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Source apportionment of particulate matter in Europe: A review of methods and results

Review

M. Viana^{a,r,*}, T.A.J. Kuhlbusch^b, X. Querol^{a,r}, A. Alastuey^{a,r}, R.M. Harrison^c, P.K. Hopke^d, W. Winiwarter^{e, f}, M. Vallius^g, S. Szidat^h, A.S.H. Prévôtⁱ, C. Hueglin^j, H. Bloemen^k, P. Wåhlin^l, R. Vecchi^m, A.I. Mirandaⁿ, A. Kasper-Giebl^o, W. Maenhaut^p, R. Hitzenberger^q

^aInstitute for Environmental Diagnosis and Water Studies (IDAEA-CSIC), Barcelona, Spain ^bIUTA e.V., Duisburg, Germanv ^cSchool of Geography, Earth and Environmental Sciences, University of Birmingham, Birmingham, UK ^dDepartment of Chemical and Biomolecular Engineering, Clarkson University, Potsdam, NY, USA ^eAustrian Research Centers—ARC, Vienna, Austria ^fInternational Institute for Applied Systems Analysis, Laxenburg, Austria ^gDepartment of Environmental Health, National Public Health Institute, Kuopio, Finland ^hDepartment of Chemistry and Biochemistry, University of Bern, Bern, Switzerland ⁱLaboratory for Atmospheric Chemistry, Paul Scherrer Institute (PSI), Villigen-PSI, Switzerland ^jLaboratory for Air Pollution and Environmental Technology, Swiss Federal Laboratories for Materials Testing and Research (EMPA), Dübendorf, Switzerland ^kLaboratorium voor Milieumetingen, RIVM, Bilthoven, The Netherlands ¹National Environmental Research Institute (NERI), Aarhus University, Roskilde, Denmark ^mInstitute of General Applied Physics, University of Milan, Milan, Italy ⁿDepartment of Environment and Planning, University of Aveiro, Aveiro, Portugal ^oInstitute for Chemical Technologies and Analytics, Vienna University of Technology, Vienna, Austria ^pDepartment of Analytical Chemistry, Institute for Nuclear Sciences, Ghent University, Gent, Belgium ^qAerosol, Bio- and Environmental Physics, University of Vienna, Vienna, Austria ^rInstitute of Earth Sciences "Jaume Almera"-CSIC, Barcelona, Spain

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Abstract

European publications dealing with source apportionment (SA) of atmospheric particulate matter (PM) between 1987 and 2007 were reviewed in the present work, with a focus on methods and results. The main goal of this meta-analysis was to provide a review of the most commonly used SA methods in Europe, their comparability and results, and to evaluate current trends and identify possible gaps of the methods and future research directions. Our analysis showed that studies throughout Europe agree on the identification of four main source types (PM₁₀ and PM_{2.5}): a vehicular source (traced by carbon/Fe/Ba/Zn/Cu), a crustal source (Al/Si/Ca/Fe), a sea-salt source (Na/Cl/Mg), and a mixed industrial/fuel-oil combustion (V/Ni/SO₄²⁻) and a secondary aerosol (SO₄²⁻/NO₃⁻/NH₄⁺) source (the latter two probably representing the same source type). Their contributions to bulk PM levels varied widely at different monitoring sites, and showed clear spatial patterns in the cases of the crustal and sea-salt sources. Other specific sources such as

* Corresponding author. Fax: +34934110012.

E-mail address: mviana@ija.csic.es (M. Viana).

biomass combustion or shipping emissions were rarely identified, even though they may contribute significantly to PM levels in specific locations.

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Keywords: PM10; PM2.5; Emission sources; Receptor modelling; Tracers; Research directions; Shipping emissions; Biomass burning

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1. Introduction

In the field of atmospheric sciences, source apportionment (SA) models aim to re-construct the impacts of emissions from different sources of atmospheric pollutants, e.g., particulate matter (PM), based on ambient data registered at monitoring sites (Bruinen de Bruin, Koistinen, Yli-Tuomi, Kephalopoulos, & Jantunen, 2006; Hopke & Song, 1997; Watson et al., 2002). There are three main groups of SA techniques:

- (a) Methods based on the evaluation of monitoring data. Basic numerical data treatment is used to identify sources. Examples are: (1) correlation of wind direction with levels of measured components to identify source locations (Henry, Chang, & Spiegelman, 2002); (2) the correlation of gaseous pollutants with PM components to identify source associations; (3) subtraction of levels measured at regional background from those obtained at urban background and/or roadside levels to identify the contributions from the regional background, the city background and the monitored street (Lenschow et al., 2001), or (4) quantification of natural PM contributions (e.g., African dust) by subtracting PM levels at regional background sites from those at urban background locations for specific days (Escudero et al., 2007). The main advantage is the simplicity of the methods and the consequent low impact of mathematical artefacts due to data treatment.
- (b) Methods based on emission inventories and/or dispersion models to simulate aerosol emission, formation, transport and deposition (Eldering & Cass, 1996; Visser, Buring, & Breugel, 2001). These models require detailed emission inventories that are not always available, and they are limited by the accuracy of emission inventories, especially when natural emissions are important. A significant advantage of these methods is that they may be used in scenario studies to evaluate the impact of emission abatement strategies on the anthropogenic contribution to ambient PM concentrations.
- (c) Methods based on the statistical evaluation of PM chemical data acquired at receptor sites (receptor models). The fundamental principle of receptor modelling is that mass and species conservation can be assumed and a mass



Fig. 1. Approaches for estimating pollution source contributions using receptor models (modified from Schauer et al., 2006). Specific models are shown in italics and with dotted arrows.

balance analysis can be used to identify and apportion sources of airborne PM in the atmosphere (Hopke et al., 2006). An overview of the wide range of statistical models and modelling approaches which are currently available in the literature is shown in Fig. 1. As shown in the graph, one of the main differences between models is the degree of knowledge required about the pollution sources prior to the application of receptor models. The two main extremes of receptor models are chemical mass balance (CMB) and multivariate models.

The CMB model assumes knowledge of the composition of the emissions for all relevant sources. If changes of the source profiles between the emitter and the receptor may be considered minimal, CMB can be regarded as the ideal receptor model. However, these requirements are almost never completely fulfilled, and thus, pure CMB approaches are often problematic. One important characteristic of CMB is that secondary aerosols must be included not as components of emission source profiles but as specific, single chemical compounds. This absence of mixture with other tracer elements is often regarded as a limitation, and may lead to misinterpretation of results.

Principal component/factor analysis (e.g., principal component analysis or PCA, positive matrix factorisation or PMF, UNMIX) attempts to apportion the sources on the basis of observations (internal correlations) at the receptor site alone. These are commonly used tools, because software to perform this type of analysis is widely available and detailed prior knowledge of the sources and source profiles is not required. The choice of the model dimension and the search for non-negative solutions by axis rotations can be based entirely on mathematical criteria. Nevertheless, it has been suggested that factor analysis attempts to get more information out of atmospheric data than is really there (Henry, 1987). Furthermore, it is a common problem that the resulting components or factors may represent mixtures of emission sources, as opposed to clearly independent source profiles. Source signatures that change with time are a limitation for this and other types of receptor models.

