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Source Apportionment of $PM_{2.5}$ in Delhi, India Using PMF Model

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Abstract Chemical characterization of $PM_{2.5}$ [organic carbon, elemental carbon, water soluble inorganic ionic components, and major and trace elements] was carried out for a source apportionment study of $PM_{2.5}$ at an urban site of Delhi, India from January, 2013, to December, 2014. The annual average mass concentration of $PM_{2.5}$ was $122 \pm 94.1\ \mu g\ m^{-3}$. Strong seasonal variation was observed in $PM_{2.5}$ mass concentration and its chemical composition with maxima during winter and minima during monsoon. A receptor model, positive matrix factorization (PMF) was applied for source apportionment of $PM_{2.5}$ mass concentration. The PMF model resolved the major sources of $PM_{2.5}$ as secondary aerosols (21.3 %), followed by soil dust (20.5 %), vehicle emissions (19.7 %), biomass burning (14.3 %), fossil fuel combustion (13.7 %), industrial emissions (6.2 %) and sea salt (4.3 %).

Keywords $PM_{2.5}$ · Organic carbon · Elemental carbon · Positive matrix factorization

It is well documented that atmospheric particulate matter (PM) significantly affects atmospheric chemistry, ambient air quality, visibility and the earth's radiation budget (Ramgolam et al. 2009; Pope et al. 2009). Several studies have revealed that aerosols, especially fine mode particles (<2.5 μm) can lead to serious human health effects like cardiovascular and respiratory disorders (Pope and Dockery 2006). Therefore, identification of PM sources is necessary to develop air quality improvement strategies in order to be able to control and reduce ambient PM concentrations through targeted action (Waked et al. 2014). To address this issue, many tools or receptor models have been used for identification and quantification of PM sources (Paatero 1997; Ulbrich et al. 2009).

In the recent past, receptor models have been applied in many studies and have shown the ability to accurately identify the potential emission sources of ambient PM at a receptor site (Waked et al. 2014). The PMF model is highly recommended when sources are not formally known, but it requires post-treatment source identification. Recently, the PMF model has been improved significantly and a new approach was developed by Paatero and Tapper (1994), using a least squares approach. PMF solves the problem arising in factor analysis by integrating non-negativity constraints in the optimization process and utilizing the error estimates for each data value as a point-by-point weight (Begum et al. 2004). The PMF model has been applied successfully worldwide (Kim and Hopke 2004; Karanasiou et al. 2009; Sharma et al. 2015). In this study, we report the chemical characteristics and source apportionment of $PM_{2.5}$ at an urban site in Delhi, India. The PMF model was applied for the source apportionment using chemical composition (organic carbon, elemental carbon, major ions, metals and trace elements) of $PM_{2.5}$ mass.

Materials and Methods

$PM_{2.5}$ samples were collected periodically at CSIR-National Physical Laboratory, New Delhi (28°38′N, 77°10′E; 218 m amsl) from January, 2013, to December, 2014. The sampling site is amenable to free wind flow from all the directions and represents a typical urban atmosphere,

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surrounded by huge roadside traffic (~100 m) and agricultural fields in the southwest direction (~500 m). Roadside vehicles, industrial emission and biomass burning could be major sources of carbonaceous aerosols and several other pollutants. The occasional occurrence of dust storms may contribute the presence of mineral dust significantly to the aerosol loading in summertime (Ram et al. 2010). The sampling area is under the influence of air mass flow from north-east to north-west in winter and from south-east to south-west in the summer. The temperature of Delhi varied from maximum in summer (March to June) to minimum in winter (November to February). The average rainfall in Delhi during monsoon (July to October) was of the order of ~900 mm. A detailed description of sampling site is available in Sharma et al. (2015).

