Short review on atmospheric aerosols source apportionment methods

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Appropriate understanding of particulate matter emission sources is required to properly establish the policies aiming at the aerosols emitters' elimination. The article provides a short review on atmospheric aerosols source apportionment methods using three computing models.
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Introduction

There is no need to convince anybody how important the environment quality is for each living organism, including human. As that is commonly known, inhaling of particulates, especially those fine-sized ones, into the lungs may have an adverse impact on health. Diseases of autoimmune, cardiovascular and pulmonology systems are the consequence of both short- and long-term exposure of organisms to the augmented concentrations of dust. In April 2014, WHO issued new information estimating that outdoor air pollution was responsible for the deaths of some 3.7 million people under the age of 60 in 2012. The Organization also emphasized that indoor and outdoor air pollution combined are among the largest risks to health worldwide [1].

As a matter of priority, causes of pollution should be identified and emissions of pollutants should be dealt with at source, in the most economically and environmentally effective manner. Therefore, an understanding of particulate matter source contributions is required in order to establish policies for reducing the emissions. Only knowing the variability of PM sources, their occurrence, strength and spatial distribution it is possible to design effective strategies for minimizing of health-related risk. In the field of atmospheric sciences, data on the identification of PM sources may be obtained by means of receptor modelling techniques [2-4]. Receptor models are mathematical procedures for identifying and quantifying the sources of ambient air pollution and their effects at a site (receptor), primarily on the basis of concentration measurements at the receptor site and generally, without need of emission inventories and meteorological data [5]. The fundamental principle of receptor modelling is that mass conservation between the emission source and the study site can be assumed, and a mass balance analysis can be used to identify and apportion sources of atmospheric pollutants. Generally, each receptor model may be boiled down to the mathematical equation as follows [6, 7]:

$$x_{ij} = \sum_{k=1}^{p}\left(g_{ik}f_{kj} + e_{ij}\right) \qquad [1]$$

where x_{ij} is the concentration of the jth species in the ith sample, g_{ik} the contribution of kth source to the ith sample, f_{kj} the concentration of the jth species in the kth source, and e_{ij} is the residua (i.e. the difference between the measured and fitted value) term.

The aim of the review is to provide short information about two source apportionment methods which enable to recognize emission sources of atmospheric aerosols.

Source apportionment approaches

The objective of source apportionment (SA) is to derive information about pollution sources and the amount they contribute to ambient air pollution levels [8,9]. Three main approaches may be distinguished within the SA methods: emission inventories, source-oriented models and receptor-oriented models [10]. This document aims at providing the knowledge about the receptor-oriented methodology (positive matrix factorization), explaining its role in the identification of sources with particular reference to particulate matter [11] and source-oriented model (mass closure).

Mass closure

Mass closure is accomplished by comparing the mass of particulate matter (PM) to the sum of the masses of the major chemical components. It is a computing analysis of accuracy of measurements carried out for the purpose of chemical characterization of atmospheric aerosols.

