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Application of Multivariate Receptor Models in Identification, Quantification, and Management of Common Pollutant Sources in Various Environmental Sectors

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ABSTRACT

Identification and allocation of various pollutant sources are crucial tools for effective prevention, control of pollution, and management of different environmental sectors. In this regard, methods used for determining, allocating, and managing pollutant emission sources in the environment can be categorized into source identification and quantitative source allocation methods. Source identification methods have limitations, including the inability to process missing data and detect values below the detection limit commonly observed in environmental data. Additionally, these methods cannot quantitatively determine the contribution of natural and human sources of pollutants. Conversely, the second type of methods is capable of quantitatively identifying and determining the share of pollutant sources, among which receptor models are particularly notable. In terms of source allocation, receptor models are mathematical computational approaches that can identify and quantify the contribution of sources based on the chemical and physical characteristics of pollutants in sources and receptors. Given the practical importance of receptor models in quantifying the contribution of various pollutant sources in the environment, the aim of this study is to introduce and compare different types of receptor models for determining pollutant emission sources in the environment. The study also highlights recent research in this area. Therefore, by presenting these models, the current research can serve as a guide and a valuable resource for the identification and quantitative determination of various pollutant sources in the environment, consequently providing a basis for environmental management and improvement.

1. INTRODUCTION

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The rapid development of industrialization, urbanization, and intensified agricultural activities are among various factors that have led to a decline in environmental quality in recent years [1]. Various pollutants originate from both natural and anthropogenic sources, but it is generally believed that the latter is the primary cause of excessive accumulation of pollutants in the environment [2]. Therefore, the identification and classification of anthropogenic sources of various pollutants are crucial tools for preventing and effectively controlling pollution in different environmental compartments [3]. For example, in the air sector, overall environmental air quality management involves identifying sources of emitted substances into the air, quantitatively estimating pollutant emissions, and recognizing the pathways of pollutant transport from their sources to nearby locations [4]. In the water resources sector, considering the current water scarcity conditions, the fundamental first step in prioritizing pollution management efforts in these systems is to identify pollution sources and provide a relative quantitative assessment of their contribution to stream pollution [5]. Similarly, in the soil sector, to address issues such as high spatial heterogeneity of soil pollutants, complexity, and diversity of emission sources, and lack of monitoring information, it has been emphasized that necessary research should be conducted to determine the sources of pollutant emissions in this sector [6]. Therefore, considering the mentioned factors, the division of pollutant emission sources in different environmental compartments is of fundamental importance, and it can pave the way for environmental management and improvement.

Current methods for determining and dividing pollutant sources can be categorized into methods for source identification and quantification [7]. In the first case, major pollution sources are identified using Geographic Information System (GIS), Principal Component Analysis (PCA), Factor Analysis (FA), and Cluster Analysis (CA) [12-8]. While in the second case, regarding the identification and quantification of sources, existing methods include Enrichment Factor (EF), stable lead isotopes, and receptor models [7]. Among the source identification methods, conventional methods such as FA and PCA provide useful results in investigating various environmental phenomena [14-13]. However, PCA and FA have serious drawbacks due to the potential existence of negative values in almost all factors. Additionally, PCA is unable to handle missing data and is below the detection limit, which is often observed in environmental data [15]. Moreover, these methods cannot determine the quantitative contribution of natural and human sources of pollutants. Therefore, for these reasons, valid solutions in this area need to be explored. Among the mentioned second-case methods, receptor models, especially in recent studies, are very prominent in identifying potential pollution sources based on the chemical composition of receptor sites [16].

Receptor models can identify and determine the contribution of sources based on the chemical and physical characteristics of pollutants in sources and receptors. In terms of source allocation and determination, receptor models are mathematical approaches developed to quantify the contributions of sources to samples based on the combination or fingerprint effect of the source. Existing receptor models include Chemical Mass Balance (CMB) [17], Positive Matrix Factorization (PMF) [18], Absolute Principal Component Analysis with Multiple Linear Regression (APCA-MLR) [19], and UNMIX [20]. Although the use of receptor models was initially created for partitioning pollutant sources in the atmosphere [23-21], currently, receptor models are used to determine the contribution of pollutant sources in the air [24], soil [25], aquatic environment sediments [26], and so on. Therefore, considering the practical importance of receptor models in determining the quantitative contribution of various pollutant emission sources in the environment, the aim of this study is to introduce and compare various types of receptor models in determining sources of different pollutants and also to highlight recent studies conducted in this field.

