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内 容 简 介

本书结合颗粒物污染防治工作的需求，系统总结了中国大气颗粒物来源解析受体模型理论的发展与应用。全书共分为七章：第一章主要介绍受体模型的发展；第二章介绍化学质量平衡受体模型（CMB）的基本原理；第三章介绍因子分析模型；第四章介绍受体模型在中国大气颗粒物来源解析中的应用；第五章到第七章分别介绍主成分分析/多元线性回归-化学质量平衡复合受体模型（PCA/MLR-CMB），非负主成分回归化学质量平衡模型（NCPCRCMB）和化学质量平衡嵌套迭代模型（CMB-iteration）。

本书适合高等院校和科研院所大气环境相关专业的研究生、教师及科研人员，环境保护部门从事大气污染防治工作的技术人员及管理人员阅读和参考。

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Preface

Source apportionment is the key stone for air pollution controlling, and can provide scientific information for air quality management. In the field of source apportionment researches, particulate matter (PM) study is the hot point over the world, due to the complex sources for PM. The classification of the source categories of PM includes primary source emission, and secondary source forming; not only the point emission (i.e. coal combustion), but also the open sources (i.e. resuspended dust), and so on. Therefore, sources of PM have been studied with the most interest in recent years over the world, especially in China.

The methods of identificating the possible sources and quantifying their contribution are called source apportionment technology. With the changing of source emission patterns and pollution characteristics, the source apportionment methods are developing accordingly. Generally, the methods can be classified into three kinds: the source emission inventory method, the air quality model, and the receptor model.

The source emission inventory method can qualitatively or semi-quantitatively analyze the major source contributions in an area, according to the emission mass (t) of particulate matters from diverse sources. However, there is high uncertainty of the result of source emission inventory method. Firstly, the emission mass (t) from one source can not be linearly related to its contribution ($\mu g/m^3$); secondly, it is difficult to obtain the emission mass (t) of some sources, such as the resuspended dust source. Thus, the emission inventory method is hard to meet the requirement of air management in recent years.

The air quality model (dispersion model) is to estimate the source contribution according to the source emission data, source strength information and meteorological data. Recently, the latest air quality model can simulate the physical transportation and chemical reactive process. However, the challenges for application of air quality model are the uncertainties of emission strength of open sources, as well as lack of the filed information for chemical mechanism and PM size distributions.

The receptor model depends on the ambient dataset, which calculates the source contribution according to the chemical profile of sources and measurement of receptor samples. The receptor model does not need the specific source emission inventory and meteorological data, so it has been playing an important role in the source apportionment researches.

The above three kinds of source apportionment methods have been applied in lots of source apportionment works in China. The receptor model, due to its convenient performance, has been developed and applied in these years. The chemical mass balance model and factor analysis model (PMF, UNMIX etc.) are two important receptor models that have been recommended by Chinese NEP and USEPA. Therefore, the aim of this book is to: (1)describe the principle and development of the receptor; (2)summarize the characteristics of different receptor models; (3)introduce some new receptor models.

We have attempted to write this book for a group of readers who are interested in the source apportionment for particulate matter.

This book includes seven chapters. Chapter 1 introduces the development of receptor models, collated by Xiaozhen Fang; Chapter 2 is about the CMB and the improved model, collated by Guirong Liu and Xiaoyu Zhou; Chapter 3 describes the factor analysis models, collated by Xing Peng; Chapter 4 summarizes the application of the receptor models in China, collated by Qili Dai and Jiao Wang; Chapters 5-7 introduce the models of PCA/MLR-CMB, NCPCRCMB and CMB-iteration, respectively.

Tan Zhu　Yinchang Feng

Contents

Chapter 1
Introduction of Receptor Model

Source apportionment is the technique to qualitatively or quantitatively study the source apportionment of PM. With the development of source apportionment, there are three source apportionment techniques: the emission sources inventory, the dispersion model and the receptor model. Emission sources inventory was first applied to source apportionment to calculate atmospheric PM. According to emission factors, this method can estimate the emissions from various source categories, and then to identify the main emitted source categories based on the emissions. There are two disadvantages: one is that there are many "open sources" which are difficult to accurately estimate their emissions, and the other is the nonlinear relationship between emissions and contributions of source categories. Therefore, as types of pollution sources increasing and diversifying, and the requirements for environmental management are more and higher, emission sources inventory has been unable to meet the requirements for source apportionment of atmospheric PM.

