



Application of Statistical Methods in Source Apportionment of Atmospheric Aerosols

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Abstract

Several thousands of articles have been published globally to apportion sources of atmospheric aerosols using various statistical tools and marker elements present in the aerosols in the urban cities of the world in designing mitigation strategies to improve the air quality. In this study, the effectiveness of various statistical methods used in source identifications and quantifications of atmospheric aerosols are discussed. This paper also discussed the importance of source identifications of aerosols chemistry using various receptor models. We have applied IMPROVE (Interagency Monitoring of Protected Visual Environments), PCA (principal component analysis) and PMF (positive matrix factorization) models on chemical species of PM_{2.5} collected at an urban site of Delhi, India, over the period January to December 2021 and explored the better and accurate source analysis of the PM_{2.5}. A comparative analysis of these models was conducted to assess their performance in resolving source contributions. The results highlight the strengths and limitations of each method and offer insights into their applicability for accurate source apportionment in complex urban environments.

Keywords: Aerosols, Source apportionment, IMPROVE, PCA, PMF

1. Introduction

The quantification of the plausible sources to the atmospheric aerosols /particulate matter (PM) is a critical step toward designing effective mitigation and improving urban air quality, especially in regions with severe pollution burdens like India [1]. Hence, accurate source apportionment enables policymakers, researchers, and urban planners to identify the major contributors to PM levels and implement targeted interventions. To achieve reliable source identification, the development and application of advanced statistical techniques have become increasingly important [2-4]. Source apportionment of aerosols is essential tool for identifications of specific aerosol concentrations to the adverse health effects [5;6]. There are several statistical methods [enrichment factors (EFs), principal component analysis (PCA), IMPROVE protocol, chemical mass balance (CMB), UNMIX and positive matrix factorization (PMF) etc.] are being used for source apportionment of pollutants around the globe [3;7;8], some of them are obsolete (EFs, PCA, and IMPROVE etc.) and some of them are relevant (UNMIX, CMB and PMF). For instance, EFs—first applied in the 1960s—offer a basic understanding of the elemental origin (natural or anthropogenic) based on crustal abundance but fail to account for regional variations in elemental concentrations [9]. It provides the simple crude information that the element is higher than the typical earth's crustal values does not account for local variations in elemental abundance [4]. Similarly, PCA, which operates on the eigenvector method, is recognized as a preliminary screening tool rather than a full receptor model due to its inability to quantify source contributions accurately [4;10]. Hopke and Jaffe (2020) has suggested that these tools (EFs and PCA) may not be used as a receptor model for source apportionment of atmospheric

aerosols because more quantitative data analytical tools had been developed and available [7].

Factor analysis tools like non-negative constrained alternating least square [8], positive matrix factorization [7] and non-negative least squares that allow proper weighting of individual data points for optimization process [4]. From last two decades, two major classes of the statistical tools that have been used worldwide as receptor models are: i) chemical mass balance (CMB) and ii) multivariate factor analysis models (PCA-APCS, UNMIX, and PMF) [11]. Even though with the same data sets and same chemical species of atmospheric aerosols (e.g. PM₁, PM_{2.5}, PM₁₀ etc.) these receptor models resolve the different sources with different apportionate due to different statistical algorithms and constraints. Hence, applying these models concurrently to the same dataset allows for a comparative evaluation of their outputs, facilitating a more precise identification of the dominant pollution sources. In the present manuscript we report the application of various tools used in the source apportionment (SA) analysis using PM_{2.5} chemical species collected in Delhi from January 2021-December, 2021. We explore the relative strengths and limitations of these models and provide insight into their performance and applicability in an urban Indian context. Our findings aim to support more informed choices in source apportionment methodology and contribute to the formulation of evidence-based air quality management strategies.

