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foR Assessment
of Integrated Strategies
At regional and Local scales

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Deliverable 2.6 Source Apportionment Methodologies

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Summary

The methodologies used in Europe to identify the sources of atmospheric pollutants are summarized and discussed. The report combines information available from surveys and scientific reviews on source apportionment models available in the literature and the results of the questionnaire carried out within the framework of the project APPRAISAL.

Version History

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Summary of Changes

Version	Section(s)	Synopsis of Change
0.1		
0.2		Natural sources, case study Portugal, contribution of SA to IAM
1.0		contribution of SA to IAM
1.1		re-arrangement of titles and annexes
[1.1] note		

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1 Introduction to Source Apportionment

In Europe, the population exposed to levels of particulate matter, tropospheric ozone or polycyclic aromatic hydrocarbons (PAH) above the reference values for the protection of human health varies between 16% and 30% (EEA, 2012). Air pollution also causes ecosystem eutrophication, acidification and vegetation oxidative stress. In addition, many air pollutants contribute also to global warming. For those reasons, air quality is considered as a major environmental issue, particularly in urban areas.

Source Apportionment (SA) is the practice of deriving information about pollution sources and the amount they contribute to ambient air pollution levels.
Information on pollution sources is essential in the design of air quality policies and, therefore, SA is required explicitly or implicitly for the implementation of the Air Quality Directives (Dir. 2008/50/EC and Dir. 2004/107/EC; hereon AQD). The following are examples of activities for which identification of sources is relevant (see also tables 3 and 4):

- Drawing up action plans
- Assessment effectiveness of abatement measures (*ex ante* and *ex post*)
- Quantification of:
  - long range transport
  - transboundary transport
  - natural sources
  - winter sanding and salting
- Identification of sources for pollutants of particular interest (e.g. PAHs, ozone precursor hydrocarbons, black carbon)

In recent years, SA was also a pre-requisite to motivate applications for postponement of attaining limit values of PM_{10} and NO_2

Different approaches are used to determine and quantify the impacts of air pollution sources on air quality. Commonly used SA techniques are:

- Exploratory methods
- Emission inventories
- Inverse modelling
- Artificial neural networks
- Lagrangian models
- Gaussian models
- Eulerian models
- Receptor models

Table 1. Types of Receptor model (modified from Belis et al., 2013)

Type of receptor model	Examples
Exploratory methods	Enrichment factor, tracer method, Lenschow approach, APEG
Chemical Mass Balance	EPA CMB 8.2
Eigenvector based models	PCA, UNMIX

Factor analysis without constraints	FA, APCFA
Positive matrix factorization	PMF2, EPA PMF v3
Hybrid trajectory based models	CPF, PSCF
Hybrid expanded models	PMF solved with ME-2, COPREM

Legend: CMB, chemical mass balance; PCA, principal components analysis; FA, factor analysis; APCFA, absolute principal component factor analysis; PMF, positive matrix factorization; ME, multilinear engine; CPF, conditional probability function; PSCF, potential source contribution function; COPREM: constrained physical receptor model, APEG: air pollution expert group approach.

- Exploratory methods use simple mathematical relationships and a number of assumptions to achieve a preliminary estimation of the source contribution (e.g. enrichment factors, Lenschow approach).
- Emission inventories are detailed compilations of the emissions from all source categories in a certain geographical area and within a specific year. Emissions are estimated by multiplying the intensity of each relevant activity (activity rate) by a pollutant dependent proportionality constant (emission factor). Even though emission inventories do not represent the actual contribution of sources to atmospheric pollution, many local governments use this information straightforward as source identification tool for the design of abatement measures (Ulrike Döring, Pilot Project, personal communication).
- In inverse modelling, air quality model parameters are estimated by fitting the model to the observations. The inverse technique constitutes a least squares optimization problem with an objective function defined as the sum of squared deviations between modelled and observed concentrations.
- Artificial neural networks (ANN) are sets of interconnected simple processing elements (artificial neurons) which can exhibit complex global behaviour. In order to produce a desired signal flow, algorithms designed to modulate the weights of the connections in the network are applied.
- Lagrangian models use a moving frame of reference to describe the trajectories of single or multiple particles as they move in the atmosphere.
- Gaussian plume models assume that turbulent dispersion can be described using a Gaussian distribution profile. This type of models is often used to estimate emissions from industrial sources.
- Eulerian models encompass equations of motion, chemistry and other physical processes that are solved at points arranged on a 3D grid.
Often, the term dispersion models or source oriented models is used to refer to the previous three categories. In this document these models are treated separately because of the relevant differences in their applications for source identification purposes.
- Receptor models (RM) are focused on the properties of the ambient environment at the point of impact as opposed to the source-oriented models that account for transport, dilution, diffusion and other processes that take place between the source and the sampling or receptor site.

2 State of the art

A group of experts produced a report on the contribution of natural sources to PM levels in Europe that summarized the outcome of the workshop that took place in Ispra in October

2006 (Marelli et al., 2007). For the purposes of compliance with air quality limit values, experts agreed that only contributions to PM from natural sources that may not be influenced by human action can be deducted from PM levels according to the indications given in the directives:
- Primary biological aerosol particles (PBAPs), including pollen, spores, and plant debris.
- Biogenic non-sea-salt sulphur aerosol.
-Volcanic activities, only occasionally causing exceedances of PM limit values in few areas.
-Desert dust long range transport (see also the Portuguese case study in Annex 1).

