

## Review

# Critical review and meta-analysis of ambient particulate matter source apportionment using receptor models in Europe



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## HIGHLIGHTS

- ▶ Receptor models evolve towards tools with refined uncertainty treatment.
- ▶ Positive Matrix Factorization and Chemical Mass Balance are the most used models.
- ▶ Gas-to-particle conversion is the main PM mass and particulate organic carbon source.
- ▶ To abate exceedances, secondary inorganic and traffic are the main sources to target.
- ▶ More long term speciated PM datasets would foster source identification studies.

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## ABSTRACT

A review was conducted of the published literature on source apportionment of ambient particulate matter (PM) in Europe using receptor models (RMs). Consistent records were identified for source contribution estimates of PM mass concentrations for 272 records and of organic carbon (OC) in PM for 60 records. Over the period 2000–2012, a shift was observed in the use of RMs from principal component analysis, enrichment factors, and classical factor analysis to Positive Matrix Factorization while Chemical Mass Balance is still topical.

Following a meta-analysis of the published results, six major source categories for PM were defined that comprise almost all individual sources apportioned in Europe: atmospheric formation of secondary inorganic aerosol (SIA), traffic, re-suspension of crustal/mineral dust, biomass burning, (industrial) point sources, and sea/road salt. For the OC fraction, the three main source categories were: atmospheric formation of secondary organic aerosol, biomass burning, and fossil fuel combustion. The geographical and seasonal variations of these sources are mapped and discussed.

A special analysis of PM concentrations that exceed the current European air quality limits indicated SIA and traffic as the most important source categories to target for abatement throughout the year together with biomass burning during the cold season.

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## 1. Introduction

The apportionment of sources for air pollution in areas where the legislation thresholds are exceeded is of utmost relevance for the development of remediation measures. Different approaches have been used in Europe: a) chemical transport models based upon pollutant emission rates and meteorological data, and b) receptor-oriented models (RMs), especially for airborne particulate matter (PM), based on statistical analysis of pollutant

concentrations measured at a sampling site (*receptor site*) to infer the source-types and estimate their contributions to the measured site concentrations. The pollutants used for RMs are referred to here as *receptor species*.

How to classify receptor models? Receptor oriented source apportionment (SA) encompasses many tools ranging from simple techniques based on elementary mathematical calculations and basic physical assumptions (e.g. enrichment factor analysis) to complex models with pre- and post-data processing and user-friendly interfaces. Although all such tools deal with measured pollutant concentrations at the receptor site, the nature of the input data and the formats vary considerable. In general, there are three kinds of data input: ambient pollutant mass concentrations, source

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profiles, and meteorological data (i.e. wind speed and direction or backward trajectories). In addition, there are extended models, which can process other kind of information like season, week-day, precipitation, etc.

The present study presents a review of the most common methods and a meta-analysis of receptor models' source contribution estimation (SCE) for PM in Europe yielding a quantitative estimation of the most relevant source types and a mapping of their spatial distributions.

In this work, the approach used by Viana et al. (2008b) and Karagulian and Belis (2012) was extended and updated. SA methods are critically described and classified, including those dealing with particulate organic carbon. The information on pollution sources is sorted, summarized, and quantitatively evaluated, taking into account the sources of uncertainty. In this research the causes of exceedances are quantified on the basis of source contributions providing inputs for policy planning.

## 2. Receptor model techniques used in Europe for source apportionment of PM

For the present study, a total of 108 peer-reviewed, European studies and air quality research centres reports were scrutinized. These publications contain 332 records (every site is a record and more datasets on the same site are considered as different records). In the majority of the records (272), the complete PM mass is apportioned while the remaining 60 records deal with sources for the carbonaceous fraction. Included in these, are studies reporting aerosol mass spectrometer (AMS) measurements. Tables 1S and 2S in Supplementary material (SM) summarize the studies considered in this review, while Tables 3S and 4S report studies published after the conclusion of this work. Common for all the utilized multivariate RMs is the basic equation that refers to the solution of the mass balance problem (Eq. (1)):

$$x_{ij} = \sum_{k=1}^p g_{ik} f_{kj} + e_{ij} \quad (1)$$

where  $x_{ij}$  is the concentration of the  $j$ th species in the  $i$ th sample,  $g_{ik}$  the contribution of the  $p$ th source to the  $i$ th sample and  $f_{kj}$  the concentration of the  $j$ th species in the  $p$ th source, and  $e_{ij}$  is an uncertainty term introduced to facilitate a statistical solution of the mass balance as opposed to an analytical mathematical solution. In the  $F$  matrix, receptor models (RMs) may use input of experimental data on the concentrations of the receptor species in the source emissions, which is often referred to as the source fingerprints. We adopt the term *source profiles*. Alternatively, RMs may derive the concentrations of the receptor species in the  $F$  matrix by iterative methods. For these, we adopt the term *factor profiles*.

The main assumptions for the mass balance in Eq. (1) are common to all RMs, although they may be less critical for some RM techniques (i.e. factor analysis) than for others. These assumptions are:

- 1) that *source profiles* do not change significantly over time or do so in a reproducible manner so that the system is quasistationary. CMB depends on them not changing or knowing the fractionation coefficients. Factor analysis depends on them changing in a uniform manner from sampling interval to sampling interval;
- 2) that receptor species do not react chemically or undergo phase partitioning (solid/gas or solid/liquid) during transport from source to receptor (i.e., they add linearly).

Two other intrinsic assumptions are i) that data are representative of the studied geographical area and consistent with the conceptual model and ii) that comparable/equivalent analytical methods are used for the receptor site(s) throughout the study as well as for the characterization of the source profiles.

The statistical solution to Eq. (1) relies on variability in impacts of sources at the receptor. Variability may derive from variations in emission rates and in the meteorologically driven advection of emissions toward or away from the receptor. If Assumption 1 is not fulfilled, the RMs may either not be able to resolve the proper source profile, resolve composite profiles, or resolve split profiles. If Assumption 2 is not fulfilled, in the best case the source contribution estimates (SCEs) of the RMs may be under/overestimated or the inferred source types may be erroneous and in the worst case no solutions to Eq. (1) exists.

Metals are excellent receptor species with respect to Assumption 2 and they have been used from the very beginning of receptor modelling. However, in the search for specific receptor species for different combustion sources (*source tracers* also called marker compounds or molecular markers), the use of organic chemical compounds has grown popular in modern SA studies even though this class of compounds often comes into conflict with Assumption 2. RMs can tolerate small deviations from the assumptions and reactive compounds may in some cases be used as receptor species (e.g. Chemical Mass Balance modelling) with additional terms to correct gains or losses during their transport from source to receptor. Semivolatile and/or reactive compounds may work well as receptor species for near-source modelling or during certain meteorological conditions that preserve these compounds to a high extent (low temperatures, low photochemical activity). Moreover, if deviations from Assumption 2 can be quantified somehow, they can be expressed as fractionation coefficients (Friedlander, 1973; Grosjean and Seinfeld, 1989), which accounts for selective loss due to processes such as gravitational settling, chemical transformation, or evaporation; or selective gains in constituent due to chemical formation or condensation. In practice, only species with a fractionation coefficient close to 1 (small gains or losses) are included in the analysis (Schauer et al., 1996). Alternatively, such information has been utilized as error-input to the type of RMs that use error weighting in the statistical procedures (Belis et al., 2011; Junninen et al., 2009; Larsen et al., 2012).

### 2.1. Explorative methods

#### 2.1.1. Incremental concentrations approach

Two studies included in Table S1 are based upon the incremental concentrations approach (AIRPARIF and LSCE, 2011; Lenschow et al., 2001). This approach was applied primarily in the evaluation of SCEs from traffic to PM<sub>10</sub> in urban agglomerations. The SCE is based upon the comparison of the main chemical components at stations in the regional and urban background and a station at the kerbside of a busy street. The approach assumes that the SCE from traffic near the roads can be computed as the concentration increment from urban background stations to kerbside stations and that the SCE from city sources can be computed as the concentration increment from rural background stations to urban background stations. The contribution of sources to primary and secondary pollution is assumed to be proportional to their emission estimations derived from emission inventories for main chemical components with some adjustments for traffic and point sources.

A similar incremental concentrations approach has also been used to derive effective traffic emission profiles to be utilized in more complex receptor modelling (Wählin et al., 2006). This approach may be useful in exploring potential urban emission

contributions to ambient PM. In addition to the uncertain fulfilment of Assumptions 1 and 2, this approach falls back on potential oversimplifications of certain processes, specifically those concerning coarse and secondary particles. Compared to more complex RMs, this approach relies heavily on emission inventories.

### 2.1.2. Enrichment factor and tracer-based methods

The concept of enrichment factor (EF) analysis was developed in the seventies (Dams and DeJonge, 1976; Lawson and Winchester, 1979) and can be used for SA of PM together with knowledge on source profiles to suggest the emission sources. The EF is the ratio between elemental ratios in the measured sample to that of a reference material (e.g. particle composition vs crustal abundance):

$$EF = \frac{x_{\text{aerosol}}/r_{\text{aerosol}}}{x_{\text{crust}}/r_{\text{crust}}} \quad \text{or} \quad EF = \frac{x_{\text{aerosol}}/x_{\text{crust}}}{r_{\text{aerosol}}/r_{\text{crust}}} \quad (2)$$

where  $x$  is the concentration of the element under consideration (usually in mass/mass units),  $r$  is the chosen reference element and the subscripts indicate the environmental media.