2. Materials and Methods

PM_{2.5} samples ($n = 59$) were collected at the experimental site of CSIR-NPL, New Delhi, over the period from January to December 2021, using a fine particle sampler in accordance with standard recommended procedures [12]. Due to the COVID-19

lockdown, sampling was not possible from May 2021 to July 2021, resulting in a data gap during this period of the study. The sampling location situated in Central Delhi (28°38'N, 77°10'E) and represents an urban background and surrounded by heavy roadside traffic [13]. Delhi, home to a population exceeding 33.8 million [14] and is ranked among the most polluted cities globally. PM_{2.5} samples were analyzed for their carbonaceous components (OC and EC), water soluble inorganic ions (WSIIs: Na⁺, Ca²⁺, Mg²⁺, K⁺, NH₄⁺, Cl⁻, F⁻, NO₃⁻ and SO₄²⁻) and elements (Si, Ti, Al, Fe, Zn, Cu, Mn, Pb, As, Br, Cr, Mo and P) using different standard instrumentations and analytical techniques [3;12]. Organic carbon (OC) and elemental (EC) concentrations of PM_{2.5} samples were determined using an OC/EC carbon analyser (DRI 2001A, Atmoslytic Inc., Calabasas, CA). WSIS were analyzed using Metrohm 930 Compact Ion Chromatography (IC). Concentrations of various elements of PM_{2.5} were analyzed using Wavelength Dispersive X-Ray Fluorescence (WD-XRF; ZSX Primus, Rigaku, Japan) [12]. Field sample blank filters were analyzed using the same procedure as exposed filters, and their average concentrations were subtracted to determine final sample values. Each sample was analyzed in triplicate to assess analytical repeatability.

The analyzed chemical species (OC, EC, WSIS and elements) are used as input for IMPROVE, PCA and PMF models. The details of model description and outcome procedures are available in our previous publications [12;15]. The IMPROVE (Interagency Monitoring of Protected Visual Environments) model is a receptor-based method used to apportion sources of particulate matter (PM_{2.5}) through mass reconstruction. It estimates source contributions by analysing concentrations of specific chemical species, categorizing PM

into components like organic matter, elemental carbon, soil dust, sea salt, ammonium sulphate, ammonium nitrate, and trace elements. Each component is calculated using marker species and conversion factors. The reconstructed mass is compared to gravimetric mass to evaluate accuracy, providing a simplified yet effective approach for air quality management [15]. PCA-APCS and PMF 5.0 models were used to identify and apportion sources of PM_{2.5} across the study period. PCA-APCS was applied using SPSS, where variables were standardized, and only principal components with eigenvalues greater than 1 were considered, following Kaiser's criteria. A KMO value greater than 0.6 confirmed the data's suitability for factor analysis, and Varimax rotation was used. Variables with factor loadings > 0.5 were used to identify sources [12;15].

Annual variations in sources were further examined using the EPA PMF 5.0 model. A seven-factor solution was found suitable for the available data. Model validation showed strong agreement between modelled and measured data ($R^2 > 0.6$), indicating good performance with $Q_{\text{true}}/Q_{\text{expected}} < 1.5$. Additional uncertainties were accounted for using DISP and bootstrap (BS) analyses. The BS (100 runs) showed no unmapped cases, and DISP analysis indicated zero factor profile swaps, confirming model reliability [12;15].

3. Results and Discussion

The mean statistics of chemical species (OC, EC, WSIS and elements) of PM_{2.5} extracted during January-December 2021 was depicted in Table 1 with standard deviation (\pm SD at $\sigma = 1$). The mean concentration PM_{2.5} was $110 \pm 54 \mu\text{g m}^{-3}$ during the sampling period. Throughout the study period, the highest average concentration of PM_{2.5} was observed during the post-monsoon season ($153 \pm 113 \mu\text{g m}^{-3}$), followed by winter ($135 \pm 49 \mu\text{g m}^{-3}$).

³), summer ($56 \pm 16 \mu\text{g m}^{-3}$), and the lowest in the monsoon season ($40 \pm 15 \mu\text{g m}^{-3}$). Similarly, the carbonaceous component i.e., OC and EC were found highest during winter ($21 \pm 10 \mu\text{g m}^{-3}$ and $9 \pm 4 \mu\text{g m}^{-3}$) respectively, followed by post-monsoon ($20 \pm 16 \mu\text{g m}^{-3}$ and $7 \pm 5 \mu\text{g m}^{-3}$), summer ($8 \pm 3 \mu\text{g m}^{-3}$ and $3 \pm 2 \mu\text{g m}^{-3}$), then in monsoon ($5 \pm 0.7 \mu\text{g m}^{-3}$ and $2 \pm 0.4 \mu\text{g m}^{-3}$). In the present study the total carbonaceous components (TC = OC+EC) were accounted for 18.6% of PM_{2.5}, whereas the total WSIS (NH_4^+ , SO_4^{2-} , NO_3^- , F^- , Cl^- , Na^+ , Mg^{2+} , K^+ and Ca^{2+}) was accounted for 36% of PM_{2.5} ($39.7 \mu\text{g m}^{-3}$). The total mass concentrations of elements extracted as $5.26 \mu\text{g m}^{-3}$ which was accounted for 5% of PM_{2.5}. These chemical species (OC, EC, WSIS and elements) are used as input for the various receptor models (IMPROVE, PCA and PMF) to evaluate their capability.