The report also reviewed the most commonly used methodologies to apportion this type of sources: 1) routine methods, 2) information in parallel from background stations, and 3) advanced tools implemented by research groups. Nevertheless, experts concluded that the estimation of natural sources on the basis of measurements monitoring networks is quite difficult, and that only few Member States (MS) routinely implement methods for the identification and quantification of a natural episode.

Within the activities of the Forum for Air Quality Modelling in Europe (FAIRMODE) Working group 1 Sub-group 2 (SG2) on the “Contribution of natural sources and source apportionment”, two surveys on the type and frequency of modelling tools that are used in Europe for source apportionment were carried out (Fragkou et al., 2012).

The first survey focused on studies to support applications for the postponement of attaining PM_{10} limit values, and the second survey collected information on model use for source apportionment of regulated pollutants and on the procedures used to evaluate the applied methodologies.

Concerning Receptor models, two reviews were published at a five-year distance (Viana et al., 2008 and Belis et al., 2013). In addition, the use of Receptor models for policy implementation is discussed by Karagulian and Belis (2012). Moreover, quantitative data on receptor model performance and uncertainty is available in the reports on the European intercomparison exercises for Receptor models performed within the framework of the JRC initiative on Receptor model harmonization (e.g. Karagulian et al., 2012).

Table 2. Modelling tools used for source apportionment by different MS for the purposes of preparing the time extension reports (from Fragkou et al., 2012).

Model type	Number of countries	% *
Lagrangian	7	41
Eulerian	10	59
Receptor	5	29
Gaussian	6	35
Computational Fluid Dynamics (CFD)	1	6
Combination of models	12	71

*Percentages do not add up to 100%, as many countries used more than one model type

The first survey was launched when the European Commission announced the decision on the applications for the time extension presented by 17 MS, including 289 air quality zones.

The study analysed the reports submitted by MS as technical annexes to support the time extension applications. The methodologies and the number of countries in which they were used are reported in table 2. The percentages for each methodology vary between 30% and 60% with Eulerian models showing the highest share. Worth to mention that the majority of the countries (71%) applied a combination of modelling approaches. The high share of

Eulerian and Lagrangian models is explained by the interest of many MS to support their claim that most of the pollution episodes have origin outside their boundaries and derive from long range transport. On the other hand, Receptor models were used to identify sources at the urban or regional level.
The second survey was based on the questionnaires distributed by the leading team of FAIRMODE SG 2 on source apportionment and natural sources. The questionnaires were distributed via e-mail among National Focal Points representatives of the European Environment Information and Observation Network (EIONET, representing 40 European countries) and 49 experts representing 17 countries. A total of 17 questionnaires corresponding to 11 EU countries were collected.
The use of the different tools for source identification ranged from less than 20% for Gaussian models to almost 60% for Receptor models (Figure 1). Lagrangian (e.g. Lagrangian particle dispersion models) and Trajectory models were less frequently used and always complementary to other models. CFD models were only reported in one case.

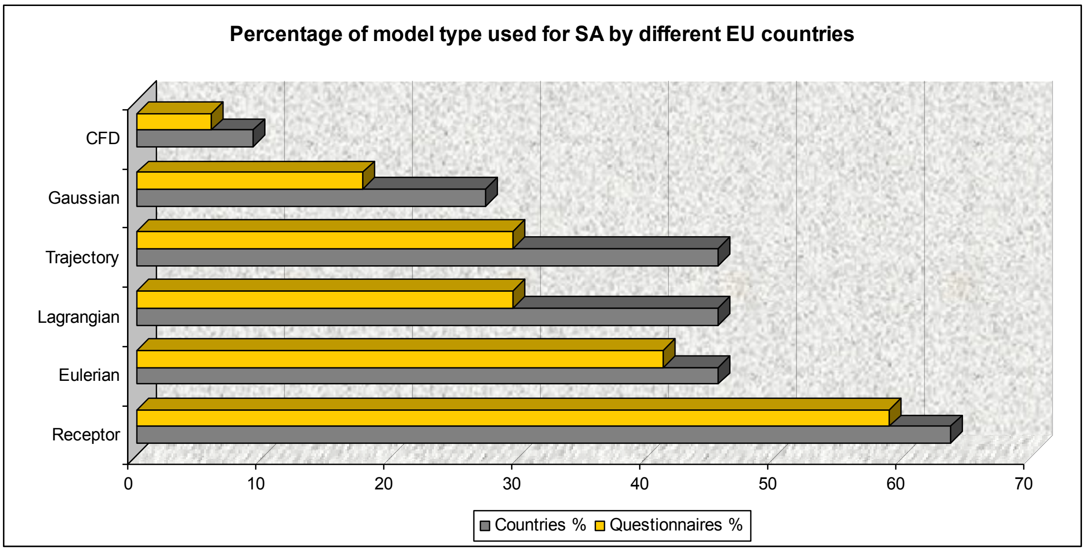

Figure 1: Percentage of model type used for SA by different EU countries (from Fragkou et al., (2012).

