Investigating the seasonal variability in source contribution to $PM_{2.5}$ and PM_{10} using different receptor models during 2013–2016 in Delhi, India

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Abstract

The present work deals with the seasonal variations in the contribution of sources to $PM_{2.5}$ and PM_{10} in Delhi, India. Samples of $PM_{2.5}$ and PM_{10} were collected from January 2013 to December 2016 at an urban site of Delhi, India, and analyzed to evaluate their chemical components [organic carbon (OC), elemental carbon (EC), water-soluble inorganic components (WSICs), and major and trace elements]. The average concentrations of $PM_{2.5}$ and PM_{10} were 131 ± 79 μg m^{-3} and 238 ± 106 μg m^{-3}, respectively during the entire sampling period. The analyzed and seasonally segregated data sets of both $PM_{2.5}$ and PM_{10} were used as input in the three different receptor models, i.e., principal component analysis-absolute principal component score (PCA-APCS), UNMIX, and positive matrix factorization (PMF), to achieve conjointly corroborated results. The present study deals with the implementation and comparison of results of three different multivariate receptor models (PCA-APCS, UNMIX, and PMF) on the same data sets that allowed a better understanding of the probable sources of $PM_{2.5}$ and PM_{10} as well as the comportment of these sources with respect to different seasons. PCA-APCS, UNMIX, and PMF extracted similar sources but in different contributions to $PM_{2.5}$ and PM_{10}. All the three models extracted 7 similar sources while mutually confirmed the 4 major sources over Delhi, i.e., secondary aerosols, vehicular emissions, biomass burning, and soil dust, although the contribution of these sources varies seasonally. PCA-APCS and UNMIX analysis identified a less number of sources (besides mixed type) as compared to the PMF, which may cause erroneous interpretation of seasonal implications on source contribution to the PM mass concentration.

Introduction

Delhi, the capital of India, has been in the limelight recently for all the wrong reasons about air pollution, bearing the

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choking level of air pollutants in the ambient atmosphere. Particulate matter (PM) (particularly $PM_{2.5}$ and PM_{10}) is one of the important atmospheric pollutants which supremely governs the air quality of the megacity. PM has adverse effects on regional air quality, visibility, atmospheric chemistry, and overall on global climate (IPCC 2013; Pui et al. 2014; Seinfeld and Pandis 2016; Singh et al. 2017). Several studies have identified and reported the harmful effects of PM pollution on human and biota health that include cardiovascular and respiratory diseases, allergies, respiratory tract inflammation, and even deaths in severe cases (Schwartz et al. 1996; Pope and Dockery 2006; Li et al. 2009; Pope et al. 2009; Tie et al. 2009; Brauer et al. 2015). A recent study revealed that air pollution causes 26% of premature deaths in India where solely $PM_{2.5}$ was accountable for around 12.4 lakh deaths in the year 2017 (Balakrishnan et al. 2019). Numerous studies in the past two decades have unveiled the significance of size,

composition, and sources of PM in health-related studies (Schlesinger et al. 2006; Kelly and Fussell 2012; Brauer et al. 2015; Pongpiachan et al. 2017), which urges the need to identify the composition and sources of PM to formulate the preventive measures and abatement strategies to reduce PM pollution.

PM has varied sources and is made up of different components (such as acids, organic and inorganic components, dust particles, metals), which have varying extents of health impacts and toxicity (Pongpiachan et al. 2013; Pongpiachan et al. 2014; Pongpiachan and Iijima 2016), highlighting the importance and necessity of employing source apportionment studies to comprehend the PM formation processes and their sources (Chuay et al. 2020). Source apportionment of PM is commonly performed through receptor or source-oriented models that include chemical mass balance (CMB), principal component analysis-absolute principal component scores (PCA-APCS), UNMIX, and positive matrix factorization (PMF). Several studies have already published comprehensive details about statistical and theoretical aspects of these models (Thurston and Spengler 1985; Paatero and Tapper 1994; Henry 2003; Brown and Hafner 2005; Song et al. 2006; Viana et al. 2008; Paatero and Hopke 2009; Paatero 1999; Belis et al. 2013) and references therein.

Source apportionment studies conducted in Delhi by Khillare and Sarkar (2012) and Pathak et al. (2013) revealed 3 major sources, i.e., crustal, vehicular emissions (VE), and industrial emissions (IE) of heavy metal pollution. Srivastava and Jain (2007) and Srivastava et al. (2008) reported soil dust (SD) and VE as 2 major sources of PM_{10} in Delhi, contributing more than 70% to PM_{10} concentration. The emission inventory prepared by NEERI (2010) also reported that SD, VE, and IE were the dominant sources of PM_{10}. Behera et al. (2011) built up emission inventory for Kanpur region for coarser particulates and reported that industries (25%), traffic (20%), fossil fuel combustion (19%), and road dust resuspension (15%) were the major sources. Nagar et al. (2017) and Jaiprakash et al. (2017) reported that VE, biomass burning (BB), and SD were the dominant sources of fine PM in Delhi. Gianini et al. (2012) and Dongarra et al. (2010) reported that the vehicular emissions are the major source of PM_{10}, where VE contributed 30% to PM_{10} mass in Bern, Switzerland (Gianini et al. 2012), and ∼ 50% in Palermo, Italy (Dongarra et al. 2010), whereas Titos et al. (2012) estimated the major contribution of mineral matter (43%) to PM_{10} in Granada, Spain. Moreno et al. (2006) also estimated that the crustal segments solely contributed 30% to PM_{10} in Puertollano, Spain. Researchers have reported varying results of the sources and their contribution to PM even over the same locations. Since different receptor models rely on different statistical procedures and constraints, it produces varying results. Therefore, applying them together on the same data sets would give the scope of comparing their results, which further

helps in explicit identification of the significant sources. The detailed description of the receptor models along with their similarities and differences can be found in Belis et al. (2013). A similar study on source apportionment of PM_{10} using three multivariate receptor models (PCA-APCS, UNMIX, and PMF) was conducted by Callén et al. (2009) in Zaragoza, Spain. In the study, they focused on the comparison of different models in order to determine which was more adequate for the apportionment.

The present work is a subsequent part (part II) of the previous work done and reference therein (Jain et al. 2020). The first part dealt with the comprehensive description of $PM_{2.5}$ and PM_{10} measurements, their chemical characterization, and seasonal variations coupled with the meteorological parameters whereas the present study encompasses the application of three different receptor models (PCA-APCS, UNMIX, and PMF) on the seasonally allocated data sets of $PM_{2.5}$ and PM_{10} and the comparative analysis of these models' results and performances. It determines the seasonal variability in source composition and contribution to both fine and coarse fractions of PM over Delhi, India, along with the analysis of the transport pathways of air mass parcels from potential source regions to the receptor site during different seasons via back-trajectory analysis. The definitive objective of this study is to confine the gaps in the understanding of seasonal variations of potential sources. The novelty of the study lies in implementing the three different receptor models on the seasonally segregated data set to obtain the mutually validated outputs, which ultimately aids in explicit quantification of the contribution of characteristic sources of different seasons. The implication of the application of the three different receptor models on 4-year-long data set of $PM_{2.5}$ and PM_{10} provides a scope to explore more about the model's constraints and limitations along with the possibility to scrutinize the degree of acquiescence between them. The present study aids in enriching the scientific auxiliary of policymakers and stakeholders and help them in understanding the attributes of $PM_{2.5}$ and PM_{10} and their dominant source regions along with the effective strategies to be presented to the policymakers to devise laws that can be formulated. The study may also help in establishing as well as updating inventories of $PM_{2.5}$ and PM_{10} along with reducing uncertainties associated with climate models. A well-defined objective of this research will be to upgrade emanation control strategies, enhance general well-being, and improve the overall quality of ambient air over the region.