Table 1. Mean concentration of PM_{2.5} and their chemical species ($\mu\text{g m}^{-3}$) in Delhi.

Chemical Species	Concentration ($\mu\text{g m}^{-3}$)
PM _{2.5}	110 ± 54
Organic carbon (OC)	14.7 ± 11.6
Elemental Carbon (EC)	5.8 ± 4.2
Ammonium (NH_4^+)	7.3 ± 6.1
Sulphate (SO_4^{2-})	9.7 ± 5.9
Nitrate (NO_3^-)	7.2 ± 5.3
Fluoride (F^-)	0.5 ± 0.4
Chloride (Cl^-)	6.3 ± 3.9
Sodium (Na^+)	2.9 ± 1.5
Magnesium (Mg^{2+})	0.3 ± 0.2

Potassium (K^+)	2.7 ± 1.8
Calcium (Ca^{2+})	2.8 ± 1.5
Aluminum (Al)	0.39 ± 0.36
Phosphorous (P)	0.08 ± 0.07
Sulphur (S)	2.01 ± 1.41
Chromium (Cr)	0.31 ± 0.17
Iron (Fe)	0.85 ± 0.61
Zinc (Zn)	0.49 ± 0.42
Copper (Cu)	0.21 ± 0.17
Molybdenum (Mo)	0.21 ± 0.19
Bromine (Br)	0.12 ± 0.11
Lead (Pb)	0.39 ± 0.28
Arsenic (As)	0.11 ± 0.09
Titanium (Ti)	0.09 ± 0.07

3.1. IMPROVE model

In order to obtain the probable empirical sources of PM_{2.5}, the chemical species of PM_{2.5} were re-constructed (RCPM_{2.5}) using IMPROVE equation [16;17]. RCPM_{2.5} was computed using Equation 1:

$$\text{RCPM}_{2.5} = [\text{AS}] + [\text{AN}] + [\text{POM}] + [\text{LAC}] + [\text{SS}] + [\text{Soil}] \quad (1)$$

Where, AS = ammonium sulphate,

AN = ammonium nitrate,

POM = particulate organic matter,

LAC = light absorbing carbon,

SS = sea salt.

The details of multipliers, procedures and explanation are available in literatures [12;16;18]. The mass difference (PM_{2.5} – RCPM_{2.5}) or unidentified mass (UM) of

PM_{2.5} was calculated by subtracting RCPM_{2.5} from measured PM_{2.5}.

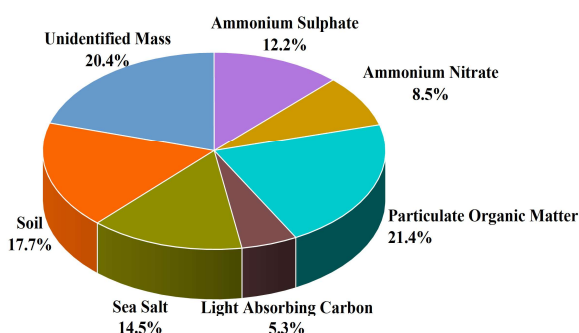


Figure 1: Re-construct mass of PM_{2.5} extracted by IMPROVE model

The percentage extracted source of PM_{2.5} by IMPROVE model depicted in Figure 1. The AS ($14.4 \mu\text{g m}^{-3}$), AN ($9.3 \mu\text{g m}^{-3}$), POM ($23.5 \mu\text{g m}^{-3}$), LAC ($5.8 \mu\text{g m}^{-3}$), SS ($16.0 \mu\text{g m}^{-3}$), Soil ($19.5 \mu\text{g m}^{-3}$) and UM ($20.3 \mu\text{g m}^{-3}$) contributed to PM_{2.5} as 12.2%, 8.5%, 21.4%, 5.3%, 14.5%, 17.7% and 20.4%, respectively. The unidentified mass (UM) of

chemical reactions of SO₂, which emits from combustion of fossil fuels (coal and diesel) whereas AN is produced through reversible reactions of gas-phase NH₃ and HNO₃, aided by the formation of oxidized nitrogen through combustion of fossil fuels and vehicular emissions [3;20]. The abundance of POM in the atmosphere are mostly coming from primary (combustion) and secondary (secondary organic aerosols formation) processes [21].