All the questionnaires report PM as the target pollutant (47% of them report PM_{10} as target). Likely, the limited availability of necessary data for source attribution of fine particles, including the characterisation of specific tracers and chemical profiles, may have contributed to the low number of SA studies targeted at the smaller PM fractions. In addition, limit values (and therefore exceedances) were only in force for PM_{10} at the time in which this survey took place. A significant number of studies reported on the questionnaires (35%) have performed SA for NO_x and NO_2, while O_3, SO_2, and CO were the target metrics in 27% of the countries. Source attribution of volatile organic compounds (VOC), metals and dioxins was examined in a very small percentage of the reported studies.
The study by Viana and co-authors (2008) overviewed source apportionment studies in Europe from 1987 to 2007 by compiling meta-data on 71 studies (see Table 1 page 831 of the above mentioned publication) based on a questionnaire and existing publications.
According to this study, PCA was the most frequently used model up to 2005 (30% of the studies), followed by the so called Lenschow approach or incremental concentrations approach (11%) and back-trajectory analysis (11%). An increase in the use of PMF (13%) and the mass balance analysis of chemical components (19%) was observed from 2006 on.
PM_{10} was the preferred target metric (46%) followed by $PM_{2.5}$ (33%) and coarse fraction ($PM_{2.5-10}$; 9%).The majority of the studies were carried out in urban background locations

(53% of the studies) while industrial or kerbside sites represented 11% and 20% of the studies, respectively.
Overall, a generally good spatial coverage of SA studies over Europe, especially regarding the northern, south-eastern and south-western dimensions, was seen.
In this review, four main source categories across Europe were identified:

- -Traffic source, characterized by Carbon/Fe/Ba/Zn/Cu often including road dust;
- -Mineral/crustal matter source with Al/Si/Ca/Fe as distinctive components;
- -Sea-salt, sea-spray and marine source associated to high *Na/Cl/Mg concentrations;*
- -Regional scale pollution and long-range transboundary anthropogenic pollution *sources* rich in either *V/Ni/sulphate or sulphate/nitrate/ammonium*.

A survey on the use of Receptor models (RM) for PM source apportionment in Europe between 2001 and 2010 including 79 studies and 243 reported records (Karagulian and Belis, 2012) evidenced a dramatic increase in the number of scientific publications on this topic during the last decade and an increasing number of ready-to-use tools (Figure 2). The highest increase rate in the number of studies coincides with the entry into force of the limit

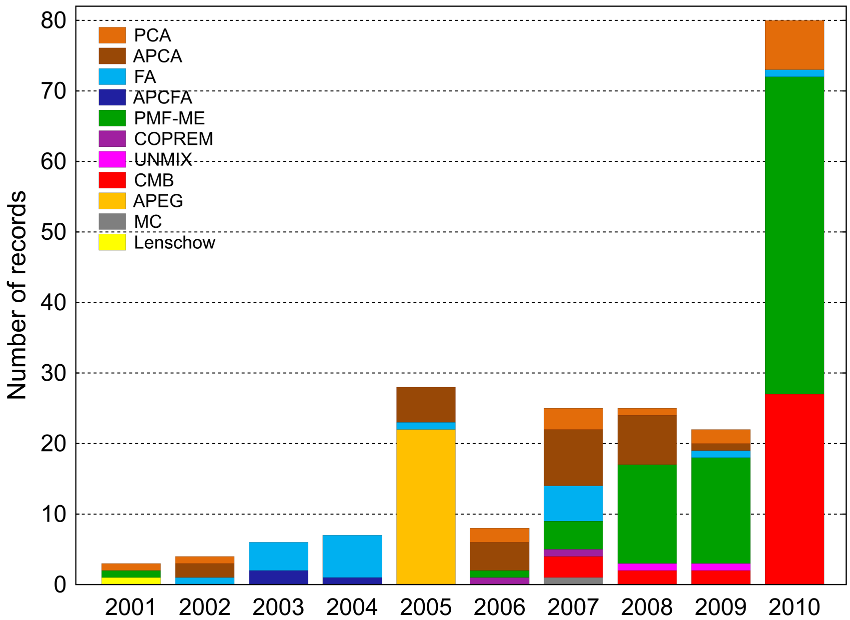

Figure 2. Time trend of RM studies in Europe between 2001 and 2010/11 (from Karagulian & Belis, 2012).

value for PM10 (1999/30/EC) and the target value for PM2.5. About 60% of the studies were carried out in urban background sites, 16% in source oriented sites, and 15% in rural sites.
In contrast with the tendency observed between 1987 and 2005, the majority of the studies were performed using Positive Matrix Factorization and Chemical Mass Balance models in the period 2001-2010 (Figure 3).
Most of the studies were conducted in Spain, Italy and the UK. Many recent studies completed or in progress were reported also in France (Figure 4).

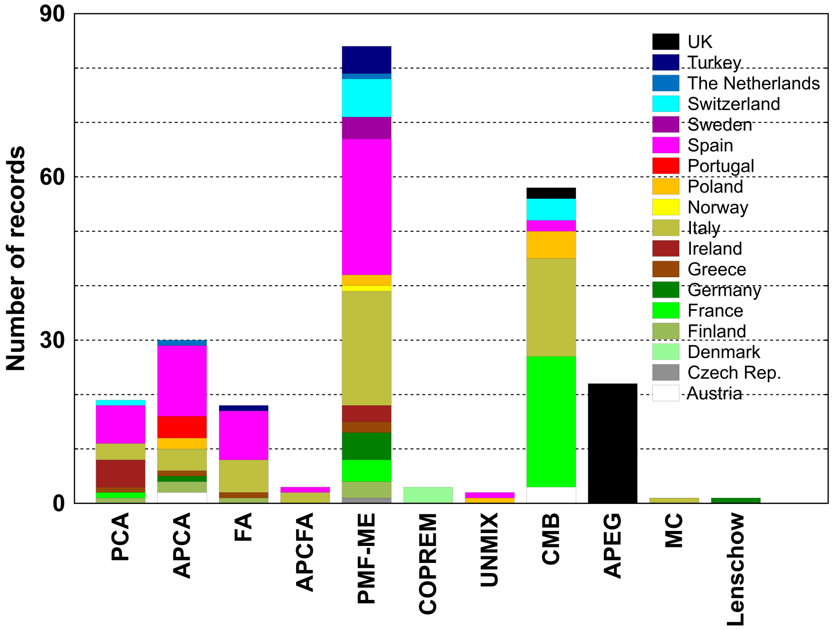

Figure 3. Grouping of European RM studies published between 2001 and 2010/11 (from Karagulian & Belis, 2012).