To combine the advantages and reduce the disadvantages of CMB and factor analysis hybrid models have been developed. Examples are confirmatory or target transformation factor analysis, which offer some control of the solutions by "fixing" or "freeing" specific parameters, set according to the theoretical expectation of the researcher (Gleser, 1997; Hopke, 1988). With a constrained physical receptor model (COPREM, Wåhlin, 2003), an initial profile matrix with the main characteristics of known sources as columns is used, and a priori knowledge about the character of the sources can be used to achieve a solution with a sufficient number of sources. The multilinear engine (ME, Paatero, 1999) can solve multilinear problems with the possibility of implementing many kinds of constraints using a script language. Like COPREM, the program allows choosing hybrid versions in the full range between PMF and CMB-type models, with the difference that individual data points can be properly weighed (not possible in any eigenvector analysis).

The main objective of receptor models is, therefore, to identify the possible sources of PM (if not assumed already from the source profiles) and to obtain data on their contributions to the bulk PM mass. Even human exposure to these pollution components has been evaluated to assess their health effects and risks (Hopke et al., 2006; Ilacqua et al., 2007; Watson et al., 2002). Furthermore, policy-makers require sound scientific knowledge of the PM sources and their contributions to atmospheric PM levels and associated health risks for the development and implementation of policies to protect human health and the environment. Thus the information provided by receptor models is key to the design of effective mitigation strategies on the local- and meso-scale.

Despite the widespread need for these data, there is little information available on receptor modelling results from different European countries, the type of models applied or the input data utilised. Furthermore, parameters such as time resolution, type of monitoring site and PM sampler, and analytical methods vary widely from one study to another. Consequently, general European-wide conclusions cannot yet be extracted. In addition to this lack of harmonisation in the published studies, a large amount of valuable material is currently available in the grey literature only.

The need to tackle this issue was detected by the COST Action 633 ("Particulate matter: Properties related to health effects") and subsequently addressed by compiling a database of SA studies available in Europe, more specifically in COST633 member countries (http://www2.dmu.dk/atmosphericenvironment/COST633/). Both meta-data on publications and data on PM sources and source contributions were collected, with the aim to obtain an overview of the results and analyse their comparability. The issue of the comparability between receptor modelling results is considered essential if SA data are to be used for the design of multinational mitigation strategies, the assessment of human exposure to PM from specific sources, and investigations of long-term changes in exposure situations over Europe.

2. Methodology

The compilation of meta-data on SA publications was carried out by means of a questionnaire, distributed via e-mail among researchers from COST633 member countries and based on already existing publications in international scientific journals or public reports. The authors were asked to report on meta-data such as the receptor modelling technique used, the location of the study area, the type of sampler or the analytical methodology used, among others.

Despite the fact that replies to the questionnaires were not obtained from all the member countries, the return may be seen as a useful overview on SA activities in Europe from 1987 up to the end of the year 2005 (date of meta-data collection). Furthermore, a literature search was carried out by the authors for SA-related papers published in 2006 and 2007 (thus extending the evaluation period to 1987–2007), the results of which are also included in this paper (Table 1). It should be noted, however, that this meta-analysis does not claim to provide an overview of all SA studies performed in Europe or the COST633 Action member countries.

3. Results and discussion

3.1. Receptor models in use in Europe

When evaluating the European publications reported in the questionnaires, PCA was the most frequently used model up to 2005 (30% of the studies), followed by the Lenschow approach (11%) and back-trajectory analysis (11%). Other models commonly used were PMF (8%), CMB (7%) and mass balance analysis (addition of chemically analysed PM components, 7%). Methods such as ME, COPREM or UNMIX were only reported in one study each until 2005. Data from 2006–2007 show a continued use of PCA (50% of the new publications) and an increase in the use of PMF (13%) and the mass balance analysis of chemical components (19%). No new publications applying CMB or the Lenschow approach in Europe were obtained for 2006–2007.

With regard to the number of research teams in Europe applying each of these receptor modelling techniques, PCA was once again the most widespread model (in use by eight different research teams), followed closely by the Lenschow approach (seven research teams) and CMB (five). Even though a relatively large number of publications were reported using back-trajectory analysis (8 publications by 2005), this approach was used by a relatively low number of research teams in Europe (three). Researchers from a number of countries (Belgium, Germany, Portugal and Spain) reported the application of more than one receptor model, the most frequent combinations being PCA-cluster analysis (CA), PCA-Lenschow, PCA-CMB, PCA-back-trajectory analysis, PMF-UNMIX-ME, and CMB-mass balance.

Table 1

Publications reported in the questionnaires (up to year 2005, and unpublished data at that point in time) and results from the literature search (marked with *, published in 2006–2007)

Results from questionnaires and literature search

Results from questionnanes i	and interature search			
Alastuey et al. (2006); Almeida, Pio, Freitas, Reis, and Trancoso (2005) ¹²	Harrison, Smith, Pio, and Castro (1997) ^{16,33}	Maenhaut and Cafmeyer (1998) ^{8,9}	Quass, Kuhlbusch, and Koch (2004) ¹⁴	Vallius et al. (2003) ³²
Almeida, Pio, Freitas, Reis, and Trancoso (2006) ¹²	Harrison, Jones, and Lawrence (2003) ¹⁶	Maenhaut et al. (1999) ^{8,9}	Querol et al. $(2001)^3$	Vallius et al. (2005) ^{28,29}
*Andersen, Wåhlin, Raaschou-Nielsen, Scheike and Loft (2007) ¹⁷	Harrison, Jones, and Lawrence (2004) ¹⁶	Maenhaut (2000)	Querol et al. (2002) ⁴	Viana, Querol, and Alastuey (2006) ⁶
Bauer et al. (2007)	Hueglin et al. (2005)	Marcazzan, Vaccaro, Valli, and Vecchi (2001) ²¹	Querol et al. $(2004)^{2,5}$	Viana, Querol, and Alastuey (2006) ³
Bauer et al. (2007b)	John, Weissenmayer, Lutz, and Kuhbusch (2004)	Marcazzan, Ceriani, Valli, and Vecchi (2003) ²¹	Raes, Hubacz, Hanssen, and Maenhaut (2005) ^{30,31}	Viana, Querol, and Alastuey (2006) ⁶
Borrego and Pio (1991)	John, Lutz, and Kuhbusch (2004)	*Marenco et al. (2006) ^{23,24}	*Ragosta et al. (2006)	Viana et al. (2007) ³
Brüggemann et al. (2000)	Kaiser and Scheifinger (2003)	*Mazzei et al. (2006)	Rodríguez, Querol, Alastuey, Viana, and Alarcón (2004) ²	Viana et al. (2008) ²⁷
*Canepari et al. (2006)	Kaiser and Scheifinger (2005)	*Mazzei et al. (2007)	*Salma and Maenhaut (2006)	Wåhlin and Palmgren (2005) ¹⁷
Castro, Pio, Harrison, and Smith (1999)	Kaiser, Scheifinger, and Langer (2002)	*Minguillón et al. (2007) ²⁷	Salvador, Artíñano, Alonso, Querol, and Alastuey (2004) ¹	Wåhlin, Berkowicz, and Palmgren (2006) ¹⁷
D'Alessandro et al. (2003) ²²	Kaiser, Scheifinger, Spangl, Nagl, and Schneider (2004)	Mossetti, Angius, and Angelino (2005) ²¹	*Salvador, Artíñano, Querol, Alastuey, and Costoya (2007) ^{1,26}	Yin et al. (2005) ^{18,19,20}
D'Alessandro et al. (2004) ²²	Kuhlbusch, John, Romazanowa, Top, and Weissenmayer (2004)	Öttl, Kurz, Hafner, and Sturm (2005) ²⁵	Schnelle-Kreis, Sklorz, and Zimmermann (2006)	Zabalza et al. (2006) ⁷
Düring et al. (2003) ¹³	*Lanz et al. (2007)	Pio, Nunes, Borrego, and Martins (1989) ¹⁰	Spindler, Müller, Brüggemann, Brüggemann, Gnauk, and Herrmann (2004) ¹⁵	
Ebert, Benker, Kandler, Müller-Ebert, and Weinbruch (2007)	*Lonati, Ozgen, and Giugliano (2007)	Pio et al. (1996) ¹¹	Stölzel (2003) ¹⁵	
*Gerasopoulos et al. (2006)	Lucarelli, Mandò, Nava, Prati, and Zucchiatti (2004) ²²	*Pratesi, Zoppi, Vaiani, and Calastrini (2007)	Szidat et al. (2004a)	
Gerwig (2005) ¹⁵	Maenhaut and Cafmeyer (1987)	Prendes, Andrade, López-Mahía, and Prada (1999) ²⁶	Szidat et al. (2004b)	

Numbers in superscript indicate the study site as listed in Fig. 6a caption.



