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Application of receptor models to airborne particulate matter

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Abstract

The human activities in their various aspects cause a change in the natural air quality. This change results more marked in very populated and in high industrialized areas. Some pollutants emitted are typical of a particular activity. Each source of pollution is identified by its profile in the composition of the emissions in the environment. Multivariate receptor models can be used in order to apportion pollutants to the different sources assessing the contribution of each source to the total pollution.

This paper deals with the application of Absolute Principal Component Scores (APCS) receptor model to data obtained from the automatic network of air quality monitoring in the city of Bari (South Italy). The parameters monitored by automatic networks, as bihourly values, are PM_{10}, NO_x, CO, Benzene, Toluene, Xilene. The data shown in this paper concerning 1 month almost of sampling in different monitoring stations of Bari Municipality during the period of time from January 2005 to April 2006. Moreover preliminary results obtained applying the APCS model to daily $PM_{2.5}$ samples collected during SITECOS PRIN project are shown. The results concerning data collected in corso Cavour (Bari) during the month of October 2005.

The results obtained by APCS receptor model seem to suggest a poor contribution of the "vehicular traffic source" and a relevant contribution of the "secondary particulate source" to particulate matter concentrations.

Keywords: Receptor models; APCS; PM_{10}; $PM_{2.5}$; Source apportionment

1. Introduction

Recent epidemiological studies have shown a consistent association of the mass concentration of urban air thoracic particles (PM_{10} particles with an aerodynamic diameter smaller than 10 μm), and its subfraction fine particles ($PM_{2.5}$ particles with an aerodynamic diameter smaller than 2.5 μm), with mortality and morbidity among cardio-respiratory patients [1].

In studies based on several severe air pollution episodes, correlations between high concentrations of particulate matter (PM) and SO_2 pollution and acute increases in respiratory and cardiopulmonary mortality had been established beginning from 1970s. In these studies lung function growth was found impaired from long-term exposure to air pollutants and improved in districts where ambient air pollution had decreased [2]. Studies performed on a population of schoolchildren show that the reduction of the lung function parameters per 10 μg m^{-3} was highest for NO_2, followed by PM_1, $PM_{2.5}$ and PM_{10}, while exposure to coarse dust (PM_{10}–$PM_{2.5}$) did not change end-expiratory flow significantly [3]. Recent researches seem to indicate that PM_{10} is associate with respiratory responses and $PM_{2.5}$ with cardiovascular diseases [4]. The chemical characteristics of the particulate fractions and biological mechanisms responsible for these adverse health effects are largely unknown as well as the aerosol parameters (mass, particle size, surface area, etc) involved in the health impacts [5].

Moreover it seems that the increase in the atmospheric aerosol burden is delaying the global warming from the increase in greenhouse gasses (GHG: CO_2, CH_4, N_2O, halocarbons). Whether the increase in GHGs since preindustrial times is producing a warming of 2.4 W m^{-2}, the overall cooling effect of aerosols might be up to -2.5 W m^{-2} [6].

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The United States Environmental Protection Agency (US EPA) have implemented a National Ambient Air Quality Standards (NAAQS) for $PM_{2.5}$ in an attempt to reduce potential adverse health and environmental effects [7]. Air quality managers at the national and local levels will soon need to characterize their particulate air quality problems and devise a strategy to reduce PM and air pollutants concentrations to comply with new standards.

Thanks to monitoring networks, air quality managers have a consolidate cycle of testing, management and interpretation, but this is not enough to develop effective control strategies. In fact airborne particles in typical urban areas are a complex mixture of inorganic and organic compounds that exist in either the solid and liquid state. Traditional strategy to reduce PM concentrations are based on reduction of the controllable emissions of compounds that form airborne particles. However it is worth to consider that a variety of sources release fine primary particulate matter to the atmosphere including combustion sources (stationary and mobile), food preparation, activities that create dust (road travel, agriculture), and natural sources such as sea spray. Moreover secondary particulate matter can also form in the atmosphere through chemical reactions that convert gaseous pollutants (NO_x, SO_x, VOC) to semi-volatile products that partition to the particle phase. Furthermore, airborne particles with aerodynamic diameter less than 2.5 μm can stay suspended in the atmosphere for long periods of time.