From the perspective of emission sources, according to the source intensity data of various source categories and meteorological data, the dispersion model can estimate the contribution of emission source to atmospheric PM, and establish the quantitative relationship between organized soot emissions and industrial dust sources with atmospheric environmental quality, in order to provide the scientific basis for organized emission sources control. However, due to unavailable intensity data, it is very difficult to estimate the contribution of open sources to atmospheric PM. Therefore, it is difficult to establish the systematical and comprehensive relationships between emission sources and atmospheric PM, which is the biggest obstacle for the dispersion model to be applied to source apportionment.

The receptor model was developed to solve the above-mentioned problems. From the perspective of receptors, the receptor model can estimate the contributions of various source categories based on the chemical and physical characteristics of atmospheric PM. In the receptor model, no information regarding source intensity and meteorological data is needed to resolve the issues encountered in emission sources inventory and dispersion model. Therefore, the receptor model has been developed rapidly and has gradually formed a technical system of source apportionment for atmospheric PM, since invented in 1970s. Now receptor model was widely used to identify the source category and estimate the contributions. It is an indispensable method for PM pollution prevention and controlling. It is also currently the most widely used as a source apportionment technique in the world[1-11].

1.1 Basic Concepts and Characteristics of Receptor Model

The receptor model is to calculate the contributions to the atmospheric PM from various emission sources based on the analysis of the chemical constituents of pollutants at the receptors and sources. The receptor model can be classified into two categories: one is the source unknown receptor model in which sources information is unavailable, the other is the source known receptor model in which comprehensive sources information and its constituents are essential[12]. The source known receptor model need to introduce both the source and receptor information into the model and then estimate the contribution of sources to the receptor by establishing a mass balance between the sources and the receptors, such as the chemical mass balance (CMB) model. Whereas the source unknown receptor model just need to be receptor information into the model. Through the analysis of extracted multiple factors which correspond to the identification of different types of sources, it can estimate the contributions of these sources to the receptor, such as the principal component analysis (PCA) , the positive matrix factorization (PMF) , the UNMIX and the multi-linear engine2 (ME2).

For the source known receptor model, both the source and receptor information is input into the model to estimate the contributions of sources to the receptor by establishing the mass balance between the sources and the receptors. Whereas, for the source unknown receptor model, only receptor information is input into the model to estimate the contributions of sources to the receptor, through the analysis on receptor to extract multiple factors in corresponding to the identification of different types of sources.

1.1.1 Source known

The fundamental principle of source-necessary receptor model is the mass balance between the sources and the receptors, as represented by the CMB model, which is applied to calculating the contribution of emitted sources to the receptor through the multiple linear regression (MLR)[13]. The basic equation is

$$C_i = \sum_{j=1}^{J} F_{ij} \cdot S_j \quad (i = 1, 2, \cdots, I; j = 1, 2, \cdots, J) \tag{1-1}$$

where C_i is the measured concentration of the ith species in samples; F_{ij} is the measured portion of the ith species in material emitted by source j; S_j is the contribution of the jth source; J is the number of source category; and I is the number of chemical species.

1.1.2 Source unknown

The source unknown receptor model is often used when the source profiles are not available, which is based on lots of measured data on the same receptor. Through the analysis on a mass of data from the same receptor, the number of source categories and source profile can be acquired[14]. Generally, the basic equation of source unknown receptor model can be expressed as the ith chemical species of the kth sample obtained from the pth source category[15].

$$x_{ik} = \sum_{j=1}^{p} g_{ip} f_{pk} + e_{ik} \tag{1-2}$$

where x_{ik} is the measured concentration of the ith species in the kth sample; g_{ip} is the measured concentration of the ith species from the pth source; the pth source profile can be represented by a matrix G; f_{pk} is the contribution of the pth source to the kth sample; and e_{ik} is the modeling fitting error.

1.1.3 The characteristics of receptor model

Source apportionment techniques of atmospheric PM are based on the relationship between various source categories and the ambient receptor. It can be used to determine the source of atmospheric PM, and provide technical support to the development of prevention and control for atmospheric PM. Through basic theoretical applications and long-term practice, main features of receptor model are summarized as follows.

1) Invertibility

From the perspective of emission sources, the receptor model, as an inverse model, establishes the relationship between receptor and source profiles, which is one of the most fundamental characteristics of receptor model and one of the most significant differences between the receptor model and the dispersion model. As an inverse model, the receptor model is to determine the results of occurred pollution incident, based on the analysis of historical observational data. Thus, it can't be used to assess and predict the possible environmental impacts caused by emission sources in the future.