$PM_{2.5}$ samples ($n = 140$) were collected on pre-combusted (at 550°C) and dessicator-stored quartz fibre filters (QM-A, Whatman, GE Healthcare UK Limited, Buckinghamshire, UK) by using a fine particle sampler (APM 550, Envirotech, Delhi, IN) at 10 m height above ground level. Ambient air was passed through the quartz filter (47 mm) at a flow rate of 1 $m^3\ h^{-1}$ (accuracy ±2 %) for 24 h. The filters were weighed before and after the sampling during the experiment in order to determine the mass of $PM_{2.5}$ collected. The quantitative elemental analysis (Mg, Al, S, Si, Cl, K, Ca, Ti, Cr, Mn, Fe, Zn, Cr, Br, As and Pb) of $PM_{2.5}$ samples was carried out first using non-destructive X-ray fluorescence spectroscopy with a Rigaku ZSX Primus wavelength dispersive X-ray fluorescence spectrometer (ZSX Primus WD-XRF, The Woodland, TX, USA). Then 6.25 cm^2 (2.5 × 2.5 cm^2) of each filter was used for analysis of water soluble inorganic ions (WSIC) by ion chromatograph (Dionex ICS-3000, Sunnyvale, CA, USA). The remainder of each filter was used for organic carbon (OC)/elemental carbon (EC) analysis with a carbon analyzer (DRI 2001A, Atmoslytic Inc., Calabasas, CA, USA). A more detailed description of the analytical methods, calibration procedures, etc. are available in our earlier paper (Sharma et al. 2015). Statistical analysis of $PM_{2.5}$ and its chemical species data was done using standard recommended methods and seasonal significance difference of chemical species of $PM_{2.5}$ was analyzed by one-way ANOVA test (Datta et al. 2010).

In the present study, PMF (v3.0) was used to quantify the contribution of various emission sources to $PM_{2.5}$ mass concentration (USEPA 2008). The model requires two input files: one of the measured concentrations of the species and another for the estimated uncertainty of the concentration. A detailed description of the PMF model has been presented in Paatero and Tapper (1994); Paatero (1997). A speciated data set can be viewed as a data matrix X of i by j dimensions, in which i number of samples and j chemical species are measured. The aim of multivariate receptor modeling (e.g., PMF) is to identify a number of factors p, the species profile f of each source, and the amount of mass g contributed by each factor to each individual sample which is given as:

$$X_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \tag{1}$$

where e_{ij} is the residual for each sample/species.

Results are constrained so that no sample can have a negative source contribution. PMF allows each data point to be individually weighed. This feature allows the analyst to adjust the influence of each data point, depending on the confidence in the measurement. For example, data below the detection limit can be retained for use in the model, with the associated uncertainty adjusted so these data points have less influence on the solution than measurements above the detection limit. The PMF solution minimizes the object function Q, based upon these uncertainties (u) as follows.

$$\boldsymbol{Q} = \sum_{i=1}^{\boldsymbol{n}} \sum_{j=1}^{\boldsymbol{m}} \left[\frac{X_{ij} - \sum_{k=1}^{p} g_{ik} f_{kj}}{u_{ij}} \right]^2 \tag{2}$$

where X_{ij} are the measured concentration (in µg m^{-3}), u_{ij} are the estimated uncertainty (in µg m^{-3}), n is the number of samples, m is the number of species and p is the number of sources including in the analysis. In this study, information on chemical properties of 140 $PM_{2.5}$ samples has been used as input to the PMF model for a total of 23 parameters. Categorization of quality of data was based on the signal to noise ratio (S/N) and the percentage of sample method detection limit (MDL). Those species which have $S/N \geq 2$ were categorized as strong in data quality. Those with S/N between 0.2 and 2 were categorized as weak in quality. These species are not likely to provide enough variation in concentration and therefore contribute to the noise in the results. Those species with an S/N ratio below 0.2 are classified as bad values and were thus excluded from further analysis. In the present case, signal to noise ratio (S/N) estimated as >0.6 and the model performance in a base run showed determination coefficient (R^2) between the modeled and experimental concentration of $PM_{2.5}$, OC, and EC of 0.97, 0.94 and 0.96, respectively and most of the other chemical species are also well reconstructed. These results are within the range of those presented in many PMF studies. For example, R^2 values of 0.71 were reported for a study in Spain (Cusack et al. 2013) and of 0.96 for a study in Germany (Beuck et al. 2011) for $PM_{2.5}$ mass reconstruction. Scaled residuals between −3 and +3 were obtained for all of the major components, and the value of Q robust is strictly identical to the value of Q true, all of these showing that no specific event was affecting the results and that the base run could be regarded as stable.

