



## Characterization of PM<sub>10</sub> and PM<sub>2.5</sub> Emission Sources at Chennai, India

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### ABSTRACT

Deteriorating ambient air quality is a substantial problem of concern for many urban agglomerations throughout the world. Particulate matter (PM) is a criteria pollutant that is of high interest in urban locations. The precise characteristics of PM in a given locale depend on the source origin, which in turn is a function of economic, social and technological factors. In order to effectively manage PM and thereby, the exposure risk to humans, it is very essential to identify the main sources and their contributions from source emissions. Receptor modelling plays a major role in identifying and apportioning sources of airborne PM across the world. Unmix model is a multivariate receptor model developed by the United States Environmental Protection Agency (U.S.EPA) based on factor analysis, which estimates the number of sources using a singular value decomposition method to reduce the dimensionality of data. In this study, Unmix receptor model version 6.0 is used to identify and quantify the sources of PM at Chennai; a metropolis in southern India. A total of 29 elements (Ag, Al, As, B, Ba, Be, Bi, Ca, Cd, Co, Cr, Cu, Fe, Ga, K, Li, Mg, Mn, Mo, Na, Ni, Pb, Rb, Se, Sr, Te, Tl, V and Zn) and ten ions (Na<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, F<sup>-</sup>, Cl<sup>-</sup>, NO<sub>2</sub><sup>-</sup>, NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup>) were analysed to find the chemical characteristics of PM<sub>10</sub> and PM<sub>2.5</sub>. Four sources were identified for both PM<sub>10</sub> and PM<sub>2.5</sub>. Vehicular pollution (11%), crustal source (27%), marine aerosol (40%) and industrial source (22%) are the sources identified for PM<sub>10</sub>. Vehicular emissions (32%), secondary aerosol (13%), marine aerosol (33%) and industrial source (22%) are the sources identified for PM<sub>2.5</sub>.

### INTRODUCTION

Over the last few centuries, many urban centres have propped up around the globe. Urbanization caused a massive population influx into minuscule parcels of land. These urban centres are therefore regions of very high anthropogenic activity. This immense human activity combined with poor planning in urban development usually results in very high pollution in these regions. Studies have shown that people exposed to higher levels of air pollution are likely to have increased morbidity and mortality (Pope et al. 1999).

The types of pollutants in the ambient atmosphere are monumental. However, a few pollutants; due to either their ease of access into the environment or their inherent toxicity or both, have been under strict scrutiny by most regulating bodies. Of these six common "criteria pollutants" (SO<sub>x</sub>, NO<sub>x</sub>, CO, O<sub>3</sub>, PM and Pb), particulate matter (PM) is considered to be the most dangerous (U.S. EPA 2016). PM is known to cause a wide spectrum of ailments ranging from coughing to cardiovascular disorders. A 10 µg/m<sup>3</sup> increase in PM is known to cause a 2.5% to 3% increase in hospital visits by patients (Gordian et al. 1999). The composition and properties of PM vary significantly with time and space. Consequently, unlike other criteria pollutants, identification of sources of PM is a colossal task. The common approach of

apportioning the sources of PM is using receptor modelling.

Receptor modelling is a process of mathematically extracting the sources of PM from the concentration data at a receptor. Receptor models are used widely throughout the world for source apportionment. Most of the receptor models lie in a spectrum between scientific and statistical models (Belis et al. 2014). Unmix is a receptor model that is developed by the United States Environmental Protection Agency (U.S.EPA), specifically for the source apportionment of PM. Unmix tries to obtain a solution to the chemical mass balance problem using multidimensional geometry, thus trying to extract enough constraints for solving the problem using the available data. Unmix has been used with varying levels of success throughout the world in identifying and apportioning PM. This paper tries to identify and apportion the sources of PM<sub>10</sub> and PM<sub>2.5</sub> from Chennai; an urban region in the southern peninsula of India using Unmix receptor model.

