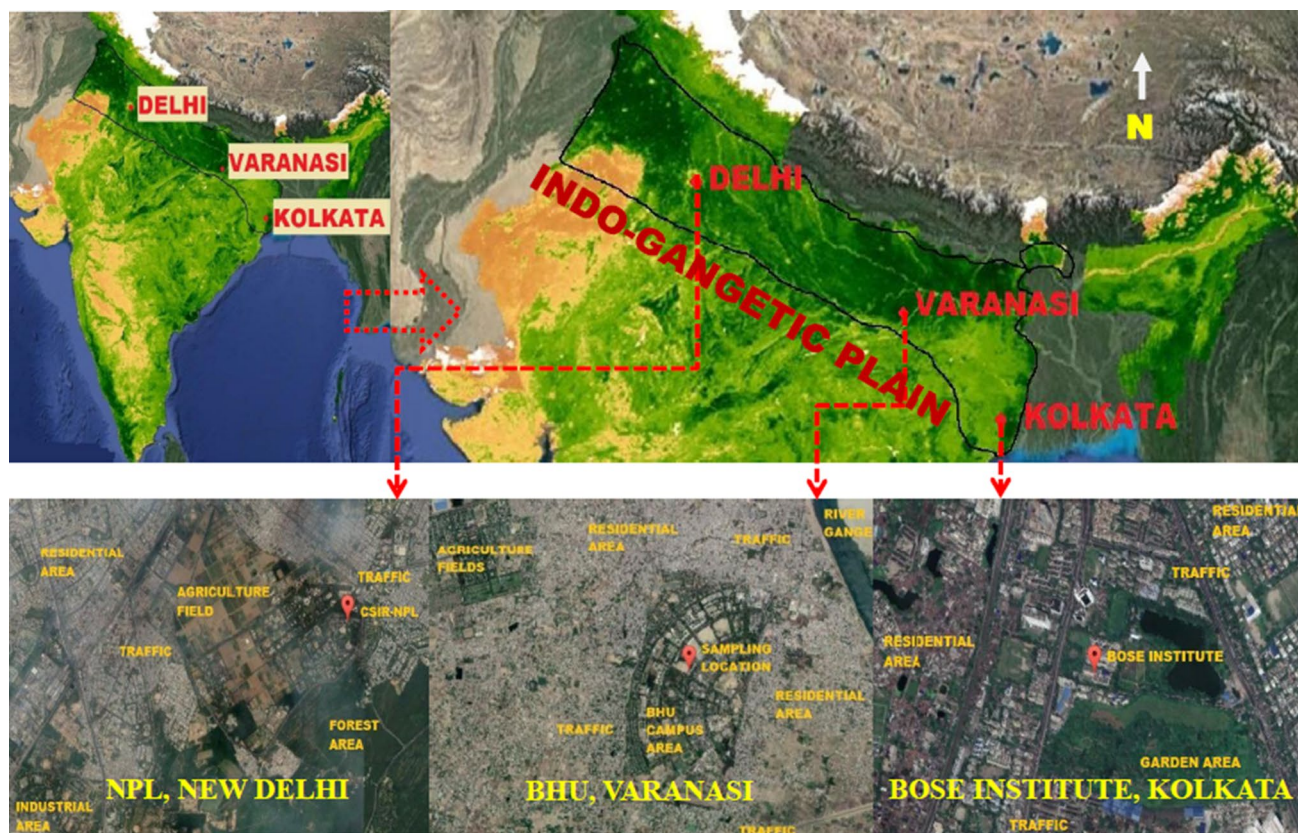
Fig. 1 Map of observational sites (Delhi, Varanasi, and Kolkata), IGP, India

Delhi

Delhi, a megacity of India, is situated in the northern part of the country and is enclosed by Himalayan ranges in the north, IGP in the east, central Plains in the south, and Thar Desert in the west. Delhi observes semiarid climatic conditions with dry and scorching summers, humid monsoons, followed by chilly winters (Goyal and Sidhartha 2002; Jain et al. 2017b). Furthermore, Delhi is notorious for its severe fog and haze conditions during winters and heavy dust storms during summers, which causes an influx of mineral dust to the aerosol loading (Ram and Sarin 2010). PM_{10} samples were collected on the roof [at a height of ~10 m above ground level (AGL)] of CSIR-National Physical Laboratory, New Delhi [28°38′N, 77°10′E; 218 m above mean sea level (amsl)]. The sampling location signifies the typical urban environment, bounded by the heavy traffic-congested area in the southeast (SE), agricultural fields in the southwest (SW), and institutional and residential area in north direction. In addition to ever-increasing vehicular growth, many large- and small-scale industries are present in and around Delhi.

Varanasi

Varanasi, the spiritual capital of India, is located on the banks of the river Ganges, in the north Indian state of Uttar Pradesh and has grown into an important industrial center in recent past years. Varanasi observes a typical humid subtropical condition with hot and dry summers (temperature range: 22–46 °C) and diurnally varied winters (warm days and downright cold nights). PM_{10} samples were collected on the roof (at ~10 m AGL) of the Department of Geophysics Banaras Hindu University (25°18′N, 83°03′E; 129 m amsl), Varanasi. The sampling area represents a characteristic urban environment enclosed by heavy traffic area on one side and residential and commercial expanse on others. Details of the site description are available in our previous paper and reference therein (Sharma et al. 2016b).

Kolkata

Kolkata is the capital of West Bengal (eastern Indian state). The city, located on the eastern bank of the Hooghly River (spread along 80 km), is the East India's prime commercial and educational center. The city with 4.5 million population lies within the lower Ganges delta of Eastern India, with

1.5–9 m elevation (amsl: above mean sea level). Kolkata observes tropical wet and dry climate with annual mean temperature of 26.8 °C. The city experiences 1800 mm of annual rainfall between June and September, which is majorly brought by the southwest summer monsoon. Samples of PM_{10} were collected on the roof (at ~ 10 m AGL) of Bose Institute (22°33′N and 88°20′E; 9 m amsl), which represents a characteristic urban environment of the megacity Kolkata.

PM_{10} Sampling

PM_{10} samples were collected periodically (4–5 samples per month) on pre-combusted (at 550 °C for at least 5 h to eradicate the organic impurities) quartz microfiber filters (QM-A; size: 20×25 cm^2) during January 2011–December 2011 at Delhi ($n = 50$), Varanasi ($n = 45$), and Kolkata ($n = 54$) of IGP, India, using respirable dust samplers (RDS) (model: PEM-RDS 8NL, Make: M/s. Polltech Instruments, Mumbai, India). The flow rate of the instrument was calibrated with top loading orifice calibrator traceable to national standard (accuracy: $\pm 1\%$ of full scale). The flow meters also were calibrated (with the accuracy of $\pm 2\%$ of full scale) with air flow calibrator traceable to national standard having the average flow rate of ~ 1.13 m^3 min^{-1}. The sampling was done for 8 h on a day and night basis [during the daytime (1000–1800 h) and nighttime (1900–0300 h)]. In general, rush hour (huge traffic influence) started from 0900 to 1100 h and 1700 to 1900 h in Delhi (Sharma et al. 2016b); hence, the sampling had been started at 1000 h (at daytime) and 1900 h (at nighttime). The sample has been collected for 8 h to follow the common protocol (i.e., 8 or 12 or 24 h) of Central Pollution Control Board (CPCB), New Delhi, India. The sample filters were weighed before and after the sampling during the experiment to determine the mass of the PM_{10} collected. The concentration of PM_{10} (μg m^{-3}) was calculated on the basis of the difference between final and initial weights of the QM-A filters (measured by a microbalance; resolution ± 10 μg) and divided by the total volume of air passed during the sampling. After collecting the samples, filters were stored under dry conditions in the deep-freezer at -20 °C before analysis.