3.2. PCA and PMF model

Figure 2 shows the percentage contribution of sources to PM_{2.5} resolved by PCA, and PMF models in Delhi, India during 2021. PCA extracted 6 sources of PM_{2.5} in Delhi, i.e. secondary aerosols (SA), vehicular emissions (VE), biomass burning + fossil fuel combustion (BB+FFC), soil dust (SD), sea salt (SS) and industrial emission. The highest contribution of SD was resolved to be 32% with prominent availability tracer elements Al, Ca, Fe, and Ti whereas lowest as SS (4%).

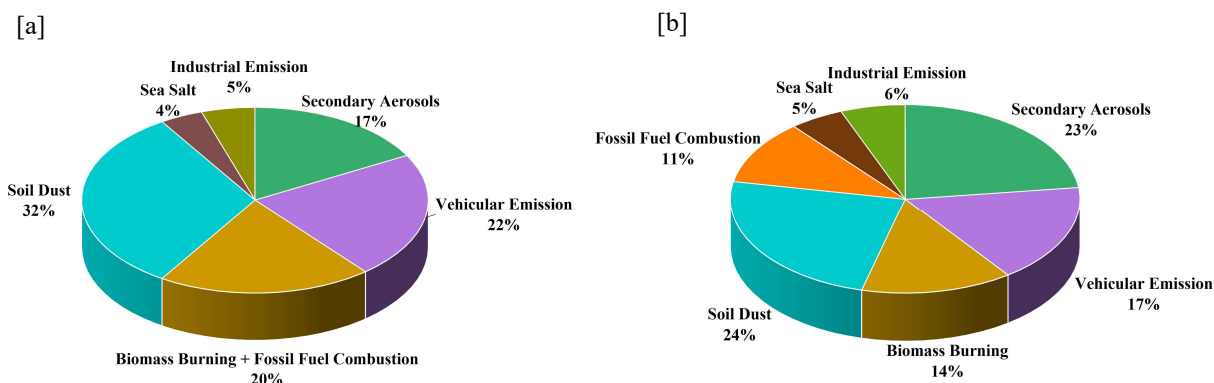


Figure 2 Source contribution (%) to PM_{2.5} resolved by a) PCA and b) PMF models in Delhi.

PM_{2.5}, estimated by reconstructing the PM_{2.5} mass, accounted for 20.4%. This could be due to the presence of carbonate-rich minerals, calcium sulfate, aluminosilicates, and other similar components in the samples [19;20]. In the urban site of Delhi, the majorly, AS is produced in the ambient air through the

PMF model resolved as 7 sources of PM_{2.5} in Delhi, i.e. SA (23%), VE (17%), BB (14%), SD (24%), FFC (11%), SS (5%) and IE (6%). PMF contributed highest loading of SD (24%) and lowest of SS (4%) to the PM_{2.5} mass concentrations. Jain et al. (2020) also examined the similar source type (common

sources: SA, VE, BB, and SD) at megacity Delhi with different percent contribution to PM_{2.5} using receptor models.