### MATERIALS AND METHODS

#### Study Site

The city of Chennai (Erstwhile Madras) is one of the biggest urban agglomerations in the southern peninsula of

India. The study location is situated at a well build residential and commercial centre of Chennai city (12°59'29.44"N, 80°14'1.03"E). The site is located adjacent to Sardar Patel road (Fig. 1), with an average traffic flow of 0.17 million vehicles per day on weekdays and 0.14 million vehicles per day on weekends during the course of the study. The study location is flanked by two intersections towards the east and west. The study site is blanketed with multiple academic and research centres like Indian Institute of Technology Madras, Central Leather Research Institute, Anna University, Adayar Cancer Centre and Central Polytechnic. Many tourist locations and recreational sites like a children's park, Kamarajar Mandapam and Gandhi Mandapam are located in and around the study site. Commercial centres like software development firms are also present in the vicinity of the sampling site. Thus, the site is rife with immense human and vehicular traffic flow.

### Sampling and Analysis

The sampling is carried from 2008 - 2009. The samples are collected from 10<sup>th</sup> of November 2008 to 12<sup>th</sup> of April 2009. A total of 63 samples were collected during the sampling period at 24-hour intervals. The samples were collected at noon to noon intervals in such a way that all days of the week were equally represented in the study. The sampling was performed at a distance of 6.5 meters from Sardar Patel Road.

**Sampling setup:** An Envirotech APM550 sampler (Envirotech Instruments India Pvt. Ltd.) was used for measuring the mass concentrations of PM<sub>10</sub> and PM<sub>2.5</sub>. A volumetric flow rate of 1 m<sup>3</sup>/hr was maintained in the instrument during the sampling interval. The unidirectional inlet of the sampler was used to cut out particles of aerodynamic diameter greater than 10 µm from entering the flow stream. Particles greater than 2.5 µm were cut off from the stream using a second WINS impactor. For measuring the concentration of PM<sub>10</sub>, the second WINS impactor was replaced with a longer down tube. The samples were collected on a Teflon filter paper.

**Sample Extraction:** Gravimetric analysis was performed on the filter papers once they were retrieved from the sampling instrument. Post gravimetric analysis, the filter papers were divided into two equal halves. One half would undergo acid digestion for extraction of elements from the sample. The other half was used for extraction of ions.

**Elemental composition:** The metallic elements were extracted using hot plate digestion (Srimuruganandam & Nagendra 2011). The sample digestion was carried out as prescribed by U.S.EPA Compendium Methods IO 3.1 (U.S.

EPA 1999a) and IO 3.5 (U.S. EPA 1999b). After hot plate digestion, the extracted sample was injected into an inductively coupled plasma optical emission spectroscopy (Optima 5300-DV ICP-OES). Twenty nine elements were measured using ICP-OES (Ag, Al, As, B, Ba, Be, Bi, Ca, Cd, Co, Cr, Cu, Fe, Ga, K, Li, Mg, Mn, Mo, Na, Ni, Pb, Rb, Se, Sr, Te, Tl, V and Zn). Of these, 21 elements were selected for PM<sub>10</sub> and 20 were selected for PM<sub>2.5</sub>. The rejected species are either completely absent or have more than two-thirds of their values missing.

**Ionic composition:** The second half of the filter paper was used for the extraction of ionic species as described by the standard operating procedure (SOP) by CPCB, New Delhi. The extracts were injected into an ion chromatography machine (Metrohm, Compact IC 761) using a syringe filter. Five cations Na<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup> and Mg<sup>2+</sup> and five anions F<sup>-</sup>, Cl<sup>-</sup>, NO<sub>2</sub><sup>-</sup>, NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup> were analysed.

### Theory of Unmix

Unmix is a multivariate receptor model that is used in source apportionment of PM. It uses N-dimensional edge detection to obtain a solution to the chemical mass balance problem. Unmix 6.0 available from U.S.EPA's web portal was used in this study. The fundamental scientific theory behind the Unmix model is chemical mass balance. The apportionment results are obtained by solving a set of mass balance equations. The concentration of a pollutant at a receptor can be described as the sum total of the product of concentration of pollutant in the emission of a given source to the contribution of that source to the pollution at the receptor under scrutiny. This can be expressed mathematically as in equation 1.

$$C_{in} = \sum_{n=1}^a x_{in} y_n \quad \dots(1)$$

Where, *i* is the pollutant under study, *n* is the source, *a* is the number of sources, *x* is the concentration of pollutant at the source emission and *y* is the contribution of that specific source to the pollution at the receptor.