Chemical Analysis

The heavy and trace metals (Al, Ti, Zn, Fe, S, P, and B) were analyzed through a quantitative elemental nondestructive method using Rigaku ZSX Primus wavelength dispersive X-ray fluorescence spectrometer (WD-XRF). WD-XRF consists of spectrometer with scintillation counter (SC) for heavy elements and flow proportional counter (F-PC) for light elements, sealed X-ray tube for excitation, end window, and an Rh target. The instrument was set to vacuum conditions and measurements were done at a temperature of 36 °C and a tube rating of 2.4 kW. Blank filter measurements were also taken, which is further used for the intensity correction of exposed filters. Based on triplicate analysis of filters, analytical error or repeatability measurement was approximated to be 5–10%.

A punch of ~ 0.536 cm^2 area of each PM_{10} filter as well as blank filter were cut and analyzed for organic carbon (OC) and elemental carbon (EC) concentrations using OC/EC Carbon Analyzer (Model: DRI 2001A, Atmoslytic Inc., Calabasas, CA). The analyzer is based on the principle of preferential oxidation of OC and EC. OC volatilizes at lower temperatures and in a nonoxidizing helium atmosphere. Hence, initially the sample is heated in pure helium at four different temperatures, i.e., 140, 280, 480, and 580 °C to estimate the OC components (OC1, OC2, OC3, and OC4). EC volatilizes by an oxidizer through combustion process; therefore, the sample is further heated in the atmosphere of 98% helium and 2% oxygen at three different temperatures, i.e., 580 °C, 740 °C, and 840 °C to estimate the EC components (EC1, EC2, and EC3). The detailed analytical procedure is available in Chow et al. (2004) and reference therein. All the samples were analyzed in triplicate to estimate the repeatability error, and it was approximated to be 3–5%.

The estimation of water soluble inorganic components (WSICs) (Li^+, Na^+, NH_4^+, K^+, Ca^{2+}, Mg^{2+}, F^-, Cl^-, NO_3^-, and SO_4^{2-}) were performed by extracting 5×5 cm^2 (of 20×25 cm^2) of filter in deionized water (resistivity 18.2 MΩ-cm), and it was ultrasonicated for 90 min. The extract was then filtered through 0.22 μm nylon membrane filters. The filterate was then transferred to polypropylene sample bottles and analyzed by using Ion Chromatograph (Model: DIONEX-ICS-3000, Sunnyvale, CA). The concentrations of anions were obtained by using an Ion Pac-AS11-HC analytical column with a guard column, ASRS-300 4-mm anion micro-membrane suppressor, 20 mM of sodium hydroxide (NaOH; 50% w/w) as eluent, and deionized water as regenerator. Concentrations of cations were estimated by using a guard column with a separation column, suppressor CSRS-300, and 5 mM of methane sulfonic acid (MSA) as eluent. Chromeleon® software was used to process the chromatograms, and data of chromatography were collected at 5 Hz. Repeatability error was measured using triplicate analysis of filters and observed to be approximately 3–7%. Comprehensive details about the analytical procedure, calibration standards used, etc., in this study are available in our previous publication and reference therein (Sharma et al. 2014a, b). The values of method detection limit (MDL) of analyzed PM_{10} components has been reported in Table 1.

Table 1 The average concentrations of constituents of PM_{10} (µg m^{-3}) over IGP and coefficient of correlation (R^2) between measured and modelled concentrations along with S/N ratio in the base run of PM_{10} and MDL (method detection limit)

Species	Delhi				Varanasi				Kolkata			
	Average	R^2	S/N	MDL	Average	R^2	S/N	MDL	Average	R^2	S/N	MDL
PM_{10}	202.3 ± 74.3	0.86	6.6	–	206.2 ± 77.4	0.89	6.7	–	171.5 ± 38.5	0.82	5.4	–
OC	22.7 ± 7.4	0.64	2.0	0.87	16.9 ± 8.6	0.79	4.5	0.87	13.7 ± 5.3	0.81	5.4	0.87
EC	8.7 ± 3.9	0.70	2.6	0.38	7.3 ± 5.1	0.78	6.7	0.38	7.8 ± 4.0	0.80	8.0	0.38
SO_4^{2-}	11.67 ± 3.7	0.74	5.9	0.23	10.7 ± 6.0	0.85	8.3	0.23	10.0 ± 2.9	0.69	8.6	0.23
NO_3^-	10.6 ± 5.5	0.85	3.9	0.37	8.7 ± 6.6	0.92	8.6	0.37	6.5 ± 2.6	0.93	8.8	0.37
NH_4^+	5.2 ± 4.2	0.78	4.6	0.27	3.4 ± 4.1	0.95	6.8	0.27	7.0 ± 3.0	0.62	7.9	0.27
F^-	0.54 ± 0.38	0.48	2.7	0.29	–	–	–	–	0.85 ± 0.66	0.40	4.1	0.29
Na^+	3.51 ± 1.86	0.76	5.0	0.34	2.92 ± 0.94	0.86	9.4	0.34	5.40 ± 1.34	0.83	9.8	0.34
K^+	1.75 ± 0.74	0.85	2.3	0.64	1.69 ± 0.79	0.76	2.7	0.64	3.07 ± 1.92	0.90	2.7	0.64
Cl^-	4.49 ± 3.49	0.68	4.3	0.26	5.84 ± 4.63	0.89	6.8	0.26	9.54 ± 3.41	0.79	9.0	0.26
Ca^{2+}	4.75 ± 1.19	0.79	9.5	0.31	3.67 ± 1.53	0.91	9.4	0.31	4.93 ± 2.52	0.85	9.8	0.31
Mg^{2+}	0.75 ± 0.21	0.92	4.4	0.18	0.61 ± 0.29	0.88	3.8	0.18	0.78 ± 0.19	0.63	5.4	0.18
Al	2.06 ± 0.86	0.88	8.9	0.18	2.10 ± 1.31	0.95	8.7	0.18	0.76 ± 0.57	0.74	5.8	0.18
Ti	0.09 ± 0.04	0.77	5.3	0.03	0.09 ± 0.12	0.99	4.1	0.03	–		–	–
Fe	0.60 ± 0.49	0.99	10.0	0.07	0.11 ± 0.08	0.93	2.4	0.07	0.43 ± 0.66	0.98	3.5	0.07
P	0.29 ± 0.18	0.99	10.0	0.02	0.11 ± 0.05	0.98	10.0	0.02	0.07 ± 0.05	0.97	10.0	0.02
S	1.98 ± 1.00	0.96	9.4	0.12	1.03 ± 0.70	0.94	7.8	0.12	0.74 ± 0.52	0.88	7.1	0.12
Zn	0.02 ± 0.01	0.74	0.6	0.06	–	–	–	–	0.02 ± 0.01	0.58	0.7	0.06
B	2.25 ± 1.05	0.98	9.8	0.04	2.63 ± 1.50	0.92	10.0	0.04	2.57 ± 1.37	0.99	10.0	0.04

Descriptions of Models

PMF

US EPA PMF (5.0) was employed in the present study to determine the sources of PM_{10} and their contributions at the sampling sites. Comprehensive description and algorithms of PMF can be perceived through number of research papers (EPA PMF 5.0 user guide; Paatero and Tapper 1994; Paatero 1999; Jain et al. 2017a, b). The obligatory characteristics of the model concomitant with the present study have been elucidated in the supplementary information ("Appendix 1").