Initially, receptor modeling methods were used for assessing air quality and suspended particles, providing information on PM levels and the relative contribution of various sources emitting PM [24, 29-27]. However, these models are currently employed to determine the contribution of pollutants from various sources in different environmental compartments, including the atmosphere, soil, water resources, and even dust [37-30]. In this section, some domestic and international studies on the use of receptor models in quantifying the contributions of various pollutant sources in the environment are highlighted.

2. MATERIALS AND METHODS

Based on the objectives of the current research, various studies, especially recent ones, in the field of investigating pollutant sources were collected to extract the methods and models used in this area. The review of multiple studies indicated that although various methods and models can be used to determine and allocate pollutant sources in the environment, receptor models are highly significant due to their ability to calculate the quantitative contribution of pollutant sources. Therefore, this section of the research discusses different receptor models used in determining the sources of various pollutants in the environment, along with their equations.

2.1. Chemical Mass Balance (CMB) Model

Chemical Mass Balance (CMB) is a fundamental receptor model based on the concept of mass balance [50]. In this model, linear regression is used to create equations for the balance of pollutants in the source and receptor using both source profiles and environmental data. It is the most accurate model, but the use of the CMB model is limited due to the lack of access to source profiles [7, 51]. Therefore, several subsequent models have been developed, and some of the most widely used models are mentioned below.

2.2. Positive Matrix Factorization (PMF) Model

PMF is a new type of receptor model performed using EPA PMF 0.5 software. This model decomposes a dataset matrix into two matrices: factor profiles and factor contributions. PMF also utilizes uncertain data in the data matrix [18]. In fact, the input variables for this model are the concentrations of rare elements and uncertain data.

The governing equation for the PMF model is as follows:

$$X=GF+E \tag{1}$$

where X (m×n) is the data matrix containing measurements of component m in n samples, G (m×p) is the factor or source profile matrix, F (p×n) is the factor score or source contribution matrix, and E (m×n) is an error matrix.

$$X_{ij} = \int_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \tag{2}$$

Where X_{ij} is the concentration of species j in sample i (mg/kg-1), g_{ik} is the contribution of source k (mg/kg-1), f_{kj} is the amount of species j from source k, eij is the error value, and p is the number of source factors.

The uncertainty data file can be based on both observations and equations. The uncertainty file is calculated based on equations for specific parameters for each sample through equations provided by the EPA [52]. Uncertainty is calculated based on the detection limit of a specific element (MDL), and the error percentage is determined by reference materials. If the measured concentration is less than the MDL for that element, the uncertainty data is calculated as follows:

$$\text{Unc}= \frac{5}{6} \times \text{MDL} \tag{3}$$

And if the concentration is greater than the provided MDL, the calculation is based on a fraction of the concentration and MDL:

$$\text{Unc} = \sqrt{(\text{error fraction} \times \text{C})^2 + (0.5 \times \text{MDL})^2} \tag{4}$$

2.3. Model (APCA-MLR) Absolute Principal Component Analysis followed by Multiple Linear Regression

The APCS/MLR model has evolved from PCA, and the source contribution is obtained by performing regression between pollutant content and APCS [53]. In essence, the flowchart of this model is such that initially, potential pollution sources are identified using PCA, and then the determination of source contributions is calculated through the MLR model. The algorithm for this model, which is performed in the SPSS software, is as follows. First, the raw initial data is normalized as follows:

$$Z_{ij} = \frac{C_{ij} - \overline{C_j}}{\sigma_i} \quad (5)$$

C_{ij} = The concentration of the jth element in the ith sample
$\overline{C_j}$ = The average concentration of the jth element for all samples
σ_i = The standard deviation of the jth element

Using the following equation, the normalized matrix is calculated from the value of the elements;

$$Z_{ik} = \sum_{j=1}^{p} W_{ij} P_{jk} \quad (6)$$

Z_{ik} is the normalized matrix of element values, j is the number of factors, W_{ij} is the factor loading matrix. This matrix contains the coefficients corresponding to each factor for the element value. P_{jk} is the factor score matrix represents the values of the source components j at each sampling site k.

(APCS)jk is the absolute principal component scores are calculated by subtracting the factor scores P_{j0} from the scores of actual samples (P_{jk}).

$$(APCS)_{jk} = P_{j0} - P_{jk} \quad (7)$$

P_{j0} is the factor score matrix is zero in the uncontaminated site, where the concentration of all elements is equal to zero. Finally, the contribution of each source can be calculated using multiple linear regression based on the following equation;

$$X_{ik} = \sum_{j=1}^{p} A_{ij} \, (APCS)_{jk} \quad (8)$$

Aij is the linear regression coefficient matrix for element i from factor j, p is the number of factors, and Aij (APCS)jk is the factor score for pth factor for Cj, along with the average value , Aij (APCS)jk represents the average factor score for the pth factor for Cj.