Depending on the number of pollutants under study, there would be a total of *i* sets of equations. The solution to these equations would yield a value for *y<sub>n</sub>*, which is the contribution of source *n*. These equations form the backbone of majority of the receptor models. The principal difference between the various receptor models is the process used to obtain the solution. These equations constitute an ill-posed problem (Henry 1987). The number of unknowns is a lot larger than the number of equations. Consequently, there would be many numbers of possible solu-

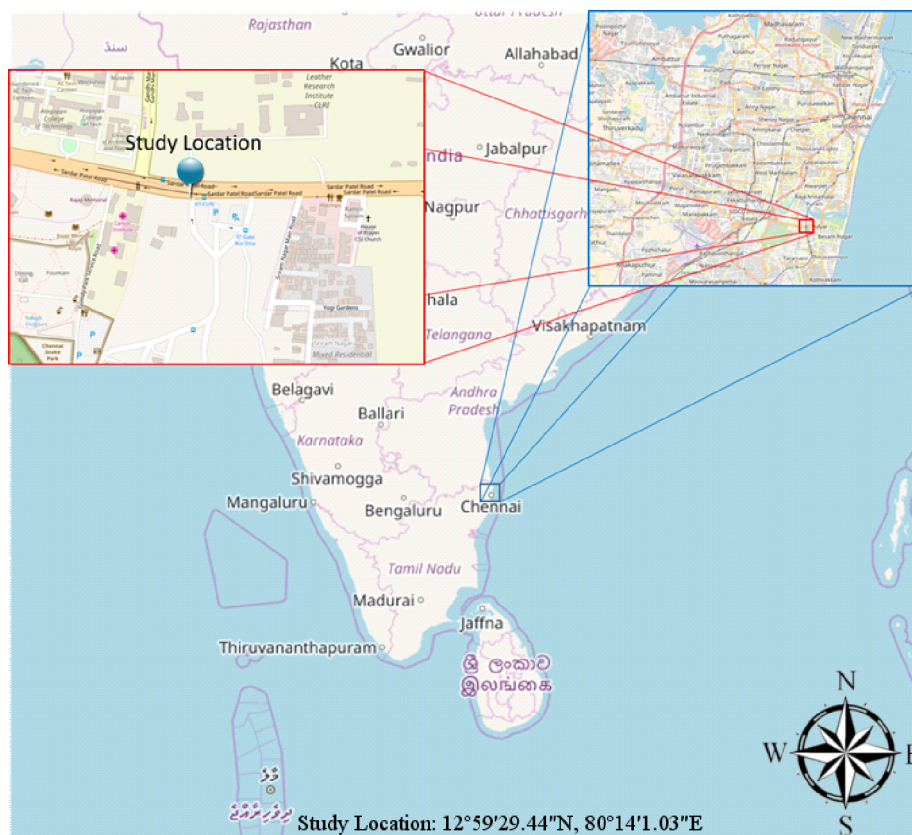


Fig. 1: Location of study site.

tions to these equations. From a mathematical point of view, these equations are unsolvable. However, there is a chance of solving these equations by introducing more constraints. Since mass concentrations can never be negative, the non-negativity constraint is one of the most common constraints used in receptor models. Often this is not enough to obtain a complete solution. Under certain conditions, the data itself provides the necessary constraints (Henry 1991). Unmix tries to obtain a total solution for the mass balance equations using these constraints.

If the data are assumed to be made up of  $M$  species, the data can then be plotted on an  $M$ -dimensional data space where the coordinates of a data point are the observed concentrations of the species during a sampling period. If  $N$  sources are present,  $N-1$ -dimensional space can be used to represent the given data. It is assumed that for each source there are some data points where the contribution of the source is not present or small compared to the other sources. These are called edge points and Unmix works by finding these points and fitting a hyperplane through them; this hyperplane is called an edge (if  $N = 3$ , the hyperplane is a line). By definition, each edge defines the points where a

single source is not contributing. If there are  $N$  sources, then the intersection of  $N-1$  of these hyperplanes defines a point that has only one source contributing. Thus, solving the equations at this point gives source composition. In this way, the composition of the  $N$  sources are found, and from this, the source contributions are calculated so as to give a best fit to the data (Henry 2003).

## RESULTS AND DISCUSSION

In this study, the source identification and apportionment of the data were carried out on the basis of both elemental and ionic concentrations. Four sources were identified by Unmix for PM<sub>10</sub>. However, SO<sub>4</sub><sup>2-</sup> and NH<sub>4</sub><sup>+</sup> show bad correlation between measured and predicted concentrations for PM<sub>10</sub>. Hence, their contributions are ignored in the source apportionment of PM<sub>10</sub>.