A simple application of EF analysis for PM source indication may be the study of heavy metals (e.g. brake-metals) at a road site. For those metals not emitted by traffic, the ratio between EF of ambient PM and the EF for mineral dust (crust) remains close to unity, while this ratio will be significantly higher than one for species like Cu. If source profiles are available for other sources, indications of their potential impact on ambient PM at the receptor site can likewise be inferred (e.g. tracers for metallurgic industries such as Zn, Fe, and Mn).

The EF method may either use: a) a multilinear regression (MLR) algorithm, b) edges of a two-dimensional scatter plot, or c) the concentration ratios of the pollutant of interest and the tracer compound in the ambient PM samples collected at a particular period when it is believed that a single source is dominant. As output, the EF method yields contributions to species from each source considered in the model; see the mathematical description by Watson et al. (2008).

EF analysis is simple and as long as the basic mass balance assumptions for RMs are well fulfilled, EF analysis can be used in data screening or to support assumptions for receptor species and sources for which little information is available. However, the existence of unique source tracers is rare. Thus, results from EF analyses should always be interpreted with caution.

The most frequent use of tracer-based methods in modern SA is found for the apportionment of organic carbon (OC) fractions in PM (see Chapter 2.6) and more than 35% of European OC SA studies use this approach (Table S2).

A particular tracer-based approach was taken by the UK Model of Air Pollution Expert Group (APEG or AQEG) to evaluate the contribution from primary combustion and secondary inorganic aerosol to the PM<sub>10</sub> mass in the UK (Stedman et al., 2001). For that purpose, the emissions from traffic, heating and power plants were assumed to be proportional to NO<sub>x</sub> concentrations. Similarly, secondary particle concentrations were assumed to be proportional to SO<sub>4</sub><sup>2-</sup> concentrations in rural sites. The remaining part of the mass was considered to derive basically from re-suspension of coarse particles and other local sources not emitting NO<sub>x</sub>. Total PM<sub>10</sub> was estimated by multilinear regression (MLR) according to:

$$x = a \cdot [\text{NO}_x] + b \cdot [\text{SO}_4^{2-}] + c \quad (3)$$

where  $x$  is the measured PM<sub>10</sub>, [NO<sub>x</sub>], [SO<sub>4</sub><sup>2-</sup>] are measured nitrogen oxides and sulphate, and  $a$ ,  $b$ ,  $c$  are the fitted MLR coefficients

(μg m<sup>-3</sup>). Therefore, the last source category could be a mixture of sources or an overestimation of re-suspension. This approach takes advantage of the information readily available from monitoring networks. It has been used by (AQEG, 2005) for the data treated in the present meta-analysis (Table 1S). However, the fulfilment of the intrinsic assumptions cannot easily be verified. More sophisticated RMs will be described in the following.

### 2.2. The Chemical Mass Balance model and related methods

Promoted by freely availability stand-alone software from the United States Environment Protection Agency (US-EPA), the Chemical Mass Balance model (CMB) has become highly popular and approximately one-fifth (18%) of European SA studies for PM (Table 1S) are based hereupon. CMB has also found applications for the OC fraction of PM (Table 2S).

CMB as described by Watson et al. (1997) uses an effective variance least squares approach to solve Eq. (1.) on the basis of known source profiles ( $f_{kj}$ ) and measured receptor species concentrations associated with ambient PM ( $x_{ij}$ ). In effective variance least squares, the weighting is inversely proportional to the square of the uncertainty in the source profiles and ambient data for each species according to Eq. (4):

$$(W_e)_{ij} = \frac{1}{\sigma_j^2 + \sum_{k=1}^p \sigma_{jk}^2 \sigma_k^2} \quad (4)$$

where  $\sigma_j$  is the measured uncertainty of the ambient concentration,  $x_i$ , and  $\sigma_{jk}$  is the measured uncertainty of species  $j$  emitted by source  $k$ .

The accuracy of a CMB modelling results relies strongly on the availability of source profiles, which ideally must be from the region where the receptor is located and that should be contemporary to the underpinning ambient air measurements. CMB requires a good knowledge of the emissions in the study area in order to assure that all relevant sources are included and to evaluate their uncertainty. Because of CMBs sensitivity to collinearity of the source profiles, which impedes the mathematical solution of the mass balance, often it is necessary to merge sources into groups of source types in order to produce composite profiles. This exercise automatically builds in intrinsic assumptions into the CMB model. Thus, it is useful to characterize the sensitivity of the produced SCEs to the lumping of sources and to the selection of receptor compounds by running a series of scenarios. In compensation for the requirement of source profiles as input, CMB can be carried out for a limited number of samples. However, small datasets may not fully characterize the source–receptor relationships at a given site.

Studies using CMB in Europe: Andriani et al. (2011), AQUELLA (2007), Belis et al. (2011), El Haddad et al. (2011b), Junninen et al. (2009), Larsen et al. (2008), Mossetti et al. (2005), Pandolfi et al. (2008), Perrone et al. (2012), Viana et al. (2008a), and Yin et al. (2010).

Methods related to CMB are Non-Negative Least Squares (Wang and Hopke, 1989) and Partial Least Squares Regression, which is a generalization of Multiple Linear Regression (MLR) suitable for analysing data with collinear, noisy, and numerous  $x$ -variables (Vong et al., 1988). These and other multivariate calibration methods have not yet found European application.

Source profiles are not readily available for most receptor sites. Thus, multivariate models that as opposed to CMB, do not require experimental source profiles as input (eigenvalue analysis and factor analysis) are widely popular and will be described. It is a common misunderstanding that such methods require less *a priori* knowledge of the modelled receptor sites. Indeed, the determination of the optimal number of factors, the assessment of

rotational ambiguity, and the interpretation of factors as potential sources are issues that require not only good command of the software tool but also a profound qualitative understanding of emissions in the study area.

### 2.3. Principal component analysis and related methods

In principal component analysis (PCA), the mass balance equation is solved using eigenvector analysis or single value decomposition. The availability of stand-alone software or plug-ins has contributed to the popularity of this method for source assignment. In Europe, approximately one quarter (24%) of European SA studies for PM were based on PCA and variants (Table S1).

#### 2.3.1. Principal component analysis

The main objective of PCA is to convert a set of observations of possibly correlated variables into a set of values of linearly uncorrelated variables, called principal components (PCs), which are subsequently interpreted by the modeller as potential source profiles. PCA uses proper orthogonal decomposition in such a way that the first principal component accounts for as much of the variability in the data as possible, and each succeeding component in turn has the highest variance possible under the constraint that it be uncorrelated with the preceding components (Roscoe et al., 1982).

PCA is sensitive to the relative scaling of the original variables and is based upon the intrinsic assumption that the dataset jointly is normally distributed, which is not always the case for environmental concentration data. The artificial positioning of variance into the first few components can be partially solved by orthogonal rotations (e.g., varimax).

In order to overcome the effects of mean-centring of PCA scores, methods have been developed in which PCs are uncentered by subtracting a zero-valued pseudosample and regressing against the total PM mass: Absolute Principal Component Scores, APCS (Thurston and Spengler, 1985), Absolute Principal Component Analysis, APCA (Swietlicki and Krejci, 1996) and PCA-MLR (Tauler et al., 2008). These methods will be referred to as APCS. The mass balance equation for the multilinear regression is shown in Eq. (5):

$$X_i = \sum_{k=1}^p z_k \text{APCS}_{ki} \quad (5)$$

where  $X_i$  is the PM mass recorded during observation  $i$ ,  $\text{APCS}_{ki}$  is the rotated absolute component score for component  $k$  on observation  $i$ , and  $z_k$  are the regression coefficients between the PCs and the pollutant mass. APCS applied to PM and combined with gaseous pollutants or meteorological parameters “*a posteriori*” have been used in support of source identification and to estimate the geographic origin of pollutants (Juda-Rezler et al., 2011; Viana et al., 2006).

The most critical step of using PCA for SA is the interpretation of the PCs and their assignment to potential emission sources. PCA does not perform explicit data uncertainty treatment (as opposed to the more sophisticated Factor Analysis described in the next section). Therefore, noise deriving from the uncertainty structure of the datasets, which often include heteroscedastic receptor species, is incorporated by PCA into the PCs (Paatero and Hopke, 2003). Moreover, the basic assumption for PCA of orthogonal component does not reflect the structure of real world data (many source profiles have a degree of collinearity). Overall, this may lead PCA to produce solutions with collinear source profiles lumped into PCs or partially split into more PCs.