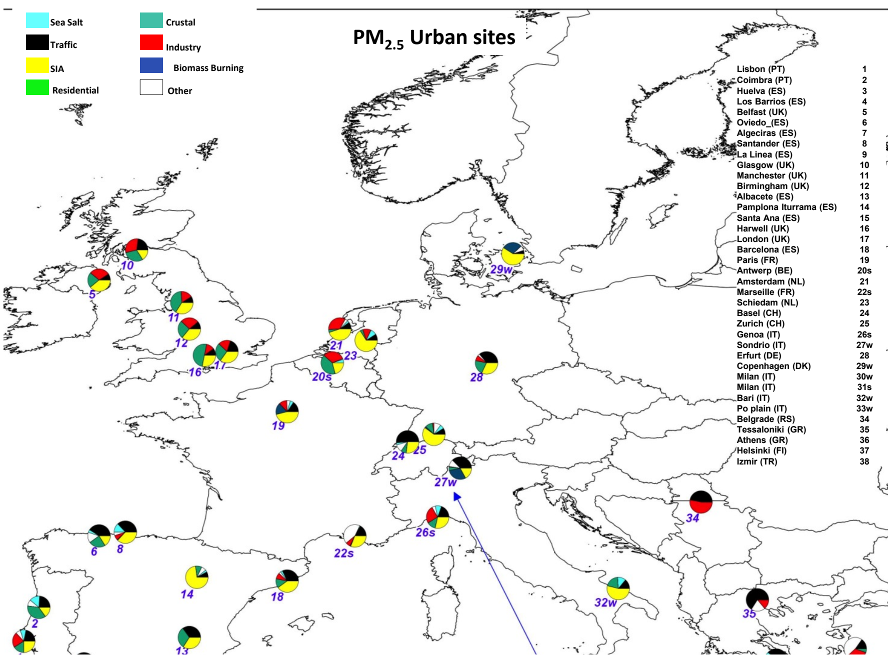

Figure 4. Geographic distribution of $PM_{2.5}$ sources in European urban sites estimated with Receptor models (from Belis et al., 2013).

In the most recent review on source identification studies carried out until 2012, a detailed meta-analysis of data available from previous studies is presented (Belis et al., 2013). In order 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. In addition, residential heating by coal (or coal substitutes) combustion was proved to be a major PM pollution source in many areas of the new EU member states. Also residential coal combustion in small stoves and boilers has

been found to be a main source of PM_{10} and benzo(a)pyrene in certain areas of Europe (Junninen et al., 2009; more details in Annex 1).
The main results of the above mentioned review show that the field of Receptor models is developing swiftly with Positive Matrix Factorization and Chemical Mass Balance, which are the most used models, evolving towards tools with refined uncertainty treatment.
The review demonstrates that, excepting mineral dust and sea/road salt, PM_{10} and $PM_{2.5}$ derive from the same sources. Secondary pollution deriving from gas-to-particle conversion is the main PM mass and particulate organic carbon source. Therefore, to reduce the concentration of these pollutants it is necessary to abate the sources of secondary inorganic aerosol mainly deriving from traffic emissions and agriculture. Also primary emissions from traffic and biomass burning have been identified as causes of exceedances, especially during the cold season.
The review authors stress the need of long term speciated PM datasets and characterization of source fingerprints to further improve source identification studies. In addition, harmonization of the different approaches would facilitate the interpretation and comparability of the results and their application in the design of abatement measures.

3 Description of the APPRAISAL Database

The APPRAISAL Database is structured in 5 main topics

- Synergies among national, regional and local approaches, including emission abatement policies
- Air quality assessment and planning, including modelling and measurement
- Health impact assessment approaches
- Source apportionment
- Uncertainty and robustness, including QA / QC

In order to populate the database, a questionnaire structured according to the relevant database fields was prepared and distributed to selected institutions or project contact persons. The questions dealing with source apportionment methodologies are presented in the following (topic 4).

To simplify data elaboration and guide the experts in filling out the questionnaire, a number of questions with multiple choice answers were included:

- What was the purpose of the source apportionment study?
- What was the used methodology?
- What were the considered source categories?
- What were the considered pollutants?
- Study Area
- Types of input data used (depending on type of SA methodology)
- Sampling design (only for receptor modelling methodology for SA)

To give experts the chance to express more freely their views on methodological aspects, a few open questions were included in the questionnaire. Nevertheless, this kind of questions requires more effort from the expert and the answers are more difficult to elaborate in a statistical manner.
Open questions in the questionnaire:

- Explain to what extent was it possible to achieve the objectives of your source apportionment study and what were the limitations of the used methodology

- Are source apportionment capacities in the institutions of your country/region/city enough to achieve reliable source identification and support air quality management?
- Is data collection in your country/region/city appropriate for source identification?

4 Analysis of questionnaire answers

In this section, the 53 questionnaire answers collected in the first phase of the APPRAISAL survey are summarized. It was accomplished by asking selected institutions to provide information on their integrated assessment modelling (IAM) activities or projects by filling up the questionnaire with the support of APPRAISAL experts. The figures in this chapter show the relative share of the different choices for every question. Quite often, the interviewed expert selected more than one answer, therefore, the percentages sum total is more than 100%.
In the second phase of the database population, the questionnaires will be filled in directly on line. This is expected to increase significantly the amount of answers improving its representativeness of the assessment of source apportionment methodologies in Europe.