**Source apportionment of PM<sub>10</sub>:** Fig. 2 shows the contribution of different species apportioned in PM<sub>10</sub>. Cd (100%), Na (51%), Cu (37%), K (38%) and Zn (37%) are the elements with the highest loading for the first source. High concentration of Cd, Zn and Cu suggests that this is a traffic related source. Presence of these elements signifies the pres-

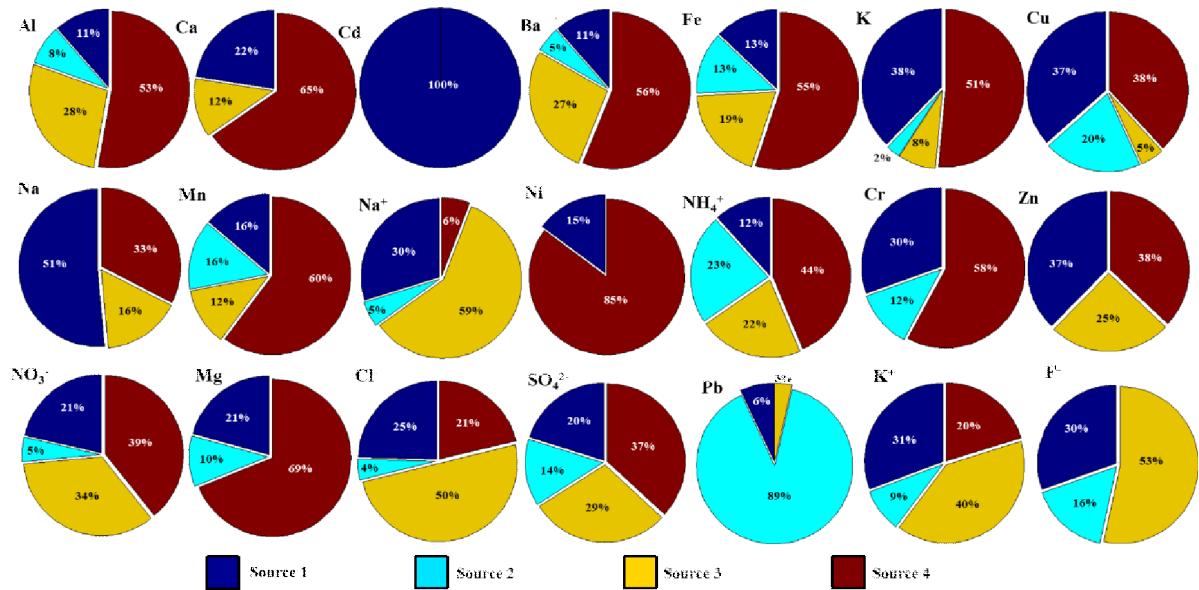


Fig. 2: Percentage contribution of chemical species in PM<sub>10</sub> mass from the Unmix results.