Fig. 2. (a) Target metric of SA studies reported in the questionnaires (1987–2005). (b) Time evolution of publications on SA of PM_{10} , $PM_{2.5}$ or PM_1 between 1987 and 2005.

3.2. Target metric, sampling and analytical methodologies, receptor sites

3.2.1. Target metric

Until 2005, PM_{10} was on average the preferred target metric (46% of the publications reported, Fig. 2a) followed by $PM_{2.5}$ (33%). Studies focusing on other size fractions were reported with much lower frequencies: $PM_{2.5-10}$ (9%), TSP (4%), PM_1 (3%), PM_2 (1%) and $PM_{0.1}$ (1%). Based on the year of the publications (Fig. 2b), the results indicate a slow but progressive increase in the use of PM_{10} from 1997 to 2003, whereas models were applied to $PM_{2.5}$ only after 2000 and increased sharply in 2003 (first year with more studies on $PM_{2.5}$ than PM_{10}). In 2006 and 2007 (not included in Fig. 2) 38% of the new studies found in the literature targeted $PM_{2.5}$ while only 29% focused on PM_{10} , thus confirming a change in focus in SA studies in Europe. This focus on the sources of fine aerosols is most probably related to stronger recent evidence on the adverse effects of fine particulates on health (Lanki et al., 2006; Pope & Dockery, 2006) as opposed to coarser particles. As a result, an increase in the number of papers published from Europe on smaller particle size fractions ($PM_{2.5}$, PM_1) is likely in the coming years. Such studies from the US are already available in the literature (Pancras, Ondov, Poor, Landis, & Stevens, 2006). The detailed SA of very fine grain size fractions (PM_1 and smaller) will also require the characterisation of specific tracers and chemical profiles for this fraction, given that source profiles should not be expected to be the same for coarse and fine aerosols.

3.2.2. Sampling methodology

A large variety of sampling methodologies was employed throughout Europe. The choice of method influences the PM and chemical data due to artefacts (magnitude and type, Turpin, Saxena, & Andrews, 2000). High-volume samplers

(typically > $30 \text{ m}^3/\text{h}$) were used in the majority of the SA studies reported (43%), with 24 h as the standard sampling period (66% of the total). This duration was the most commonly used as it allows for the comparison with the daily limit value for PM concentrations. Longer sampling durations (48 h) were also reported, although less used (5%) due to the larger influence of (mostly negative) sampling artefacts. The second widespread method of sampling was for 24 h with a low-volume sampler (typically $1 \text{ m}^3/\text{h}$, 21%) both in terms of number of studies and of groups applying this methodology. Semi-hourly automatic data represented 10% of the studies reported, mostly used in combination with back-trajectory data, and were essentially used by two research teams. In the last two years (2006–2007), the number of studies using 24 h low-volume samplers increased (31% of the total, five new studies) while there were only two new studies (13%) using high-volume samplers. The use of new sampling and monitoring methodologies (e.g., streaker sampler, particle number, particle size distribution, etc.) were reported for 50% of the 2006–2007 studies.

Factor analysis methods depend on having a series of samples containing material from the same set of sources in differing proportions. Increasing the time resolution of the measurements will typically provide samples that have greater between-sample variability in the source contributions than samples integrated over longer time periods. Lioy, Zelenka, Cheng, Reiss, and Wilson (1989) showed that one could resolve more sources with principal components analysis if one had more highly time resolved data. The US EPA Supersites Program provided such highly time resolved data that has permitted the increased resolution of sources in Pittsburgh PA (Zhou, Hopke, et al., 2004; Zhou, Kim, Hopke, Stanier, & Pandis, 2005) and Baltimore, MD (Ogulei, Hopke, & Wallace, 2006; Ogulei et al., 2005, 2006). In some of these analyses, combined data on particle composition, size distributions, and gaseous constituents were analysed together, allowing identification of atmospheric processes such as nucleation in addition to major source types (Ogulei, Hopke, & Wallace, 2006; Ogulei et al., 2006; Zhou, Hopke, et al., 2005; Zhou, Kim, Hopke, Stanier, & Pandis, 2006).

3.2.3. Analytical methodology

A broad spectrum of techniques was described. Ion chromatography (IC) was most commonly used for the determination of ionic species (22% of the studies), while major and trace elements were determined in similar proportions (9–12%) by ICP-AES (inductively coupled plasma atomic emission spectrometry), ICP-MS (inductively coupled plasma mass spectrometry), PIXE (Particle induced X-ray emission spectrometry) and XRF (X-ray fluorescence). Discrimination between organic (OC) and elemental (EC) carbon was only carried out in 5% of the studies. The low percentage of OC/EC analyses, as well as the almost complete absence of data on speciation of organic aerosols (OA) in these studies, implies an evident difficulty to detect and interpret sources of organic PM, such as different vehicular sources (e.g., diesel vs. gasoline vehicles). As novel applications, it is essential to highlight the use in the years 2006–2007 of aerosol mass spectrometry (AMS) and ¹⁴C analyses. These techniques allow for the distinction of primary and secondary organic fractions and the distinction of fossil versus non-fossil particulate carbon, respectively.

The extent and quality of the chemical data is also a determining factor. SA studies may utilise PM composition data from measurements that were not specifically designed for this purpose. As an example, the general lack of appropriate uncertainty estimates is a clear limitation when applying models such as PMF, COPREM or CMB. The missing uncertainty data must be estimated using algorithms based on detection limits or reproducibility of the results, which may result in the restriction of the potential of the models to resolve the sources. The importance of this issue has already been acknowledged (Kim & Hopke, 2007). The general lack of uncertainty data in models results in the transfer of the effect of uncertainties in the original values to unquantifiable and ambiguous uncertainties in the final SA results.

Another example is the absence of major tracer species in the compositional data sets, when certain PM components are not analysed or their analyses are not reliable enough using a given technique (e.g., Na concentrations when analysed by XRF, Götschi et al., 2005; Viana et al., 2007). Even though these tracers may be determined in parallel using correction factors or by additional techniques (in the case of Na, Maenhaut & Cafmeyer, 1998; Rhodes & Hunter, 1972), this might introduce a bias in the analysis. Sensitivity studies on the influence of the uncertainty on model results are rarely mentioned in the literature even though often studied to ensure the robustness of results.

Finally, one other key issue is the size of the data sets. The number of independent samples as well as analysed compounds defines the model's ability to resolve source profiles, and given that this number is often smaller than the optimal, many of the source profiles reported in the literature often describe a mixture of sources. Consequently, the size of the data sets deserves special attention when attempting to compare results throughout Europe.