Results and Discussion

The temporal variation in mass concentration of $PM_{2.5}$, OC, EC and inorganic ions during the study are depicted in Fig. 1. The average concentration of $PM_{2.5}$ was 122 ± 94.1 μg m^{-3} (range 25.1–430 μg m^{-3}) from January, 2013, to December, 2014. The mass concentrations of almost all the chemical constituents were highest during the months of January, 2013, and January, 2014. The seasonal variation and average mass concentrations of OC, EC, WSIC and major and trace elements (Na, Mg, Al, S, Cl, K, Si, Ca, Cr, Ti, Fe, Zn, Mn, Br and Pb) of $PM_{2.5}$ with maxima and minima are summarized in Table 1. The average concentration of OC and EC of $PM_{2.5}$ was recorded as 19.9 ± 14.3 μg m^{-3} (~15 % of $PM_{2.5}$) and 10.4 ± 8.0 μg m^{-3} (~8 % of $PM_{2.5}$), respectively. The annual average of total carbon (TC = OC + EC) concentration contributed ~23 % of $PM_{2.5}$. Perrino et al. (2011) reported similar percentage contributions of OC (~12 % of PM_{10}) and EC (~3 % of PM_{10}) of PM_{10} at Delhi,

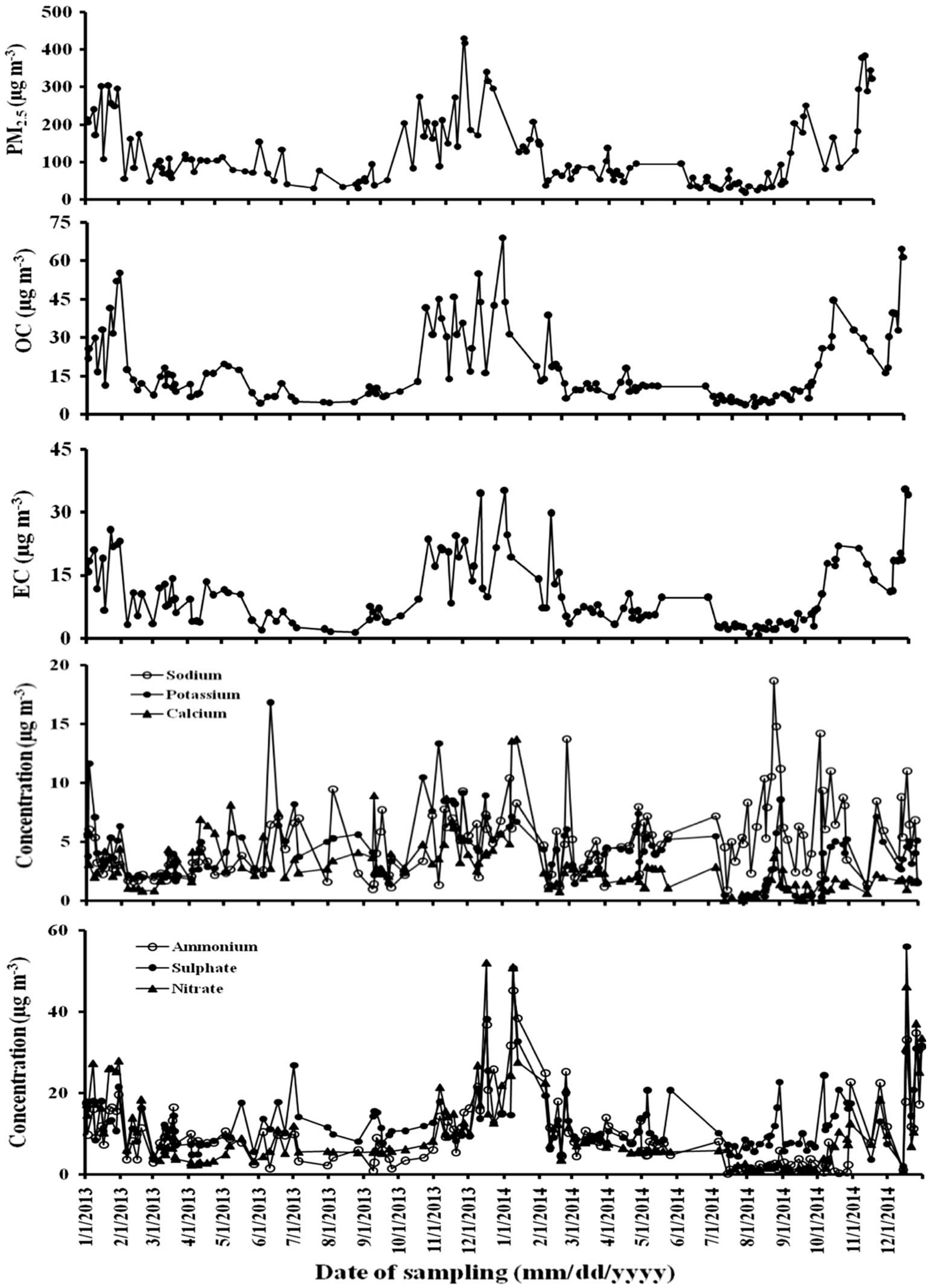


Fig. 1 Time series of OC, EC and WSIC in $PM_{2.5}$ along with $PM_{2.5}$ mass

Table 1 The average concentrations of particulate matter ($PM_{2.5}$), OC, EC, WSIC and trace elements of $PM_{2.5}$ ($\mu g\ m^{-3}$) in Delhi

Species	Average (n = 140)	Range	Seasons		
			Winter (n = 48)	Summer (n = 45)	Monsoon (n = 47)
$PM_{2.5}$	122 ± 94.1	25.1–430	$216^{a} \pm 93.2$	$81.8^{a} \pm 24.9$	$67.9^{a} \pm 56.1$
OC	17.9 ± 14.3	3.25–69.1	$31.0^{a} \pm 15.0$	$11.4^{a} \pm 3.72$	$10.1^{a} \pm 9.10$
EC	10.4 ± 8.04	0.85–35.3	$17.9^{a} \pm 7.77$	$7.15^{a} \pm 3.05$	$5.58^{a} \pm 5.41$
F^-	0.91 ± 0.69	0.05–3.72	$1.06^{b} \pm 0.80$	$0.89^{b} \pm 0.51$	$0.74^{b} \pm 0.72$