For the purpose of the mass closure study, components are divided into several classes as following: elemental carbon, organic matter (OM), mineral components or crustal materials, secondary inorganic aerosols, sea salt and unidentified mass fraction [12-16]. Some of scientists include in their considerations de-icing salt [17], industrial tracers [18] and non-sea salt components [19] as well. OM is usually estimated on the base of organic carbon values multiplied by a factor of 1.4 as proposed for urban environments [20, 21]. A value of 1.2– 1.4 is almost universally used to estimate the average organic molecular weight per carbon weight [22]. The mass of crustal fraction must also be estimated from elements, as these are frequently present as oxides or carbonates. Mineral components fraction is generally composed from aluminum, silica, carbonate ions, non-sea salt calcium (calculated as $[Ca]–[Na^+]$ x 0.038 [23]) and non-sea salt magnesium (calculated as $[Mg^{2+}]–[Na^+]$ x 0.12 [23]). Carbonate ions (CO_3^{2-}) are frequently estimated on the base of stoichiometric ratio as given in Calcium carbonate, assessing that all Ca^{2+} ions are present as $CaCO_3$ [24]. Silicon as a component of the main elements group is estimated by multiplying the aluminum concentration by a factor 3.0 [24]. Secondary inorganic aerosols is calculated as a sum of ammonium, nitrate and sulphate ions [25]. Sea salt contribution is estimated depending on the region for which the emission sources are apportioned. There are two ways of sea salt share estimation; either to assume that the total sodium ions are solely derived with the sea salt ($SS = [Na^+] + [SS-Cl^-] + [SS-Mg^{2+}] + [SS-K^+] + [SS-Ca^{2+}] + [SS-SO_4^{2-}]$) [13, 15, 23], or to base on the sea water chemical composition (55% Cl^-, 31% Na^+, 8% SO_4^{2-}, 4% Mg^{2+}, 1% K^+, 1% Ca^{2+}, <1% other) [26]. Marenco et al., 2006 estimated the sea salt contribution accordingly with the following relationship: $[sea\ salt] = 1.46[Na^+] + [Cl^-]$. The 1.46 factor in front of Na^+ stands for other ionic constituents (representing 14 % of sea salt), and Cl^- is added separately [27]. Sea salt contribution may be calculated from Na^+ only, using the appropriate molar correction factor [28]. However, this approach may results in large overestimation since Na and Cl seldom occur in the same proportion as in seawater. The reason is that Cl volatilizes by reacting with acidic species [23] or with ammonium nitrate on the samples [29]. Cheng et al., 2005 proposed the formulas for estimation of secondary derived $(NH_4)_2SO_4$ and NH_4NO_3 [30]: $[(NH_4)_2SO_4]=1.38[nss-SO_4^{2-}]$ and $[NH_4NO_3]=4.44[ex-NH_4^+]$, where $nss-SO_4^{2-}$ stands for non-sea salt sulphates and $ex-NH_4^+$ is the concentration of excess ammonium. The mentioned relationships were also applied by Wioletta Rogula-Kozłowska for source apportionment of size-segregated urban particulate matter [31]. The unidentified matter is obtained as the difference between the gravimetrically measured aerosol mass and the reconstructed mass (i.e., the sum of quantified chemical components). The remaining unidentified mass fraction can be related to aerosol-bond water, differences in the ox-idation level of organic carbon (deviations from the 1.4 calculation factor used for estimation of OM) and from analytical errors [19].

Mass closure is a frequently undertaken by scientists results analysis method aiming at the presentation of the aerosol chemical composition variations depending on sampling season, sampling locations or particulate matter fraction size. Besides that it primarily derives information about PM emission sources distribution. Figure 1-3 present the examples of mass closure for atmospheric aerosols from Los Angeles and from some countries from Europe. For the purpose of mass closure study chemicals are divided differentially on each figure. Sillanpää et al. [14] included the iron-rich dust and calcium salts as well presenting the mass closure for four size fractions of particulate matter (Figure 1). Yan et al. [32] presented the contribution of different types of aerosol, including soil, non-soil dust and sea salt, to TSP and PM2.1 (Figure 2). Yin and Harrison [18] divided chemicals into ten fractions, including sea salt and soluble/insoluble soil (Figure 3). The relative contributions
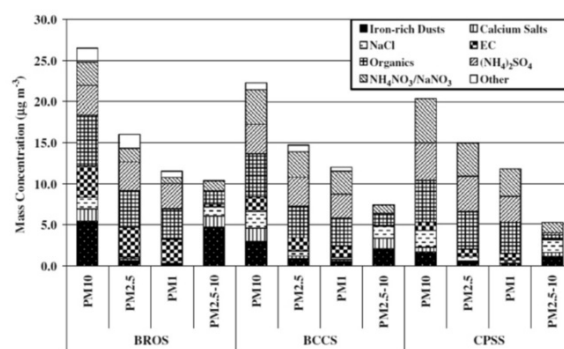

Figure 1. Mean chemical composition of the four size fractions PM10, PM2.5, PM1, PM2.5-10 at three sites in England (BROS – Bristol Road Site; BCCS – Birmingham City Centre site; CPSS – Churchill Pumping Station site) [18].
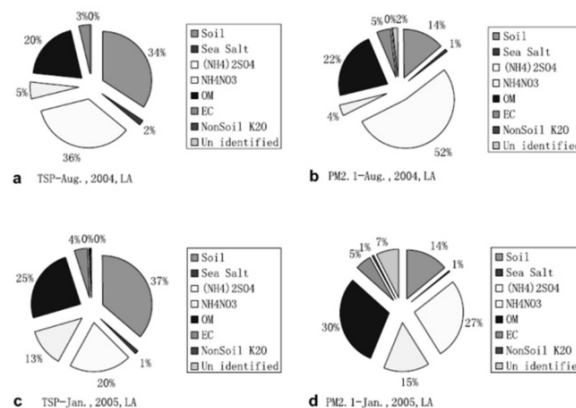