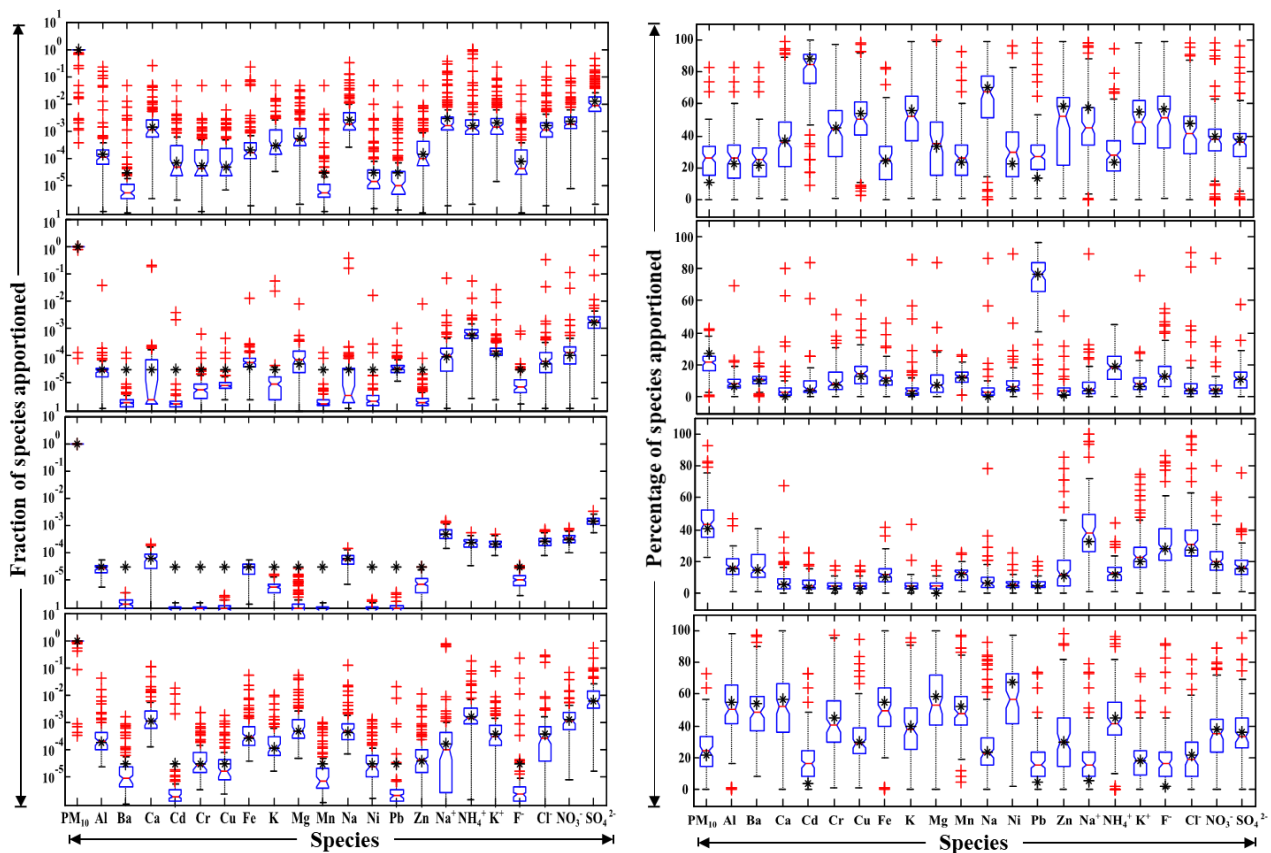


Fig. 3: Fraction and percentage of species apportioned in 24-hr averaged PM<sub>10</sub> mass.