2) Without constraints of source intensity and meteorological data

In the receptor model, the migration and diffusion processes of the pollutants after emitted from the sources are not considered. Aerosol samples are collected directly from both sources and the atmosphere to analyze and compare their chemical compositions. Therefore, no information concerning source intensity and meteorological conditions is required. Moreover, this feature enables the receptor model to calculate the contribution of fugitive emission sources, which is quite difficult for the dispersion model to do so.

3) Emphasizing on source category

It should be noticed that the results from the receptor model are not the contribution of a particular source, but the contribution of a particular source category. So, the receptor model cannot determine the contribution of a particular source to the receptor. For example, analytical calculation of the contribution of coal source is the sum of coal emissions in the city, rather than the contribution of a specific coal-fired boiler. This characteristic of receptor model has a unique advantage for urban environmental management to enable decision-makers and managers to understand the contributions of different source categories and outlining the strategies of prevention and control for atmospheric PM from the prospect of urban development.

4) Requiring accurate interpretation of analytical results

During source apportionment in receptor model, the identification and classification of source categories are different from traditional classification methods. In general, the source categories are classified by industry, such as building materials, metallurgy, chemical industry, which are convenient for statistical analysis and management. For example, the control measures are mainly concentrating on smoke prevention and dust control for these stationary sources in China. However, the sources of ambient PM are very complicated. It is not comprehensive to simply divide PM sources into a few industries, such as metallurgy, chemical industry and so on, which utilize coal burning and emit coal soot dust in the production process. Meanwhile, there also exist a large number of exposed raw coal piles to emit a small part of coal soot dust into ambient air. Although unlike coal dust, raw coal is bound to affect the carbon content of ambient PM and to affect the source apportionment of coal soot dust, consequently. Thus, the source of ambient PM can't be simply allocated to a certain industry. Another case is concerning steel dust which can be largely resulted from steel corrosion. Therefore, steel dust in ambient PM can't be simply apportioned to steel industries. As a result, it should be objective and comprehensive when interpreting analytical results of source apportionment in the receptor model.

1.2 Classification of Receptor Models

There are many types of receptor models, such as chemical mass balance, principle component analysis–multivariate linear regression, positive matrix factorization, partial least squares, neural networks, and so on. But, it is slightly different from the classification of receptor model set by various scholars. According to Hopke[12], receptor models can be divided into two categories: source unknown and source known, as described in Section 1.1.1 and 1.1.2.

According to Pant[16], receptor models can be divided into two broad categories: microscopic and chemical. Microscopic type includes optical microscope, scanning electron microscope (SEM) and automated SEM analysis. It is mainly based on the analysis of morphological features of many individual particles in ambient air and often used to analyze aerosols with more distinct morphological features. Generally, it can only be used for quantitative or semi-quantitative analysis. For quantitative analysis, a large number of individual particles should be analyzed to acquire representative results, in which a huge database of sources (microscopic inventory) should be established. However, this type of receptor model is not suitable for large-scale applications because it cannot provide quantitative results in most cases. The chemical method can be used to identify and analyze the source apportionment of ambient PM by analyzing chemical composition of airborne PM in ambient. It includes many models, such as enrichment factor (EF), time series analysis, PCA, PMF, UNMIX, sequence analysis of species, and multi-linear engine (ME2) [17-21]. The fundamental principle of chemical method is to perform source apportionment on trace elements, elemental carbon/organic carbon and organic molecular markers. Over time, these models are often used as analytical methods for PM source apportionment. The common receptor models are summarized in Table 1-1.

Table1-1 Various types of receptor model

Methods		Technical content and features
Optical microscope		• According to particle features (size, shape, color and other optical properties) to determine the category • Analyze aerosol particles with aerodynamic diameter >1 μm
SEM		• Analyze aerosol particles with aerodynamic diameter <1 μm • High cost, and can't be used in amorphous material • Time consuming to limit the number of measurements
Computer controlled scanning electron microscopy (CCSEM)		• According to the particle size, morphology and chemical composition to identify particles, and the accurate classification is the basis of CCSEM • Fast analyzing speed, most of the particles can be measured, and the fine structure of particles can be displayed • High cost, and can't be applied to organic particles
CMB		• The fundamental principle is conservation of mass, clear and easy for people to accept, and the most widely used method • Regularly monitor the source and receptor sample, and list the emission inventories • Quantitatively evaluate the contributions of various sources with high cost
Multivariate analysis	Eigenvector analysis(PCA)	• Using sample testing relationships among variables to analyze • Identify factor category without prior knowledge of emission sources • Any secondary and short-term materials can be included in the analysis, such as particle size, wind and temperature • In order to obtain the contribution of specific compositions to the receptor, emission composition is needed
	Multiple linear regression(MLR)	• Find the relationship between variables and multiple variables, and understand the related changing rules, through linear least squares fitting • These variables should be independent, usually *p* variables are chosen as *p* sources before the factor regression analysis • Estimate emission sources profiles
EF		• Identify emission sources according to the change ratio of component. It is the result from the double normalized data processing • Reduce the impact of variables in sampling process, such as wind speed, wind direction, the amount of samples and so on. And the result interpretation is more precise and reliable than the absolute concentration • It can only provide qualitatively the degree of contamination of elements • Emission composition of emission source is needed, and cannot be applied to some specific source categories
Spatial patterns		• Compare with the concentrations among samples collected from different geographic points • From the spatial distribution of chemical composition in samples, the relationship between the source position and the chemical composition, and the effects of sources can be determined • This method is simpe but indirect to presume sources, but cannot deduce information about the emission sources