Fig. 3. Receptor sites reported in the questionnaire (1987–2005, included): total mean (top, % and total number) and classified by country (bottom, only countries with > 5 sites reported are shown).

3.2.4. Receptor sites

Receptor sites where the models were applied also constitute a differentiating factor. As shown in Fig. 3 (top), urban background locations were clearly preferred (53% of the studies) probably due to the fact that SA studies aim mostly to represent maximum population exposure. However, the issue remains whether monitoring and/or SA studies should be carried out at locations where a smaller proportion of the population is exposed, but where PM levels and composition present potentially higher health risks, e.g., industrial or kerbside sites. These types of sites were the target of 11% and 20% of the studies reported, respectively. The opposite situation was found regarding rural sites (16%), given that rural environments are not generally representative of the population's exposure but are nevertheless extremely useful locations to investigate the origin and properties of background and long-range transported aerosols. The studies which applied the Lenschow approach included at least two site-types, given that in this method the spatial variability of PM concentrations is used.

However, this distribution is dependent on the country (Fig. 3, bottom). The dissimilarities in the distribution of monitoring site types between national networks in Europe have already been noted (Querol, Alastuey, Ruiz, et al., 2004), referring to the overall number of urban, industrial or rural sites. When looking at SA studies, these differences are again evident and reflect both the structure of national monitoring networks and living patterns such as the proximity of residential areas to heavy traffic (urban topography). Furthermore, the proportional differences could also be partly due to the still rather low total number of SA studies per country (thus proportions are sensitive), and the fact that in some countries SA studies may be more of a side-product of other type of studies (e.g., epidemiological) while in others they may have been specifically planned as SA studies, thus possibly affecting the selection of the sites.

According to the meta-data reported, > 75% of the SA studies in Austria were carried out at urban background locations, and < 15% at kerbside sites. In Germany, Italy or Spain, with similar distributions, there is a lower proportion



Fig. 4. Location of monitoring sites reported: Madrid, Spain (1); Onda, Spain (2); Barcelona, Spain (3); Huelva, Spain (4); Monagrega, Spain (5); Llodio, Spain (6); Alsasua, Spain (7); Ghent, Belgium (8); Waasmunster, Belgium (9); Estarreja, Portugal (10); Areao, Portugal (11); Lisbon, Portugal (12); Leipzig, Germany (13); Duisburg, Germany (14); Dresden, Germany (15); Birmingham, UK (16); Copenhagen, Denmark (17); Dublin, Ireland (18); Ballinsloe, Ireland (19); Cork, Ireland (20); Milan, Italy (21); Florence, Italy (22); Modena, Italy (23); Mt. Cimone, Italy (24); Klagenfurt, Austria (25); A Coruña, Spain (26); L'Alcora, Spain (27); Amsterdam, The Netherlands (28); Erfurt, Germany (29); Birkenes, Norway (30); Skreådalen, Norway (31); Helsinki, Finland (32); and Coimbra, Portugal (33).

of urban background sites (< 50%) and a larger number of kerbside (20–30%) and industrial sites (5–20%). The largest proportion of SA studies carried out at rural sites was reported for Belgium (40%).

3.3. SA results in Europe

The entire set of publications reported in the questionnaires was collected and analysed in search of the sources and source contributions described by the authors. The aim of this analysis was to compare PM sources throughout Europe (monitoring sites plotted in Fig. 4), and to evaluate the geographical variability of the common sources and the specificity (if any) of sources detected only in certain regions. The results of these analyses are plotted in Figs. 5 and 6 and summarised in Table 2a and b. The monitoring sites in Figs. 5 and 6 are ordered from south (left) to north (right) for each site type (rural, urban background, etc.). We included all reported SA if reasonable methods were applied. Including secondary particles in receptor model analysis was seen critical but not generally as wrong. Certainly care has to be taken when interpreting the results of such model approaches as this is generally valid for receptor models.



Fig. 5. Contribution to PM_{10} (a) and $PM_{2.5-10}$ (b) of the four main sources common to most European studies: carbon/Fe/Ba/Zn/Cu, Al/Si/Ca/Fe, Na/Cl/Mg, V/Ni/SO₄²⁻ and SO₄²⁻/NO₃⁻/NH₄⁺ sources. R: rural site. UB: urban background site. T: traffic site. IND: industrial site.

Overall, a generally good spatial coverage of SA studies over Europe, especially regarding the northern, south-eastern and south-western dimensions, was seen. The fact that no data are shown for certain countries (e.g., France) does not imply that no SA activities have been conducted, but rather that none were reported. Fig. 4 shows the location of the monitoring sites, both for PM_{10} (28 sites) and $PM_{2.5}$ (20 sites). The spatial coverage is thus clearly better for PM_{10} than for $PM_{2.5}$ due to the fact that fewer case studies are published for $PM_{2.5}$. Limited data were also available for $PM_{2.5-10}$ (Fig. 5b), PM_2 and $PM_{0.95}$. The discussion of the results, however, focuses on PM_{10} and $PM_{2.5}$.

The preliminary account of the sources described by the authors indicated the difficulty to compare source categories throughout Europe, given their large variability (> 20 sources and source categories were found). Furthermore, some authors reported sources which were interpreted as combinations of tracer species (e.g., a source characterised by OC, Cu and Ba and interpreted as "traffic"), whereas others reported chemical components (e.g., a source defined as "EC"). Finally, sources identified by similar labels may not include the same types of emissions (e.g., one "traffic" source may include exhaust emissions and tyre and brake wear, whereas another may refer only to exhaust emissions).





Fig. 6. Contribution to PM_{2.5} of the four main sources common to most European studies: carbon/Fe/Ba/Zn/Cu, Al/Si/Ca/Fe, Na/Cl/Mg, V/Ni/SO₄²⁻ and SO₄²⁻/NO₃⁻/NH₄⁺ sources. *: PM₂; **: PM_{0.95}. R: rural site. UB: urban background site. T: traffic site. IND: Industrial site.

Clearly, these differences limit the comparability of the results. Within the sources reported as combinations of tracer elements, different authors provided different interpretations or source labels to the same or similar underlying sources, based on informed and subjective decisions affected by each researcher's knowledge of local circumstances (e.g., meteorological phenomena or specific emission sources in the region). The most evident example of this problem is the source characterised mainly by SO_4^{2-} levels, identified in most of the studies and interpreted with such different labels as "industrial emissions", "secondary aerosols", "regional background", "transboundary pollution" or even "general pollution". This heterogeneity in source categories was also observed in the contrast between very specific (e.g., "domestic heating") and very general (e.g., "general pollution") source categories.