Methodology

Sampling

Sampling of $PM_{2.5}$ and PM_{10} (350 samples of $PM_{2.5}$ and PM_{10} each) were carried out periodically from January 2013

to December 2016 at an urban site of Delhi, India. The sampling station is located inside the institutional campus of CSIR-National Physical Laboratory, New Delhi (28° 38′ N, 77° 10′ E), at 216 m above mean sea level (amsl) (Fig. S1, in supplementary information). The simultaneous sampling of $PM_{2.5}$ and PM_{10} were carried out (at least twice in a week) on pre-combusted quartz microfiber filters at a height of 10 m above ground level (AGL). Further details on samplers and sampling procedures can be found in our previous publication (Jain et al. 2020). To estimate the seasonal variations of different sources of fine and coarse mode PM, a year was divided into four different seasons as per the India Meteorological Department (IMD), i.e., winter (January–February), summer or pre-monsoon (March–May), monsoon (June–September), and post-monsoon (October–December).

Analytical procedures

Analytical procedures used for chemical characterization of both $PM_{2.5}$ and PM_{10} have been summarized in Fig. S2 (in supplementary information). The complete details of the analytical procedure used in this study have already been discussed in part I of the study (Jain et al. 2020); hence, brief information has been added here. A total of 700 samples of PM ($n = 350$ for $PM_{2.5}$ and PM_{10} each) were collected and characterized for their chemical components. Initially, a non-destructive method was employed for the analysis of major and trace elements (Al, Ti, Fe, Cr, Mn, Zn, Cu, As, Pb, Br, and Mn) using wavelength-dispersive X-ray fluorescence (ZSX Primus, Rigaku, Japan). The OC and EC analyses were carried out using OC/EC carbon analyzer (DRI 2001A, Atmoslytic Inc., Calabasas, CA) following US EPA's IMPROVE-A Protocol. The estimation of water-soluble inorganic species/components (Na^+, NH_4^+, K^+, Ca^{2+}, Mg^{2+}, F^-, Cl^-, NO_3^-, and SO_4^{2-}) in $PM_{2.5}$ and PM_{10} samples were carried out by extracting filters and analyzed it using ion chromatograph (model: DIONEX-ICS-3000, Sunnyvale, CA, USA). The complete details regarding analytical procedures, underlying principles, calibration procedures, standards used, and repeatability are available in our previous publications and reference therein (Jain et al. 2017, 2020).

The same analytical procedure was used to analyze the field sample blank filters as of for exposed filters. Average blank filter concentration (background) was subtracted from all the exposed filter data to get the final concentration. All the samples were analyzed three times to calculate the analytical error/repeatability error. The repeatability error in analysis of trace elements, OC-EC, and WSICs were estimated to be ~ 5–10%, 3–5%, and 3–7%, respectively. The method detection limits (MDL) for all the instruments were calculated as three times the standard deviation of 10 repetitions of blank filter analysis. MDL value determined the quality of the data/measurements that will be useful to understand the quality of

the data and recapitulated in Table S1 (in supplementary information).

Data analyses

The allocation of sources of $PM_{2.5}$ and PM_{10} was done on a seasonal basis using three different receptor models, i.e., PCA-APCS, UNMIX 6.0, and PMF (version 5.0). Since, the several studies have been published detailing the theoretical aspects, algorithms, and working procedures of these models (Thurston and Spengler 1985; Paatero et al. 2013; Henry 2003; Viana et al. 2008; Pant and Harrison 2012; Belis et al. 2013; Sharma et al. 2016b; Jain et al. 2017, 2018, 2019) and references therein, hence, the significant details and parameters applied in the present study are explicated below.

PCA-APCS

PCA-APCS was employed on the analyzed and seasonally apportioned data sets of $PM_{2.5}$ and PM_{10} to identify characteristic sources present during different seasons. PCA-APCS model was performed using the software package SPSS statistics version 22.0. All the components were characterized and transformed before statistical evaluation into a standardized dimensionless form. Once the results were achieved, they were assessed only for PCs with eigenvalues > 1 and reviewed according to Kaiser's criteria. The value of Kaiser-Meyer-Olkin (KMO) ranges from 0 to 1, where value > 0.6 is acceptable in terms of data suitability for factor analysis (Li et al. 2004). Varimax rotation accompanied by orthogonal transformation has been applied in the present study. Chemical variable with > 0.5 factor loadings was exerted for source distribution.

UNMIX

In the present study, USEPA UNMIX 6.0 version was employed, which was freely available at EPA website (http://www.epa.gov/heasd/products/unmix/unmix.html). A detailed explanation of its algorithms and theoretical prospects is available in Song et al. (2006) and Jain et al. (2017). The best-fit species were selected through the inbuilt function of the model "select initial species" and thereafter using the second function called "suggesting additional species" to incorporate suitable components for analysis. The present study also incorporated the model's suggestion on species exclusion and diagnostic indicators of solutions that will be discussed in the section "Source apportionment." UNMIX was applied to the seasonal data set of $PM_{2.5}$ and PM_{10} to reveal the possible variations of sources with seasons. $PM_{2.5}$ and PM_{10} were considered as the total mass in their respective runs.

PMF

Seasonal variation in sources of $PM_{2.5}$ and PM_{10} was investigated by running the PMF 5.0 model for all the seasons (summer, monsoon, winter, and post-monsoon). Since the detailed description of the PMF model and its working procedure had already been explained in the first part (part I) of this study (Jain et al. 2020), hence, only the obligatory information is given here. The associated uncertainties with the data of analyzed chemical species were estimated by the PMF model (EPA PMF 5.0 user manual). In the present analysis, all the species of $PM_{2.5}$ and PM_{10} were found to be fit by the model. An eight-factor solution was obtained for both $PM_{2.5}$ and PM_{10} from the base run of the PMF model. Q robust and Q true values were observed to be in good agreement that indicated the data's stability and model's ability to fit all the species. Extra modelling uncertainties were also added to the data sets of both fine and coarse fractions of PM, which is described in the section "Source apportionment." A significant correlation (R^2) was observed between the modelled and measured concentrations of $PM_{2.5}$ and PM_{10}, signifying the results obtained were well reconstructed. R^2 observed for $PM_{2.5}$ was 0.86 and 0.78 was obtained for PM_{10}. Displacement (DISP) and bootstrap (BS) tools were applied to estimate the uncertainties associated with output factor profiles. The results of BS and DISP analysis for different seasons for both $PM_{2.5}$ and PM_{10} are summarized in Tables S16–S23 (in supplementary information). The BS analysis (100 runs) was carried out with all the data sets of $PM_{2.5}$ and PM_{10} and < 10% unmapped cases were observed that suggested the PMF solutions were well mapped. The DISP analysis was also conducted for all the data sets and reliable results were observed, i.e., < 5% swap of factor profile was obtained.

Results and discussions

Seasonal variability of chemical constituents of $PM_{2.5}$ and PM_{10}

The four-year average concentrations of $PM_{2.5}$ and PM_{10} were 131 ± 79 μg m^{-3} and 238 ± 106 μg m^{-3}, respectively. The temporal variation of $PM_{2.5}$ and PM_{10} has been shown in Fig. S3 (in supplementary information). The mean and seasonal concentrations of all the analyzed chemical species/ components of $PM_{2.5}$ and PM_{10} are given in Table S2 (in supplementary information) and the Wilcoxon-Mann-Whitney test was applied to identify the seasonal differences along with differentiating the nonsignificant and significant species and the results are recapitulated in Tables S3–S4 (in supplementary information) for $PM_{2.5}$ and PM_{10}, respectively. The highest concentrations of $PM_{2.5}$ and PM_{10} were observed in January (417 μg m^{-3} and 537 μg m^{-3},