Cl^-	7.77 ± 5.72	0.89–31.3	$10.9^{a} \pm 6.68$	$5.64^{a} \pm 3.00$	$6.48^{a} \pm 5.19$
SO_4^{2-}	12.9 ± 8.08	1.97–56.2	$16.9^{b} \pm 11.2$	$10.3^{b} \pm 3.85$	$11.3^{b} \pm 5.13$
NO_3^-	10.0 ± 9.82	0.21–52.2	$18.9^{a} \pm 11.4$	$5.82^{a} \pm 2.03$	$4.18^{a} \pm 3.16$
NH_4^+	9.40 ± 8.59	0.17–45.3	$16.2^{a} \pm 10.4$	$8.34^{a} \pm 2.97$	$3.43^{a} \pm 3.75$
Na^+	5.05 ± 3.09	0.94–18. 7	$5.10^{b} \pm 2.85$	$3.84^{b} \pm 1.74$	$6.09^{b} \pm 3.84$
K^+	4.10 ± 2.70	0.31–16.8	$5.21^{b} \pm 2.51$	$4.05^{b} \pm 2.44$	$2.94^{b} \pm 2.60$
Mg	0.96 ± 0.88	0.11–4.28	$1.32^{b} \pm 1.60$	$0.44^{b} \pm 0.25$	$1.06^{b} \pm 0.63$
Ca	2.83 ± 2.21	0.35–13.7	$3.40^{b} \pm 2.76$	$2.99^{b} \pm 1.73$	$2.05^{b} \pm 1.69$
Al	2.86 ± 0.97	0.27–5.29	$3.43^{b} \pm 0.82$	$2.39^{b} \pm 0.80$	$2.70^{b} \pm 1.06$
S	2.81 ± 1.62	0.13–5.62	$3.41^{a} \pm 1.59$	$1.27^{a} \pm 0.98$	$1.36^{a} \pm 0.84$
Si	1.74 ± 0.66	0.42–4.64	$2.17^{a} \pm 0.55$	$1.52^{a} \pm 0.59$	$1.27^{a} \pm 0.47$
Cr	0.04 ± 0.13	0.03–0.92	$0.09^{b} \pm 0.19$	$0.02^{b} \pm 0.01$	$0.01^{b} \pm 0.01$
Ti	0.12 ± 0.22	0.01–0.98	$0.22^{b} \pm 0.31$	$0.05^{b} \pm 0.04$	$0.05^{b} \pm 0.11$
Fe	0.25 ± 0.43	0.08–2.01	$0.49^{b} \pm 0.58$	$0.11^{b} \pm 0.17$	$0.06^{b} \pm 0.14$
Zn	0.13 ± 0.21	0.02–1.36	$0.27^{b} \pm 0.27$	$0.03^{b} \pm 0.02$	$0.04^{b} \pm 0.08$
Mn	0.02 ± 0.02	0.002–0.12	$0.02^{b} \pm 0.02$	$0.01^{b} \pm 0.02$	$0.01^{b} \pm 0.02$
Cu	0.04 ± 0.06	0.002–0.13	$0.08^{b} \pm 0.20$	$0.01^{b} \pm 0.02$	$0.01^{b} \pm 0.02$
As	0.06 ± 0.16	0.002–0.68	$0.13^{b} \pm 0.23$	$0.01^{b} \pm 0.02$	$0.01^{b} \pm 0.02$
Br	0.03 ± 0.11	0.003–0.41	$0.06^{b} \pm 0.17$	$0.01^{b} \pm 0.01$	$0.04^{b} \pm 0.06$
Pb	0.02 ± 0.03	0.001–0.14	$0.04^{b} \pm 0.03$	$0.01^{b} \pm 0.01$	$0.01^{b} \pm 0.06$