In order to make the data comparable across Europe the results provided by the authors were sorted and rationalised according to the tracer elements used to identify the source-related components. Therefore, sources were classified as a function of combinations of common tracer elements instead of source names or labels given by the authors of original papers. Four main sources were found to be common to most of the studies reported, both for PM_{10} and $PM_{2.5}$ (Figs. 5a and 6, respectively):

Carbon/Fe/Ba/Zn/Cu source: Interpreted as a vehicular source ranging from vehicle exhaust to traffic, combinations of its major markers were also found jointly with Br and NO₃⁻, and less frequently with Cr and Mn. If interpreted as total traffic, other tracers were crustal elements such as Fe, Ca or Al from road dust. In these cases, this source could be clearly differentiated from the Al/Si/Ca/Fe source (described below) due to the presence of trace elements linked to brake abrasion (Cu, Ba, Sb; Sternbeck, Sjödin, & Andréasson, 2002; Schauer et al., 2006) and combustion of lubricating oil (Zn, Ca). As expected, the contributions from this source showed a dependency with traffic emissions, with higher values at kerbside and urban sites (Figs. 5a and 6). The largest contributions in PM_{10} were recorded in Milan (27–55%, Marcazzan et al., 2003; Mossetti et al., 2005), Dresden (36%, Stölzel, 2003), Duisburg (36%, Quass et al., 2004), Madrid (34%, Querol et al., 2004) or Barcelona (32%, Querol et al., 2001), while the lowest values were reported for rural sites in Ireland (4%, Yin et al., 2005) and Portugal (1%, Pio et al., 1996). The low value reported for Copenhagen is linked to the fact that this contribution strictly refers to road dust. In Birmingham and Ireland elemental carbon (EC) and organic matter (OM) were reported separately. The contribution from the C/Fe/Ba/Zn/Cu source at these sites was calculated as the sum of "traffic dust" (when available) and EC (conservative approach), and thus should be considered a minimum value. In PM2.5, the spatial distribution of contributions from this source showed a similar pattern dependent on traffic volume (with the exception of A Coruña). The highest contributions were obtained for Barcelona (49%, Rodríguez, 2002), Dresden (43%, Gerwig, 2005), Erfurt (36%, Vallius et al., 2005) and Amsterdam (30%, Vallius et al., 2005), while the lowest were reported for Ireland (7%, Yin et al., 2005) and Birkenes (5%, Raes et al., 2005). Marcazzan et al. (2001) explained the surprisingly low contribution reported for Milan (5%), as exclusively

Table 2

Cr. Ni. Cu

As, Cu, Pb, Ba, Sb

Cr, Ni, Mo, Cu, Co, As

As, Pb, Zn, Zr, Tl, Cs

Mn, Zn, Pb, Cd

Si, Al, Fe, Li, Sc

K, Carbon, O₃

K, As

Estarreja, PT

Huelva, ES

Llodio, ES

L'Alcora, ES

L'Alcora, ES

Copenhagen (DK)

Monagrega, ES

Llodio, ES; Alsasua, ES

Characteristics of the four main sources identified in the present study as common to most of the European studies reported

(a) Source interpretations: source interpretations provided by the different authors								
PM	Source signatures	No. of studies	Source contribution range (%)	Trend	Source interpretations			
PM10	Carbon, Fe, Ba, Zn, Cu	23/28	1 (Areao, PT)	Dependent on volume and mixture of traffic emissions	• Traffic			
PM _{2.5}	Carbon, Fe, Ba, Zn, Cu, Pb	19/20	55 (Milan, IT) 5 (Birkenes, Norway) 49 (Barcelona, ES)	Chilissions	• Vehicle exhaust			
PM ₁₀	Al, Si, Ca, Fe	24/28	5 (Duisburg, DE)	Marked North to South	• Mineral matter			
PM _{2.5}	Al, Si, Ca, Fe, K	19/20	37 (Huelva, ES)5 (Helsinki, FI)30 (Huelva, ES)	gradent	• City dust			
PM ₁₀	Na, Cl, Mg	20/28	< 5 inland sites	Gradient from Atlantic coastal sites to conti- nental inland sites	• Marine aerosol			
PM _{2.5}	Na, Cl, Mg	15/20	88 (Areao, PT) < 5 inland sites 15 (Areao, PT)		• Sea spray			
PM ₁₀	V, Ni, SO ₄ ²⁻	21/28	8 (Barcelona, ES)	No clear trend	Oil combustion Industry			
PM _{2.5}	V, Ni, SO ₄ ^{2–}	16/20	11 (Amsterdam, NL) 43 (Barcelona, ES)		• mustry			
PM ₁₀	$SO_4^{2-}, NO_3^-, NH_4^+$	9/28	12 (Cork, IE) 46 (Milan, IT)	No clear trend	 Secondary aerosols Regional background 			
PM _{2.5}	SO ₄ ²⁻ , NO ₃ ⁻ , NH ₄ ⁺	10/20	17 (Huelva, ES) 57 (Duisburg, DE)		• Long-range transport			
(b) Sour	ce signatures reported for specifi	c sources identified	l at single locations					
Source signatures Location			Source interpretation	Study				
Cu, K, C	Ca Estarreja, I	РТ	Cu smelter	Pio et al. (1989)				

due to leaded gasoline fuel, which accounted for a small fraction of the total vehicle fleet; other important contributions due to traffic were not resolved in this study and likely included in secondary aerosol and not-explained components. On very specific occasions the tracers of the vehicular source were reported clustered with a mixture of more general anthropogenic tracers, especially V, Ni or SO_4^{2-} , in which cases the source was interpreted as anthropogenic pollution or industry/traffic.

Oil combustion

Frit production

Pigment production

Ceramic production

Biomass burning

Biomass burning

Cu smelter

Metallurgy

Pio et al. (1989)

Alastuey et al. (2006) Querol, Viana, et al. (2007)

Zabalza et al. (2006)

Andersen et al. (2007)

Rodríguez (2002)

Viana, Querol, Alastuey (2006)

Querol, Minguillón, et al. (2007)

Viana, Querol, Alastuey, et al. (2006)

Minguillón, Querol, Alastuey, Monfort, and Miró (2007)

Al/Si/Ca/Fe source: Additional tracers (Ti, Mg and Sr) were not common to all the source profiles in both size fractions. This combination was mostly interpreted as mineral/crustal matter (local or regional re-suspension, city dust, crustal material, road dust, etc.). In Southern Europe, this source would also include African dust contributions. The contributions from this source to PM_{10} showed a marked spatial pattern (Fig. 5a) with highest contributions in South-Western (Portugal, Spain, 20-37%, Almeida et al., 2005; Querol, Alastuey, Viana, et al., 2004; Viana, Querol, & Alastuey, 2006; Viana, Ouerol, Alastuey, et al., 2006) and South-Eastern Europe (Italy, 16–32%, Lucarelli et al., 2004; Mossetti et al., 2005), and lower contributions in UK (7%, Harrison et al., 2003), Ireland (7–16%, Yin et al., 2005), Germany (5%, Querol et al., 2004) and Denmark (10%, Andersen et al., 2007). This source was not identified in Austria or certain studies from Germany. As for PM_{2.5} (Fig. 6), contributions were lower on average but still larger in Portugal and Spain (15-30%) than in UK, Ireland, Germany or Finland (< 5%). It is important to remember, however, that the different studies did not determine the levels of exactly the same tracer species, and this will particularly influence mass reconstruction (e.g., some studies may reconstruct the mass from this source based on Ca, Si and Al levels, whereas others may do so based on Ca only). Furthermore, many studies do not present bulk contents of mineral dust but only soluble fractions or extractable levels of typical crustal elements, thus biasing the mineral dust contribution. Assessing bulk levels of crustal material requires specific analytical approaches (XRF, PIXE or wet chemistry using HF: HNO₃: HClO₄ dissolution).

Na/Cl/Mg source: This source showed the smallest variations regarding marker elements (mostly Na, Cl and/or Mg), and only rarely included Br, K, SO_4^{2-} or Sr. The main interpretations were sea-salt, sea-spray and marine source. Mg was mostly reported as a tracer in Central and Northern European sites, while it was grouped with the crustal elements in Southern Europe (influence of dolomite in the aerosol). Contributions from this source showed a spatial pattern in PM₁₀, with highest levels at Atlantic coastal sites in Western Europe such as Portugal (88%, rural site, Pio et al., 1996) and Ireland (56%, Yin et al., 2005), levels ranging between 5% and 15% at Mediterranean and Northern Atlantic locations, and < 5% at inland sites. In PM_{2.5} the effect of this marine source was <5% at all sites except for locations in Ireland and Portugal (5–15%, Pio et al., 1996; Yin et al., 2005), thus confirming the pattern observed for PM₁₀.