The standard uncertainty estimation (uncertainty associated with the analyzed data set) was calculated using the given equation (as per EPA PMF 5.0 user guide):

weighting factors (Jeong et al. 2017). In this study, species with less than 0.2 S/N ratios were excluded from analysis. Species with > 2 and between 0.2 to 2 S/N ratio represent strong and weak values (data quality), respectively. Species with weak data quality correspondingly contributes to the noise in the results (Sharma et al. 2016a, b). MDL values and PM_{10} concentration of each location are summarized in Table 1. Additionally, calculated S/N ratios and regression coefficients (R^2) between the measured and modeled values by the PMF model are also given in Table 1. The performance of the model was determined on the basis of model base run determination coefficient (R^2) between the modeled and experimental concentration of PM_{10}, OC, EC, WSICs, and trace metals. In the present study, most of the chemical species were well reconstructed (Table 1), except

$$\text{Uncertainty} = \sqrt{(\text{Error fraction} \times \text{concentration})^2 + (0.5 \times \text{MDL})^2} \tag{2}$$

MDL refers to mean detection limit and is calculated as three times of the standard deviation of the 10 replicates of blank analysis. Error fraction is computed by dividing the standard deviation of exposed filter analysis by the square root of the number of analyses. Furthermore, on the basis of signal-to-noise ratio criteria, those species displaying higher noise than signal were consigned with down

for some of the trace elements. These results were within the range of those presented in many PMF studies (Beuck et al. 2011; Cusack et al. 2013) for PM_{10} mass reconstruction. Scaled residuals between − 3 to + 3 are obtained for all of the major components, and the value of Q robust is strictly identical to the value of Q true. It shows that no specific event is affecting the results and that the base run can be regarded as stable.

The default "robust" mode of the PMF was applied to minimize the impact of extreme values. In PMF, the major challenge lies with small data sets (number of cases close to 50), where rotational ambiguity influences the solution significantly, thereby increasing its overall uncertainty. To resolve this issue, the latest version of PMF (i.e., PMF 5.0) allows handler to assess rotational ambiguity by providing advanced methods, such as displacement and bootstrap displacement, whereas the prior version proffered only bootstrapping tool. The combination of both bootstrapping and displacement tools help to assess rotational ambiguity as well as random errors. The determined factors might get swapped during displacement and bootstrap displacement run if the solution carries high rotational ambiguity. These factor swaps are displayed in diagnostic results. Moreover, the bootstrap displacement reveals the number of accepted resamples (Manousakas et al. 2017).

UNMIX

The UNMIX model is a multivariate analysis method, which takes the geometric approach to exploit the covariance of the data set and employs self-modeling curve resolution technique to determine the source profiles and source contributions with nonnegativity constraints. The present study employs stand-alone US EPA UNMIX 6.0 version. Simplified graphical representation of UNMIX model was given by Henry (1997) and detailed algorithms can be referred from Song et al. (2006) and Jain et al. (2017a, b). Brief details and characteristics of the model associated with the present study have been elucidated in the *Supplementary Information* ("Appendix 1").

PCA-APCS

PCA is a sophisticated technique to analyze the structure and patterns in the multivariate data sets. It transforms the large number of variables into smaller and possibly correlated sets of variables called principal components, which retains most of the variance in perceived variables. The detailed theoretical and mathematical information has been elucidated in many studies (García et al. 2006; Sharma et al. 2016b; Jain et al. 2017a, b) and brief details about the model have been given in the *Supplementary Information* ("Appendix 1").

PCA-APCS was applied on the datasets of PM_{10} of three receptor locations using software package SPSS statistics version 22.0 with 50 samples including 18 constituents/species (OC, EC, SO_4^{2-}, NH_4^+, NO_3^-, Na^+, Ca^{2+}, Mg^{2+}, K^+, Cl^-, Al, Ti, Zn, Fe, S, P, F^-, and B) for Delhi region, 45 samples with 16 constituents (OC, EC, SO_4^{2-}, NH_4^+, NO_3^-, Na^+, Ca^{2+}, Mg^{2+}, K^+, Cl^-, Al, Ti, Fe, S, P, and B) for Varanasi region and 54 samples with 17 species (OC, EC, SO_4^{2-}, NH_4^+, NO_3^-, Na^+, Ca^{2+}, Mg^{2+}, K^+, Cl^-, Al, Zn, Fe, S, P, F^-, and B) for Kolkata region. All variables were characterized and modified into normalized dimensionless form preceding statistical analysis. The lowest eigenvalue was confined to more than one for extracted factors, and the results were appraised in accordance with the Kaiser's criteria. Kaiser–Meyer–Olkin (KMO) describes the suitability of the data for factor analysis (sampling adequacy). It distributes the value between 0 to 1, designating values greater than 0.6 to be acceptable (Li et al. 2004). KMO values for the present analysis for Delhi, Varanasi, and Kolkata were 0.71, 0.68, and 0.70, respectively. Chemical variable with factor loadings greater than 0.5 (> half of the species affiliation) was utilized for source allocation (Kothai et al. 2008). Varimax rotation is administered along with orthogonal transformation to maximize the variance of squared loadings of the factors. It obviates the changing of total variance in the models and ensures the furnishing of maximal PCs having > 1 eigen values in the factor loading matrix.

Results and Discussion

PM_{10} Mass and Elemental Concentrations

The average concentration of PM_{10} at Delhi, Varanasi, and Kolkata was found to range from 63.3–354.7 µg m^{-3}, 53.3–392.1 µg m^{-3}, and 92.9–382.6 µg m^{-3} with annual average of 202.3 ± 74.3, 206.2 ± 77.4, and 171.5 ± 38.5 µg m^{-3}, respectively (Table 1). Mass concentration of PM_{10} and its chemical constituents measured at each location is reported in Table S1 (in the *Supplementary Information*) as a range and averaged values over the entire sampling duration and for each season. All the three sites, i.e., Delhi, Varanasi, and Kolkata, have recorded the highest average concentration of PM_{10} during winters: Delhi (263.8 µg m^{-3}), Varanasi (290.5 µg m^{-3}), and Kolkata (203.9 µg m^{-3}), which could be attributed to increased anthropogenic activities, more stable atmosphere, and lower boundary layer height (Sharma et al. 2016b). Lowest average concentration of PM_{10} was observed during monsoon season for Delhi (133.8 µg m^{-3}) and Varanasi (148.1 µg m^{-3}) but not for Kolkata. Kolkata, in the coastal region, observes intrusion of sea salt along with monsoonal rains that might have increased the PM_{10} concentration in the region. Lowest average concentration of PM_{10} at Kolkata was observed during the summer season (140.7 µg m^{-3}). The average concentration of PM_{10} at all the sampling sites exceeds the prescribed limit of National Ambient Air Quality Standards (NAAQS) (100 µg m^{-3} for 24 h and 60 µg m^{-3} for annual) of India. Similar studies about ambient PM_{10} concentration over Delhi region by Perrino et al. (2011) (183.0 µg m^{-3}), Sharma et al. (2003) (191.4 ± 45.5 µg m^{-3}), Tiwari et al. (2013) (161.0 ± 80 µg m^{-3}), and Sharma et al. (2014a)