± Standard deviation

[a] Significantly different ($p < 0.05$)

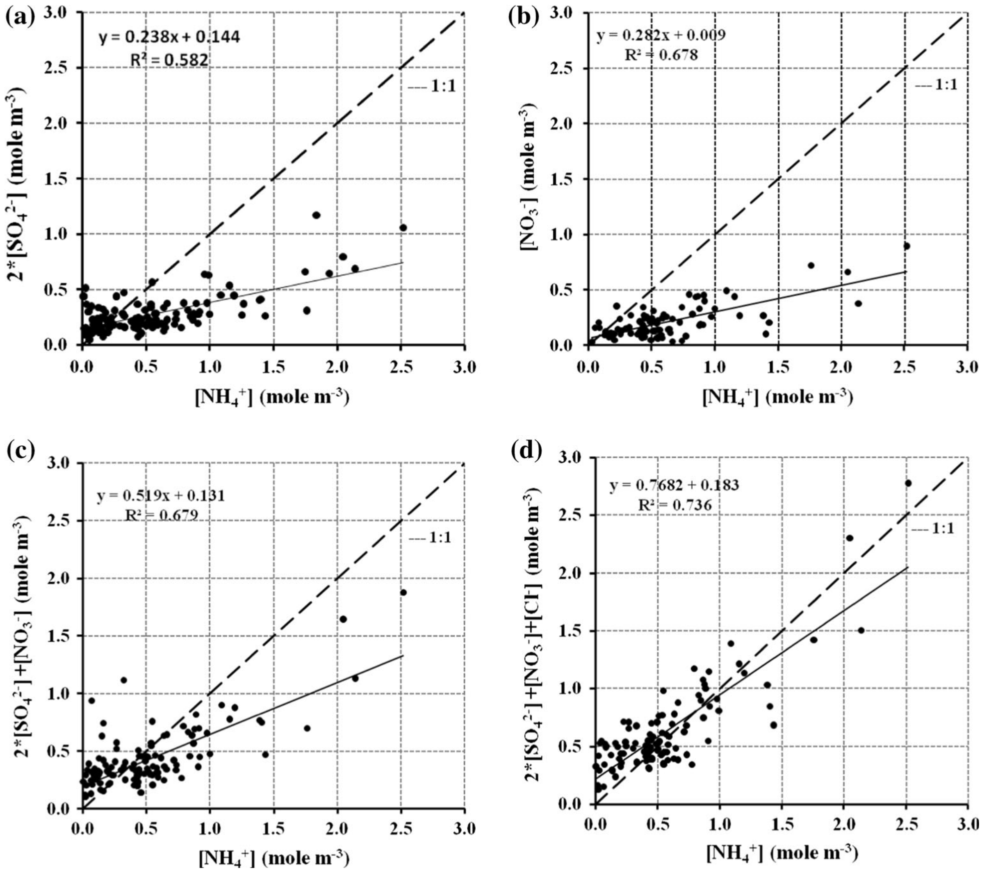
[b] Not significantly different ($p > 0.05$)

whereas Mandal et al. (2014) reported higher percentage contributions of OC (28 % of $PM_{2.5}$) and EC (9 % of $PM_{2.5}$) of $PM_{2.5}$ in an industrial area of Delhi. In the present study, the average concentration of major and trace elements in $PM_{2.5}$ was recorded as $29.0 \pm 2.3\ \mu g\ m^{-3}$ (22.3 % of $PM_{2.5}$).