 $V/Ni/SO_4^{2-}$ and $SO_4^{2-}/NO_3^{-}/NH_4^+$ sources: The rationalisation and interpretation of SO_4^{2-} -related sources revealed the largest complexity, and therefore they have been grouped in a single category but separated as two individual sources. The first combination (SO_4^{2-} , V and Ni) was more frequent than the second (SO_4^{2-} , NO_3^- and NH_4^+ , Table 2b). The $V/Ni/SO_4^{2-}$ source was occasionally found in combination with trace elements such as Pb or Cu (interpreted as regional-scale pollution), NH_4^+ and Na, OC and K, or Zn and Pb (long-range transport or anthropogenic pollution). Most authors interpreted $V/Ni/SO_4^{2-}$ as fuel-oil/petcoke combustion or industrial emissions based on the characteristic V/Ni signature of crude oil and its derivates (e.g., shipping emissions), but this interpretation was seen to be quite subjective given that the same authors during different studies labelled this combination as industrial or regional background even though the tracers were the same, based on their knowledge of the monitoring sites. On the other hand, the second combination was interpreted by most authors as secondary aerosols, regional background or long-range transport. The results from this analysis underline the difficulty to clearly discriminate between these two sources, probably because such differences are difficult to define conceptually. Secondary aerosols are by definition not emitted directly into the atmosphere by a single source, they are subject to mixing with primary or secondary particles during long-range transport, and they share the same marker species with anthropogenic emissions on the local- and meso-scale. In conclusion, the discrimination between background levels of secondary aerosols and local anthropogenic contributions is extremely complex. In terms of mass contribution, the overlapping between these two sources is evident in cases such as Estarreja and A Coruña, where the added contributions of both sources provide similar results (24 vs. 14 + 15%), or Modena and Florence where the sources are labelled differently but their contributions add up to similar proportions of the PM₁₀ mass (37% and 39%; Lucarelli et al., 2004; Marenco et al., 2006). The same is observed in PM_{2.5}. No clear spatial pattern is observed for these sources, which account for high proportions of PM_{10} and $PM_{2.5}$ (> 30%) at most of the monitoring sites.

In addition to these four main sources common to the majority of the studies reported, a number of site-specific sources were also detected (Table 2b). These emissions were mainly linked to industrial processes. The only industrial process observed repeatedly at four monitoring sites in two different countries was interpreted as metallurgy. Finally, biomass burning was detected in Denmark and Spain. The fact that biomass burning was rarely detected in studies included in this report while recent studies have found relatively large contributions (in Switzerland, Szidat et al., 2006; Szidat, Prévôt, et al., 2007, but also during the Carbosol project, Puxbaum et al., 2007) suggests that this specific source was not discriminated by the models and thus incorporated in other carbon-containing sources.

The majority of the studies reported an undetermined fraction ranging from 1% to 16% of the PM_{10} mass and 2–32% of $PM_{2.5}$. This fraction includes the contribution from unidentified sources (e.g., those of organic PM, biomass burning, biogenic secondary aerosols, etc.), the analytical uncertainties of the measurements, and components not determined in the PM mass such as water and often Si. When the models did not include an undetermined fraction (25–30% of those reported) it may be assumed that the contribution of the included sources was probably overestimated.

Despite the relevance of presenting error estimates, none of the studies described in this work provided specific information on the degree of confidence with which their SA results should be evaluated. Models such as PMF, ME or UNMIX are able to provide uncertainty estimates by applying a bootstrapping method (US-EPA, 2005), the validity of which has already been tested (Hemann et al., 2008). Such uncertainty estimations should thus be applied in future European SA studies. Another possible approach to evaluate the robustness of SA models is sensitivity analysis, based on average absolute errors (AAE) such as the one suggested by Chow et al. (2007) and applied at a receptor model inter-comparison by Viana et al. (2008). In this study, factor analytical models (PMF and PCA) provided AAEs of approximately 15–20% (average of the absolute percent differences between the estimated and experimental PM mass data, Viana et al., 2008). Additionally, tests of robustness of factors and quantitative source attribution are often done when using PMF by changing the number of allowed factors, as well as uncertainty levels for the input data (e.g. in Quass et al., 2004). These tests show variances around 20–30% for robust factors.

3.4. Current research trends

Based on the analysis of the available SA data until 2005, and in comparison to those published in 2006–2007, four main current research trends were identified.

3.4.1. Inter-comparison of SA methods

The comparison of SA results from different European regions is complex. However, this complexity is not only detected when comparing geographical regions, but also when different receptor models are applied to a single data set. Recent publications focus on this issue (Almeida et al., 2006; Ilacqua et al., 2007; Vallius, Ruuskanen, & Pekkanen, 2008; Viana, Zabalza, et al., 2006; Viana et al., 2008), most of them agreeing on the difficulty to obtain coinciding results with different receptor models for the same data set. The main reason for these disagreements is the different theoretical approaches behind the models. The basic problem is the rotational ambiguity (the fact that the output matrices may be linear combinations of the true ones, and thus the solution to the system is not unique) in combination with the subjective choice of model dimension. Even for constrained models, with a sufficient number of sources, rotational ambiguity will exist if the number of suitable tracers is insufficient. SA models and, if feasible, also data collection and analytical schemes, should thus be evaluated, harmonised and standardised, and further research is needed to ensure that SA methods are robust enough for their application to PM health effects assessment. The combined application of different types of receptor models could possibly solve the limitations of the individual models, by constructing a more robust solution based on their strengths. Inter-comparison exercises between different receptor models have recently also been carried out in the US (Hopke et al., 2006; Lane, Pinder, Shrivastava, Robinson, & Pandis, 2007; Rizzo & Scheff, 2007) and China (Song et al., 2006).

3.4.2. Receptor modelling of particle size distribution and meteorological data

The application of factor analytical techniques to size distribution data (Ruuskanen et al., 2001; Wåhlin, Palmgren, Dingenen, & Raes, 2001) suggested that particle size data can be quantitatively apportioned to sources. Particle size distributions in the atmosphere are dynamic with the potential for coagulation and size dependent deposition. Zhu, Hinds, Kim, and Sioutas (2002) and Zhu, Hinds, Kim, Shen, and Sioutas (2002) have made studies near major highway sources, suggesting that within a few hundred metres of the roadway, there are significant changes in the size distributions as the smallest sizes coagulate and further dry deposit. However, at a reasonable distance from the road, these processes slow significantly as the particle number concentrations decrease and thus, a quasi-stationary profile can be anticipated. The first quantitative apportionment was performed by Kim, Hopke, Larson, and Covert (2004) for Seattle, who identified factors associated with surrounding particle sources of wood burning, secondary aerosol, diesel emissions, and motor vehicle emissions. Zhou et al. (2005) and Zhou, Hopke, et al. (2005) applied PMF to particle size data from the Pittsburgh Supersite, finding that it is important to analyse relatively limited time interval data (one to two months) because of the changes in size arising from changing physical and chemical conditions. Correlations with simultaneously

measured gaseous pollutants help to identify the likely source types. PMF was also applied to particle size distribution data in Rochester (Ogulei, Hopke, Chalupa, & Utell, 2007) and Buffalo, NY (Ogulei, Hopke, Ferro, & Jaques, 2007) and to particle size distributions measured in indoor air (Ogulei et al., 2006). Absolute principal components analysis has been applied to particle size distributions and gaseous data from sites in Southern Ontario (Chan & Mozurkewich, 2007).