Figure 2. Contributions of the different types of aerosols to TSP and PM2.1 in August 2004 and January 2005 at Los Angeles site [32]
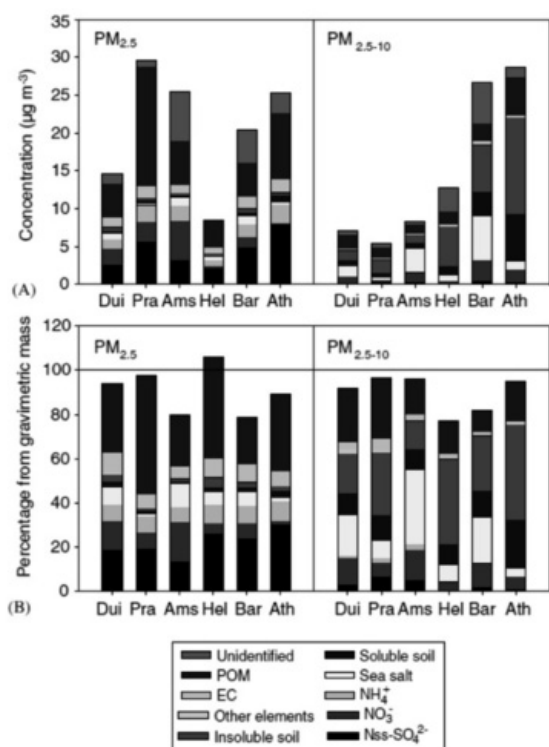
Figure 3. The mean mass concentrations: (A) and relative contributions; (B) of the nine chemical components and unidentified matter to PM2.5 and PM2.5-10 of the six sampling campaigns (Duisburg, Prague, Amsterdam, Helsinki, Barcelona, Athens) [14]

of particulate components fractions reflect differences in emission sources and processes (e.g., transformation, rain) controlling the atmospheric aerosols composition.

Positive matrix factorisation

Positive matrix factorization (PMF) is considered as a specific type of factor analytical method which uses experimental uncertainty for scaling matrix elements. Detailed knowledge on PMF computing methods were presented by Adam Reff and Shelly I. Eberly, who pointed out to the numerous procedural decisions which must be made and algorithmic parameters which must be selected while analysing PM by PMF [3]. Estimation of uncertainties is a very significant step in the PMF analysis. Uncertainties are usually estimated on the base of analytical uncertainties. The samples are size-segregated, which helps to apportion the sources. In order to find the solution, a dataset with a rather large amount of data consisting of chemical constituents (such as elemental concentrations) gathered from a number of observations (samples) is required. The larger the data matrix, the higher the chances that the model will identify distinct factors that can be identified as sources. PMF model is generally boiled down to the corresponding matrix equation:

$$x_{ij} = \sum_{k=1}^{p}\left(g_{ik}f_{kj} + e_{ij}\right) \qquad [2]$$

Where: X is a n×m data matrix with n measurements and m number of chemical species; E is a n×m matrix of residuals; G is a n×p source contribution matrix with p sources; F is a p×m source profile matrix [10]. It is well known that factor analysis can give a number of possible solutions, all mathematically correct. The choice of the best solution in PMF analysis, e.g. the number of factors that best represent the real case under study, shall be supported by quantitative indicators. A detailed description of the positive matrix factorisation may be found in the Pattero's study [33]. The computational tool for source apportionment by the PMF was developed by Paatero and Tapper [34,35].

The PMF model was already applied in several studies: for source apportionment of ambient particles in the San Gorgionio wilderness [36], for indicating inorganic and organic species as tracers for ambient particulates from Rochester (U.S.) [37], impact of biogenic aerosols and biomass combustion on the urban particulate matter were assessed in Berlin [38], PM emission sources were apportioned in Genoa (Italy) [39] and in Izmir (Turkey) [40].