Studies using PCA and related methods included in the meta-analysis (more details in Table 1S): Almeida et al. (2005, 2006),

Amato et al. (2010b), Astel (2010), Bruno et al. (2008), Callén et al. (2009), Contini et al. (2010), D’alessandro et al. (2006), Larsen et al. (2008), Manoli et al. (2002), Marengo et al. (2006), Moreno et al. (2006), Negral et al. (2008), Querol et al. (2004), Salvador et al. (2007), Samara et al. (1994), Sánchez de la Campa et al. (2007, 2010), Vallius et al. (2005), and Viana et al. (2007, 2008a).

#### 2.3.2. The UNMIX model

A few European studies are based upon the UNMIX model (Callén et al., 2009; Larsen et al., 2008; Mijić et al., 2009) (Table 1S), which solves the chemical balance equation (Eq. (1)) using eigenvalue analysis to reduce the dimensionality of the dataset without centring the original data (Henry, 2003). The number of PCs is estimated by the NUMFACT algorithm (Henry, 1997) relying on the signal to noise ratio of PCs in advance. An edge-finding algorithm based on Self-Modelling Curve Resolution (SMCR) techniques is then applied. Edges are hyperplanes determined by points in which a source profile is absent or has a very low relative contribution. Edges are used as explicit physical constraints to define a region of the real solution where source contributions are greater than or equal to zero. As output UNMIX yields a set of factors (PCs) of co-varying receptor species, which are subsequently interpreted by the modeller as potential source profiles.

UNMIX does not incorporate errors into the analysis and suffers from some of the same concerns as PCA. Although the software is freely available from the US-EPA (current version 6.0), it has not gained much popularity in Europe. This may be linked to a limitation of the model that not always find a mathematical solution to the mass balance and to the characteristic of UNMIX that it is able to resolve the most intense sources while the weakest sources often show poor agreement between the expected and estimated source contributions (Henry, 2003).

### 2.4. Factor analysis

More than 40% of the European SA studies for PM have applied Positive Matrix Factorization (PMF) and 7% some other type of factor analysis without non-negative constraints (Table 1S). A few European PMF applications are also found for the OC fraction (Table 2S). PCA and FA are similar in the way they operate linear transformation of the original variables to create a new set of variables, which better explain cause–effect patterns. However, there are many differences between them. While PCA aims to maximize the variance by minimizing the sum of squares, FA relies on a definite model including common factors, specific factors and measurement errors. PCA has a unique solution while factors in FA are not exact linear functions of  $x$ . In PCA, variables are almost independent from each other while common factors (communalities) contribute to at least two variables (Hopke et al., 1976). FA is considered more efficient than PCA in finding the underlying structure of data (Jolliffe, 2002). However, PCA and FA produce similar results when there are many variables and their specific variances are small.

#### 2.4.1. Positive Matrix Factorization

Positive Matrix Factorization (PMF) resolves a weighted factorization problem with non-negativity constraints using known experimental uncertainties as input data thereby allowing individual treatment (scaling) of matrix elements (Paatero, 1997). PMF solves the mass balance equation (Eq. (1)) by minimizing the object function in Eq. (6).

$$Q = \sum_{i=1}^n \sum_{j=1}^m \left( \frac{x_{ij} - \sum_{k=1}^p g_{ik} f_{kj}}{u_{ij}} \right)^2 \rightarrow Q = \sum_{i=1}^n \sum_{j=1}^m \left( \frac{e_{ij}}{u_{ij}} \right)^2 \quad (6)$$

where  $e_{ij}$  represents the model uncertainties and  $u_{ij}$  the measured uncertainties.

The original version used the alternating least squares iterative method (Paatero and Tapper, 1993), but convergence was very slow and a faster algorithm was developed by computing  $G$  and  $F$  matrices simultaneously (PMF2). US-EPA PMF v 3.0 includes tools for input data pre-treatment, outlier detection, missing value correction, and uncertainty estimation. The most recent versions of the US-EPA software solve the PMF problem via the Conjugate Gradient algorithm (see Chapter 2.5.2) and contain routines to estimate the optimal number of factors (analysis of  $Q$ , analysis of residuals), to test the rotational ambiguity ( $FPEAK$  vs  $Q/Q_0$ ) and to introduce small constraints to limit this uncertainty ( $Fkey$ ). The uncertainty and stability of the solution is estimated by bootstrapping. Identification of the optimal number of factors, rotational uncertainty and factor source relationships are the areas for future development of this model.

Studies carried out using PMF (Table 1S): AIRPARIF and LSCE (2011), Alastuey et al. (2006), Aldabe et al. (2011), Alleman et al. (2010), Amato et al. (2009), Andriani et al. (2011), AQUELLA (2007), Belis et al. (2011), Bernardoni et al. (2011), Beuck et al. (2011), Callén et al. (2009), Chavent et al. (2009), Dogan et al. (2008), Fabretti et al. (2009), Furuşjö et al. (2007), Gietl and Klemm (2009), Gu et al. (2011), Healy et al. (2010), Hellebust et al. (2010), Huang et al. (2001), Junninen et al. (2009), Karanasiou et al. (2009, 2011), Koçak et al. (2009), Krecl et al. (2008), Larsen et al. (2008), Laupsa et al. (2009), Lucarelli et al. (2011), Mazzei et al. (2008), Mijić et al. (2009), Mooibroek et al. (2011), Moreno et al. (2011), Nicolás et al. (2008), Pandolfi et al. (2008, 2011), Pey et al. (2009), Raatikainen et al. (2010), Richard et al. (2011), Rodríguez et al. (2011), Santacatalina et al. (2010), Thimmaiah et al. (2009), Vestenius et al. (2011), Viana et al. (2008a, 2009), Yatkin and Bayram (2008), Yubero et al. (2011), Yue et al. (2008), and Zabalza et al. (2006).

Studies using factor analysis other than PMF included in the meta-analysis (more details in Table 1S): Harrison et al. (1997), Ilacqua et al. (2007), Koçak et al. (2009), Marczazan et al. (2003), Querol et al. (2002), Salvador et al. (2004, 2007), and Van Borm and Adams (1987).

## 2.5. Hybrid methods

Two categories of hybrid methods have been used for SA of PM: i) Constrained or expanded receptor models; ii) Trajectory based receptor models. The latter category utilizes pollutant concentrations and wind speed/direction measured nearby the receptor site or backward trajectories generated with a Lagrangian model (see SM for details). Among the data covered in the meta-analysis, only the first category of hybrid methods have been used, so the detailed discussion will be focused on these. Such models utilize multivariate factor analysis methods and accept explicit introduction of additional information (in addition to wind and trajectories) to reduce the rotational ambiguity of the solution.

### 2.5.1. The Constrained Physical Receptor Model (COPREM)

A few European SA studies for PM (Table 1S) are based upon COPREM (Andersen et al., 2007; Glasius et al., 2006; Wählin et al., 2006), which combines the features of chemical mass balance models and non-negative factor analysis (Wählin, 2003). The model is solved by minimizing a  $\chi^2$  function using a two-step iterative procedure. In COPREM, the modeller can use background knowledge to direct the iteration to a rational result by choosing, for example, vectors that are proportional to known source profiles, and by setting up constraints that keep parts of source profiles or whole profiles constant and hereby prevents unwanted mixing of the source vectors. The outputs of the model are the source

strength matrix, the profile matrix, plus  $\chi^2$  and the degrees of freedom ( $\nu$ ). A “one-factor” analysis is performed on the residues to reveal a possibly ignored source. The uncertainties of the source profiles are estimated with a specific module by linear regression. The calculated uncertainties represent, however, only lower bound values, because the rotational ambiguity and the uncertainties of the independent variables are ignored.

### 2.5.2. Extended Factor Analysis models

The classical bilinear factor analysis performed on a two dimensional matrix of samples by receptor species (two way model) has been further “extended” to solve more complex multilinear equations ( $n$ -way models) using the Multilinear Engine (ME) platform (Paatero, 1999). ME generates a table that specifies the model and then solves it using the Conjugate Gradient Algorithm. In the extended models the rotational ambiguity of factor analysis is reduced by including in the model additional constraints such as: known source profiles, known source contributions (or lack of contribution), meteorological variables or week-day. ME flexibility has been exploited to develop dataset specific models and to process heterogeneous data like aerosol composition and size distribution, data with different time resolution or multisite/multi type samples (Hopke, 2010).

European applications of the extended PMF model (Table 1S): Amato et al. (2010a), Amato et al. (2009), Escrig Vidal et al. (2009), and Yli-Tuomi et al. (2003).

## 2.6. Receptor modelling applications for the carbonaceous fraction of PM

The carbonaceous fraction is one of the main components of PM deriving from direct emissions (elementary carbon, EC and primary OC, POC) as well as from atmospheric gas-to-particle conversion of other pollutants through condensation processes, driven by temperature and dilution effects, and oxidation processes (secondary OC, SOC). Since most of the emitted POC is semivolatile and some gas-to-particle processes take place shortly after emission, some authors like Donahue et al. (2012), consider the distinction between primary and secondary OC obsolete. In this work, this terminology is utilized for coherence with the reviewed literature and to lay emphasis on the difference between sources (POC) and processes (secondary OC) which is relevant for the development of abatement measures. Like PM itself, the OC fraction is composed of a multitude of chemical compounds with a range of physical–chemical properties and it is practically impossible to identify and quantify more than a minor fraction. The reactivity, volatility and to some extent the hygroscopicity of compounds in the OC fraction may compromise the basic assumptions for RMs and make it unfeasible to apportion the sources for the precursor compounds for SOC. Therefore, efforts have been focused on apportioning the carbonaceous PM fraction into different classes with less emphasis on the pollution sources.