1. What was the purpose of the source apportionment study?

The answers to this question confirm there are many motivations for performing source apportionment analysis within the framework of integrated assessment studies. The main reasons are associated to obligations deriving from the AQD: to design air quality plans or action plans, to identify the causes of exceedances, and to identify the contribution from other countries (transboundary pollution; Figure 5).
Other motivations for SA studies are the evaluation of geographic origin within a country (not transboundary), application for postponement of attainment and assessing the effectiveness of measures.

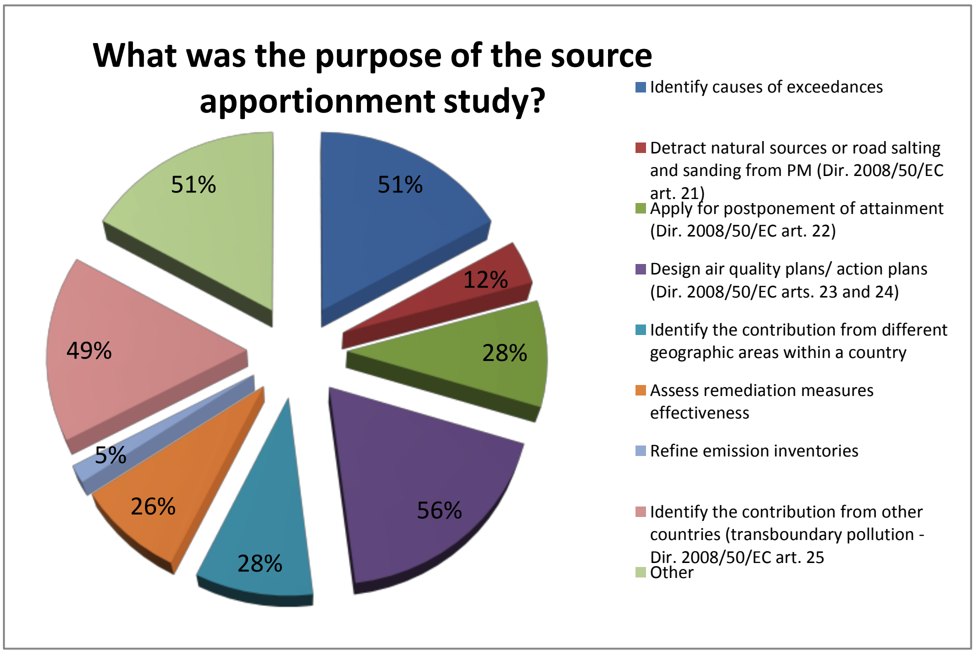

Figure 5. Answers to the question: What was the purpose of the source apportionment study?

2. What was the used methodology?

As already observed in previous studies, Receptor models, Lagrangian models, Eulerian models and Gaussian models are all used for the identification of sources. Objective estimation and inverse models are used marginally for this task (Figure 6). Worth to mention that one third of the answers report the combined use of more than one methodology.

3. What were the considered source categories?

The most frequent activity sectors/source categories identified in the studies are combustion in the energy sector and road transport (more than 70% of the studies), followed by combustion in industry, non industrial combustion and agriculture (Figure 7). Interestingly, many of the studies (40%) focus only on one single activity sector/source category. The frequency of activity sectors/source categories reflects the most commonly encountered pollution sources. Nevertheless, this is also influenced by the availability of source characterization studies and the existence of mandatory emission registers.

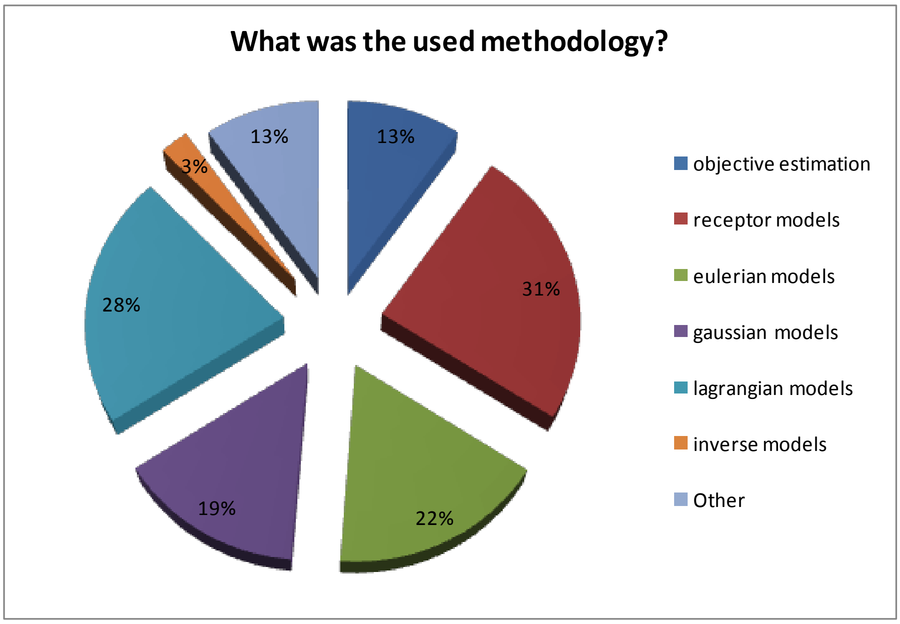

Figure 6. Answers to the question: What was the used methodology?