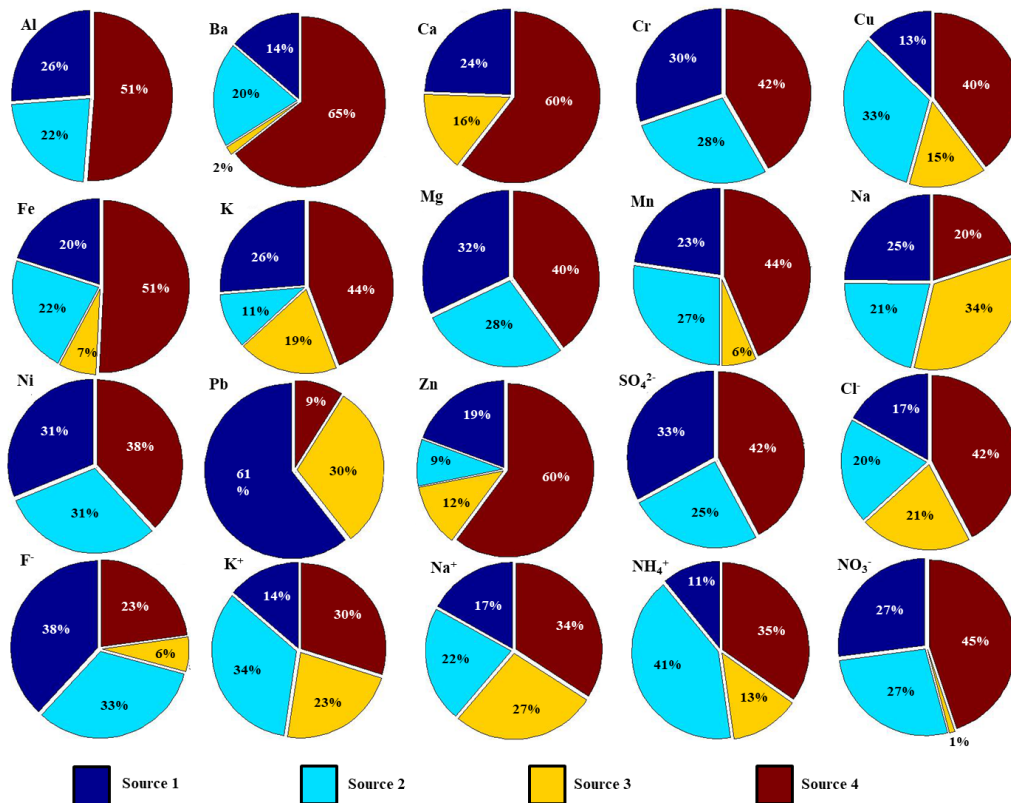


Fig. 4: Fraction and percentage of species apportioned in 24-hr averaged PM<sub>2.5</sub> mass.

ence of particles generated from tyre and brake wear. Combustion products from vehicular exhaust however cannot be identified because concentrations of EC and OC are not weighed up in this study. As per a study done in 2001, Cu and Zn are the major components of brake lining dust (Westerlund 2001). In another separate study, Zn, Pb, Cr and Cu were found to be the major heavy metal contents in surface runoff water from urban roads (Legret & Pagotto 1999). Brake linings are a major contributor of Cu. Zn is mostly generated by tyre wear. The reason Pb is low in this source is because lead emission from vehicles reduced significantly since the ban on tetraethyl lead, an anti-knock additive that is commonly added to petrol.

The second source shows high concentration in Pb (90%), Cu (20%), Mn (15%) and Fe (13%). This could be an industrial source. Chennai has a large number of industries in its vicinity. The major ones being the automobile and electronics industries. Although it is extremely difficult to pinpoint the exact origin of the second source, it could be fairly assumed that this is probably an industrial source. Cr and Cu may be generated from electroplating industry, while the source of Pb is probably from the manufacturing and

handling of lead acid batteries. Pb can also be generated from iron and steel foundries.

Major loading of Na<sup>+</sup> (59%), Cl<sup>-</sup> (50%), F<sup>-</sup> (54%) and K<sup>+</sup> (40%) signifies the presence of marine aerosol in the third source (Cheng et al. 2000, Guo et al. 2009, Li et al. 2015). Chennai is a coastal city and the study location is 5 kms away from the Bay of Bengal. The location is thus subjected to significant amount of sea breeze. An easterly prevailing wind direction, especially during winter due to the retreating monsoons, will cause an abundance in marine aerosol in the atmosphere. The generation of marine aerosol is by the action of wind. Wind generates stress in the surface of the sea, creating air bubbles. These air bubbles later burst releasing a jet of small particles (Blanchard & Woodcock 1980). Another source of large spray droplets is spume drops generated by the mechanical disruption of wave crests by the wind (Wang & Street 1978). Most of the marine aerosol is likely to be coarse while their influence in fine particulate matter is likely to increase with proximity to a marine body. This is most likely the reason why the contribution from this source is much greater for coarse fraction when compared to fine fraction. The presence of NO<sub>3</sub><sup>-</sup> with marine