respectively) whereas, the minimum concentrations of the fine and coarse fractions of PM were recorded in September (17 μg m^{-3} and 33 μg m^{-3}, respectively). The highest seasonal concentrations of both $PM_{2.5}$ (186 ± 90 μg m^{-3}) and PM_{10} (320 ± 97 μg m^{-3}) were observed during the post-monsoon season (October–December) and the lowest concentrations of both $PM_{2.5}$ (69 ± 28 μg m^{-3}) and PM_{10} (152 ± 75 μg m^{-3}) were found during the monsoon season (June–September). The stable atmosphere, lower boundary layer height, frequent fog and haze events, and elevated biomass and agricultural burning activities (prominently in Punjab and Haryana states during October and November) during cold seasons are the major reasons for the increased concentration of PM and aerosol loading over a region (Ravindra et al. 2019), whereas atmospheric particles were scavenged off by the rainfalls and strong winds during the monsoon season (Jain et al. 2017). Other studies conducted over Delhi also reported the similar results with the maximum concentration of PM during cold seasons and minimum in monsoon season (Kandlikar 2007; Mandal et al. 2014; Gopalaswami 2016; Sharma et al. 2016b; Sharma and Dikshit 2016; Jain et al. 2017; Gupta et al. 2018; Sharma et al. 2018b; Bannoo et al. 2020; Agarwal et al. 2020). More than 80% of the measured data of $PM_{2.5}$ and PM_{10} were observed to be exceeding the 24-h averaged National Ambient Air Quality Standards (NAAQS) defined by Central Pollution Control Board (CPCB) of India ($PM_{2.5}$, 60 μg m^{-3}; PM_{10}, 100 μg m^{-3}, averaged over 24 h), which is in accordance with other studies on PM over Delhi (Perrino et al. 2011; Tiwari et al. 2013; Mandal et al. 2014; Sharma et al. 2014; Tiwari et al. 2013; Trivedi et al. 2014; Panda et al. 2016; Sharma et al. 2016a, b; Gupta et al. 2018; Jain et al. 2017, 2018; Sharma et al. 2018b; Gadi et al. 2019; Jain et al. 2019; Banoo et al. 2020).

The average concentrations of OC and EC of $PM_{2.5}$ was found to range from 2.7 to 69.1 μg m^{-3} (average ± standard deviation, 15.7 ± 12.7 μg m^{-3}) and 0.8 to 35.3 μg m^{-3} (7.31 ± 6.17 μg m^{-3}), respectively, while the average concentrations of OC and EC of PM_{10} were observed to range from 4.6 to 71.9 μg m^{-3} (26.6 ± 14.7 μg m^{-3}) and 1.1 to 35.6 μg m^{-3} (9.31 ± 6.56 μg m^{-3}), respectively during the entire sampling period. The temporal variation of OC and EC of $PM_{2.5}$ and PM_{10} has been shown in Fig. S4 (supplementary information). Total carbononaceous species/aerosol (TCA=(1.6*OC) + EC) (Sharma et al. 2018a, b) were observed to contribute to ~ 28% and 24% to the total $PM_{2.5}$ and PM_{10} mass concentration, respectively. Similar studies carried out in Delhi also reported the high contribution (%) of carbonaceous aerosols to PM (Mandal et al. 2014; Sharma et al. 2014; Bisht et al. 2015; Gupta et al. 2018; Sharma et al. 2018a, b; Gadi et al. 2019). The seasonal trend of OC and EC was observed in the following manner: post-monsoon season > winter season > summer season >

monsoon seasons, and summarized in Table S2 (in supplementary information). It is to be noted that besides the stable meteorological conditions during cold seasons, the elevated crop residue burning in the states of Punjab and Haryana emits the large number of particulates that further travel towards Delhi region and causes the loading of carbonaceous aerosols during the post-monsoon season over the receptor site (Gupta et al. 2018). The total four-year average and seasonal concentrations of WSICs are summarized in Table S2 (supplementary information). Among analyzed WSICs, the dominant anionic and cationic species of $PM_{2.5}$ and PM_{10} were SO_4^{2-}, NO_3^-, Cl^- and NH_4^+, K^+, respectively. The four-year average concentrations of SO_4^{2-} of $PM_{2.5}$ and PM_{10} were 13.8 ± 8.7 μg m^{-3} and 20.1 ± 16.1 μg m^{-3}, respectively. The average concentration of NO_3^- was observed to be 11.6 ± 11.6 μg m^{-3} in $PM_{2.5}$ and 17.2 ± 17.9 μg m^{-3} in PM_{10}. Both SO_4^{2-} and NO_3^- showed significant seasonal variability.

The average quantification of chemical components of $PM_{2.5}$ was 26.8% secondary inorganic aerosols (SIA) (NH_4^+, NO_3^-, and SO_4^{2-}), 13.9% sum of other WSICs, 11.9% OC, 5.5% EC, 6.6% crustal elements, and 1.2% trace metals. Similarly in PM_{10}, 21.2% SIA, 14.1% crustal elements, 11.2% OC, 10.7% sum of other WSICs, 3.9% EC, and 1.7% trace metals were observed. The distribution of these chemical components in $PM_{2.5}$ and PM_{10} mass has been shown in Fig. S5 (in supplementary information). It is to be noted that the metals constitute a very small part of PM, and they have deleterious health effects on humans, e.g., Pb causes anaemia, fatigue, kidney, and brain damage in humans (Assi et al. 2016). In the present study, the measured concentration of Pb in PM_{10} was found to be exceeding both USEPA standards (0.15 μg m^{-3}, annually) and NAAQS given by CPCB, India (0.5 μg m^{-3}, annually) (NAAQS 2009). The highest concentration was observed in the winter season (0.78 μg m^{-3}) while the lowest in monsoon season (0.29 μg m^{-3}). However, the Pb concentration in $PM_{2.5}$ lies within the limit set by NAAQS, India, but exceeds USEPA standards for ambient air quality, with maximum concentration in the winter season (0.41 μg m^{-3}) and minimum in monsoon season (0.20 μg m^{-3}).

Source apportionment

The present study encompasses the results (source profiles and their seasonal contributions) of PCA-APCS and UNMIX. These results were compared with the result given by the PMF model, reported previously (Jain et al. 2020). A summary of source apportionment of $PM_{2.5}$ and PM_{10} resolved by these three models has been given in Tables S5–S14 (in supplementary information). In total, 22 species (OC, EC, Al, Ti, Fe, Cr, Mn, Zn, As, Pb, Br, Mn, Cu, Na^+, K^+, NH_4^+, Ca^{2+}, Mg^{2+}, F^-, Cl^-, NO_3^-, and SO_4^{2-}) of $PM_{2.5}$ and PM_{10} were

incorporated into the model for factor analysis. The three receptor models (PCA-APCS, UNMIX, and PMF) were run individually on the data sets of $PM_{2.5}$ and PM_{10} for winter, summer, monsoon, and post-monsoon seasons.

Source identification and quantification by PCA-APCS

The factor profiles of all the sources of $PM_{2.5}$ and PM_{10} for different seasons are given in Tables S5–S14 (in supplementary information). The comparative analysis of the contribution of different sources to fine and coarse fractions of PM during different seasons is shown in Fig. 1. The estimated value of KMO and Barlett's test ensures fitting of the data of $PM_{2.5}$ were 0.71, 0.73, 0.71, and 0.70 for winter, summer, monsoon, and post-monsoon seasons, respectively, while 0.73, 0.79, 0.75, and 0.75 were observed for winter, summer, monsoon, and post-monsoon seasons' data sets, respectively, for PM_{10}. Hence, all the estimated values of KMO and Barlett's test were > 0.6, suggesting the suitability of the data for factor analysis. PCA-APCS evaluated 6-source solution for $PM_{2.5}$ for all the seasons except for post-monsoon season where it estimated 7-source solution. In the post-monsoon season, one source was unidentified due to the unavailability of any dominant tracer species. For PM_{10}, PCA-APCS analysis estimated 5-source solution for summer, winter, and post-monsoon seasons and 6-source solution in monsoon season. Mixed sources were also obtained for both fractions of PM.
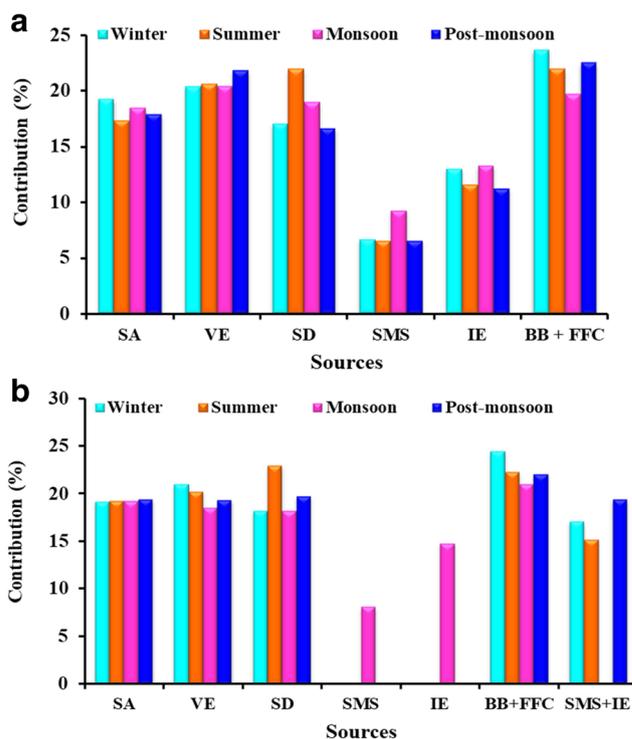
Fig. 1 Seasonal variability in source contribution to a) $PM_{2.5}$ and b) PM_{10} as determined by PCA-APCS over Delhi during 2013–2016