More complete SA were possible when the particle size distributions could be combined with highly time resolved particle composition measurements (Zhou, Hopke, et al., 2005; Ogulei, Hopke, Zhou, et al., 2006). Zhou, Hopke, et al. (2005) showed several types of nucleation events. One type of event occurred in the late morning/early afternoon and was associated with sulphate. The other nucleation event type occurred just before sunrise and was associated with nitrate. Similar results were obtained for Baltimore (Ogulei, Hopke, Zhou, et al., 2006) using similar highly time resolved particle species data, showing that with particle size alone, only a limited number of factors could be resolved. The source identification improved with the addition of the pollutant gases as variables in the analysis. This is in agreement with an earlier work from Helsinki (Finland, Vallius et al., 2003), where the most complete and plausible SA results were obtained when both gaseous pollutants and particle size distribution data were combined with the particulate chemical speciation data.

3.4.3. SA analysis of specific PM fractions

3.4.3.1. Carbonaceous aerosols using aerosol mass spectrometry An improved understanding about the sources of OA was identified as one of the main future topics related to the environmental effects of OA (Fuzzi et al., 2006). Currently, the general absence of speciated data on the organic fraction of aerosols has resulted in the difficulty to detect and interpret sources of organic particulates. High-time resolution data of the chemical composition of aerosols as provided by aerosol mass spectrometers (AMS) have been used in several SA studies. In contrast to most SA studies, the AMS allows for an SA in time-resolution of minutes compared to typically daily samples.

Using AMS data, Zhang et al. (2005) and Zhang, Worsnop, Canagaratna, and Jimenez (2005) developed and applied a technique based on two mass spectral tracers and an iterative linear regression technique to apportion the organic carbon in PM₁ into hydrocarbon-like aerosol (HOA) and oxygenated organic aerosol (OOA). HOA and OOA result mainly from combustion-related emissions and from secondary processes, respectively. Marcolli et al. (2006) applied a cluster analysis to an AMS data set and found different classes of organic particles that could be interpreted in terms of sources and origin. PMF was then applied to AMS data from an urban site in Zurich (Switzerland) to quantify main sources and processes of organic carbon in PM₁ (Lanz et al., 2007). It was found that only a small (< 10%) fraction of the organic PM₁ originates from primary fuel combustion emissions, while other identified primary sources were wood burning (10%) and charbroiling (10–15%). The dominating fraction (60–69%) of organic PM₁ was OOA, which is mainly secondary organic aerosol (SOA).

An interesting aspect of utilising AMS data for SA is the fact that a rich library of reference spectra for aerosols from a variety of sources and near-source measurements exists. These reference spectra are extremely helpful for SA studies using multivariate receptor models, because the similarity between estimated factors and reference spectra can be tested (Lanz et al., 2007), and the interpretation of retrieved factors can be justified.

In a follow-up SA study by Lanz et al. (2008), a hybrid receptor modelling approach using ME which incorporates a priori known emission source profiles was applied to AMS data. During the considered time period in winter, wood burning and fossil fuel combustion accounted on average for 35–40% and 3–13% of the organic PM₁, respectively. The major component of organic PM₁ was again OOA (52–57%). Collocated radiocarbon measurements of organic carbon indicated that the dominating fraction of OOA originated from non-fossil sources.

The combination of different SA concepts like the combination of the multivariate receptor model and the radiocarbon method by Lanz et al. (2008) is very promising, because (a) additional insight about sources of organic aerosols can be gained, or (b) the results of either method can be cross-checked.

3.4.3.2. Carbonaceous aerosols using isotopic mass balance (IMB) and ¹⁴C As a unique and powerful IMB tracer, radiocarbon (¹⁴C, $T_{1/2} = 5730$ years) offers the possibility for unambiguous apportionment of fossil and non-fossil sources for carbonaceous aerosols (Currie, 2000; Szidat et al., 2004b). Whereas ¹⁴C has completely disintegrated in fossil material, primary and secondary organic PM originating from fresh biomass is on the contemporary radiocarbon level. By separated analysis of ¹⁴C in EC and OC (Szidat et al., 2004a), these fractions are quantified: EC_{wood} (wood burning, the only major origin of non-fossil EC), EC_{fossil}, OC_{fossil}, and OC_{non-fossil}. The last fraction comprises OC from wood burning as well as primary biogenic OC and biogenic SOA. In order to assess their contributions to $OC_{non-fossil}$, OC_{wood} is estimated from EC_{wood} using an average EC/OC emission ratio and OC_{bio} is calculated by subtraction (Szidat et al., 2006).

Results obtained for Zurich (Szidat et al., 2004a,b) showed that,whereas EC stemmed nearly exclusively (94%) from fossil sources, OC was dominated by biogenic SOA. For winter, a substantial (25%) contribution of wood burning from residential heating was discovered for EC (Szidat et al., 2006), which is considerably higher than for an urban background site at Gothenburg, Sweden (10% of EC originated from wood combustion, Szidat et al., 2008). For a Swiss Alpine valley, at which wood burning is a traditionally popular method for residential heating, the relative contribution of this source to EC emissions exceeded that at Zurich by a factor of ~ 3 (Szidat, Prévôt, et al., 2007).

This ability to directly quantify fossil and non-fossil sources is unique among other apportionment approaches. A detailed attribution of emissions, however, is only possible in combination with other techniques, especially when high time resolutions are necessary. Here, most promising are combinations with methods enabling relative SA information, which needs to be calibrated independently by ¹⁴C. Three examples of such combinations with high time resolutions (a few minutes) are: (a) joint determinations of ¹⁴C and m/z 60 by AMS (Alfarra et al., 2007), (b) verification of PMF receptor modelling results with ¹⁴C(Lanz et al., 2008) and (c) parameter analysis of wavelength-dependent aethalometer measurements using the EC_{fossil} vs. EC_{wood} distinction of ¹⁴C (Sandradewi et al., 2008).

3.4.3.3. Mineral dust Many major, minor and trace elements are associated with the earth's crustal material. Most notable amongst them are Al, Si, Fe and Ca, but other elements such as K, Ti, Sr and Mn are present, and soils also contain organic matter. Abrasion products from road traffic also contain elements such as Fe (brake wear), Al (car body and engine parts), OC and EC (tyre wear) and Ca, Fe, Al, Si and carbon (road surface wear). Furthermore, Sb, Cu, Zn and Ba are also found in many studies as tracers of emissions from tyre and brake abrasion. Additionally, road traffic leads to the resuspension of road dust, of which soil is a major component. The precise elemental content of road dusts is location-dependent, due to differences in local geochemistry, the composition of road surface materials, and the composition of brake pads (F. Amato, unpublished data). Harrison, Jones, and Lawrence (2004) in UK found a roadside enhancement of Fe, but not Ca, whilst Wåhlin, Berkowicz, and Palmgren (2006) in Denmark found a roadside enhancement in Ca which appears to have derived from road surface materials.