Three main methods have been followed in the vast majority of European studies (Table 2S):  $^{14}\text{C}/^{12}\text{C}$  isotopic ratios combined with macro-tracer enrichment factors (IRMEF; 35% of the records); aerosol mass spectrometry (AMS) combined with PMF of the non-resolved mass spectra (29% of the records); and CMB with specific molecular marker compounds (23% of the records).

El Haddad et al. (2011a) have used linear regression of macro-tracers to first apportion primary carbonaceous aerosol sources based on the known source profiles and subsequently to assign the non-apportioned mass to secondary organic aerosol (SOA).  $^{14}\text{C}/^{12}\text{C}$  ratios have been used to distinguish fossil from non-fossil carbon in PM (Szidat et al., 2007) and when  $^{14}\text{C}/^{12}\text{C}$  ratios are combined with the use of other tracers, resolution can be obtained of

anthropogenic from natural PM, and of primary from secondary aerosols (Gelencsér et al., 2007). A number of tracers and EFs have been employed in European studies with this approach: Levoglucosan as tracer for biomass combustion together with levoglucosan/OC ratios and elementary carbon (EC) to OC ratios; carbon monoxide as tracer for primary fossil fuel combustion together with OC/EC ratios; ratios of arabitol and mannitol saccharide concentrations to numbers of fungal spores as tracer for primary OC from biogenic sources (Gelencsér et al., 2007; Gilardoni et al., 2011; Larsen et al., 2012; Szidat et al., 2007). It should be noted that recent probabilistic uncertainty characterizations have demonstrated that results obtained with such trace-based methods may have high uncertainties (Larsen et al., 2012).

A relatively new method to identify OA sources uses AMS spectra. These instruments quantify the mass concentration of non-refractory species (including organic matter) via thermal vaporization and electron impact ionization. Because most molecules undergo extensive fragmentation, the AMS spectra provide information on the bulk OA with limited molecular detail. However, deconvoluting the mass spectral matrix using multivariate factor analysis (PMF, ME) yields distinct factors. A comprehensive review of this method has recently been published by Zhang et al. (2011). The most common factors obtained with this technique are a) hydrocarbon organic aerosol (HOA) that is considered to represent POA from fossil fuel combustion; b) cooking-related OA (COA) which has spectral features similar to those of POA from cooking emissions and a distinctive diurnal pattern; c) biomass burning (BBOA) correlated with biomass burning emission tracers and elevated peaks at  $m/z$  60 and 73 in the mass spectra; d) the low volatile oxidized organic aerosol (LV-OOA), which is interpreted as surrogate for regional, aged OA; and e) the semivolatile oxidized organic aerosol (SV-OOA) that is interpreted as surrogate for less photochemically aged OA (Zhang et al., 2011). The last two categories put the emphasis on the aerosol volatility and oxidation irrespective of its primary or secondary nature.

Also CMB has been used to apportion the carbonaceous fraction of PM to specific sources using molecular markers (Section 2.2). The latter was accomplished either directly by including oxidized species not present in primary emissions as molecular markers or indirectly by including only primarily emitted receptor compounds and attributing all non-apportioned OC mass to SOC. A number of oxidized species have been proposed such as terpene oxidation products, aromatic and linear aliphatic dicarboxylic acids, branched aliphatic ketones, and oxy-PAHs (Subramanian et al., 2007). However, the list can never be exhaustive and parts of SOC will remain undetected with this CMB approach. On the other hand, the attribution of non-apportioned OC is also not straightforward since this may derive from unknown primary sources, decay of molecular markers, sampling artefacts, and mathematical residuals in the statistical models. In order to validate the relationship between SOA and unexplained OC the EC-tracer method has been used (Subramanian et al., 2007).

Studies using IRMEF: Gelencsér et al. (2007), Genberg et al. (2011), Gilardoni et al. (2011), Heal et al. (2011), Larsen et al. (2012), and Yttri et al. (2011). Studies using AMS-PMF: Allan et al. (2010), Favez et al. (2010), and Lanz et al. (2010). Studies using CMB-MM: El Haddad et al. (2011a), El Haddad et al. (2011b), and Perrone et al. (2012). Studies using other models: Glasius et al. (2006), Saarikoski et al. (2008), and Sciare et al. (2011).

### 3. Results of European source apportionment of PM by receptor modelling

The majority of the studies considered in the present review focus on the PM<sub>10</sub> (56%) and PM<sub>2.5</sub> mass fractions (37%) and a few

studies address PM<sub>1</sub> (6%) and total suspended particles (TSP, 1%). About 67% of the studies were conducted at urban background sites, out of which 7% located in suburban and residential areas. The remaining part was carried out at source-oriented sites (18%), rural background sites (13%), and remote sites (4%). The source-oriented sites were focused on traffic, industrial, or harbour emissions. The monitoring sites are distributed over 20 European countries, with the highest number of records deriving from Italy, Spain, UK, and Poland (Table 5S; Figs. 5, 1S and 2S). Many of these studies have been undertaken to assess the sources in areas where concentrations of PM exceed the limit values or where they are cause for concern of harmful health effects and detrimental environment impacts of atmospheric pollution. Therefore, it is possible that the picture emerging from this review may over-represent critical situations and under-represent conditions where pollution levels are not a matter of concern.

A total of 243 records were retained for the statistical analysis while 29 had to be discounted for reasons such as evident over- or under-estimation of the total mass, unexplained fractions higher than 40%, and apportionment of the variance rather than the mass. The 243 records derive from studies carried out with different sampling duration and strategies. Most (61%) are based on data collected with either regular sampling throughout the calendar year or in series of short-term campaigns spanning all seasons. Other studies were conducted in only parts of the year (28% cold season, 11% warm season) in order to capture periods when sources of interest were active. This has been kept in mind when comparing data.

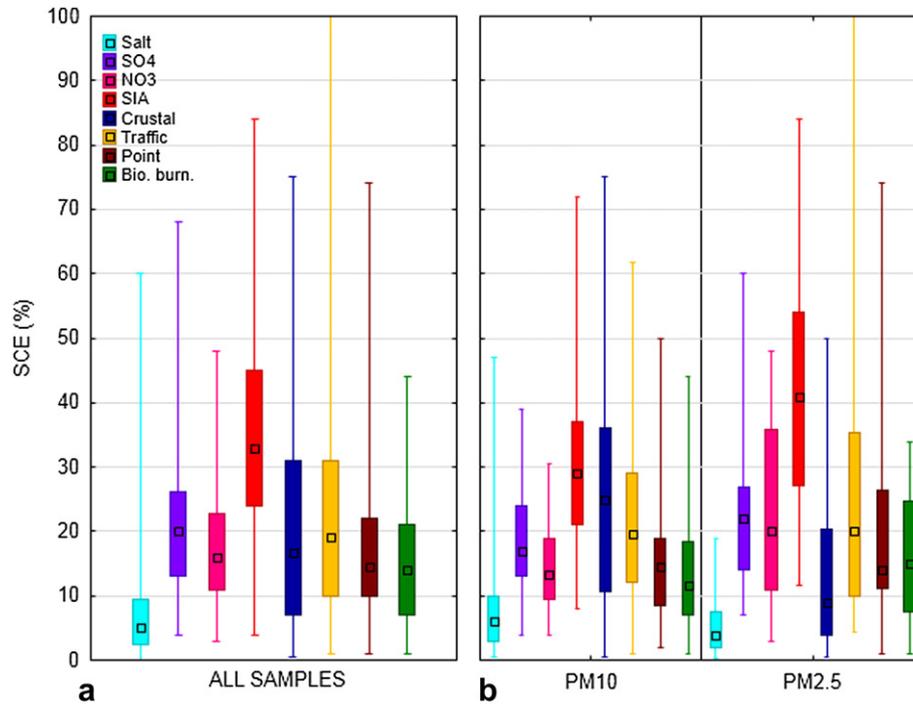
In the following, a descriptive statistical analysis is given for the published SA results and discussed in relation to sources for PM and its' organic fraction. The data do not follow normal distributions. Thus, testing of hypotheses is accomplished with the rank ANOVA non-parametric test for multiple values (Kruskal and Wallis, 1952).

#### 3.1. Contributions from six major source categories to the PM mass concentrations over Europe

The studies considered in the present review have been carried out with varying objectives and the SAs have been obtained with a range of different techniques, which do not necessarily produce directly comparable results. Often contributions have been estimated from mixed sources (e.g. by composite profiles in CMB and by mixed factors in PMF). In order to be able to compare all the SA results and to attain useful conclusions, sources have been pooled into six major categories covering those most frequently observed in the individual studies: Sea/Road Salt, Crustal/Mineral Dust, Secondary Inorganic Aerosol (SIA), Traffic, Point Sources, and Biomass Burning. An overview is given in Figs. 1 and 2 for the relative contribution from each source category and discussed in details as follows.