Factor 1 The first factor of both $PM_{2.5}$ and PM_{10} represents the source of secondary aerosols (SA) due to the high loading of NO_3^-, SO_4^{2-}, and NH_4^+. The NH_4NO_3 and $(NH_4)_2SO_4$ are formed from their gaseous precursors NO_3^- and SO_4^{2-}, respectively (Seinfeld and Pandis 2016; Saraswati et al. 2019; Kotnala et al. 2020). In $PM_{2.5}$, SA was maximum during the winter season (19.2%) followed by other seasons that have more or less similar contributions. Similarly, to PM_{10}, the contribution of SA during all the seasons did not vary much. The most probable reason could be that secondary nitrate (SN) is thermally unstable during warm seasons whereas strong solar radiations, high temperature, and high relative humidity during warm seasons favor the formation of secondary sulfate (SS). Contrarily, low temperature favors the formation of SN more than SS (Querol et al. 2008; Goel et al. 2018a; Saraswati et al. 2019). Since SA is composed of both SN and SS, hence, it did not show much seasonal variations. In total, PCA estimated a higher contribution of SA in PM_{10} (21%) as compared to $PM_{2.5}$ (18%) (Fig. S7, in supplementary information).

Factor 2 The second factor represents the important source of carbonaceous aerosols, i.e., OC and EC, which are globally considered as tracers for vehicular emissions (VE) (Sharma et al. 2016b; ChooChuay et al. 2020a). Along with OC and EC, other elements were also present in significant (Mn and Zn) and small amounts (Al and Pb), indicative of motor vehicles and road traffic emissions (Begum et al. 2011). Zn and Mn are markers of brake and tire wear as well as used in fuel additives (Kothai et al. 2008; Pant and Harrison 2012). The presence of Al can be indicative of emanation from wearing of brake lining and pistons (Jain et al. 2018). Similar studies conducted over Delhi also reported VE to be a major source of PM (Mandal et al. 2014; Sharma et al. 2016a, b, 2017; Jain et al. 2017, 2018, 2019; Gupta et al. 2018). PCA-APCS estimated almost similar annual contribution of VE to $PM_{2.5}$ (21.0%) and PM_{10} (20.6%) (Fig. S2, in supplementary information). The seasonal differences in the contribution of VE were not very prominent as it persists most of the year, which is also reported by Jaiprakash et al. (2017). The contribution of VE to $PM_{2.5}$ during post-monsoon, summer, winter, and monsoon seasons were 21.8%, 20.6%, 20.4%, and 20.4%, respectively, whereas to PM_{10}, the maximum contribution of VE was during the winter season (21.0%) followed by summer (20.3%), post-monsoon (19.3%), and monsoon (18.5%) seasons.

Factor 3 The third factor is dominated by the crustal elements like Al, Ti, Fe, Mn, Ca^{2+}, and Mg^{2+} that mark the factor as soil/road dust (SD) source (Balachandran et al. 2000). The crustal elements signify the major fraction of soil dust and re-suspended road dust and largely contribute to coarser particles (Khillare et al. 2004), which was further supported by enrichment factor (EF) analysis (Sharma et al. 2020). Ti, Fe,

Mn, Ca, and Mg in both coarse and fine fractions were observed to have low EFs for all the seasons and thus concluded to have arrived from crustal/soil dust sources (Fig. S6, in supplementary information) (Sharma et al. 2020). The detailed description of the analysis of EFs is given in Appendix-1 (in supplementary information). The considerable presence of OC, EC, and some other metals like Cu, Cr, Mn, and Zn in this factor indicated the combined emanations from road dust (Suryawanshi et al. 2016). However, the results obtained from PCA-APCS analysis of the total data set did not show much difference in the contribution of SD to $PM_{2.5}$ (16.3%) and PM_{10} (16.5%) (Fig. S7, in supplementary information). Regarding seasonal variations, the results showed a striking difference between summer and other seasons. The maximum contribution of SD to $PM_{2.5}$ was during summer (21.9%) season while the contributions in monsoon, winter, and post-monsoon seasons were 18.9%, 17.1%, and 16.2%, respectively. Similarly to PM_{10}, the highest contribution of SD was in the summer season (23.0%) followed by other seasons. The maximum contribution of SD was observed during the summer season accredited to the frequent and intense dust storms (traversing from the Thar Desert in Rajasthan) that occur in the Delhi region particularly in the summer season (Singh et al. 2016). The back-trajectory analysis also confirmed the influx of air mass parcel from arid regions of Rajasthan during the summer season (Fig. 4b).

Factor 4 The fourth factor is identified as sodium and magnesium salt (SMS) source, due to the presence of the high concentrations of Na^+ and Mg^{2+}. The species like Na^+, Mg^{2+}, and Cl^- are used as global tracers for marine aerosols and sea salt (Kumar et al. 2001; Pant and Harrison 2012). The sampling location is not surrounded by any sea or coastal region and emanations from this source could be ambiguous as sources like sea salt, rivers, soil/road dust, and other waterways could contribute together; therefore, referring this factor as SMS instead of sea salt was more appropriate (Jain et al. 2019). In the monsoon season, SMS source contributed ~ 8% to PM_{10}, whereas for all other seasons, the model evaluated mixed-type source that will be discussed in the fifth factor. For $PM_{2.5}$, PCA-APCS analysis apportioned this source in all the seasons, being dominant in monsoon season (9.2%) which is probably due to the intrusion of sodium and magnesium salt-laden aerosols by heavy monsoonal rainfall along with winds flowing from the Arabian Sea and Bay of Bengal (Jain et al. 2017, 2019) to the sampling region (Fig. 4c).

Factor 5 The fifth factor is identified as industrial emission (IE) source due to the presence of the high concentration of mainly Cr, Cu, and As and lower concentrations of elements like Zn, Br, Mn, and Pb. These elements could be originated from various small- to medium-scale industries, metal

processing industries, industrial effluents, and coal-fired thermal power plants (Sharma et al. 2016a; Jain et al. 2017). PCA-APCS could not apportion this source type separately for all seasons in PM_{10} and resulted in mixed-type source (IE + SMS) due to incursion of marker species of SMS source like Na^+ and Mg^{2+} except in monsoon season where IE contributed ~ 15% to PM_{10}. The annual contribution of IE + SMS to PM_{10} was 18.4% and seasonal contribution was maximum in post-monsoon season (19.4%). For $PM_{2.5}$, PCA-APCS analysis differentiated this source in all the seasons, being highest in the winter season (13.2%) due to higher emission rates and stable meteorological conditions during the cold seasons.