The mass concentrations of $PM_{2.5}$, OC and EC varied significantly during winter, summer and monsoon seasons at Delhi (Table 1). During winter, the concentrations of $PM_{2.5}$, OC and EC were recorded as being more than twice the concentrations during the summer and monsoon seasons. This may be due to the source strength and prevailing meteorological conditions at the sampling site. Significant lowering of mixing height of the boundary layer during winter season may also contribute to the higher concentration of $PM_{2.5}$ during winter (Datta et al. 2010). Strong positive linear relationships between OC and EC ($R^2 = 0.91$; at $p < 0.05$), OC versus $PM_{2.5}$ ($R^2 = 0.87$) and EC versus $PM_{2.5}$ ($R^2 = 0.82$) were recorded during the study. A significant linear correlation between OC and EC of $PM_{2.5}$ is usually indicative of their common sources like vehicular traffic and biomass burning (Salma et al. 2004; Sharma et al. 2014).

The average concentration of WSIC and its seasonal variability are summarized in Table 1. During the study, the concentrations of NH_4^+, SO_4^{2-} and NO_3^- of $PM_{2.5}$ was recorded as 9.4 ± 8.6, 12.9 ± 8.1 and $10.0 \pm 9.8\ \mu g\ m^{-3}$, respectively. The water soluble inorganic ionic species (WSIC) accounted for ~40 % of $PM_{2.5}$ concentration during the study with seasonal variability (Table 1). The study revealed positive linear correlations of molar mass between NH_4^+ and SO_4^{2-} ($R^2 = 0.65$), NH_4^+ and NO_3^- ($R^2 = 0.69$) of $PM_{2.5}$, as well as for charge balance between SO_4^{2-} and NH_4^+; NO_3^- and NH_4^+; SO_4^{2-}, NO_3^- and NH_4^+; and SO_4^{2-}, NO_3^-, Cl^- and NH_4^+. The above correlations indicate the possible formation of secondary aerosols [$(NH_4)_2SO_4$, NH_4NO_3 and NH_4Cl] at the sampling site. NH_4^+ generally combines with NO_3^- and SO_4^{2-} in the atmosphere to form NH_4NO_3 and $(NH_4)_2SO_4$, respectively. Figure 2 shows the charge balance between SO_4^{2-} and NH_4^+ ($R^2 = 0.58$); NO_3^- and NH_4^+ ($R^2 = 0.67$); SO_4^{2-}, NO_3^- and NH_4^+ ($R^2 = 0.67$); SO_4^{2-}, NO_3^-, Cl^-

Fig. 2 Charge balance: **a** between SO_4^{2-} and NH_4^+; **b** between NO_3^- and NH_4^+; **c** between SO_4^{2-}, NO_3^- and NH_4^+; **d** between SO_4^{2-}, NO_3^-, Cl^- and NH_4^+



and NH_4^+ ($R^2 = 0.73$) in Delhi during the study. The charge balance (in Fig. 3 a-c) is well below a 1:1 relationship, indicating an excess of NH_4^+ compared to anions (Behra and Sharma 2010). The charge balance between SO_4^{2-}, NO_3^-, Cl^- and NH_4^+ ($R^2 = 0.73$) in Fig. 3d was closer to the 1:1 line confirming that most of the time sufficient NH_4^+ was present to neutralize the acidic components (H_2SO_4, HNO_3 and HCl) to form $(NH_4)_2SO_4$, NH_4NO_3 and NH_4Cl (Sharma et al. 2015).

The PMF was applied to the analyzed data set consisting of 23 species and 140 $PM_{2.5}$ samples collected at sampling site. For the final analysis, PMF was applied to the data sets using factors and the resultant change in the Q values was examined. In this study, the theoretical Q value was to be approximately 3220 (i.e., 140 × 23). In seven factor solutions, more than 95 % of Q values were quite close to 3220. Based on an evaluation of the model results for the Q value variations, a seven-factor solution provided the most feasible results. The descriptions of the model and source apportionment of PM have been discussed in detail in our previous paper (Sharma et al. 2015). The mass fraction distribution of species was used to identify the sources, which were soil dust, vehicular emission, sea salt, industrial emission, secondary aerosol, biomass burning and fossil fuel combustion for $PM_{2.5}$ mass (Fig. 3).