##### 3.1.1. Estimates of the relative source contributions for PM

**Uncertainty of individual SCEs.** Although it is becoming more common to include uncertainty considerations into source apportionment studies, only 35% of the SCE records in this review contain associated uncertainties (57% of data published since 2010). Mostly, the reported uncertainties vary between 2 and 8% from source to source, with maximum values in the range of 10–60%. Considering that SA by RM is based upon chemical measurements, which often have higher uncertainties than 2–8% and considering that partial fulfilment of the intrinsic assumptions for RM also contribute to the uncertainties of the final SA results, most of the reported uncertainties in the studies included in this review seem very optimistic and may not comprise all potential error sources such as e.g. the source profile variability. Larger uncertainty estimates are obtained

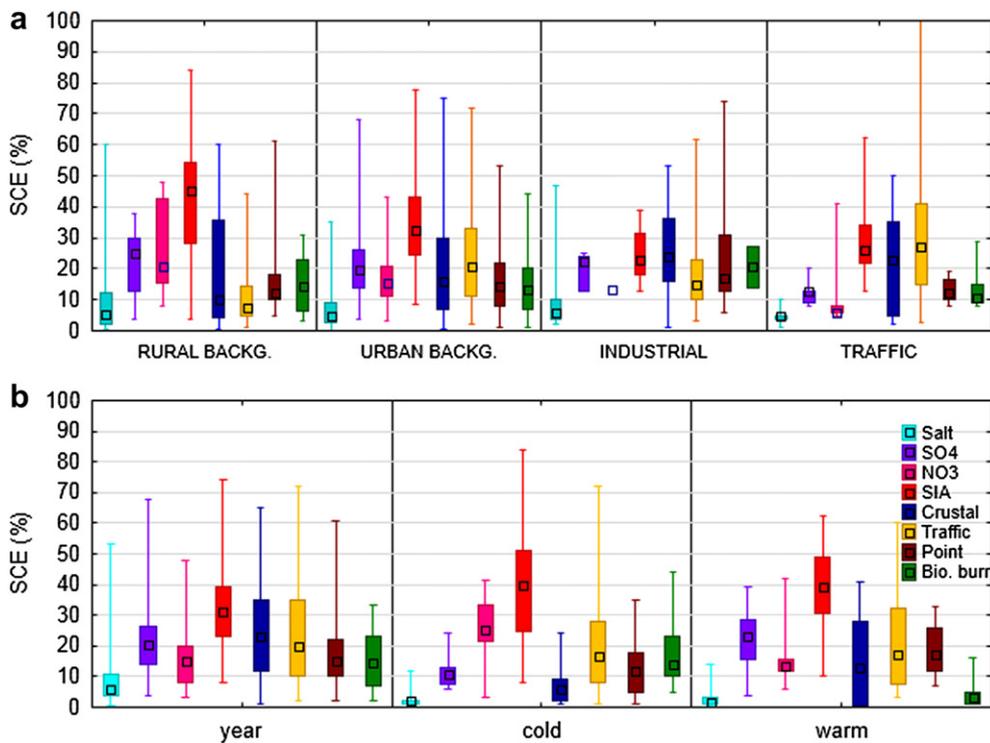


**Fig. 1.** Median, interquartile range (boxes), minimum and maximum (whiskers) relative contributions from each source category to: (a) total PM mass, and (b) PM<sub>10</sub> and PM<sub>2.5</sub> in Europe (all records). Salt: sea/road salt, SO<sub>4</sub>: ammonium sulphate, NO<sub>3</sub>: ammonium nitrate, SIA: secondary inorganic aerosol (SO<sub>4</sub> + NO<sub>3</sub>), Crustal: re-suspended dust, Traffic: mobile sources, Point: point sources, Bio. burn.: biomass burning.

when SA results from the same site with different RMs are compared (Belis et al., 2011; Junninen et al., 2009).

Uncertainties deriving from most PMF and CMB studies include only the measurement error and the quality of the fit to Eq. (1). To

account for the uncertainty associated with selection of fitting species and source profiles, some authors have produced results for a number of different CMB models, each of which is fit to the entire dataset using a different combination of source profiles and fitting



**Fig. 2.** Median, interquartile range (boxes), minimum and maximum (whiskers) relative contributions from each source category to PM in Europe split (a) by site type and (b) by season. Salt: sea/road salt, SO<sub>4</sub>: ammonium sulphate, NO<sub>3</sub>: ammonium nitrate, SIA: secondary inorganic aerosol (SO<sub>4</sub> + NO<sub>3</sub>), Crustal: re-suspended dust, Traffic: mobile sources, Point: point sources, Bio. burn.: biomass burning.

species (Subramanian et al., 2007). Other approaches are to use randomisation tests like the latin hypercube sampling (Gelencsér et al., 2007) or Monte Carlo permutations (Gilardoni et al., 2011). In addition, a quantitative evaluation of the output uncertainty with bootstrap methods is standard in US-EPA PMF 3, but allows only a partial evaluation of the rotational uncertainty (Paatero et al., 2002). A new tool to improve this step is at an advanced development stage (Paatero, pers. comm.).

Error propagation and probabilistic uncertainty with Monte Carlo simulations have been applied to estimate the uncertainty of combined CMB, PMF, and EF outputs (Larsen et al., 2012). Owing to the lack of uncertainty data for most of the studies included in the present review, such computations cannot be carried out and attempts of estimating the uncertainties of the relative SCEs from the six major source categories have not been done. Thus, in the following the data is given as median  $\pm$  half of the interquartile range (difference between 3rd and 1st quartiles).

**Sea/Road Salt** includes profiles characterized by Na, Cl and Mg that can be attributed either to sea salt in areas close to the coast or to road salt in continental areas of Central and Northern Europe. Since these two sources are not clearly distinguished in the areas where both of them are likely to contribute to PM they have been pooled into a single category. This source category is reported in more than half of the monitoring sites ( $n = 133$ , 55%). Although SCEs may be high at specific sites (Fig. 2), this source category is on average the lowest contributor to the PM mass concentrations ( $5 \pm 3\%$ ) among the six source categories (Fig. 1). In urban areas it is higher for  $PM_{10}$  ( $7 \pm 4\%$ ) than for  $PM_{2.5}$  ( $4 \pm 3\%$ ;  $p = 0.015$ ) in agreement with previous studies that report NaCl from road salting in the coarse PM fraction and in the super-micrometric fraction for marine aerosol (Peré-Trepat et al., 2007). Cities on the Atlantic coast show contributions higher than those on the Baltic coast (Fig. 5).

**Crustal/Mineral Dust** is characterized by elements abundant in the earth's crust rocks and the soils like: Al, Si, Ca and Fe. This

component of the PM is associated with the re-suspension from fields or bare soils by local winds. Also long-range transport during Saharan dust events is frequently reported in Mediterranean countries (Querol et al., 2008; Sillanpää et al., 2005).

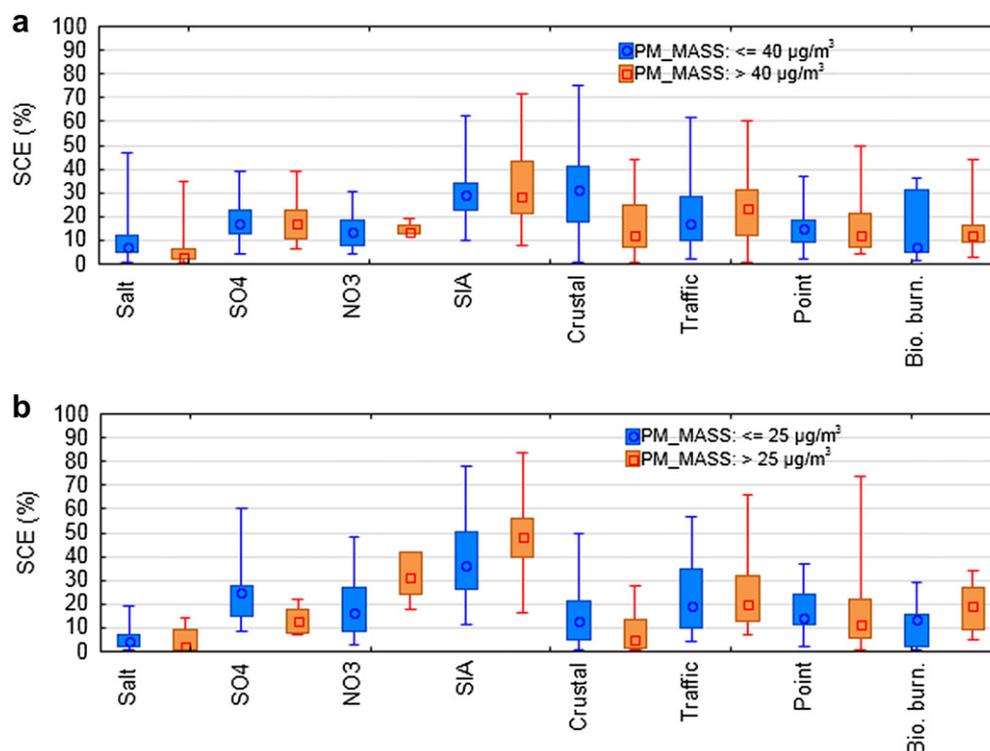
In order to assess the contribution of traffic as a whole, in this review road dust, when reported, is included in the traffic source category. Even though, re-suspension of natural soils and road-dust re-suspended by vehicular traffic can be distinguished using markers of brake, tyre, and road wear (or analysing the diurnal profiles), separating these two contributions may be challenging. Such collinearity should be taken into account when attributing and interpreting the uncertainty in mineral dust and traffic source categories.