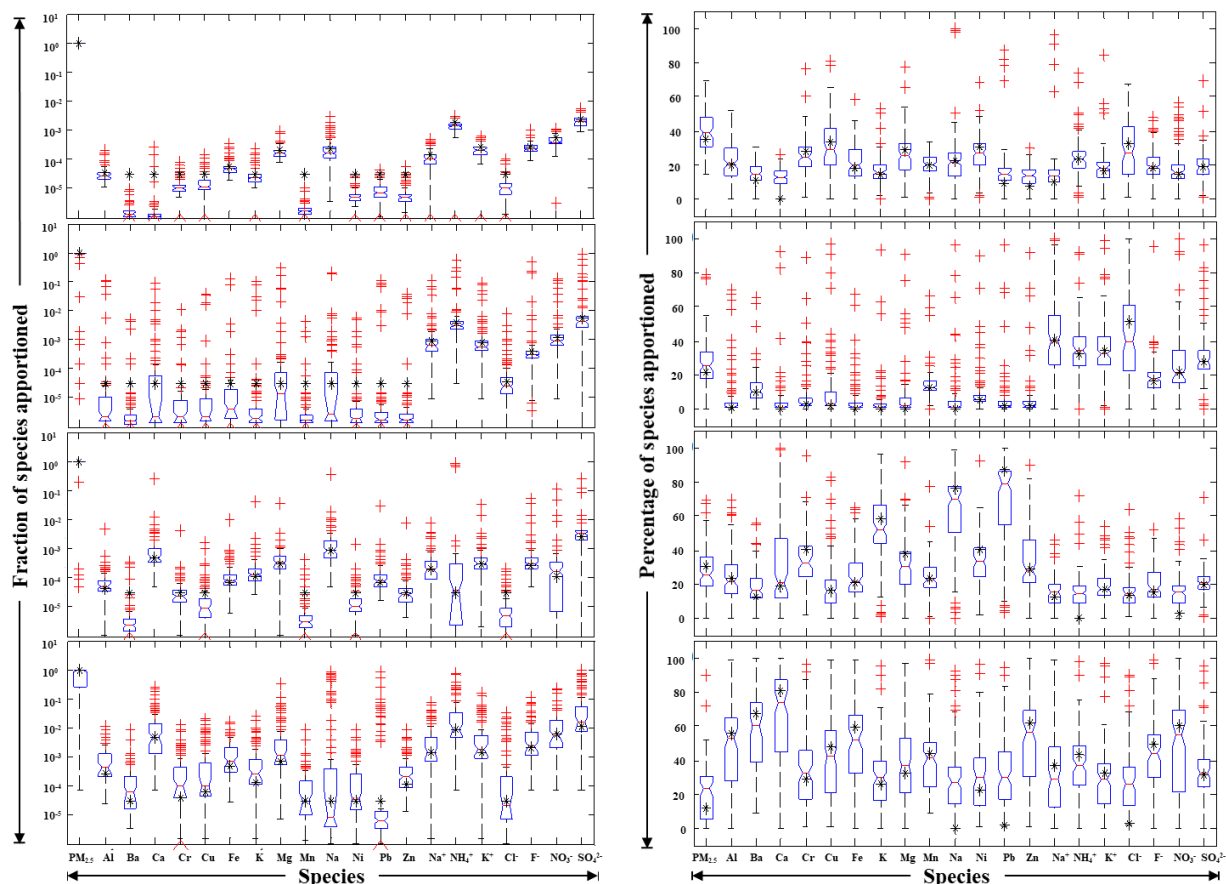


Fig. 5: Fraction and percentage of species apportioned in 24-hr averaged  $PM_{2.5}$  mass.

aerosols is most likely due to the generation of secondary PM on reacting with marine aerosol.

The presence of Ni (85%), Mg (69%), Ca (65%), Mn (59%), Cr (58%), Fe (55%) and Al (58%) could signify a crustal origin for this source. This includes both resuspended road dust and wind-blown soil dust. Road dust is generated by the continuous pounding of road surfaces by heavy vehicles. The quick loading and unloading cycle causes the road surface to crack and release particulates. Ca (65%), Al (53%) and Fe (55%) are the major indicators for contamination from road pavement dust. Presence of  $K^+$  (20%) shows the presence of biological materials in the crustal source. Significant contribution of Na (33%) and K (52%) from this source would likely be due to the presence of settled marine dust. The contribution of this source to  $PM_{10}$  is comparatively low due to the absence of Si, which is a major component of crustal dust, from this study (Watson et al. 2002). Fig. 3 shows the percentages and fractions of species apportioned to each different sources of  $PM_{10}$  in a box whisker diagram. Source 1 is at the top. The different points plotted

in the graph indicates the apportionment result for each day during the observation period. The median, first quartile and third quartile values of percentage and fractions of species apportioned to individual sources are shown in the respective plots.

**Source apportionment of  $PM_{2.5}$ :** Similar to  $PM_{10}$ , Unmix provided a four source solution for  $PM_{2.5}$ . Fig. 4 shows the contribution of different species apportioned in  $PM_{2.5}$ . The first source has very high loadings for Pb (61%),  $SO_4^{2-}$  (33%), F<sup>-</sup> (38%), Mg (32%), Cr (30%) and Ni (31%). This suggests an industrial source of origin. The high loading of Pb (61%) suggests emissions from battery refurbishing plants in the region. Lead based electrical solder used by the electronic industries could also be a major source of Pb. These plants could also be responsible for the presence of sulphate, as  $PbSO_4$  is used extensively in the manufacture of lead acid batteries. Cr and Ni could be generated from electroplating industries.