Wind-driven suspension of surface soils and desert dusts gives rise to airborne particles whose composition has much in common with road dusts, the main difference being a different trace element profile. Thus trace elements such as Ba and Cu (brake pads) and Zn (tyres) are enriched in traffic-generated particles relative to natural soils and dusts (Birmili, Allen, Bary, & Harrison, 2006; Hjortenkrans, Bergback, & Haggerud, 2007). However, there are such strong overlaps between the elemental composition of natural and anthropogenic crustal matter-based dusts that receptor models based upon elemental profiles have great difficulty in distinguishing them. Rodriguez et al. (2007) estimated mineral dust concentrations in PM_{2.5} from the sum of Al₂O₃ + SiO₂ + Ca + CO₃²⁻ + Ti + Fe + Sr, and found major differences in concentrations between Barcelona ($3.7 \,\mu g/m^3$ in PM_{2.5}, highest concentrations), Milan and London ($0.6 \,\mu g/m^3$ in PM_{2.5}, lowest). Mineral dust contributions to background sites in Germany not directly influenced by human activities were reported to be around $2 \,\mu g/m^3$ or 18% of PM₁₀ (yearly average; Kuhlbusch, John, Romazanowa, Top, & Weissenmayer, 2004).

One other type of natural dust contribution is the transport of mineral dust from the African continent, which affects especially Southern European countries. Due to the overlap between the elemental composition of local soil dust and anthropogenic mineral dust, the identification and quantification of these contributions is complex. Methodologies for addressing this issue were presented by Collaud Coen et al. (2004) and Escudero et al. (2007). Escudero et al. (2007) obtained the daily net African dust load in PM_{10} by subtracting the daily regional background (RB) level from the PM_{10} concentration value at an RB station. In turn, the daily RB level was estimated by applying a monthly moving 30th percentile to the PM_{10} time series at an RB station after a prior extraction of the days with African dust transport. The daily PM_{10} RB level obtained is only subtracted on days when the occurrence of African dust outbreaks was demonstrated (by means of meteorological analysis and dust map interpretation), the difference being the daily net African PM event without the need for PM speciation.

Finally, coal fly ash has also a composition that may resemble that of soil dust, and this contribution may be important at some sites in Europe. In certain study areas this could represent an additional source of bias in the receptor modelling results (Viana et al., 2007).

3.4.4. Receptor modelling for epidemiological and health-related studies

Historically, epidemiological studies on the associations between PM and health effects have relied on fixed site measurements of mass concentrations (TSP, PM_{10} and, more recently, $PM_{2.5}$). However, during the last decade an increased pressure has emerged to uncover the specific PM constituents responsible for the observed health effects. If specific constituents were found to be more harmful than others, this would inevitably lead to new legislation and mitigation actions, and more efficient control of the sources of these constituents. When correctly linked to epidemiological studies, SA of PM can therefore play a significant role in attempts to answer this crucial question.

The use of receptor modelling in epidemiological PM health studies is still very young and therefore it is difficult to detect clear trends in this research area. Reports from epidemiological studies utilising PM SA results as input have been published during the last decade especially from Europe (Andersen, Wåhlin, Raaschou-Nielsen, Scheike & Loft, 2007; Lanki et al., 2006; Yue et al., 2007) and North America (Laden, Neas, Dockery, & Schwartz, 2000; Mar et al., 2005). In the studies performed in Europe, SA analyses have been applied to investigate the associations of PM with cardiac but also respiratory endpoints, all of which studies indicated that the adverse health effects were primarily associated with exposure to traffic-related particles.

Ambiguity in source classification and misclassification of sources is a major problem in contemporary SA analyses and may have serious implications with regard to the use of SA results in epidemiological studies. Implications of inadequacies of SA methods and the use of results from SA analyses in epidemiological research have been discussed by Grahame & Hidy (2007).

More precise estimates for human exposure and, if possible, personal exposure to source specific PM fractions are a prerequisite for obtaining new information on the potentially differential effects of PM from different sources. This goal can only be achieved by incorporating in epidemiological studies extensive air pollution measurements which allow efficient application of the latest and most powerful SA models.

4. Gap analysis and possible research directions

The review of existing data and their meta-analysis, presented in this work, evidenced the following gaps, (a)–(f), and upcoming research directions, (g)–(l), which should be taken into account for the design of future SA studies

- (a) Calculation of uncertainty estimates for SA results: by means of bootstrapping or other methods. Uncertainty estimates are currently unavailable from European SA studies.
- (b) Quantification of natural source contributions (e.g., African or windblown dust, secondary organic material): even though certain methodologies are already available to quantify African dust contributions (Collaud Coen et al., 2004; Escudero et al., 2007), further research is still necessary. As regards biogenic secondary organic material, its composition is not well known and cannot be traced by inorganic compounds.
- (c) Apportionment of specific anthropogenic emission sources: certain anthropogenic combustion emission sources (e.g., shipping emissions, diesel/gasoline exhaust) were not discriminated by any of the SA studies described above. This is most likely due to the absence of marker species in the input data sets and/or the inability of the models to separate sources with common tracers. Additional data on source-specific marker species (specifically, speciation of OA for sources of organic PM) and their combined application with meteorological parameters (wind speed, wind direction, etc.) and/or particle number and size distribution data may provide new insights to this problem.
- (d) Differentiation between SO_4^{2-} -containing sources: with the aim to discriminate sources such as secondary regionalscale aerosols vs. local- or meso-scale anthropogenic emissions such as industry or shipping.
- (e) Identification of the biomass combustion source: events are generally episodic, thus complicating their identification on longer time-scales. The use of more specific tracers such as levoglucosan and/or the detailed speciation of the organic fraction would help identify the biomass combustion source. New methods and instrumentation are also available (¹⁴C, AMS, multiwavelength light absorption). It will be important to compare these emerging techniques with organic tracer methods and potassium (Sandradewi et al., 2008).
- (f) Identification of the nitrate component: receptor models have been very poor in identifying a nitrate component. Identification of this component is a high priority for the following reasons: (1) Western European secondary inorganic aerosol is progressively becoming dominated by nitrates, whereas sulphates were historically more important (AQEG, 2005); (2) whilst sulphate and nitrate concentrations are frequently well correlated, there are occasions when the two components are entirely uncoupled (Abdalmogith & Harrison, 2006); and (3) work in

the UK shows that nitrate makes a much greater contribution to $PM_{2.5}$ and PM_{10} composition on peak episodes, relative to all days (Harrison, Jones, & Lawrence, 2004).

- (g) Choice of SA tools: CMB-type approaches are ideal when the compositions of the emissions for all relevant sources are known and do not change with time. Factor analytical models are used when certain sources or source profiles are unknown. Hybrid models avoid the main limitations of CMB-type and factor analytical methods, although the issue of the variability of source profiles with time is not solved by these models either.
- (h) Comparability between SA methodologies: further research should be carried out on the inter-comparison of SA models.
- (i) Evaluation and harmonisation of data collection and analytical schemes: in order to allow better comparability across individual sites. Measurements should aim to provide data on PM chemical composition (tracers and source profiles) which facilitate the separation of complex source components.
- (j) Use of single particle aerosol mass spectrometry: in addition to AMS, there is considerable potential in the use of single particle aerosol mass spectrometers in SA of inorganic and organic constituents. The use of single particle mass spectrometry methods within Europe is relatively limited, but Dall'Osto & Harrison (2006) identified five main classes of particles in the atmosphere of Athens, from the mass spectra of 166,000 particles. Whilst the results were not used to derive a quantitative SA, they were qualitatively comparable with data obtained by traditional chemical methods. Because of the fast response of the technique, diurnal variations in semi-volatile aerosol components were readily identified.
- (k) SA of indoor air concentrations, and relation to exposure: as a consequence of the current increase in publications reporting indoor air particle concentrations and composition.
- (1) Combination of back-trajectory modelling with SA analyses: the further utilisation of back-trajectories (TS and PSCF, Lupu & Maenhaut, 2002) holds a large potential (e.g., identification of African dust).

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