The contribution of crustal/mineral dust to PM is reported for 77% of the monitoring sites ( $n = 188$ ). On average,  $17 \pm 12\%$  of the PM derives from this source category, which is the second highest contributor to  $PM_{10}$  and the second to last for  $PM_{2.5}$  (Fig. 1). In urban sites, its' contribution is much higher for  $PM_{10}$  ( $24 \pm 11\%$ ) than for  $PM_{2.5}$  ( $9 \pm 8\%$ ;  $p < 0.001$ ). The highest contributions to  $PM_{2.5}$  are reported in Harwell (UK) with yearly mean values close to 50% and in Antwerp (BE) with values around 41% during summer.

The median contribution of crustal/mineral dust to PM varies from  $24 \pm 10\%$  at industrial sites over  $23 \pm 14\%$  at traffic sites and from  $16 \pm 11\%$  at urban sites to  $10 \pm 16\%$  at rural sites. The high levels for industrial sites may be explained by: the presence of milling process, spills from piled materials exposed to the wind, and re-suspension by lorries circulating in unpaved areas.

A marked seasonal trend is evident for this source category ( $p < 0.001$ ) with lower contribution during the cold season consistent with frequent periods of thermal inversion, low wind speeds and snow cover typical of intermediate latitudes (Fig. 2).

There is a considerable average relative contribution from crustal/mineral dust to  $PM_{10}$  in the Mediterranean region (Fig. 5a) due to Saharan events as well as by local re-suspension.



**Fig. 3.** Median, interquartile range (boxes), minimum and maximum (whiskers) relative contributions from each source category at PM concentrations below and above the limit value (Directive 2008/50/EC): (a)  $PM_{10}$  (annual average limit value:  $40 \mu\text{g m}^{-3}$ ) and (b)  $PM_{2.5}$  (annual average limit value:  $25 \mu\text{g m}^{-3}$ ).

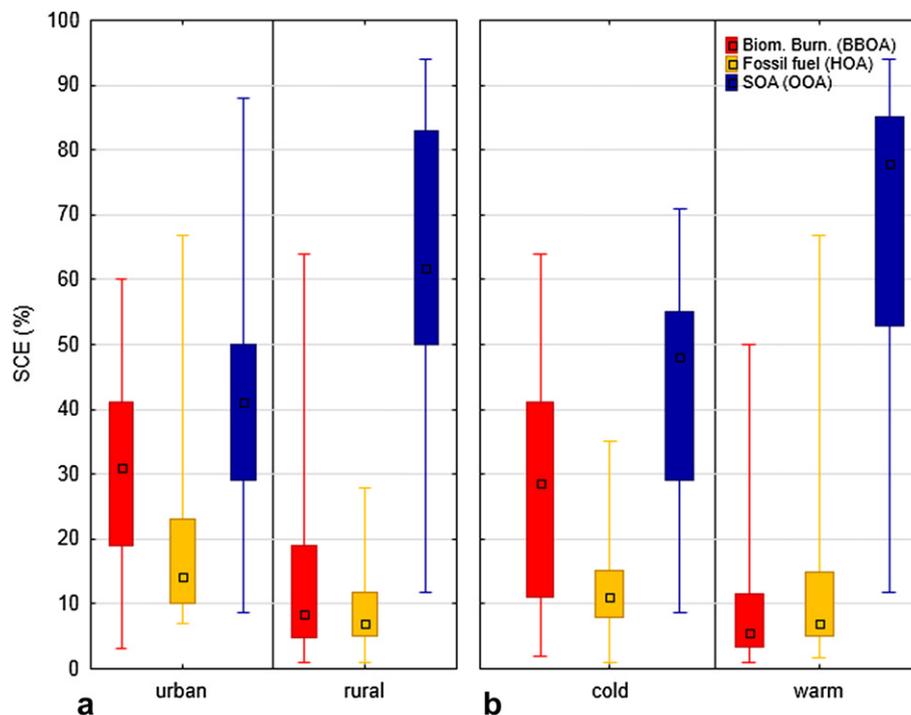


Fig. 4. Median, interquartile range (boxes), minimum and maximum (whiskers) relative contributions from each source category to organic carbon (OC) split by (a) site type and (b) by season. Biom. Burn. (BBOA): biomass burning, Fossil fuel (HOA): fossil fuel/hydrocarbon organic aerosol, SOA (OOA): secondary organic aerosol/oxidized organic aerosol.

**Secondary inorganic aerosol (SIA)** is mainly composed of ammonium- sulphate and nitrate deriving primarily from the gaseous precursors ammonia ( $\text{NH}_3$ ), sulphur dioxide ( $\text{SO}_2$ ), and nitrogen oxides ( $\text{NO}_x$ ). The sources of these gaseous precursors are not apportioned in the studies included in the present review because the RMs applied cannot deal with sources for compounds that are not present in significant concentrations in primary emissions. In CMB studies, all ammonium- sulphate and nitrate are assumed to be SIA with the risk of overestimating the contribution from this “source”. In factor analysis, the source with a chemical composition including OC and dominated by the mentioned species is often referred to as sulphate, nitrate or SIA.

$\text{NH}_3$  mainly derives from emissions from farming activities (breeding and the use of synthetic N-fertilizers) (EEA, 2011). Nevertheless, in urban areas, catalysed gasoline engines may also constitute a relevant source of  $\text{NH}_3$ .  $\text{NO}_x$  are emitted in combustion processes and emission inventories attribute ambient air levels mainly to road transport, energy production, and energy use by industry; the contribution from the first being predominant in most urban areas (EEA, 2011).

The main sulphate precursor ( $\text{SO}_2$ ) is emitted in the studied areas by the combustion of sulphur containing liquid and solid fuels (e.g. fuel oil and coal). However, sulphate may also derive from dimethyl sulphide (DMS) emitted in the oceans (Seinfeld and Pandis, 2006). Examples of primary sulphate sources are known such as sea salt, gypsum dust particles and to some extent exhaust PM from combustion of sulphur-rich fuels in diesel engines on ships. Therefore, in urban areas next to harbours, the sources characterized as “SIA” may contain a non-negligible fraction of primary sulphate (Dominguez et al., 2008; Moldanová et al., 2009).