$(191.4 \pm 45.5 \ \mu g \ m^{-3})$ have reported lower values than that recorded in the present study, whereas Kulshrestha et al. (2009) $(219.0 \pm 84 \ \mu g \ m^{-3})$, Mandal et al. (2014) $(285.7 \pm 26.3 \ \mu g \ m^{-3})$, Tiwari et al. (2012) $(219 \pm 84 \ \mu g \ m^{-3})$, Sharma et al. (2014b) $(213.1 \pm 15.0 \ \mu g \ m^{-3})$, and Jain et al. (2017b) $(249.7 \pm 103.9 \ \mu g \ m^{-3})$ reported higher values. For middle IGP region, Pandey et al. (2013) at Lucknow $(168.1 \pm 29.1 \ \mu g \ m^{-3})$, Murari et al. (2015) $(176.1 \pm 85.0 \ \mu g \ m^{-3})$, and Sen et al. (2014) $(139.6 \pm 68.0 \ \mu g \ m^{-3})$ at Varanasi observed lower values of PM_{10} compared with the contemporary study. For lower IGP region, Das et al. (2006) $(303.8 \pm 49.4 \ \mu g \ m^{-3})$ and Gupta et al. (2007) $(288.9 \pm 123.5 \ \mu g \ m^{-3})$ over Kolkata perceived much higher concentration of PM_{10} as collated with the present study. The average concentrations and seasonal variability of OC, EC, and WSICs of PM_{10}, as well as PM_{10} concentration at Delhi, Varanasi, and Kolkata of IGP are discussed in detail in our previous paper and referenced therein (Sharma et al. 2016b).

Source Apportionment

In total, 18 species (OC, EC, SO_4^{2-}, NH_4^+, NO_3^-, Na^+, Ca^{2+}, Mg^{2+}, K^+, Cl^-, Al, Ti, Zn, Fe, S, P, F^-, and B) of 50 PM_{10} samples of Delhi, 16 species (OC, EC, SO_4^{2-}, NH_4^+, NO_3^-, Na^+, Ca^{2+}, Mg^{2+}, K^+, Cl^-, Al, Ti, Fe, S, P, and B) of 45 PM_{10} samples of Varanasi, and 17 species (OC, EC, SO_4^{2-}, NH_4^+, NO_3^-, Na^+, Ca^{2+}, Mg^{2+}, K^+, Cl^-, Al, Zn, Fe, S, P, F^-, and B) of 54 PM_{10} samples of Kolkata were analyzed and used as an input of PCA-APCS, UNMIX, and PMF models. The compendium of extracted sources of PM_{10} by applying these three receptor models for the monitoring sites is presented in Tables S2, S3, and S4 (in *Supplementary Information*) for Delhi, Varanasi, and Kolkata, respectively. The average percent contribution of observed sources to PM_{10} for the receptor sites is compiled and depicted in Fig. 2 and Table 2.
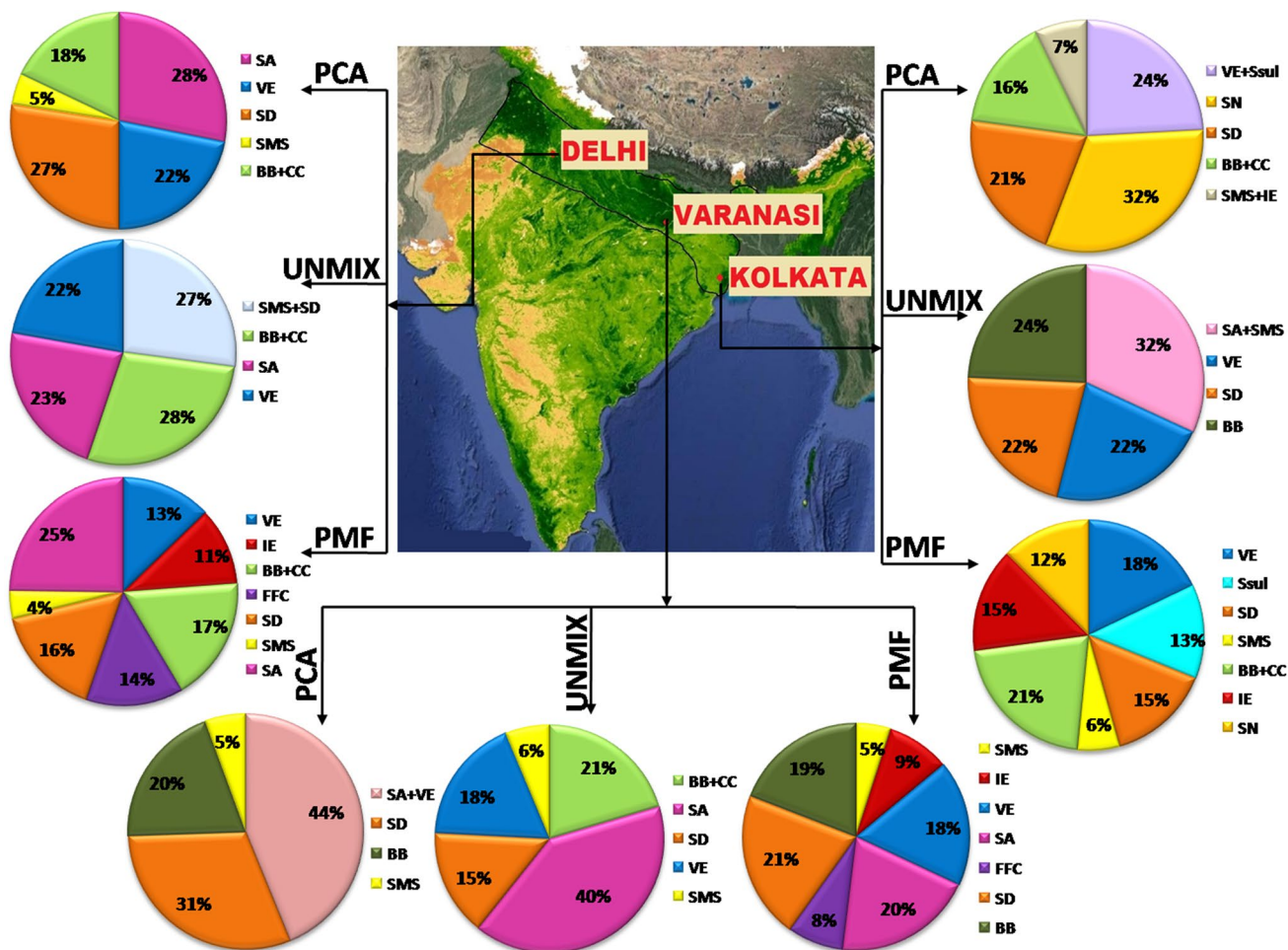