Factor 6 The dominant presence of K^+, Pb, Br, and Cl^- marked this factor to be of mixed type, including emissions from biomass burning (BB), wood burning, fossil fuel combustion (FFC), and coal combustion. Biomass comprises cow dung, dry leaves, fuel wood, residential and agricultural wastes, and post-harvest residue (Almeida et al. 2006). The significant amount of Cl^- presented in this factor revealed the intrusion from sources like coal combustion (CC), and wood burning (Pant and Harrison 2012). Presence of Br along with Cl^- signified CC emanations (Sharma et al. 2016b). Furthermore, the occurrence of OC and EC along with K^+ indicated the FFC emissions (Cesari et al. 2018). The significant presence of Pb in this factor (particularly in cold seasons) corroborated that BB (including solid waste burning) and FFC activities emit lead particles which are in agreement with other studies conducted by Koppmann et al. (2005), Xu et al. (2012), and Chen et al. (2017). BB + FFC annually contributed 23% and 21% to fine and coarse mode PM, respectively. It is to be noted that BB + FFC substantially contributed to $PM_{2.5}$ than PM_{10} which is consistent with the fact that combusted particles contribute largely to fine PM (ChooChuay et al. 2020b; Callén et al. 2009). Seasonal contribution of BB + FFC was maximum during winter (23.6% to $PM_{2.5}$ and 24.5% to PM_{10}) followed by post-monsoon (22.5% to $PM_{2.5}$ and 22.2% to PM_{10}), summer (21.9% to $PM_{2.5}$ and 22.1% to PM_{10}), and monsoon (19.7% to $PM_{2.5}$ and 21.0% to PM_{10}) seasons. The elevated biomass and agricultural burning, domestic heating activities, and stable meteorological conditions during winters are the major factors for the high BB and FFC emanations (Sharma et al. 2020). During winter and post-monsoon seasons, the majority of the trajectories were observed to be coming from Punjab and Haryana probably carrying aerosols emanated from the burning of agricultural residues (Fig. 4a, d).

Source identification and quantification by UNMIX

The comparative analysis of the contribution of different sources to $PM_{2.5}$ and PM_{10} during different seasons is shown in Fig. 2. Diagnostic indicators of the solution (R^2 and S/N ratio) were found to agree with the model's recommendations
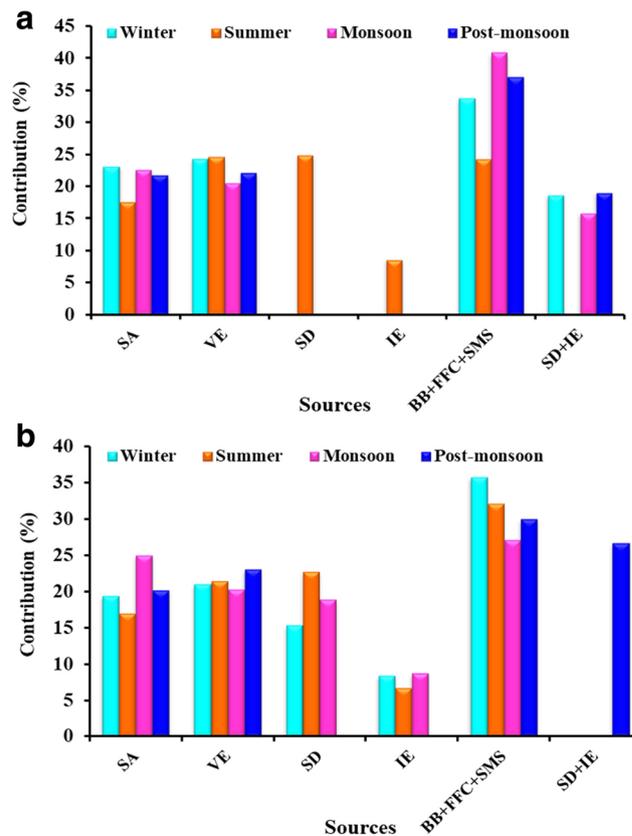
Fig. 2 Seasonal variability in source contribution to **a)** $PM_{2.5}$ and **b)** PM_{10} as determined by UNMIX over Delhi during 2013–2016

in all the seasons for both $PM_{2.5}$ and PM_{10}. Each run was made to re-sample the data 100 times before providing any solution. As per the model's function of "suggested exclusion," fluoride (F^-) was rejected from further processing of the data in all the seasons for both $PM_{2.5}$ and PM_{10}. Furthermore, the model rejected S in monsoon season and Mn in post-monsoon season from any further processing of the $PM_{2.5}$ data. For annual source apportionment analysis, model discarded Mn for PM_{10} while Na^+, Mg^{2+}, Br, and Fe for $PM_{2.5}$ from further calculations. It is recommended in the UNMIX 6.0 fundamentals and user guide that species with more than 50% of the variance due to error or specific variance (SV) be considered for exclusion from further UNMIX modelling. UNMIX analysis estimated 4-factor solution for $PM_{2.5}$ in winter, monsoon, and post-monsoon seasons while 5-factor solution in summer season including mixed-type sources in all. And for PM_{10}, the model evaluated 5-factor solution in winter, summer, and monsoon seasons whereas post-monsoon season has 4-factor solution including mixed-type sources in all.

Factor 1 Factor 1 of both fine and coarse fractions of PM is characterized by a high share of NO_3^-, SO_4^{2-}, and NH_4^+, which makes it a mixture of SN and SS sources. SA

contributed higher in $PM_{2.5}$ (23%) than PM_{10} (21%) annually. Seasonally, UNMIX resulted in a higher contribution of SA in monsoon season to both $PM_{2.5}$ and PM_{10}, which could be accredited to the fact that high humid conditions favor new particle formation through the transformation of NO_2 to NO_3^- and SO_2 to SO_4^{2-} during monsoon season (Saraswati et al. 2019) and also act as cloud condensation nuclei (Goel et al. 2018b).

Factor 2 The high percentage shares of OC and EC along with the presence of a significant amount of Mn, Al, Zn, and Pb marked this factor as VE. In the total data set analysis, the model gave an elevated percent contribution of VE to $PM_{2.5}$ (23.2%) as compared to PM_{10} (19.8%). It is to be noted that VE contributed higher in finer than coarser PM. The higher contribution of VE to $PM_{2.5}$ was during the winter season (24.6%) followed by summer (24.3%), post-monsoon (22.2%), and monsoon (20.5%) seasons, whereas to PM_{10}, the contribution of VE during post-monsoon, summer, winter, and monsoon seasons was 23.1%, 21.5%, 21.0%, and 20.3%, respectively.

Factor 3 It is represented by the dominant presence of crustal elements like Al, Ti, Fe, Mn, Ca^{2+}, and Mg^{2+}, which mark the factor as SD. The results obtained from UNMIX analysis of the annual data set showed a slightly higher contribution of SD to PM_{10} (19.2%) than $PM_{2.5}$ (18.7%) (Fig. S6, in supplementary information). Regarding seasonal variations, the results showed a striking difference between summer and other seasons. The maximum contribution of SD to PM_{10} was during summer (22.7%) followed by monsoon (18.9%) and winter (15.4%) seasons; however, in post-monsoon season, SD was found to be mixed with IE. SD contributed ∼ 25% to $PM_{2.5}$ during the summer season while for all other seasons, UNMIX resolved mixed-type source (SD + IE), combining species like Cu, As, Zn, Cr, and Mn with Al, Ti, Fe, Mn, Ca^{2+}, and Mg^{2+}. SD + IE contributed 19.1%, 18.7%, and 15.8% to $PM_{2.5}$ in post-monsoon, winter, and monsoon seasons, respectively.

Factor 4 This factor is identified as IE due to the high concentration of mainly Cu and As. Other heavy metals like Cr, Zn, Br, Pb, and Mn were also present in this factor in lower but significant concentrations. UNMIX model estimated the 8.7%, 8.4%, and 6.7% contribution of IE to PM_{10} in winter, monsoon, and summer seasons, respectively, while in post-monsoon season, UNMIX resulted in a mixed source (SD + IE), contributed ∼ 27% to PM_{10}. The contribution of IE to $PM_{2.5}$ in the summer season was estimated to be 9%. The source apportionment of annual data set of $PM_{2.5}$ and PM_{10} estimated 10.3% and 5.1% contribution of IE to $PM_{2.5}$ and PM_{10}, respectively.