1.3 The Development of Receptor Models

1.3.1 Initiating stage (1960s-1970s)

Source apportionment of atmospheric PM was initiated with the dispersion model which is based on emission from sources. The dispersion model was also called "source model". It uses the mathematical formulas to describe the behavior of particles after emission, and a variety of factors. Its core part is the atmospheric dispersion model, which can be used to simulate the transport, diffusion and dilution of pollutants in the atmosphere, and to simulate dry or wet deposition and chemical conversion of pollutants experienced during the dispersing process.

With the development of source apportionment, it is found that open source is a major emission source of PM. However, it is very difficult to obtain the reliable information of emission intensity due to large number of open sources and wide range of emission patterns. Moreover, it is not usually a linear relationship between the emissions and the contribution to the receptor of one source category. Therefore, the emission inventory and dispersion model cannot estimate the contribution of source categories to the receptor. To deal with this problem, the receptor model appeared. From the aspect of receptor, it can estimate the contributions of different source categories to the receptor based on the chemical, physical characteristics and other information of PM in the ambient air.

In 1967, Blifford and Meeker[22] firstly shifted the focus from emission source to receptor (a local atmosphere affected by emission sources) and inferred the origin of PM by analyzing the samples collected from ambient air. In 1972, Miller et al.[23] firstly set the equation of chemical element balance and named it as the chemical element balance (CEB) method. In 1980, Cooper and Watson[24] renamed it as the chemical mass balance (CMB) method. After that, the model gradually developed into a very influential source apportionment receptor model. During this phase, the dispersion model was the dominant model, while the receptor model was just starting and had not been widely used. The characteristics of dispersion model and receptor model are shown in Table 1-2.

Table 1-2 The comparisons of dispersion model and receptor model

Patterns	Input	Output	Problems
Dispersion model	1) emission parameters (such as emission rates, chemical composition) ; 2) exhaust parameters; 3) meteorological parameters (wind speed, atmosphere stability) and some aerosol dynamics parameters factors (such as dispersion coefficient, the growth, cohesion, and settlement of PM)	The contribution of emission source to a receptor	1) There are a variety of assumptions in the dispersion model. 2) The required input parameters are not often accurately obtained, such as the fluctuation of the characteristics of emission sources, the complexity and randomness of atmosphere process. 3) The model is not appropriate for dealing with a lot of very important source, including natural PM (such as soil sand particles, marine aerosol) , secondary particles produced by the atmosphere process and long-distance transmission particles, etc. 4) As a very important content in environmental standards, particle size is not included in emission source data required by source model. 5) The dispersion model is not capable to deal with variable, carcinogenic and toxic particles, visibility drop, climate change and other issues.
Receptor model	1) Chemical profile of the emission sources; 2) Physical and chemical information of receptor (ion distribution, morphology, chemical composition, etc.)	The contribution and rates of each emission category to the receptor	1) Can only determine the contribution of each source category, but not an emission source. 2) Particle migration process is not taken into account to avoid the difficulties encountered in source model. 3) Unable to forecast air quality. 4) Unable to distinct emission sources with similar chemical or morphological characteristics. 5) Limited to specific receptors.

1.3.2 Developmental stage (1980s-1990s)

Since 1980, the methodology of CMB has attracted many scholars' attention to put forward a variety of algorithms. In 1982, "least squares fitting" was proposed by Henry to promote the development of CMB model algorithm[25]. In 1984, "effective variance weighted least squares method" was proposed by Watson[26], which was recommended and incorporated in the series of source apportionment techniques by the United States Environmental Protection Agency (USEPA) and had been adopted ever since. Over the past few years, a lot of manpower and resources were invested in researches on CMB and other receptor models, and the development of a more mature application package, in US. Now, the CMB model is upgraded to CMB 8.2.