Size-segregated samples are apportioned accordingly with factors denoted for specific sources. During the PMF analysis the number of source factors and the factor rotation has to be carefully considered. The number of factors should be chosen by observing the objective function of PMF analysis. Thus, for instance Carvahlo and Freitas, 2011 [41] and Pražnikar et al., 2014 [42] use the trace elements composition of particulate matter for source identification. In both experiments the factors are suited to the climate and location conditions. The first one explains: Fe, K and Na as *soil factor*, Na, K, Fe and Br – as *sea salt factor*; Zn, Fe, K and Na – a *industrial factor*; Sb and Hf – as *paved road dust*, including the vehicle wear; K, Br, Zn and As – as *fuel oil combustion factor*. The second one considers: V and Ni – as *ship and heavy machinery combustion factor*; Ca, Ti and Sr – as *mineral dust factor*; Cl and Br – as a *marine-derived aerosols*; Fe and Mn – as *Iron ore manipulation in harbours*; S and K – as *coal and biomass combustion*. Wagener et al. [38] indicated factors as following: 2-methyl-tetrols as a tracer for *isoprene-derived SOA* (secondary organic aerosols) [43]; levoglucosan and residuals of EC were denoted as tracers for *biomass burning* [44]; cholesterol which contributes to the *wood combustion* [45]. The factor with EC is named *combustion fossil*. The factor with large amounts of fatty acids is named *bio/urban primary*, as fatty acids are known to derive from biogenic sources like plant detritus or other plant fragments, and anthropogenic sources like cooking or combustion processes [46]. Figure 4 presents the example of final results of the PMF analysis. Briefly, the fatty acids in PM10 were arising from biogenic emissions rather than from cooking emissions, and in
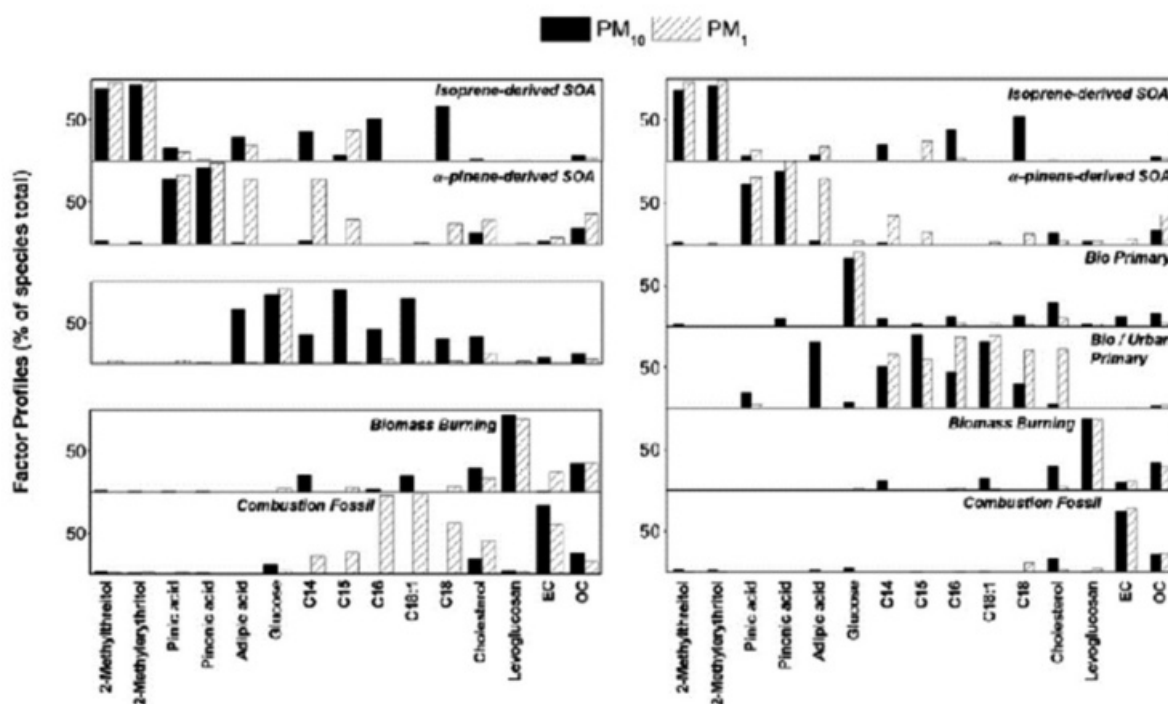
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Figure 4. Factor profiles for the PMF analysis in Berlin [38]

PM10 they were rather attributed to cooking emissions than to fossil combustion. As already suggested by tracer analysis, biomass burning dominates the organic fraction in the cooler months at sites that are not directly exposed to traffic and dwellings. Thus biomass burning still represents a source not to be underestimated also in industrialized countries [38].