Factor 5 This factor is characterized by tracers of BB and FFC like K^+, Cl^-, Pb, and Br along with markers of SMS, i.e., Na^+ and Mg^{2+}, hence called as mixed-type (BB + FFC + SMS) source. Also, OC and EC were present in significant amounts in association with K^+ and Cl^-, indicating the FFC emanations (Jain et al. 2019). The seasonal contribution of BB + FFC + SMS to PM_{10} was observed to be highest in the winter season (35.7%) whereas to $PM_{2.5}$, it was higher during monsoon season (40.8%). However, the mixed type of sources could not correctly estimate the seasonal contributions as different sources show different contributions during different seasons. For example, SMS shows a higher contribution during the monsoon season (Sharma et al. 2016a) while BB shows a higher contribution during the winter season (Jain et al. 2017); therefore, the percent contributions got mixed up in the mixed type of sources.

Source identification and quantification by PMF

Seasonal source apportionment of both fine and coarse mode PM using the PMF model has been reported in detail in the first part of this study (Jain et al. 2020); here, the results of PMF are being briefed for the comparative purpose. The comparative analysis of sources of $PM_{2.5}$ and PM_{10} with their contribution in percentage is shown in Fig. 3 and contributions to mass concentration are summarized in Table S15 (in supplementary information). The annual average contributions
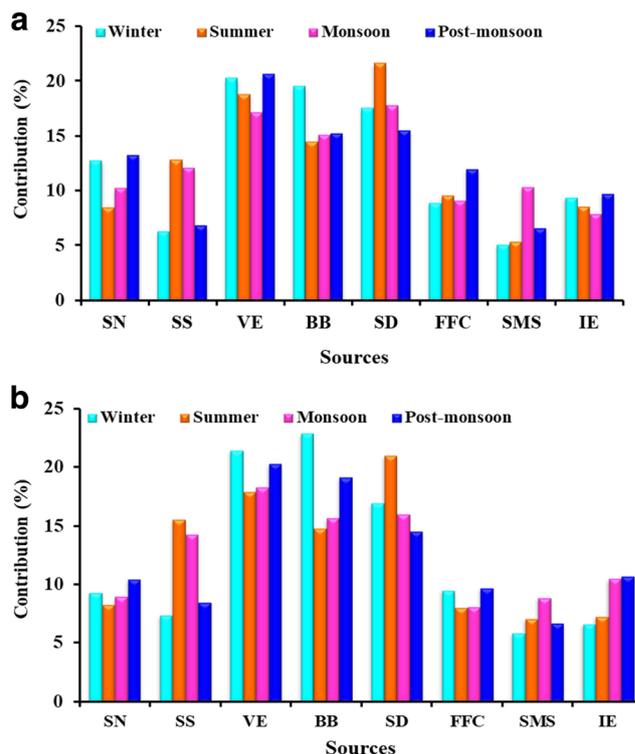
Fig. 3 Seasonal variability in source contribution to **a)** $PM_{2.5}$ and **b)** PM_{10} as determined by PMF over Delhi during 2013–2016

for $PM_{2.5}$ and PM_{10} are analogized in Fig. S7 (in supplementary information). PMF estimated eight-factor solutions for both $PM_{2.5}$ and PM_{10} in all the seasons.

Factor 1 The first factor of both fine and coarse fractions of PM was dominated by NO_3^- and NH_4^+, thus inferred as SN. The contribution of SN to both $PM_{2.5}$ and PM_{10} followed the same seasonal trend. The contribution of SN to $PM_{2.5}$ was 13.2% > 12.8% > 10.2% > 8.4% while to PM_{10} was 10.5% > 9.3% > 8.9% > 8.3% during post-monsoon, winter, monsoon, and summer seasons, respectively. Annualy, SN contributed to 11.7% to $PM_{2.5}$ and 11.8% to PM_{10}.

Factor 2 The second factor of $PM_{2.5}$ and PM_{10} was inferred as SS due to the dominant presence of SO_4^{2-}. The contribution of SS to both $PM_{2.5}$ and PM_{10} was highest during the summer season, i.e., 12.9% to $PM_{2.5}$ and 15.5% to PM_{10}, while minimum during the winter season, i.e., 6.4% to $PM_{2.5}$ and 7.4% to PM_{10}.

Factor 3 The presence of a high concentration of key markers like OC, EC, Zn, Mn, and Al inferred the source of VE. VE contributed annually ~ 16% to $PM_{2.5}$ and 21% to PM_{10}. The seasonal variations in the contribution of VE were not evident since it persists most of the year. The contribution of VE to $PM_{2.5}$ during post-monsoon, winter, summer, and monsoon seasons was 20.7%, 20.3%, 18.8%, and 17.1%, respectively. The highest contribution of VE to PM_{10} was during the winter season (21.4%) while lowest during the summer season (18%).

Factor 4 The profusion of markers like K^+, Pb, and the considerable presence of Cl^-, Br, OC, and EC marked this factor to be of BB source. Furthermore, S was also present in a small amount in some seasons indicating both BB and CC emissions (Andreae 1985). The annual contribution of BB to fine and coarse mode PM was 23% and 19%, respectively (Fig. S7, in supplementary information). It was observed that BB contributed more to $PM_{2.5}$ than PM_{10}. BB source contributed highest in the winter season while lowest in the summer season.

Factor 5 The abundance of crustal elements like Al, Fe, Ti, Mn, Ca, and Mg marked this factor as the source of SD. PMF analysis estimated that SD contributed annually 13% to $PM_{2.5}$ and 17% to PM_{10}, which is consistent with the fact that SD contributes largely to coarser particles. The maximum contribution of SD to $PM_{2.5}$ and PM_{10} was during summer and minimum in the post-monsoon season.

Factor 6 The sixth factor is represented as FFC due to the presence of a high concentration of Cl^-, F^-, Cr, and Br. Coal-fired power plants and oil combustion are the significant sources of Cr in the atmosphere besides IE (Fishbein 1981;

Galbreath et al. 1998; Cheng et al. 2009). PMF analysis estimated the annual contribution of FFC to be of 10% and 7% to $PM_{2.5}$ and PM_{10}, respectively. It was observed that FFC contributed more in fine PM than the coarser one (Saraswati et al. 2019). The highest contribution of FFC was in the post-monsoon (12% to $PM_{2.5}$ and 9.7% to PM_{10}) season.

Factor 7 It is inferred as the source of SMS because of the presence of Na^+ and Mg^{2+} in high concentration. The annual contribution of SMS to $PM_{2.5}$ and PM_{10} was 6% and 7%, respectively. The highest contribution of SMS to both $PM_{2.5}$ and PM_{10} was in the monsoon season, i.e., 10.4% and 8.9%, respectively, while the lowest during the winter season, i.e., 5.1% to $PM_{2.5}$ and 5.8% to PM_{10}.

Factor 8 Due to the presence of a high concentration of As and Cu and lower concentration of Zn, Pb, and Cr, this factor is inferred as IE. PMF estimated the annual contribution of IE was 10.3% to $PM_{2.5}$ and 9.9% to PM_{10}. The contribution of IE was maximum during the post-monsoon season, where IE contributed 9.8% to $PM_{2.5}$ and 10.7% to PM_{10}.

The results of the uncertainty (BS and DISP) tools as provided by PMF 5.0 are documented in Tables S16–S23 (in supplementary information) for both $PM_{2.5}$ and PM_{10} for different seasons. For $PM_{2.5}$, sources like IE and FFC have high uncertainties during winter, summer, and post-monsoon seasons while BB and IE sources show high uncertainty during monsoon season. However, the rotational tools indicate stable results for SN, SS, VE, and BB sources during winter, summer, and post-monsoon seasons whereas during monsoon season, SN, SS, VE, and SMS sources show stable results. Uncertainty tools suggested steady results for SN, SS, VE, BB, and SD sources during winter, summer, and post-monsoon seasons for PM_{10} while SN, VE, BB, SD, and SMS sources show higher stability during monsoon season.

Source comparison

The results of PCA-APCS and UNMIX were compared to the PMF output based on the identified source numbers, source type, and the contribution of each source to $PM_{2.5}$ and PM_{10} for different seasons (Table S24, in supplementary information). Seasonal variations of identified sources using these receptor models were also compared. The seasonal source apportionment study helps in quantifying the contribution of potential sources of PM in different seasons as well as season-specific sources can also be identified but for doing so, the large data set is required for each season to carry out the source apportionment analysis (Pant and Harrison 2012).