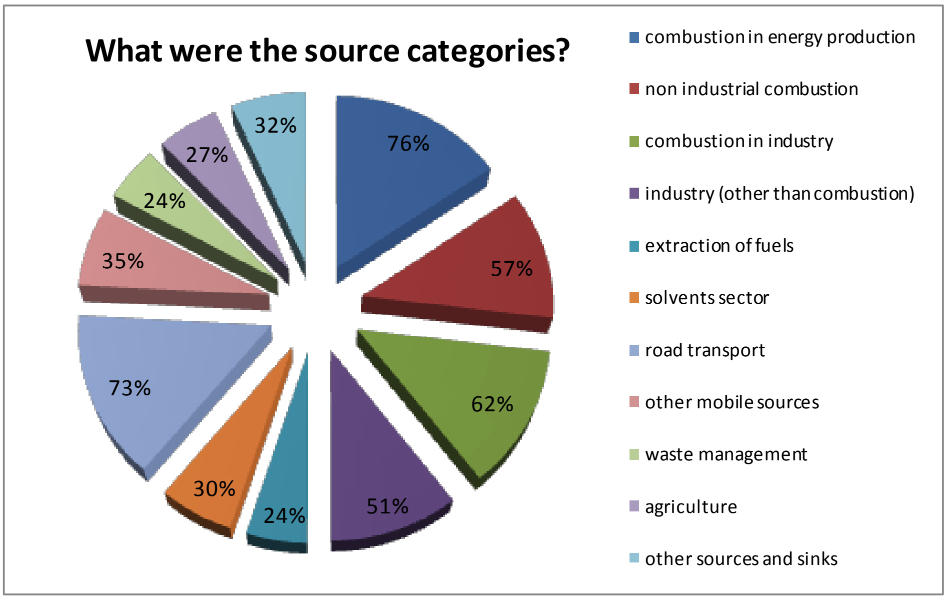

Figure 7. Answers to the question: What were the considered source categories?

4. What were the considered pollutants?

The most important pollutants considered in source apportionment studies are PM_{10} and nitrogen dioxide following by two pollutants associated to them: $PM_{2.5}$ and nitrogen oxides, respectively (Figure 8). All the other pollutants are treated in less than 10% of the studies.

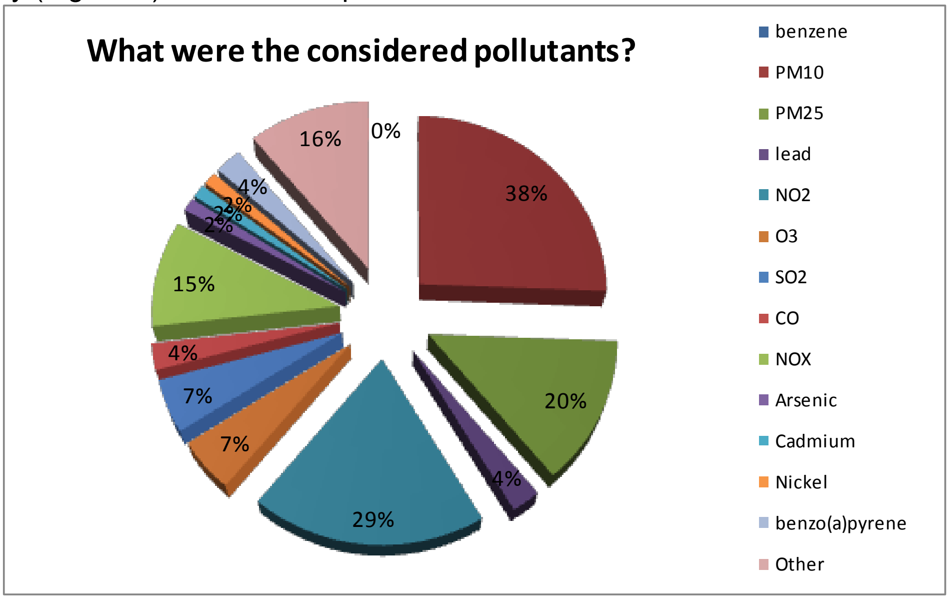

Figure 8. Answer to the question: What were the pollutants considered?

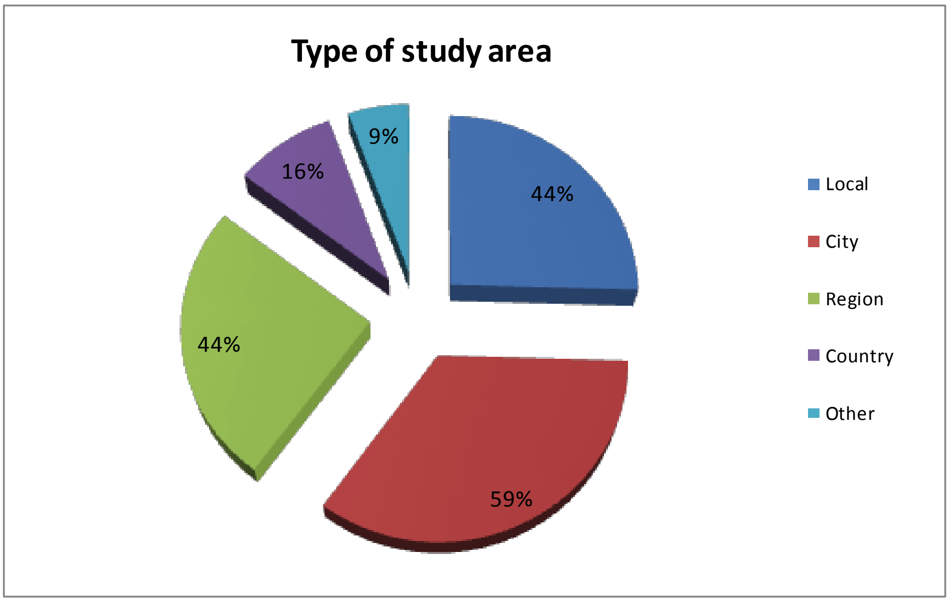

Figure 9. Answer to the question: Study Area

5. Study Area

The great majority of the studies focus on the city level (59%) while local (lower than city) and regional scales represent a 44% each (Figure 9).