Faced with this complexity, decision makers need a set of tools that clearly show the relationship between emissions sources and airborne particle concentrations [8].

Source apportionment techniques for airborne particulate matter are generally defined as any method that quantifies the contribution that different sources make to airborne particulate matter concentrations at receptor locations in the atmosphere. Source apportionment techniques are valuable tools that aid in the design of effective emissions control programs to reduce particulate air pollution. A lot of literature about different source apportionment techniques for airborne particulate matter is available [9–13].

2. Materials and methods

2.1. Receptor model used: absolute principal component analysis

The aim of the application of the receptor models is the apportionment of the pollutant's sources. The two main approaches of receptor models are Chemical Mass Balance (CMB) and multivariate factor analysis (FA). CMB gives the most objective source apportionment and it needs only one sample; however, it assumes knowledge of the number of sources and their emission pattern. On the other hand, FA attempts to apportion the sources and to determine their composition on the basis of a series of observations at the receptor site only [10]. Among multivariate techniques, PCA is often used as an exploratory tool to identify the major sources of air pollutant emissions [14–17]. The great advantage of using PCA as a receptor model is that there is no need for a priori knowledge of emission inventories [18].

In our experience Factor Analysis performed by APCS (Absolute Principal Component Scores) method is the most forceful in the reconstruction of the source profile and contribution matrices [19].

The mass balance equation can thus be extended to account for all *m* elements in the *n* samples as the sum of the contributions by *p* independent unknown sources,

$$C_{ij} = \sum_{K=1}^{P} M_{ik} A_{kj} \tag{1}$$

where C_{ij} is the *i*th concentration measured in the *j*th sample, M_{ik} is the concentration of the *i*th parameter in the *k*th source, and A_{kj} is the fraction of the *k*th source contributing to the *j*th sample.

In matrix form Eq. (1), taking into account the data error, becomes:

$$X = \mathrm{FA} + E \tag{2}$$

where *X* (n_m) is the measured concentration matrix. *F* (n_p) is the source contribution matrix. *A* (p_m) is the source profile matrix. *E* is the residue matrix, that takes in account the reconstruction error. The solution of Eq. (2) is not unique but, in any case, the solutions must be found according to a set of physical constraints. For example, all the terms of the matrices *F* and *A* must be non-negative numbers.

One of the most used methods to decompose the concentration matrix in the product of the source pattern, and contribution matrices is the APCS. The starting point is the matrix *X* (samples_parameters). In APCS the first step is the search of the Eigenvalues and Eigenvectors of the data correlation matrix *G*. Only the most significant p Eigenvectors (or factors) are taken into account. Generally p Eigenvectors are taken into account until the sum of their Eigenvalues reaches at least 80% of the total variance. The *p* Eigenvectors are then rotated by an orthogonal or oblique rotation. The most used rotation algorithm is Varimax, which performs orthogonal rotation of the loadings. The goal of this strategy is to obtain a clear pattern of loadings, that is, factors that are somehow clearly marked by high loadings for some variables and low loadings for others. After the rotation all the components should assume positive values; small negative values are set zero. An abstract image of the source contributions to the samples can be obtained by multivariate linear regression:

$$Z = PCS{*}V^T \tag{3}$$

where *Z* is the scaled data matrix, PCS is the principal component scores matrix, and V^T is the transposed rotated loading (Eigenvectors) matrix.

In order to pass from the abstract contributions to real ones, a fictitious sample Z_0, where all concentrations are zero, is built [17,20].

Using the matrix V^T and the Eq. (3) the vector PCS_0, corresponding to Z_0, is calculated and subtracted from all the vectors that form PCS. The matrix obtained in this way is