A total of seven common sources were identified by all the three models, be it an individual (single)-type or mixed-type source. For annual source apportionment analysis (using 4-year data sets), two common sources (single source) were

identified by all the three models for both $PM_{2.5}$ and PM_{10}: soil dust and vehicular emission. UNMIX resolved the overestimated contribution of VE to $PM_{2.5}$ as compared to PMF analysis while all the three models estimated similar contribution of VE to PM_{10} (Fig. S7, in supplementary information). PCA-APCS resolved similar contribution of SD to both fine and coarse modes of PM while PMF estimated higher contribution of SD to PM_{10} as compared to $PM_{2.5}$, which is in consistency with other related studies (Ho et al. 2003; Cesari et al. 2018; Banerjee et al. 2015; Sharma et al. 2016a; Jain et al. 2019). PMF evaluated 8-factor solution for both $PM_{2.5}$ and PM_{10}. However, PCA-APCS and UNMIX resolved 5- and 6-factor solution for PM_{10}, respectively, and 6- and 5-factor solution for $PM_{2.5}$, respectively. PMF distinguished SN and SS distinctly while PCA and UNMIX models resolved them as SA source (combining both SN and SS). PCA analysis of the PM_{10} data set resulted in a mixed-type source of BB + FFC, probably due to their similar tracers (Sharma et al. 2020). However, UNMIX evaluated the BB source in individual factor while mixing the markers of FFC with SMS source. For $PM_{2.5}$, PCA identified the sources of SMS and IE distinctly while UNMIX could not identify SMS source at all (due to exclusion of Na^+ and Mg^{2+} by the model). UNMIX attributed the dominant contribution of SA and VE to both $PM_{2.5}$ and PM_{10} while PMF analysis identified VE and BB as dominant sources.

Similar source apportionment study conducted on heavy metals over Delhi by Khillare and Sarkar (2012) estimated the contribution of crustal, VE, and IE to be 49–65%, 26–31%, and 4–21%, respectively, to metal concentration. The emission inventory of different sources of PM_{10} for the Delhi region was also prepared by NEERI (2010). It was found that PM_{10} was dominated by re-suspension of road dust to the extent of 52% while IE, VE, and area sources (cooking emissions, waste incineration, construction activities, etc.) contributed 22%, 7%, and 19%, respectively, to PM_{10}. Tiwari et al. (2013) estimated a total of 4 sources of PM_{10} (SD, VE, SA, and sea salt) where SD and VE were the major sources, contributing 37% and 23%, respectively, to PM_{10}. Patil et al. revealed some interesting results about the PM speciation profile. They reported many other sources of $PM_{2.5}$ and PM_{10} over Delhi besides SD, VE, and BB like paved and unpaved road dust, coal-fired power plants, solid waste open burning, and kerosene generators. A similar study was also conducted by Nagar et al. (2017), wherein, addition to expected primary sources (VE, SD, BB) of $PM_{2.5}$, the newly identified sources like municipal solid waste (MSW) burning, coal, and fly ash were also reported. Similar sources of $PM_{2.5}$ and PM_{10} (with slightly varying contributions) over Delhi were reported by Sharma et al. (2016b) and Jain et al. (2017, 2018), which is consistent with the present study. Jaiprakash et al. (2017) identified the sources of $PM_{1.0}$ using PMF over Delhi and revealed 6 major sources, i.e., SA, VE,

BB, SD, IE, and secondary chloride. SA (38.6%) and secondary chloride (19.4%) were found to be the principal contributors of $PM_{1.0}$. Other source apportionment studies conducted over Delhi region were with organic compounds, which also revealed similar sources of the fine and coarse mode of PM like VE, BB, cooking emissions, solid waste burning, biogenic, and industrial emissions (Gupta et al. 2018; Gadi et al. 2019). The comparison of the contribution (in %) of major sources of $PM_{2.5}$ and PM_{10} reported by recent source apportionment studies in Delhi is shown in Table S25 (in supplementary information).

Three common sources, i.e., SA, VE, and SD, were identified by all the receptor models for both $PM_{2.5}$ and PM_{10} in all the seasons but with varying seasonal contributions. UNMIX analysis overestimated the contribution of SA in monsoon season for both fine and coarse mode PM while PCA attributed almost similar contribution of SA in all the seasons. However, PMF differentiated SA source into its two components, i.e., SN and SS, where SS was higher during warmer seasons while SN was higher during colder seasons. Regarding the traffic source, PMF output showed slight seasonal variations with a high contribution in the winter season and low in monsoon season for both sizes of PM and all three models agreed that VE source does not show any specific seasonality. PMF and PCA-APCS estimated the highest contribution of SD to $PM_{2.5}$ and PM_{10} in the summer season and displayed significant seasonal trends whereas UNMIX analysis resulted in mixed-type source (SD + IE) in some seasons and thus seasonal trend could not be traced for the UNMIX output. Fe-rich soil dust that primarily originated from Aravalli hills of Delhi prevail over the region all year round along with frequent dust storms that come from the Thar Desert of Rajasthan that increase the loading of soil dust over the region particularly in summer season (Banerjee et al. 2015; Goel et al. 2018c, 2020). Similar studies also revealed the predominance of crustal components (Al, Ca, Mg, and Fe) in PM particularly in summer season majorly due to the prevalence of high wind speed and dry soil surface (Shukla and Sharma 2008; Chakraborty and Gupta 2010). Emissions from biomass burning were found to be high in winter and post-monsoon seasons for both fractions of PM as proffered by PMF output whereas PCA and UNMIX were not able to differentiate this source separately. Emissions from BB are one of the major problems in the Delhi region that cause devastating effects on human health, especially during post-monsoon and winter seasons. PCA-APCS combines the markers of BB with FFC source in all the seasons while UNMIX mixed three sources, i.e., BB + FFC + SMS, in a single factor, probably due to their overlapping tracers. PCA-APCS analysis assigned the highest contribution of BB + FFC to $PM_{2.5}$ and PM_{10} in the winter season while minimum in monsoon season, without displaying striking seasonal differences. However, the UNMIX model resolved the highest contribution of BB +

FFC + SMS source type to PM_{10} in winter and to $PM_{2.5}$ in monsoon season, giving erroneous seasonal distribution. It is to be noted that these types of mixed sources could not identify the seasonal variations since different sources show different contributions in different seasons as each source is characteristic of a particular season. Hence, models producing the mixed type of sources could not be relied on for determining and quantifying seasonal sources.

NEERI (2010) also published the seasonal dominance of potential sources of PM_{10} for the Delhi region. Construction and soil dust sources dominate in the summer season while solid waste burning and fuel combustion sources were major contributors in post-monsoon and winter seasons (NEERI 2010). Ram et al. (2012) in Kanpur distinguished fine-mode particulates as generally anthropogenic during winter as opposed to that from crustal amid summer. Jaiprakash et al. (2017) also studied the seasonal variations in the contribution of $PM_{1.0}$ sources over Delhi and reported comparable results. The contribution of SA and BB was higher in the winter season while SD was dominant in the summer season. VE is not showing any seasonal variation, while Nagar et al. (2017) studied the seasonal variation of $PM_{2.5}$ sources over Delhi and identified the seasonal distribution of potential sources similar to what has been observed in the present study. BB (30%) and SA (26%) were the major contributors during the winter season while SD (27%) and airborne fly ash (26%) were significant contributors of $PM_{2.5}$ during the summer season (Nagar et al. 2017). Different studies on comparison of source apportionment results furnished by applying different models on the same data sets also agreed that the contribution and number of sources of PM as analyzed by different receptor models may be different (Favez et al. 2010; Callén et al. 2009; Sharma et al. 2016b; 2020; Jain et al. 2017, 2019). The variations in the results arise due to the incorporation of different theoretical approaches and procedures in the models used for source apportionment. Conclusively, PMF apportioned PM sources distinctively probably because PMF employs point-by-point error estimation of the data by permitting down-weighting of outliers and missing values. PMF (version 5.0) model helps to obtain the rotational ambiguity with better representation of associated uncertainty component (Manousakas et al. 2017), whereas PCA-APCS and UNMIX estimated mixed type of sources as well. Mixing of tracers of different sources together hinders the essence of source apportionment by quantifying inappropriate contribution and thus causes difficulty in understanding to the users. Most importantly, PCA and UNMIX do not include uncertainties of the experimental data into analysis while PMF includes the uncertainty of each species and every sample into consideration and has a provision of incorporating extra modelling uncertainty as well. This function assists PMF in conferring more accurate and consistent results (Paatero et al. 2013).