6. Types of input data used

The types of input data strongly depend on the adopted methodology (Figure 10). Monitoring networks are the most frequent source of information (45 % of the studies) due to the fact that it is common for many types of models. Emission inventories and meteorological fields are both equally represented (36%). Dedicated field campaigns represent one fourth of the answers.

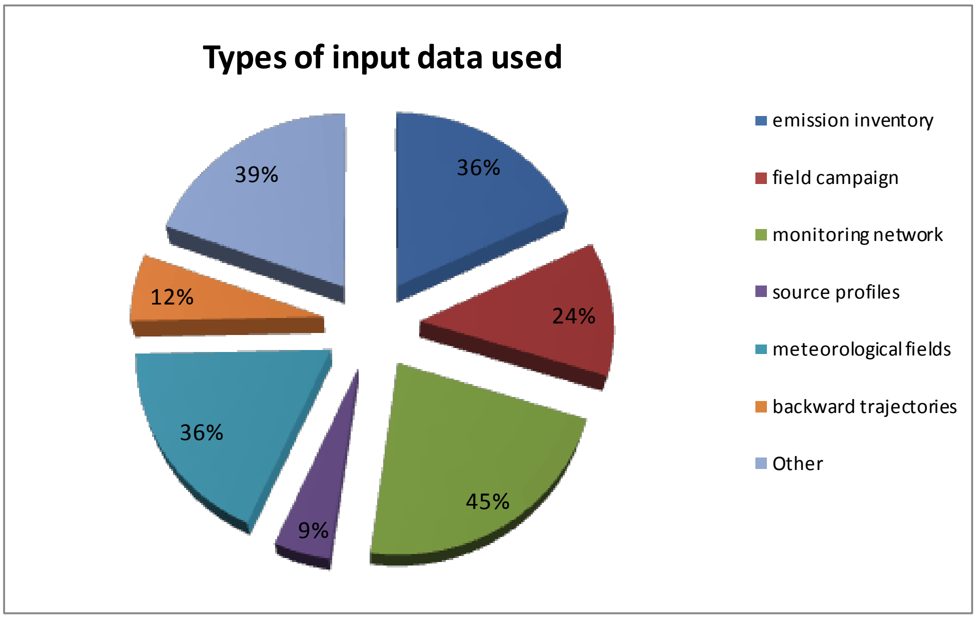

Figure 10. Answer to the question: Types of input data used

7. Sampling design (only for receptor modelling methodology for SA)

Only few experts answered this question, likely due to fact that specific technical information was requested. The number of sites (4 answers) varies between 3 and 22 and the number of samples (2 answers) between 17 and 31. Sampling period spans from seasonal (winter, winter and summer) to yearly sampling strategy (4 answers). The reported sampling equipment (3 answers) is: CEN low volume sampler, HiVol PM_{10} Sampler 1200, and beta-absorption FH 62 I-R.
There were no answers to the open questions aiming at obtaining information about the methodological aspects and the institutional capacity to deal with source apportionment studies.

5 Limitations of the current assessment and planning tools and key areas for future research and innovation

5.1 Strengths and limitations of the different approaches

When it comes to overall source identification and quantification in a given area, Receptor models and Eulerian models with tagging modules are the most suitable tools. The other techniques cited in this document are used either for exploratory analysis, for the quantification of the contribution of specific sources, or as a complement of the above mentioned techniques. Nevertheless, it must be clear that even the most advanced tools have strengths and limitations.
Therefore, the most robust approach to deal with source identification is the use of different models on the same data or on different data of the same area to mutually validate the results and assess the quality of the output quantitatively. For all the tools, experienced users and tested operational protocols including validation steps are required to achieve acceptable performances.

Receptor models

Strengths
- Derive from real-world measurements on one or more sites.
- Appropriate for urban areas and source-oriented sites, but also for regional scale.
- Good output uncertainty estimation.
- Can be used to identify main source categories even when there is poor information about source chemistry and location.
- Mainly used for PM, but also for VOC, PAH and gaseous pollutants.
- Combination with trajectories or wind analysis makes it possible to track the geographic origin of pollution.

Limitations
- Time series of pollution measurements and chemical characterization are needed.
- Not appropriate for reactive species.
- Provide limited information on secondary inorganic aerosol sources.
- Need for harmonization of methodological steps like estimation of the number and definition of source categories.
- Difficulty to estimate accurately the different sources of carbonaceous fractions due to the limited knowledge of its molecular composition, atmospheric processes and characteristic emission profiles (Pio et al., 2011).

Eulerian models

Strengths

- Reproduce complex physical and chemical atmospheric processes in a simplified manner.
- Provide estimation for every cell in the grid with hourly time resolution.
- Deal with reactive species and, therefore, are suitable to estimate the sources of secondary pollutants.