A discussion follows of the seven major sources of $PM_{2.5}$ air pollution at the sampling site.

Source 1 Present PMF analysis shows that secondary aerosols have contributed to about 21.3 % for $PM_{2.5}$ mass concentrations, respectively. Secondary aerosols are mainly composed of ammonium sulphate and nitrate deriving primarily from the gaseous precursors NH_3, SO_2 and NO_x. The abundance of gaseous NH_3, SO_2 and NO_x at Delhi (Sharma et al. 2012) supports the presence of secondary aerosols over the region. The key markers of secondary aerosols are NO_3^-, SO_4^{2-} and NH_4^+, as shown in Fig. 3.

Source 2 PMF analysis showed that soil dust contributed 20.5 % of aerosol mass in $PM_{2.5}$ at sampling site. Soil dust includes most of the crustal elements and has high concentrations of Fe, Ca, Na, Mg, Al and K (Lough et al. 2005). The concentration of Ca in $PM_{2.5}$ is associated with its resuspension from agricultural fields or bare soils by local winds. Crustal elements typically used as tracers for soil and/or crustal resuspension include Al, Si, Ca, Mg, Fe and Na (Begum et al. 2006). A whole array of element tracers has been used in India for identification of this source type, including Al, Si, Ca, Ti, Fe, Pb, Cu Cr, Ni, Co and Mg (Sharma et al. 2014).

Source 3 Vehicle exhaust is generally dominated by elemental carbon, Cu, Zn, Ba, Sb, Pb, Mn, Mo and Ni