Back-trajectory analysis

In order to identify and trace the trans-boundary movement of particulate matter from their potential source of origin to the receptor site, 120-h backward air mass trajectories for each experimental day, starting at 0500 h Universal Coordinated Time (UTC) at the height of 500 m above ground level (AGL) at the sampling site, were plotted employing the National Oceanic and Atmospheric Administration (NOAA) Air Resource Laboratory's (ARL) Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (http://ready.arl.noaa.gov/HYSPLIT.php) with the Global Data Assimilation System (GDAS) data as input (Draxler and Rolph 2003). It can be observed that the majority of the air mass parcel during the study period is approaching to receptor site from regions of Pakistan, Punjab, Haryana, Uttar Pradesh, IGP region, and its surrounding areas during the winter and post-monsoon seasons (Fig. 4a, d). During the summer season, the air mass parcel was coming from the arid landscapes of Rajasthan (Thar desert), regions of Pakistan, and Afghanistan to the receptor location (Fig. 4b) whereas during monsoon season, the approaching air mass was majorly transported from IGP region and the Arabian Sea through Rajasthan. However, some of the trajectories were seen to be coming from the Bay of Bengal passing through the vast stretch of IGP region (Fig. 4c). The trajectories have been plotted to trace the origin and transport pathways of air masses as they are ascribed to letting the influx of pollutants and their precursors in the city. These groups of trajectories have been also clustered together to display the major transport pathways of the polluted air mass flow, so as to provide a picture of dominant source regions precisely. It can be discerned that the source regions are both trans-boundary and locally originated from the continental landmass.

Conclusions

The present study focuses on determining the seasonal variations in the source profile and source contribution to $PM_{2.5}$ and PM_{10} concentrations over Delhi from January 2013 to December 2016. Two receptor models (PCA-APCS and UNMIX) were applied on the seasonally segregated data sets of $PM_{2.5}$ and PM_{10} and the results were compared with the PMF model's output for the same data sets. The three applied models have their own sets of merits and demerits and thus help in producing conjointly corroborated results to allow better cognizance about the potential sources of $PM_{2.5}$ and PM_{10} as well as the comportment of these sources with respect to different seasons. The annual average concentrations of $PM_{2.5}$ and PM_{10} were 131 ± 79 μg m^{-3} and 238 ± 106 μg m^{-3}, respectively. The maximum concentrations of both $PM_{2.5}$ (186 ± 90 μg m^{-3}) and PM_{10} (320 ± 97 μg m^{-3}) recorded
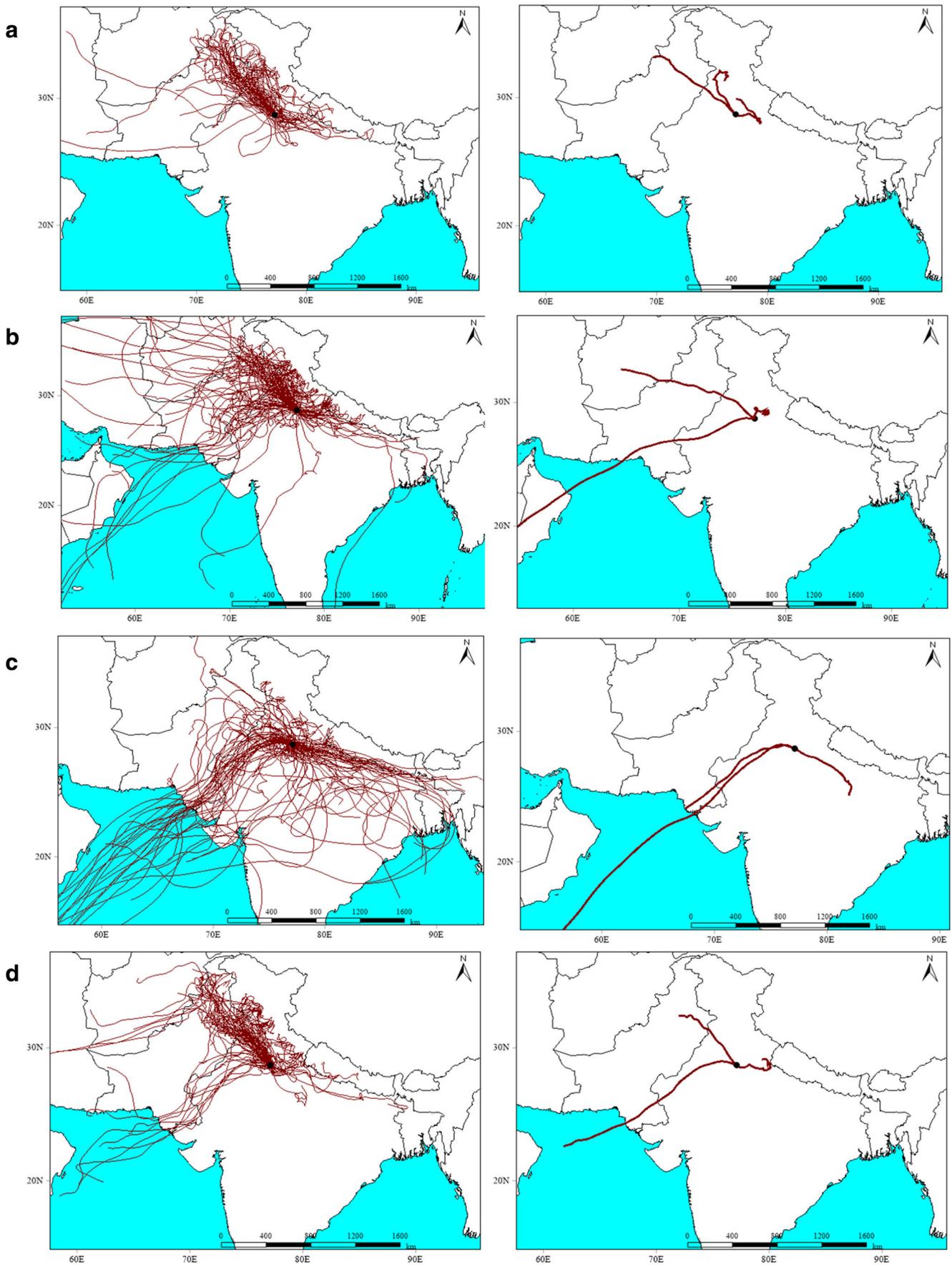
◀ **Fig. 4** The 120-h air mass HYSPLIT back trajectories and cluster analysis plots for **a)** winter, **b)** summer, **c)** monsoon, and **d)** post-monsoon seasons (2013–2016) at Delhi, India

during the post-monsoon season (October–December), whereas the minimum average concentrations of both $PM_{2.5}$ (69 ± 28 µg m^{-3}) and PM_{10} (152 ± 75 µg m^{-3}) were observed during monsoon season (June–September). All the three models extracted similar sources (i.e., SA, VE, BB, SD, FFC, IE, and SMS) but in different nature and contribution to PM mass. The three applied models mutually confirmed the dominant presence of 4 significant sources over Delhi, viz., SA, VE, BB, and SD, although the contribution of these sources varies seasonally. PCA-APCS and UNMIX estimated a less number of sources, including the mixed type of sources as well, which may cause erroneous interpretation of seasonal implications on source contribution to PM, whereas PMF identified the highest number of sources (8) for both fine and coarse fractions of PM in all the seasons without combining precursors of different sources that helps in determining the accurate percent contribution of different sources in different seasons, thus displaying significant seasonal variations.

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