Second source is high in  $NH_4^+$  (41%), F<sup>-</sup> (33%),  $K^+$  (40%), Ni (31%), Mg (28%),  $NO_3^-$  (27%) and  $SO_4^{2-}$  (25%). The pres-

ence of NH<sub>4</sub><sup>+</sup> and SO<sub>4</sub><sup>2-</sup> is generally indicative of secondary PM formation and photo-chemical reactions (Cheng et al. 2000). H<sub>2</sub>SO<sub>4</sub> formed from the dissolution of primary sulphates in water vapour can react in both gas and aqueous phases. The primary gas phase reaction is with the hydroxyl radical, while the primary reaction in the aqueous phase is from oxidation of dissolved ozone. Nitrates and nitrites are formed in the atmosphere by reaction of fixed soil nitrogen compounds with atmospheric H<sub>2</sub>SO<sub>4</sub>. Generally F<sup>-</sup> in the atmosphere is attributed to vaporization of HF formed in the atmosphere.

The third source has high loadings for Na (34%), Pb (30%), Na<sup>+</sup> (27%), K<sup>+</sup> (23%) and Cl<sup>-</sup> (21%). These could be indicators for marine aerosol. The fraction of marine aerosol in PM<sub>10</sub> is found to be greater compared to that in fine PM; 13% for PM<sub>2.5</sub> compared to 40% for PM<sub>10</sub>. This is expected because the fine fraction of marine aerosol is found to decrease with increase in distance from a water body (Wang et al. 2016). Generation of marine aerosol is similar in many respects to that of wind blown dust. They are generated by winds that interact with air bubbles that escape from the sea.

The final source for PM<sub>2.5</sub> is found to contain Ba (64%), Ca (60%), Zn (60%), Al (60%), Fe (51%), K (45%) and Mn (44%). These elements are characteristics of a vehicular combustion source. High loading of barium is due to the use of organometallic fuel additives (Truex et al. 1980). Premature gas to particle conversion within the engine results in the deposition of barium as grey or white particles in engine valves. The primary source of Al is from piston wear. Presence of Fe can be due to both the wear of metal particles in the exhaust system and emissions from ferrocene additives in diesel fuels (Kasper et al. 1999). However, owing to small particle size, the latter is expected to be the case. Fig. 5 shows the percentages and fractions of species apportioned to each different sources of PM<sub>2.5</sub> in a box whisker diagram.

## CONCLUSION

PM<sub>10</sub> and PM<sub>2.5</sub> samples were collected for this study at Sardar Patel road in Chennai city from November 2008 to April 2009. A total of 29 elements (Ag, Al, As, B, Ba, Be, Bi, Ca, Cd, Co, Cr, Cu, Fe, Ga, K, Li, Mg, Mn, Mo, Na, Ni, Pb, Rb, Se, Sr, Te, Tl, V and Zn) and ten ions (Na<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup> and Mg<sup>2+</sup>, F<sup>-</sup>, Cl<sup>-</sup>, NO<sub>2</sub><sup>-</sup>, NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup>) were analysed to find the chemical characteristics of PM and run the Unmix receptor model. Various sources and their corresponding contribution to PM<sub>10</sub> and PM<sub>2.5</sub> was extracted using Unmix receptor model. A total of four factors were identified for both PM<sub>10</sub> and PM<sub>2.5</sub>. Twenty one species were selected for the analysis of PM<sub>10</sub>. The model extracted four sources, i.e.,

vehicular pollution (11%), crustal source (27%), marine aerosol (40%) and an industrial source (22%). Cd was removed from analysis of PM<sub>2.5</sub> due to low signal to noise ratio. The minimum R<sup>2</sup> value was found to be 0.79 and minimum signal to noise ratio to be 2.22 for PM<sub>10</sub>. For PM<sub>2.5</sub>, the minimum R<sup>2</sup> value was found to be 0.84 and minimum signal to noise ratio was found to be 2.47. A total of 20 species were selected for the analysis. Like PM<sub>10</sub>, four sources were extracted for PM<sub>2.5</sub>. Vehicular emissions (32%), secondary aerosol (13%), marine aerosol (33%) and industrial source (22%) are the four sources identified by Unmix.

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