2.4. Modified model (PCA-MLRD) Principal Component Analysis and Multiple Linear Regression with Distance

The modified model is based on the assumption that the total pollution content is equal to the sum of the contributions from each source. Since this pollution, originating from pollutants, expands as the distance from emission sources decreases, the contribution of sources can be determined by specifying the distance between sampling sites and emission sources. The flowchart of this model is as follows: PCA is used to identify potential sources, and the identification and determination of source contributions are done through MLR, metal concentrations, and the distance from the source [7].

$$X_{ik} = \sum_{j=1}^{p} B_{in} \, D_{nk} \quad (9)$$

D_{nk} represents the distance from sampling sites to pollution emission sources, $\mathbf{B_{in}}$ is the regression coefficient matrix from distance to sources, and n is the number of sources.

2.5. Unmix Model

The Unmix model is a relatively new multi-variable receptor model based on preliminary analysis [50] and non-negativity constraints. The primary method employed in Unmix is an algorithm for identifying convex hulls in N-dimensional space [55]. This model assesses the number of sources by reducing the dimensionality of the information space from m to p using Singular Value Decomposition (SVD). Unmix seeks to address the general mixing problem where the assumption is that the data is a linear combination of hidden sources whose mixture is unknown, and in each sample, it contributes with an unknown weight [58-56]. The Unmix model can be represented by the following equation:

$$C_{ij} = \sum_{i=1}^{p}\left(\sum_{k=1}^{p} U_{ik}D_{ki}\right)V_{ij} + \varepsilon_{ij} \quad (10)$$

where; U, D, and V are diagonal matrices of sizes n × p, p × p, and a matrix of size p × m, respectively. ξ_{ij} is the term "error" represents the observation error, and C_{ij} is the observed concentration of species i in sample j. To perform this model, Unmix 6.0 is utilized, which is a mathematical receptor model developed by EPA researchers. The use of this model helps reduce the number of variables in complex analytical datasets to a combination of species called source types and their contributions.

3. DISCUSSION AND CONCLUSION

In general, various receptor models have different advantages and disadvantages. Chemical Mass Balance (CMB) is a fundamental receptor model based on the concept of mass balance [50]. In this model, linear regression is used to create equations for balancing pollutants in both the source and receptor using both source profiles and environmental data. It is the most accurate model, but its use is limited due to the lack of access to source profiles [51]. Therefore, considering the limitations of the CMB model, other multi-variable receptor models such as Absolute Principal Component Analysis/Multiple Linear Regression (APCS/MLR) and Positive Matrix Factorization (PMF) have been developed to solve the chemical mass balance (CMB). In fact, APCS/MLR and PMF models, unlike the CMB model, do not rely on the previous effects of sampling and measurement; therefore, APCS/MLR and PMF are more suitable and efficient than traditional CMB. Among these models, PCA-MLR is a pattern recognition method that analyzes the relationship between measured variables to explain the variance of large datasets with reduced-dimensional factors [59]. The PMF model includes the maximum information from the dataset, which includes sampled and measured values and analytical uncertainties [61-60]. This model is a type of receptor model that decomposes a dataset into two matrices of factor profiles and factor contributions, which then need interpretation by users [62]. Meanwhile, Unmix uses a Singular Value Decomposition (SVD) method to determine the number of influential sources on the total dataset [55]. In general, receptor models such as PCA-MLR and PMF are often preferred options for source apportionment studies because: (1) the use of extensive monitoring programs aiming to create a large database has recently become a common practice, (2) these models, unlike CMB, do not require source profiles, and (3) the models' capacity to handle large monitoring datasets [63].

In fact, these receptor models encompass various mathematical and physical constraints. The differences in theoretical methods lead to diverse results in terms of the number and types of sources obtained from different receptor models [53, 56]. Therefore, to provide a stronger source apportionment and better interpretation, the sources extracted by different receptor models should be compared and evaluated together. For this reason, multiple studies have examined various receptor techniques based on single-site source apportionment [63, 64-66]. The assumption of comparing different receptor models based on single-site source apportionment is that the obtained source emission patterns should be consistent for all receptor techniques. Therefore, given the different results from the use of various receptor models, it can be said that comparing these models is essential for understanding and selecting appropriate receptor techniques for single-site source apportionment to develop management actions for point source pollution.

Declaration

We acknowledge that we used ChatGPT to enhance the academic writing of our manuscript while ensuring the originality and integrity of our work.

Transparency Statement

The data supporting this study are available upon reasonable request to the corresponding author, subject to ethical and confidentiality considerations.

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Declaration of Interest

The authors declare that they have no competing interests.

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