Fig. 2 Sources (SA: secondary aerosols; VE: vehicular emissions; SD: soil dust; BB: biomass burning; SMS: sodium and magnesium salt; FFC: Fossil fuel combustion; Ssul: secondary sulphate; SN: secondary nitrate; CC: coal combustion) and their contributions (%) to PM_{10} mass as calculated by the three receptor models

Table 2 Comparison of average source contributions (percentage) to PM_{10} mass concentration among the PCA/APCS, UNMIX, and PMF models

Identified sources	Delhi			Varanasi			Kolkata		
	PCA/APCS	UNMIX	PMF	PCA/APCS	UNMIX	PMF	PCA/APCS	UNMIX	PMF
Soil dust	27	–	16	31	15	21	24	22	15
Vehicular emissions	22	22	13	–	18	18	–	22	18
Industrial emissions	–	–	11	–	–	9	–	–	15
Secondary aerosols	28	23	25	–	40	20	–	–	–
Biomass burning	–	–	–	20	–	19	–	24	–
Sodium and magnesium salt	5	–	4	5	6	5	–	–	6
Fossil fuel combustion	–	–	14	–	–	8	–	–	–
Biomass burning + coal combustion	18	28	17	–	21	–	20	–	21
Secondary aerosols + sodium and magnesium salt	–	27	–	–	32.2	–	–	32	–
Secondary aerosols + vehicular emissions	–	–	–	44	–	–	–	–	–
Secondary nitrate	–	–	–	–	–	–	7	–	12
Secondary sulfate	–	–	–	–	–	–	–	–	13
Vehicular emissions + secondary sulfate	–	–	–	–	–	–	32	–	–
Industrial emissions + sodium and magnesium salt	–	–	–	–	–	–	17	–	–

PM_{10} Sources in Delhi

A total of 18 species were processed in the UNMIX model, where PM_{10} was considered as total mass as well as norm species. According to the suggested exclusion given by the model, fluoride (F^-) was discarded. Solution's diagnostic indicators were $R^2 = 0.89$ and S/N = 2.37 (S/N ratio should be > 2), which were observed to be reconcilable with the recommendations. In PMF, robust mode is used to run the model to minimize the influence of extreme values on the result. To identify the optimal number of sources (factors), parameter of goodness of fit "Q" was estimated to make the solution lie in the F-PEAK range. The influence of outliers or extreme values is not controlled in true Q value but is minimized in robust Q value. The theoretical Q value of this study (Delhi region) was 900 (i.e., 50×18). For seven factor solutions, more than 95% of Q values were close to 900, obtained from incorporating 5% error constant while performing 100 runs. The diagnostic indicators manifest that the observed Q value was the global minimum, thus establishing the feasibility of seven factor solution (Jain et al. 2017b).

PMF The combined presence of nitrate (NO_3^-), sulfate (SO_4^{2-}), and ammonium (NH_4^+) in the major proportions signify the source 1 to be mixture of secondary nitrate (SN) and secondary sulfate (Ssulf). Secondary sulfate formed through photochemical reactions, favored by strong solar radiations and high temperature, whereas secondary nitrate is formed by NO_x oxidation, which is encouraged by low temperature (Seinfeld and Pandis 2016). Principally $(NH_4)_2SO_4$ and NH_4NO_3 comprise secondary aerosols, procured from their gaseous precursors, i.e., NO_x, SO_2, and NH_3. Secondary aerosol (SA) plays a significant role in the formation of fog (Jaiprakash et al. 2017). SA have major contribution in PM_{10} mass over Delhi, which is approximately 25%. A comparable study on source apportionment of PM_{10} over Delhi by Sharma et al. (2015) reported similar percent contribution of SA, i.e., 21%. A second source has been designated as vehicular emissions (VE), because it is rich in OC, EC, Zn, Al, and B. OC and EC credibly derived from motor vehicle and traffic emissions (Pant and Harrison 2012; Jain et al. 2017b). EC is primarily emanated from combustion sources and is profoundly used as a tracer for diesel exhaust (Yin et al. 2010). Combustion and vaporization of fuel and other solvents encompass the primary sources of OC (Ho et al. 2003). The abundance of Zn signifies the contribution from tire wear and two stroke engines (Zn is used as fuel additive) (Kothai et al. 2008). The presence of Al indicates the influences from wear and tear of brake lining and pistons (Srimuruganandam and Nagendra 2012). The PMF analysis indicated that VE were responsible for 13% of total PM_{10} emissions, which is in concordance with the previous studies that highlighted VE as a major source of PM_{10} and has become a predicament for Delhi region (Sharma et al. 2015, 2017; Jain et al. 2017a, b). Source 3 is inferred as soil/road dust (SD) due to high share of crustal elements that include Al, Ti, Fe, Ca^{2+} Mg^{2+}, and OC. The extensive range of crustal elements (Al, Ca, Si, Ti, Mg, Pb, Zn, Na, and K) is being employed as marker species for soil/road dust source in India (Balachandran et al. 2000; Khillare et al. 2004; Banerjee et al. 2015). These crustal elements represent significant integrant of soil and road dust, which majorly contribute to coarser PM (Jaeckels et al. 2007; Begum et al. 2011). The occurrence of Fe with OC is suggested marker for road dust (Gupta et al.

2007; Banerjee et al. 2015). PMF results showed the contributions from crustal/soil/road dust to PM_{10} mass to be 16%. Source 4 is identified as biomass burning (BB) source due to the dominant contribution of K^+, F^-, and Cl^-. Ionic species of potassium (K^+) are frequently used as a marker of biomass burning besides levoglucosan in India and Europe (Robinson et al. 2006; Shridhar et al. 2010). In India, biomass or refuse burning is an amalgamation of cow dung, fuel wood, and post-harvest agricultural residue burning, including wildfires. The abundance of Cl^- indicates the intrusion from coal combustion (CC) and wood-burning sources (Pant and Harrison 2012). The presence of NH_4^+ along with K^+ and SO_4^{2-} with Cl^- confirms the emissions from biomass burning and coal combustion respectively (Khare and Baruah 2010; Jain et al. 2017b). Furthermore, the significant contribution of the EC in this source also indicates the contribution of biomass burning to EC, because the K^+/EC ratio for Delhi region in the present study was 0.20, which indicates the emissions from biomass burning (K^+/EC ratios more than zero signifies BB source) (Andreae and Merlet 2001). BB contributes approximately 17% to PM_{10} mass in the present study. Source 5 represents the sodium and magnesium salt (SMS) source (can be called sea salt) due to the abundance of precursors, such as Na^+ and Mg^{2+}. The use of these markers are often baffling, because they can be originated from both sea salt and crustal type sources (Jain et al. 2017b). The sampling site in Delhi is not surrounded by coastal area, therefore referring this contribution from sodium and magnesium salt source would be more appropriate than sea salt. In literature, markers, such as Na^+, Mg^{2+}, Cl^-, Ca^{2+}, and K^+, have been used for identifying marine aerosols (Kumar et al. 2001; Kothai et al. 2008; Sharma et al. 2014b). This source type contributes approximately 4% to PM_{10} mass in the present study. In Source 6, the presence of tracers, such as Cl^-, Al, Zn, and SO_4^{2-}, suggest the source of fossil fuel combustion. Cl^- along with SO_4^{2-} have been widely used as a marker of coal combustion (Pant and Harrison 2012; Sharma et al. 2016a). Tracers, such as Zn, Se, Te, and As, have been used internationally to signify coal and fuel burning (Lee et al. 2002; Gupta et al. 2007). PMF analysis revealed that fossil fuel burning has contributed 14% to PM_{10} mass over the Delhi region, which is bounded by three coal-fired thermal power plants. The exceptional abundance of Fe in source 7 indicates the emissions from iron industries. Several small- and medium-scale industries of metal processing and steel strip rolling in and around Delhi along with steel ingots manufacturing industry in neighboring states (Haryana) are potentially contributing Fe at the sampling location. Iron source contributes approximately 11% to PM_{10} mass in the present study.