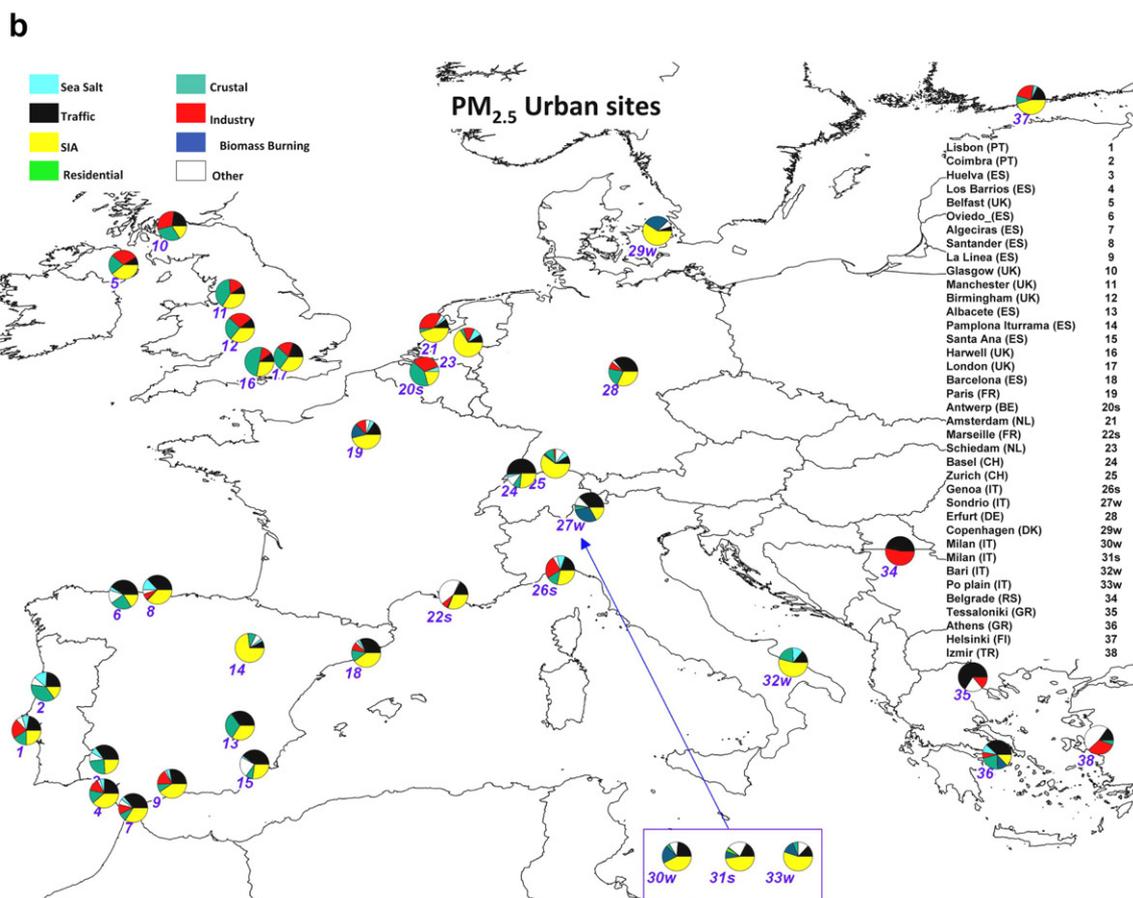
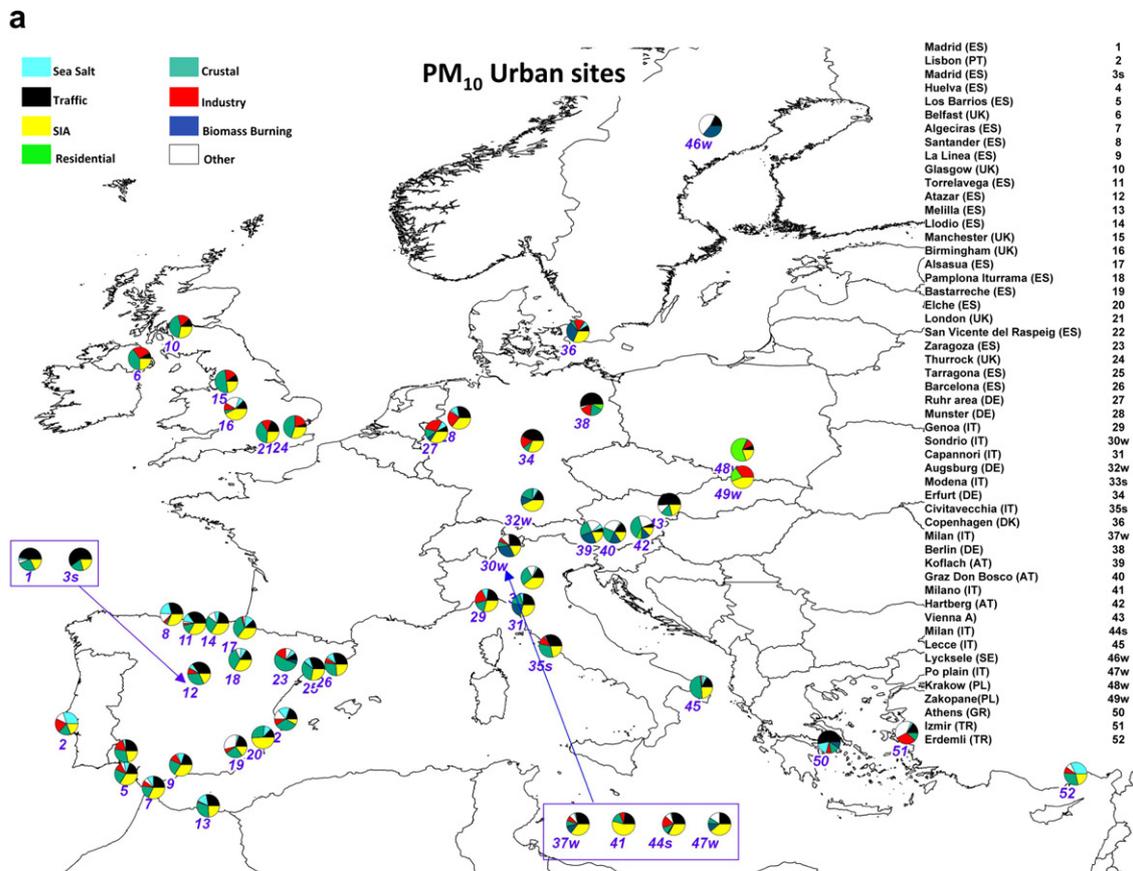
In most cases PMF and PCA distribute ammonium- sulphate and/or nitrate into profiles associated with other components (dust minerals, NaCl, soot or metals deriving from point sources such as V and Ni), which has led to the attribution of these profiles as “long-range transport”, “marine sources”, “dust”, or “combustion”. Since

the residence time of sulphates and nitrates in the atmosphere is between 3 and 9 days (Lelieveld et al., 1998; Seinfeld and Pandis, 2006), it is not surprising to find these compounds associated with species deriving from aerosol ageing (regional background emissions).

In this section, sulphate and nitrate contributions are discussed only for the studies that report them separately (about one third), whereas these species are discussed as a single source category (SIA) for the studies with ammonium sulphates and nitrates lumped together (Figs. 1 and 2).

Sulphate is reported in 87 records and constitutes  $20 \pm 7\%$  of the PM mass concentrations, while nitrate is present in 72 records and constitutes  $16 \pm 6\%$ . In general, the relative sulphate contributions to PM are highest during the warm season ( $p = 0.005$ ) explained by the combination of a faster photochemistry and a lower wet deposition during that part of the year (Ménégoz et al., 2009). Contributions in industrial areas are not very different from those observed in urban and traffic sites. On the other hand, the relative contribution from nitrate is highest during the cold season when they overtake sulphates. Increased emissions during winter from combustion of fuels not containing sulphur may explain this fact in some regions (e.g. methane and biomass burning for house heating). However, also atmospheric chemistry and physics in winter compared to summer may contribute e.g.  $\text{NO}_2$  winter photochemistry more relevant than the one of  $\text{SO}_2$  and a more favourable partition of ammonium nitrate into the particle phase at lower temperatures.

As a whole, SIA is reported in 86% of the studied sites ( $n = 208$ ) and constitutes on average  $33 \pm 11\%$  of PM mass concentrations (Fig. 1). As expected from the scarce presence of primary sources in rural areas, the median relative contribution from SIA is higher here ( $43 \pm 14\%$ ) than in industrial sites ( $23 \pm 6\%$ ;  $p < 0.009$ ) and urban sites ( $33 \pm 9\%$ ;  $p < 0.05$ ). Relative SIA contributions are higher for  $\text{PM}_{2.5}$  than for  $\text{PM}_{10}$  ( $p < 0.001$ ) and regardless of the mass fraction considered (Fig. 1), it is by far the main component of PM. In urban



**Fig. 5.** a. Contributions to PM<sub>10</sub> mass (%) in urban background sites in Europe. The pie charts represent the average relative contribution of the main source types. The sites are reported on the right. b. Contributions to PM<sub>2.5</sub> mass (%) in urban background sites in Europe. The pie charts represent the average relative contribution of the main source types. The sites are reported on the right.

areas, the median relative contribution is  $40 \pm 11\%$  to  $PM_{2.5}$  and  $30 \pm 6\%$  to  $PM_{10}$ , which can be explained by the previously discussed higher relative contributions from crustal/mineral dust and road (sea) salt to  $PM_{10}$ . The highest SIA shares are found in the Po Plain and the Netherlands (Fig. 5).

**Traffic** is a source category that encompasses different kinds of emission deriving from many different vehicle types and associated processes. In addition to the primary PM emissions from exhaust and the emissions of organic and inorganic gaseous PM precursors from the combustion of fuel and lubricant, vehicles emit significant amounts of particles through the wear of brake linings, clutch, and tyres. These are deposited onto the road and subsequently re-suspended by vehicle traffic together with crustal/mineral dust particles and road wear material. Traffic source profiles contain elemental carbon, Fe, Ba, Zn, Cu and Pb (Viana et al., 2008b) as well as hopanes and steranes that can be used in RMs to distinguish exhaust from gasoline and diesel powered engines together with the certain OC fractions (Watson et al., 2008). Cu, Zn, Mn, Sb, Sn, Mo, Ba, and Fe are markers of brake wear and can serve as indicators of traffic re-suspension (Amato et al., 2010a; Schauer et al., 2006).

Similarly to SIA, there are studies in which different traffic sources are treated separately and others in which generic terms like “traffic”, “vehicle emissions”, “road”, “mobile sources”, have been used with no detailed explanations of the considered emissions. In order to form an overview of the primary traffic emissions as a whole, all of the published SCEs (mainly exhaust and road dust re-suspension including brake- and tyre wear) are incorporated in the following into the Traffic source category.