PMF does not require source profiles as model inputs but does require knowledge of source profiles to interpret the factors derived from the model as air pollution sources.

Chemical mass balance (CMB)

The Chemical Mass Balance (CMB) model is one of several receptor models that has been applied to air quality problems over the last decades. The method was developed and applied i.e. by Kowalczyk et al. in 1978 who used a weighted least-squares regression analysis to fit six sources with eight elements for ten ambient samples. Finally, the chemical-element-balance method served for the interpretation of emission sources of ambient particles collected at four sites in Washington in 1974 and analysed for 27 elements. Emission sources were apportioned into six components: coal (I, As, Se), oil (V, Ni) and refuse combustion (Zn, Cd, Sb), marine aerosols (Na), soil (K, Mn, Mg) and motor vehicle emissions (Pb, Br) [47]. Unlike the positive matrix factorisation method where only the qualitative information on sources is sufficient, the chemical mass balance estimation requires a detailed quantitative knowledge of the emission sources profiles. In comparison to other receptor models, the CMB is able to apportion a larger number of

sources. However, while computing the source apportionment, the CMB meets some limitations like: 1) source profiles need to be experimentally determined; 2) unknown sources are unable to be identified; 3) secondary aerosols linked to the input emission sources are unable to apportion. Besides that, the CMB requires both speciation and uncertainty data, plus emission profiles with uncertainties [9]. The CMB was already applied by many scientists as summarized by Watson et al. [11]. Several method of estimation were used, but the application of the variance least squares estimation method seems to be most effective and is most commonly used. The reason is that this method incorporates precision estimates for all of the input data into the solution and propagates these errors to the model outputs [11]. Nevertheless, the method is frequently used for the source apportionment process. Watson and Chow applied the CMB for the source characterisation of major emission sources in the Imperial and Mexicali Valleys along the US/Mexico border [48]. They acquired source measurements of geological sources, motor vehicle exhausts, vegetative burning and industrial sources. Samples of PM10 fraction were analysed for ~50 chemical species. Source profiles resulted in elevated abundances of crustal components (Al, Si, K, Ca, Fe) from geological material, carbon (OC, EC) and trace elements (Br, Pb) from vehicle exhausts, carbon (OC, EC) and ions (K^+, Cl^-) from vegetative burning, ions (SO_4^+, NH_4^+, Na_+, K_+, Cl^-) and elements (Cl, Se) from a manure-fuelled power plants, and sulphur and trace elements (Na^+, Pb, Se, Ni, V) from an oil-fuelled glass plant [48]. In turns, Chow et al. described

sources profiles as emissions from paved and unpaved roads (Ca), natural soil dust, motor vehicles (EC-to-OC ratio), vegetative burning (K$^+$), coal-fired power plants (SO4+, Se), coal fly ash, a petroleum refinery catalytic cracker (SO$_4^+$, V, Ni, La, Sb), cement kilns (SO$_4^+$, Ca, total K, OC) and meat cooking (OC) [49].

Conclusions

Receptor models serve for the apportionment of emission sources of pollutants. They require to determine the chemical characterization consisted of a wide range of chemical species (metals, non-metals, ions, organic and elemental carbon, organic species) both for source and receptor samples. Application of receptor modelling for the source apportionment requires to perform several steps as defined by Watson et al. [11]: 1) formulation of the conceptual model; 2) compilation of emission inventory; 3) characterization of source emissions; 4) analysis of ambient samples for mass, elements, ions, carbon, and other components from sources; 5) confirmation of source types with multivariate model; 6) quantification of source contribution; 7) estimation of profile changes and limiting precursors; 8) reconciliation of source contributions with other data analyses and source models.

Understanding of particulate matter source contributions is required in order to establish policies for reducing the emissions. Accordingly with Watson et al. [50] considerations, the costs of implementing emissions reduction strategies that did not result in the desired effects on air quality would be hundreds of times the cost of the receptor modelling study and would prolong human exposure in the area to excessive particle concentrations until the effective control strategies were identified.

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