UNMIX The presence of high concentrations of NH_4^+, SO_4^{2-}, and NO_3^- mark the source 1 to be of mixture of secondary nitrate and secondary sulfate. UNMIX analysis showed that secondary aerosol has contributed 23% to PM_{10} mass. Source 2 was identified as vehicular exhaust emissions due to the presence of OC and EC. UNMIX analysis revealed that vehicular emission has contributed 22% to PM_{10} mass. Source 3 is designated as mixed type source combining precursors of soil dust and sodium and magnesium salt. Higher concentrations of Al, Ti, Fe, S, Na^+, and Mg^{2+} indicate the same. Higher concentrations of Al, Ti, and Fe suggest it to be source of soil/road dust, whereas abundances of Na^+ and Mg^{2+} indicate the intrusion from the source of sodium and magnesium salts or sea salt. However, Mg^{2+} is believed to be emitted from soil dust also, which creates the ambiguity in interpreting the sources (Pant and Harrison 2012). UNMIX analysis shows 27% contribution of this mixed type source to PM_{10} in the present study. Source 4 has a high concentrations of K^+, Cl^-, SO_4^{2-}, and Ca^{2+} indicate the mixed source of biomass burning and coal burning. Cl^- along with SO_4^{2-} have been used as a tracer for coal combustion while K^+ used as a tracer for biomass burning, including cow dung and agricultural residue burning (Khare and Baruah 2010; Shridhar et al. 2010). UNMIX analysis shows 28% contribution of this mixed type source to PM_{10} in the present study.

PCA-APCS Source 1 is dominated by the key markers of secondary aerosols, i.e., NH_4^+, SO_4^{2-}, and NO_3^-. It explained 23.6% of the variance and contributed approximately 28% to PM_{10} mass in the present study. Source 2 is rich in OC, EC, Zn, and B, which are considered markers for vehicle emissions. This factor explained 17.8% of the variance and contributed approximately 22% to PM_{10} mass in the present study. Source 3 has high concentrations of Al, Ti, Fe, and P, which indicate the source of soil/road dust. This factor explained 15.7% of the variance and contributed approximately 27% to PM_{10} mass in the present study. Source 4 is characterized as biomass burning, wood burning, coal combustion, and vegetative burning source due to the presence of K^+ and Cl^-. This factor explained 12.7% of the variance and contributed approximately 18% to PM_{10} mass in the present study. Source 5 is dominated by the Na^+ and Mg^{2+} precursors, suggesting the source of sodium and magnesium salt. This factor explained 8% of the variance and contributed approximately 5% to PM_{10} mass in the present study.

PM_{10} Sources in Varanasi

Total 16 species were processed in the UNMIX model, where PM_{10} was considered as total mass as well as norm species. Solution's diagnostic indicators were $R^2 = 0.93$ and S/N = 2.05, which were observed to be reconcilable with the recommendations. In PMF, to identify the optimal number of sources (factors), parameter of goodness of fit "Q" was

estimated to make the solution lie in FPEAK range. The theoretical Q value for this study (Varanasi region) was 720 (i.e., 45×16). For seven factor solutions, more than 95% of Q values were close to 720, obtained from incorporating 6% error constant while performing 100 runs. The diagnostic indicators manifest that the observed Q value was the global minimum, thus establishing the feasibility of seven factor solution (Jain et al. 2017b).

PMF Source 1 is characterized as SA due to the presence of higher share of tracers of SA, i.e., NO_3^-, SO_4^{2-}, and NH_4^+. PMF analysis shows that SA has contributed approximately 20% to PM_{10} mass concentration. Source 2 is identified as VE due to the dominant share of OC and EC, which are key markers for traffic emissions (Belis et al. 2013). VE contributed approximately 18% to PM_{10} mass in the present study as receptor site being bounded by the heavy traffic areas; hence, VE is a significant contributor to PM pollution. Source 3 is characterized as soil/road dust due to the abundant presence of crustal elements, i.e., Al, Ti, Fe, S, and B. SD has contributed approximately 21% of aerosol mass in PM_{10} at the sampling site. Source 4 is characterized as BB source due to maximum share of K^+. K^+ is the inorganic tracer for biomass burning sources, including vegetative residue burning and wildfire (Guttikunda et al. 2013). PMF analysis revealed the contribution of BB to PM_{10} mass was 19%. The higher percent share of Na^+, Mg^{2+}, and Ca^{2+} distinguished the source 5 to be of sodium or magnesium salt or can be referred as sea salt (Raman and Ramachandran 2010). However, these markers of sea salt source, such as Mg^{2+} and Ca^{2+}, can possibly originate from crustal emissions. Moreover, Varanasi is a noncoastal region, therefore mentioning it as sodium and magnesium salt source would be more accurate than sea salt. SMS accounts 5% of PM_{10} mass in the present study. Source 6 represents emissions from coal combustion due to the dominance of markers, such as Cl^-, along with the presence of SO_4^{2-}, Fe, and Al indicate possible emissions from coal combustion (Sharma et al. 2016b). PMF results showed that CC contributes approximately 8% to total PM_{10} mass. The exceptionally high share of Ti in source 7 might have been coming from titanium-based industry. Many small- and medium-scale pulp and paper factories and handicraft industries in and around Varanasi may have contributed Ti to the receptor site. IE has contributed approximately 9% of aerosol mass in total PM_{10} mass in this study.

UNMIX Source 1 is characterized by high concentrations of SA markers, i.e., NH_4^+, NO_3^-, and SO_4^{2-}. UNMIX analysis showed the contribution of SA to PM_{10} mass was 40%. Source 2 represents the vehicular emission source due to high concentration of OC and EC. VE has contributed 18% to PM_{10} mass in the present study. Source 3 is dominated by crustal elements, i.e., Al, Ti, Fe, P, S, and Ca^{2+}, indicat-

ing the source of soil/road dust. SD has contributed 15% to the total mass of PM_{10} as identified by UNMIX analysis. Source 4 has a high concentration of K^+ and Cl^-, suggesting the mixed source of biomass burning and coal combustion. Significant contributions of OC with K^+ and SO_4^{2-} with Cl^- confirm the presence of biomass burning and coal burning sources (Chakraborty and Gupta 2010; Pachauri et al. 2013). BB accounts for 21% of PM_{10} mass concentration in the analysis done by UNMIX model. Source 5 is characterized due to high concentrations of Na^+ and Mg^{2+}, which suggests sodium and magnesium salt source type. It accounts for 6% of PM_{10} mass concentration in the present study.