The sources in this category have been reported for 89% of the sites ( $n = 216$ ). The median relative contribution from traffic to PM is  $19 \pm 11\%$ . Traffic is the third most intense source for  $PM_{10}$  and the second for  $PM_{2.5}$  in Europe (Fig. 1). In urban sites (Fig. 2), the median relative contribution from traffic to  $PM_{10}$  ( $23 \pm 9\%$ ) is not significantly higher than the one to  $PM_{2.5}$  ( $21 \pm 12\%$ ). Unsurprisingly, there is no clear geographical pattern in the traffic contributions to PM. However, as expected, rural sites display relative contributions ( $7 \pm 4\%$ ) significantly lower than urban ( $22 \pm 11\%$ ;  $p < 0.001$ ) and traffic sites ( $27 \pm 12\%$ ;  $p < 0.001$ ). Annual PM concentrations are dominated by traffic in many European cities (Fig. 5).

**Point sources** is a rather heterogeneous category including mainly emissions from oil combustion in power plants together with emissions from different types of industry emissions (petrochemical, metallurgic, ceramic, pharmaceutical, IT hardware, etc.) and from harbour areas. The sources are sometimes mixed with unidentified combustion sources or traffic.

The V and Ni ratios together with sulphate have been used as oil combustion or ship emissions tracers (Viana et al., 2008b), while Sc, Se, Co and PAHs have been used as tracers for coal combustion (Chow and Watson, 2002). Metallurgic industries release specific metals like Cu, Fe, Mn, or Zn and examples of tracers for non-metallurgic industries are As, Pb, Zn, Zr, Tl and Cs for frit production (Minguillón et al., 2007) as well as Cr, Ni, and Mo for pigment production (Viana et al., 2006).

The sources in this category are present in 55% of the records ( $n = 134$ ) and their median relative contribution to PM is  $15 \pm 6\%$ , which makes point sources the fourth most important contributor for  $PM_{10}$  and the third most important contributor for  $PM_{2.5}$ . As expected, the median relative contribution from this source category is highest at the industrial sites. High contributions are observed in cities influenced by industrial facilities (e.g. Izmir, Belgrade) or by harbours (e.g. Antwerp, Genoa).

**Biomass burning** has recently started to cause concern due to the impact that domestic wood burning and open fires may have on

atmospheric PM levels. Until the first half of the past decade, K was the most common species used to trace biomass burning. The identification of more specific tracers for this source category such as levoglucosan (Simoneit, 2002), and the increased availability of source profiles, e.g., Hays et al. (2005) and Schauer et al. (2001), paved the way for targeting biomass burning in SA studies. The high variability in the emissions factors of levoglucosan combined with its imperfect atmospheric stability has recently been demonstrated to induce high uncertainties in the SCE from biomass/wood burning, especially when this compound is used as single marker as in EF analysis (Belis et al., 2011; Piazzalunga et al., 2011).

Although this source type is present in only 37% ( $n = 89$ ) of the records, the number of studies reporting the contribution of biomass burning to PM is continuously growing. The mentioned uncertainty of the emission factors for the different types of fuels and combustion conditions and the scarcity of sites in which biomass burning has been identified permit only a preliminary assessment of the role of this source at the European scale. As expected, the relative contribution of biomass burning to  $PM_{2.5}$  ( $15 \pm 7\%$ ) is slightly higher than to  $PM_{10}$  ( $12 \pm 5\%$ ) although not statistically significant (low number of studies available). The median relative contribution of biomass burning to PM is  $14 \pm 6\%$  of PM (Fig. 1), which makes biomass burning the fifth highest contributor to  $PM_{10}$  and the fourth highest contributor to  $PM_{2.5}$  (comparable in strength to point sources). Considering that biomass burning in urban areas is associated with domestic heating, the median relative contributions during the cold season (14%) as expected are higher than those in the warmer part of the year (3%;  $p < 0.001$ ). The highest relative contributions from biomass burning are observed in the Alps and in Northern Europe (Fig. 5a,b).

### 3.1.2. Sources contribution estimates for PM during exceedances of air quality limits

An important purpose of SA studies is to identify the causes for exceedance of legislated thresholds such as the ones set in Directive 2008/50/EC for  $PM_{10}$  and  $PM_{2.5}$ . In order to get an overview of the main causes for exceedance in Europe the distribution of SCEs from the six source categories are compared for average PM concentrations below and above the annual European limit values in Fig. 3. It appears from this analysis, that efficient abatement strategies must target SIA and traffic, first of all because they amount to a large proportion of the  $PM_{10}$  and  $PM_{2.5}$  mass concentrations and secondly because their relative contributions increase significantly when PM concentrations are above the limit values. SIA (especially ammonium nitrate that in urban areas derives mainly from traffic) is more pertinent for  $PM_{2.5}$  ( $p < 0.05$ ) and traffic (including road dust) for  $PM_{10}$  ( $p < 0.001$ ). Second, Fig. 3 shows that the relative contributions from biomass burning to  $PM_{2.5}$  are significantly higher for sites that exceed the mentioned air quality limits ( $p < 0.05$ ) and abatement of this source category, which is most intense during winter, is expected to act more effectively on the 24-h air quality limits ( $50 \mu\text{g m}^{-3}$  of  $PM_{10}$ ).

### 3.2. Apportionment of the carbonaceous PM fraction

Organic matter is one of the most important components of ambient PM. In Europe it represents on average 15–26% of the  $PM_{10}$  and 22–25% of the  $PM_{2.5}$ , depending on the geographic area (Putaud et al., 2010). Due to the specificity of the methodologies to apportion OC (see Chapter 2.6), the SA results have to be analysed separately. The dataset contains 60 records deriving from 14 studies in 12 European countries (Table 2S). In more than 80% of the records, three main sources have been reported: biomass burning (including wood burning), fossil fuel combustion, and SOA. To form an overview, these categories have been considered comparable to

the AMS factors BBOA, HOA and OOA, respectively. This choice implies the assumption that OOA is mainly secondary, even though, as explained in Section 2.6, AMS factors do not distinguish between oxidized primary and secondary contributions.

Overall, SOC makes up the highest proportion of OC ( $52 \pm 17\%$ ) regardless of aerosol size fraction and site location. The median and spread of the relative contribution from biomass burning ( $20 \pm 17\%$ ) are higher than the one from fossil fuel combustion ( $11 \pm 5\%$ ). The proportion of SOC is higher in  $PM_1$  than in  $PM_{10}$  ( $p < 0.05$ ). This result is consistent with formation by atmospheric photochemical oxidation of gaseous precursors (followed by nucleation, coagulation and condensation) and also confirmed by the seasonal variation of the SOC proportion of OC, which is higher during summer ( $p < 0.01$ ; Fig. 4). Unsurprisingly, the opposite seasonal trend is seen for biomass burning, which point to domestic heating as a main source ( $p < 0.01$ ; Fig. 4). No significant seasonal trend can be observed in the contribution from fossil fuel combustion, which may indicate that traffic exhaust is a main source for this fraction.

In agreement with some studies, SOC prevails in rural sites ( $p < 0.001$ ) while OC deriving from primary sources (fossil fuel and biomass burning) is higher in urban sites, where there is a higher density of sources ( $p < 0.01$ ; Fig. 4). In the literature there is contrasting evidence on this point. Sheesley et al. (2004) claim OC in urban areas is mainly deriving from primary sources, especially in winter, while studies conducted with AMS suggest that SOA dominates OC levels, even in urban areas (Zhang et al., 2007).

#### 4. Conclusions and recommendations

In Europe, source apportionment of PM and its organic fractions has been conducted over the past two decades with a variety of receptor models shifting from principal component analysis techniques, enrichment factors and classical factor analysis towards models able to handle uncertainties on the input and output such as e.g. Positive Matrix Factorization. A wider use of advanced factor analysis techniques able to deal with heterogeneous and complex data and to provide improved uncertainty estimations should be promoted. On the other hand, PCA technique should be preferably used for qualitative or preliminary estimations. The Chemical Mass Balance model is still topical and has gained new impetus from the application of molecular markers for the apportionment of the carbonaceous fraction. Nevertheless, the scarcity of measured source profiles for European sources and the lack of long term, speciated PM series, especially in urban areas, should be dealt with at the earliest to remove the limiting factors that hamper the development of receptor model studies in Europe. The definition and documentation of the source categories in Europe has improved swiftly but there is still a need of harmonization of the different approaches in order to facilitate the interpretation and comparability of the results and their application in the design of abatement measures. In particular, more efforts and methods are needed to estimate and constrain the uncertainties of the resulting source contribution estimates and more studies are required with a widespread geographical distribution to improve the estimations of biomass burning contribution to PM.

Gaseous precursors emitted by combustion sources (in most urban areas traffic is the dominating source for nitrogen oxides) and agriculture undergo gas-to-particle conversion. The results of the present meta-analysis stress that such atmospheric process is the strongest source for PM mass concentrations over Europe (SIA) and the strongest contributor to the organic PM fraction (SOA). The results demonstrate that over Europe, most sources contribute equally to  $PM_{10}$  and  $PM_{2.5}$  except for crustal/mineral dust resuspension and sea/road salt, which prevail in the coarse fraction.

In order to abate exceedance of air quality limits, SIA and traffic are the most important source categories to target throughout the year together with biomass burning during the cold season.

#### Appendix A. Supplementary material

Supplementary material related to this article can be found at <http://dx.doi.org/10.1016/j.atmosenv.2012.11.009>.

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