In India, identifying the sources of particulate matter (PM) pollution involves analysing specific elemental signatures. Soil dust is typically traced using elements like Al, Si, Ca, Ti, Fe, along with trace metals such as Pb, Cu, Cr, Ni, Co, and Mn [12; 22; 23; 24]. Biomass burning is a major contributor to air pollution, especially in the post-monsoon season. Water-soluble K is a widely recognized marker for this source [25]. In India, K⁺ has proven to be particularly effective in identifying emissions from crop residue and wood burning [20]. The health effects are severe—long-term exposure can lead to respiratory diseases, headaches, dizziness, and in extreme cases, premature death. Fossil fuel combustion, particularly from coal, is another critical source. Elements like As and Cl are commonly released during coal burning and serve as key indicators of this process [26; 27]. Prolonged exposure to these emissions raises the risk of respiratory and cardiovascular diseases and contributes significantly to environmental damage and global warming. In urban areas, vehicular emissions add to the problem. Exhaust releases pollutants like NO_x, CO, SO₂, VOCs, and particulate matter containing metals such as Pb, Zn, and Cu [28; 29]. Beyond exhaust, non-exhaust sources like brake wear, tire degradation, and road dust also release harmful particles. Zn, Pb, and Mo are typically associated with these non-exhaust emissions [26; 30; 31]. Industrial activities, especially in Delhi, contribute further by releasing hazardous metals like V, Cd, and Pb, often from burning refuse oil and improper battery disposal [32]. Other metals such as Zn, Mn, Co, Cu, and Cr have been linked to emissions from metal-based industries [3]. Key particulate pollutants also include black carbon (BC) and organic

carbon (OC). BC absorbs sunlight and contributes to atmospheric warming, while OC scatters light, impacting visibility and climate. These are often accompanied by CO, NO_x, SO₂, and greenhouse gases like CO₂ and CH₄ [33], highlighting the complex mix of pollutants affecting both human health and the environment. SO₄²⁻, NO₃⁻, and NH₄⁺ are key indicators of secondary aerosol formation processes [34], while Na⁺, K⁺, and Mg²⁺ serve as primary tracers for sea salt contributions [23].

Recent global assessments, including the comprehensive review by Hopke et al. (2020), have identified several dominant sources contributing to ambient particulate matter (PM) concentrations in India. These sources include secondary inorganic aerosols, sea salt, vehicular emissions, industrial activities, biomass burning, coal and oil combustion, as well as secondary organic aerosols. Understanding the origin and characteristics of PM, particularly PM_{2.5} and PM₁₀ is crucial for atmospheric scientists, public health researchers, policymakers, and other stakeholders involved in air quality management. Source apportionment studies play a vital role in elucidating the contribution of local and regional emission sources to ambient PM levels. These studies help characterize the physical and chemical properties of aerosols, offering valuable insight into source-specific influences on air quality. The information derived from such analyses is instrumental in formulating targeted mitigation strategies aimed at reducing pollutant levels and improving overall air quality, both regionally and globally. Moreover, quantifying source-specific PM concentrations is increasingly important in epidemiological research, as it enables the identification of pollutant sources most strongly associated with adverse health outcomes. This targeted approach supports the development of more effective public health interventions and regulatory policies.

We evaluated the performance of different receptor models in identifying and quantifying sources of PM_{2.5} and found that while all models detected similar source types, the number of sources and their contributions varied at sampling site. Among the models, PMF demonstrated several advantages over PCA. Unlike PCA, which may yield negative values in source profiles and have limited efficiency in source identification, PMF ensures non-negative solutions and provides more meaningful results. Although PMF requires a relatively large dataset, it is robust against data gaps and low concentrations below detection limits by incorporating experimental uncertainties and assigning weights based on measurement confidence. This enhances the reliability of PMF outputs, allowing it to resolve sources more accurately by effectively linking marker species to their respective sources. In contrast, PCA use only concentration data without uncertainty weighting and apply relatively coarse methods for handling outliers and missing data, often leading to the mixing of distinct source signals and less precise source apportionment. Given these strengths, PMF offers more accurate and interpretable estimates of source contributions, as supported by comparisons in our earlier studies [12; 35].

4. Conclusions

In this study, we demonstrated the applications of various simple to complex tools of receptor (IMPROVE, PCA and PMF) models on same chemical species of PM_{2.5} to access source contribution to PM_{2.5}. The IMPROVE model computed the various sources PM_{2.5} with a simple established multiplying factors whereas PCA extracted the sources through dimensionality reduction method. PMF functions on the principle of decomposing a data matrix into two smaller matrices, one representing source profiles and the other representing source

contributions. In the present case, both PCA and PMF revealed comparable source categories, though with varying contribution estimates, while the IMPROVE approach provided fixed source types based on standard classifications. This comparative analysis highlights the utility and limitations of different receptor models in identifying and quantifying PM_{2.5} sources.

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