PCA-APCS Source 1 is observed to be a mixed type source of SA and VE with higher concentrations of key markers of SA, i.e., NO_3^-, SO_4^{2-}, and NH_4^+ along with tracers of VE, i.e., OC and EC. This mixed type source explained 34.3% variance and contributed approximately 44% to PM_{10} mass concentration. Source 2 is characterized by higher share of crustal elements, i.e., Al, Ti, Fe, P, B, and Ca^{2+}, indicating the source of soil/road dust. SD source explained the 21.3% variance and accounted 31% of total PM_{10} mass concentration in PCA analysis. Source 3 has dominant tracers for biomass burning and coal burning, i.e., K^+ and Cl^- respectively. The presence of OC with K^+ and SO_4^{2-} with Cl^- assert the BB and CC source type, respectively. PCA analysis revealed the contribution of this source type to PM_{10} mass concentration was 20%. Source 4 is identified as sodium and magnesium salt source due to higher concentration of Na^+ and Mg^{2+}. This factor contributed approximately 5% to PM_{10} mass in the present study.

PM_{10} Sources in Kolkata

Total 17 species were processed in the UNMIX model, where PM_{10} was considered as total mass concentration as well as norm species. According to the suggested exclusion given by the model, species with more than 50% of the specific variance (SV) or variance due to error should be excluded from further statistical analysis. Accordingly, UNMIX discarded calcium (Ca^{2+}) from further analysis. Solution's diagnostic indicators were $R^2 = 0.79$ and S/N $= 2.50$, which were observed to be reconcilable with the recommendations. In PMF, to identify the optimal number of sources (factors), parameter of goodness of fit "Q" was estimated to make the solution lie in F-PEAK range. The theoretical Q value for this study (Kolkata region) was 918 (i.e., 54×17). For seven factor solutions, more than 95% of Q values were close to 918, obtained from incorporating 9% error constant while performing 100 runs. The diagnostic indicators manifest that the observed Q value was the global minimum, thus establishing the feasibility of seven factor solution (Jain et al. 2017b).

PMF The abundance of S, SO_4^{2-} with NH_4^+ in source 1 mark the source of secondary sulfate (SSulf). SSulf being in the form of $(NH_4)_2SO_4$, formed from its gaseous precursors, i.e., SO_2 and NH_3, through photochemical reactions, which is further favored by strong solar radiations and high temperature (Saraswati et al. 2017). Moreover, high relative humidity accelerates its dry conversion (Song et al. 2006). During the summer season, the high concentration of secondary sulfate and ammonium could form higher share of PM_{10} (Zhang et al. 2013). In India, sulfate has been employed as a tracer for wood burning and coal combustion (probably coal fired power plants), whereas ammonium has been attributed to agricultural and industrial activities (Pant and Harrison 2012). PMF analysis showed the contribution of SSulf to PM_{10} mass was approximately 13%. The occurrence of a higher share of NO_3^- and NH_4^+ identifies source 2 to be of secondary nitrate. SN is in the form of NH_4NO_3, formed from its gaseous precursors, i.e., NO_x and NH_4^+, through NO_x oxidation, which is encouraged by low temperature (Li et al. 2004). NO_x is primarily emitted from diesel and gasoline engines as well as electricity generating plants (Song et al. 2006). SN is semivolatile at high temperatures, therefore, generally has high concentrations during the winter season. PMF analysis revealed that SN contributes approximately 12% to total PM_{10} mass concentration. Source 3 has been designated as vehicular emissions, because it is rich in OC, EC, Zn, and B. OC and EC are attributed to motor vehicle and traffic emissions. OC is majorly emitted through gasoline engines, whereas EC is chiefly produced from diesel exhausts (Sharma et al. 2018a, b; Karar and Gupta 2007). PMF results showed the contribution of VE in PM_{10} mass was 18%. Source 4 is characterized as soil/road dust due to the abundant presence of crustal elements, i.e., Al, Fe, S, and P. SD has contributed approximately 15% of aerosol mass concentration in PM_{10} at the sampling site. Source 5 has high concentrations of K^+, OC, and Cl^-, suggesting the mixed source of biomass burning and coal combustion. Significant contribution of OC with K^+ confirms the presence of biomass burning while Cl^- possibly coming from coal/fuel combustion. A similar study by Pachauri et al. (2013) reported that the increased biomass burning emissions could attribute to significant correlation between K^+ and OC. The K^+/EC ratio for the Kolkata region in the present study was observed as 0.36, which suggests the presence of BB source in EC (Cesari et al. 2016) and the significant contribution of the EC in this source confirms the same. BB accounts for 21% of PM_{10} mass concentration in the analysis done by PMF model. The higher percent share of Na^+ and Mg^{2+} distinguished source 6 to be sodium or magnesium salt, which can be referred to as sea salt. A significant share of Cl^- also suggests the source to be sea salt. Because Kolkata is a coastal city, intrusion from sea salt spray to the receptor location is more obvious. SMS accounts 6% of PM_{10} mass concentration in the present study. The exceptionally high share of Zn in source 7 might have been coming from zinc-based industries. Several small- and medium-scale industries of metal processing, galvanizing, electroplating, and metallurgy in and around Kolkata are potentially contributing Zn at the sampling location. Zinc source contributes approximately 15% to PM_{10} mass concentration in the present study.

UNMIX Source 1 is observed to be a mixed type source of SA and SMS with higher concentrations of key markers of SA, i.e., NO_3^-, SO_4^{2-}, and NH_4^+, along with tracers of the SMS or sea salt, i.e., Na^+, Mg^{2+}, and Cl^-. UNMIX analysis showed the contribution of this mixed type source to PM_{10} mass concentration was 32%. Source 2 represents the vehicular emission source due to high concentration of OC and EC. VE has contributed 22% to PM_{10} mass concentration in the present study. Source 3 is dominated by crustal elements, i.e., Al, Fe, P, S, and B, indicating the source of soil/road dust. SD has contributed 22% to the total mass concentration of PM_{10} as identified by UNMIX analysis. Source 4 has a high concentration of K^+ and OC, suggesting the source of biomass burning. BB accounts for 24% of PM_{10} mass concentration in the analysis done by the UNMIX model.