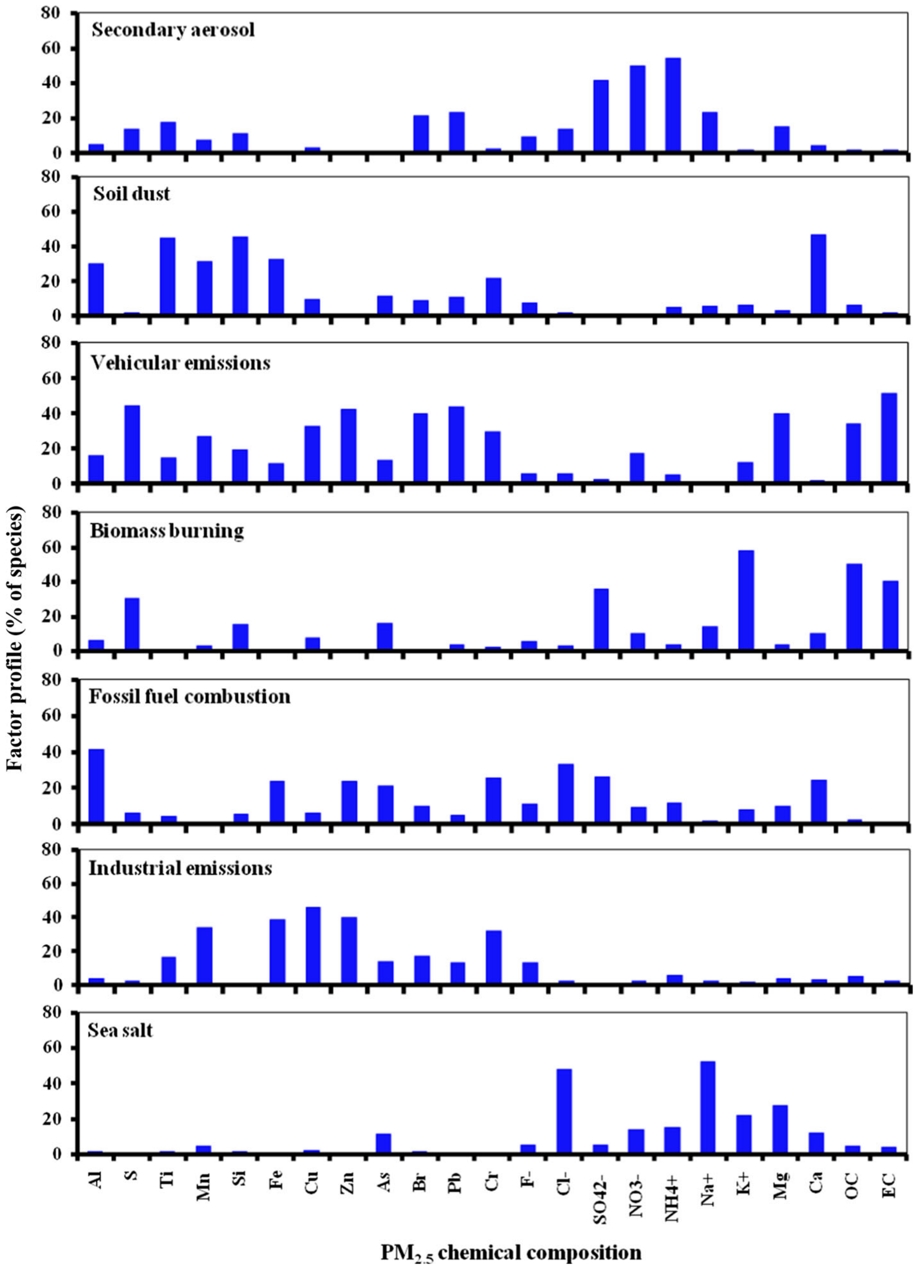


Fig. 3 PMF factor profiles of soil dust, secondary aerosol, industrial emissions, biomass burning, vehicle emissions, fossil fuel combustion and sea salt of $PM_{2.5}$

which are widely used as markers of vehicular sources. In the present study, Cu, Zn, Mn, Pb and EC contributed 32.7 %, 42.6 %, 26.9 %, 43.5 % and 51.3 %, respectively, indicating vehicle emissions as the source. PMF analysis indicated that vehicle emissions contributed 19.7 % in $PM_{2.5}$ at Delhi. Internationally, EC (Lee and Hopke 2006) is used extensively as a marker for diesel exhaust. In India, Cu, V, Mn, Co, Pb and Zn have been used as tracer elements for identification of vehicular emission. Vehicular emissions are a major source of the PM and research indicates that they contribute 10 %–80 % to PM in cities across India.

Source 4 Biomass burning, wood burning and vegetative burning have been characterized as having high concentrations of K^+ and SO_4^{2-} by various source studies (Wu et al. 2007). The potassium ion has been used in many source apportionment studies conducted in Europe and Asia as an indicator of biomass burning (Pant and Harriso 2012). The PMF analysis showed that biomass burning contributed 14.3 % for $PM_{2.5}$ mass in the present study. In India, K^+ has been used as a key marker for biomass/wood combustion for TSP, PM_{10} and $PM_{2.5}$ (Shridhar et al. 2010), whereas levoglucosan is the key organic marker (Chowdhury et al. 2007).

Source 5 The higher concentrations of Al, Cl, Fe, Zn, Cr and SO_4^{2-} at the sampling site clearly indicate the source of fossil fuel combustion of $PM_{2.5}$. The PMF analysis showed that fossil fuel burning contributed 13.7 % for $PM_{2.5}$ in the present study. In international studies, a key marker for coal combustion includes As, Se, Te and SO_4^{2-} and it has contributed 6 %–30 % to PM in different studies (Gupta et al. 2007; Sharma et al. 2007).

Source 6 The results of the PMF analysis show that industrial emissions accounted for about 6.2 % for $PM_{2.5}$ mass concentration. Begum et al. (2006) used Ni, Pb and S as markers for industrial emission, whereas Song et al. (2006) used Ni, Cr, Fe and Mn, and Tauler et al. (2009) used Zn, Fe, Mn and Cd. Generally Zn, Cu, Mn, S, Ni, Cd, Fe, Mo and Cr are used as tracers for industrial emission IE in India (Sharma et al. 2014).

Source 7 Higher concentrations of Na, K and Cl (52.2 %, 27.2 % and 47.6 %, respectively) in $PM_{2.5}$ indicate the possible contribution of sea salt, which is supported by PMF analysis. The use of K offers possible confusion with wood/biomass burning combustion and Cl with coal burning, but a combination of the four elements (Na, K, Cl^- and Mg) should provide a reliable signature. In the present study, PMF analysis shows that sea salt contributed to about 4.3 % for $PM_{2.5}$ mass concentration.

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