PCA-APCS Source 1 is observed to be a mixed type source of VE and SSulf with a higher concentration of key markers of VE, i.e., OC and EC along with tracers of SSulf, i.e., SO_4^{2-}, NH_4^+, and S. PCA analysis showed the contribution of this mixed-type source to PM_{10} mass concentration was 32% and explained 21.5% variance. Source 2 is identified as a secondary nitrate source due to higher concentration of NO_3^- and NH_4^+. This factor explained 6.9% of the variance and contributed approximately 7% to PM_{10} mass concentration in the present study. Source 3 is characterized by a higher share of crustal elements, i.e., Al, Fe, S, P, B, and Ca^{2+}, indicating the source of soil/road dust. SD source explained the 21.3% variance and accounted 24% of total PM_{10} mass in PCA analysis. Source 4 has dominant tracers for biomass burning and coal burning, i.e., K^+ and Cl^-, respectively. PCA analysis revealed the contribution of this source type to PM_{10} mass concentration was 20% and explained variance was 18.8%. Source 5 has a higher concentration of key markers, such as Na^+ and Mg^{2+}, with the presence of Cl^- indicating the source of sodium and magnesium salt or sea salt. Also, an exceedingly high concentration of Zn is present in this source, potentially coming from zinc-based industries (metal processing, galvanizing, electroplating, and metallurgy) in Kolkata, asserting that this factor to be a mixed-type source of SMS and IE. PCA analysis revealed the contribution of this source type to PM_{10} mass concentration was 17%, and the explained variance was 13.3%.

Model Comparisons

The present work intends to effectuate an intercomparison of PM_{10} source apportionment results observed at three receptor sites of IGP: Delhi (upper IGP), Varanasi (middle IGP), and Kolkata (lower IGP) using three receptor models (Table 2). The intercomparison were designed to investigate their performances in source identification, quantification of source contribution, and the extent of acquiescence between them. PMF agreed on seven individual sources (roughly similar source type) for all sampling sites based on the availability of tracer species, whereas PCA-APCS and UNMIX varied between four to five sources (including mixed type) for all measurement sites. The three applied models agreed on the similar source types but varied source numbers and contributions for the sampling locations.

For Delhi region, all three models identified dominant sources contributing to PM_{10}, i.e., SA, SD, VE, and BB. However, PMF identified two additional but crucial sources, i.e., IE and FFC. Furthermore, the quantification of the contribution of different sources to PM_{10} varied for the different models. For all regions, PCA-APCS and UNMIX over quantifies the contribution due to the limited number of source identification, whereas PMF effectively distributes the tracer species to corresponding sources and thereby offers a precise percent contribution of different sources. Similarly, for Varanasi and Kolkata regions, the identified major sources by three models were SA, VE, BB, and SD. PMF revealed two more sources, i.e., IE and FFC in Varanasi and was able to differentiate between secondary sulphate and secondary nitrate sources in Kolkata, which other models failed to do. Moreover, PCA-APCS and UNMIX results create ambiguity in source identification by mixing precursors of two or more sources (mixed-type source), whereas PMF restraints mixing of two source types in single factor and thus identified individual sources. Different studies on the comparison of source apportionment results furnished by applying different models on the same data sets agreed that the contribution and number of sources of PM as analysed by different receptor models may be different (Favez et al. 2010; Amato et al. 2009; Contini et al. 2012; Cesari et al. 2016; Sharma et al. 2016b; Jain et al. 2017a, b). The variations in the results arise due to incorporation of different theoretical approaches and procedures in the models used for source apportionment. The variations between the results of three applied models for the sampling sites and their comparative source profiles have been well illustrated in Figs. S1, S2, and S3 for Delhi, Varanasi, and Kolkata respectively (in *Supplementary* information).

PCA-APCS has basic analytic method that generalizes the original data set based on statistical affiliation of data set (Thurston and Spengler 1985; Banerjee et al. 2015). UNMIX effectively differentiates the significant sources, but is incompetent in estimating weaker sources (Belis et al. 2013). UNMIX applies self-modeling curve resolution technique, whereas PCA-APCS employs orthogonal linear transformation to derive considerable factors. Both PCA-APCS and UNMIX cannot process data with missing as well as below detection level values and have no provision of externally incorporating uncertainties in the measurements as well as in the model (Song et al. 2006). Conversely, PMF implements point-by-point least squares minimization technique, which allows the direct comparison of untransformed profile to input matrix. It excludes negative entries and deals with the below minimum detection level values. The results of the uncertainty (BS and DISP) tools as provided by PMF 5.0 are documented in Table S5, S6, and S7 (in *Supplementary* information) for three sampling sites: i.e., Delhi, Varanasi, and Kolkata respectively. For Delhi region, sources, such as IE and SMS, have high uncertainties, whereas the rotational tools indicate stable results for SA, VE, and BB sources. Uncertainty tools suggested steady results for VE and BB sources while darting results for SD source in Varanasi region. For Kolkata region, SD shows high uncertainty, whereas VE and SMS sources show lower uncertainties. PMF and UNMIX employs nonnegative restrains unlike PCA-APCS; however, the combination of APCS with PCA minimizes the reconstructed concentration matrix error (Chan and Mozurkewich 2007). PMF has an additional feature of graphical representation of source contributions on a daily basis (Ogundele et al. 2016).

Conclusions

The present work focuses on the application of different receptor models, including PCA-APCS, UNMIX, and PMF, on the data sets of three different sampling locations of IGP region, i.e., Delhi, Varanasi, and Kolkata of India to obtain the mutually validated outputs and results. The study attempts to intercompare the receptor models, majorly focusing on identification of sources, quantification of contribution of sources to PM_{10}, the precision of the results, and furnishes the following points.

- The arithmetic mean concentration of PM_{10} at Delhi, Varanasi, and Kolkata was found to range from 63.3–354.7 µg m^{-3}, 53.3–392.1 µg m^{-3}, and 92.9–382.6 µg m^{-3} with annual average of 202.3 ± 74.3 µg m^{-3}, 206.2 ± 77.4 µg m^{-3}, and 171.5 ± 38.5 µg m^{-3}, respectively. Maximum concentration of PM_{10} in Delhi, Varanasi, and Kolkata was observed during the winter season.

- The three applied models agreed on similar source, although different source numbers and percent contribution; the majority are SD, SA, VE, and BB for Delhi region. PCA/APCS extracted five sources of PM_{10} [SA, SD, VE, BB + CC, and SMS], whereas UNMIX model identified four sources of PM_{10} [mixed type (SS + SD), mixed type (BB + CC), SA, and BB]. PMF model revealed seven different sources of PM_{10} [SA, SD, VE, mixed type (BB + CC), FFC, IE, and SMS].

- For Varanasi region, the identified sources by PMF were SA, SD, VE, BB, IE, FFC, and SMS, whereas PCA-APCS and UNMIX resolved four and five source types, respectively, that include mixed type [SA + VE, BB, SD, and SMS] as resolved by PCA-APCS while SA, VE, BB + CC, SD, and SMS were revealed by UNMIX. Major sources for the Kolkata region as identified by three models were: PMF identified seven different sources, i.e., secondary sulphate, SN, SD, VE, BB + CC, IE, and SMS. UNMIX identified four sources, i.e., mixed type (SA + SMS), VE, SD, and BB. PCA-APCS distinguished five sources, i.e., mixed type (VE + Ssulf), mixed type (SMS + IE), SD, mixed type (BB + CC), and SN.

- The present study aids in enriching scientific auxiliary of policymakers and stakeholders and help them to understand the attributes of respirable PM_{10} and their dominant source regions. The effective strategies to be presented to the policy makers to devise laws that can be formulated. A well-defined objective of this research will be to upgrade emanation control strategies, to enhance general well-being, and to improve the overall quality of ambient air.

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