Limitations

- The complexity of the model makes it difficult to estimate the uncertainty of the output.
- The output depends on the quality and resolution of the emission inventories.
- Current versions do not focus much on the contribution of species that have a small share of the total pollution mass (e.g. heavy metals, PAHs).
- Models using brute force (BFM) or zero out (ZO) methods to identify sources have high computational intensity, and the smaller concentration changes between the simulations may be strongly influenced by numerical errors (Koo et al., 2009). In addition, BFM results depend on the case scenario and the model response may be nonlinear or non-additive making difficult to compare the base case and the scenario case.

5.2 State of the art currently not implemented in the collected projects or plans

In the following, advanced technologies, the application of which in European studies is still at the beginning but growing swiftly, are discussed.

- Hybrid trajectory based Receptor models combine analysis of wind direction or backward trajectories with Receptor models. They are used to determine the geographical origin of pollutants and are suitable to investigate domestic and transboundary pollution. The first type of models estimate the probability that a given source contribution from a given wind direction will exceed a predetermined threshold criterion (e.g. Kim et al., 2003). Trajectory based Receptor models make it possible to calculate the probability of a trajectory circulating over a cell and reaching the receptor site when the pollutant concentrations or source contributions are above a selected threshold (e.g. Ashbaugh et al., 1985). Even if this kind of models has been available for a long while, they have found little application in Europe.
- Carbonaceous compounds and elemental carbon are among the most important components of aerosol from both the quantitative and the qualitative points of view. The elemental carbon derives mainly from combustion processes and is associated to impacts on health (affinity with toxic organic pollutants) and on climate (positive radiative forcing). On the other hand, the organic fraction of the aerosol is a complex mixture of substances that are of interest because of their possible toxic effects (e.g. PAHs) or because they can be used as tracers for specific pollution sources (e.g. levoglucosan). There are three methods used to apportion the carbonaceous aerosol: isotopic ratios combined with macrotracer enrichment factors (IRMEF; e.g. Gelenccsér et al., 2007), organic molecular markers processed with CMB (e.g. El Haddad et al., 2011) and aerosol mass spectrometry (AMS; Zhang et al., 2011). About 20% of the European studies with Receptor models used these techniques (Belis et al., 2013).
- Hybrid receptor models utilize multivariate factor analysis methods and accept explicit

introduction of information (in addition to the chemical composition) to reduce the ambiguity of the solution (e.g. Amato et al., 2009). Application of hybrid models takes advantage of information on pollutant physical and chemical properties and on the processes that influence them. At present, ready to use tools, that allow a flexible implementation of these models depending on the available data, are at expert's disposal.

- The aethalometer receptor model is used to quantify the contribution of traffic and biomass burning to aerosol on the basis of wavelength absorption ratios (e.g. Sandradewi et al., 2008). Source identification with this simple technique is likely to grow consistently in the near future thanks to the increasing number of measures performed with aethalometers.

None of these advanced methodologies have been reported in the answers to the questionnaire.

- Among the Eulerian models used for source identification, those with source tagging modules are providing the most reliable results since the calculation of the origin for selected pollutants are carried out at every step of the simulation and therefore fully integrated in the model algorithm. Examples of this type of tools are: CAMx PM Source Apportionment Technology (PSAT), CMAQ Tagged Species Source Apportionment (TSSA) and LOTOS-EUROS with labelling. Only one of the studies in the questionnaire reported having implemented one of these advanced methodologies.

6 What is the contribution of SA to the Air Quality Directive and to Integrated Assessment Modelling

Information on pollution sources is essential for air quality management. The implementation of the Air Quality Directives encompasses many steps or activities for which identification of sources is relevant (Tables 3 and 4).

The effectiveness of any type of remediation measure strongly depends on the reliability of pollution source identification and quantification. Hence, the use of methodologies with the least feasible biases and uncertainties certainly contributes to convey valuable resources and time on abating only those sources which actual contribution in the area of interest is significant.

A straightforward way to identify pollution sources is simply taking the information from emission inventories as representative of the contribution of sources to ambient air in a given geographic area. Although emission inventories are essential in understanding the relevance of sources, it must be considered that a number of factors, like physical and chemical processes or advection, determine the actual levels of pollutants in the atmosphere at a given point.

In other cases, information about sources derives from previous studies that support "a priori" decisions on what are the activity sectors to focus on (e.g. traffic, point sources) or provide quantitative input for other models.

In the questionnaire presented in this report, only 60% of the IAM studies have explicitly identified sources using one of the methodologies listed among the multiple choices. This suggests that the identification of relevant sources is in many cases embedded in the steps of the modellistic chains that are used to accomplish the complex set of tasks leading to the final output. Nevertheless, the explicit application of source apportionment models could significantly contribute to the overall performance of the IAM by optimizing an early step in the process that is relevant in steering the following ones into the appropriate direction.

In the overall IAM framework, source apportionment methodologies can bring added values at different stages of the process:

- During the set-up phase of an IAM framework the identification of the key emission sources in the area of interest would allow a better delimitation of the problem and therefore to allocate resources to study in depth the identified more relevant sectors of activity (e.g. no need to invest resources to get details on emission sectors which are of minor importance)

- One of the key aspects determining the overall robustness of the IAM system is the evaluation of the air quality modelling system used to derive the source-receptor relationships. Although the information retrieved from source apportionment studies is not always fully compatible with the output of AQ models, the comparison of the two approaches will certainly result in a better quality and understanding of the whole system.

- SA could be also used to determine the boundary conditions, in particular the amount of pollution originating from outside the considered domain where the IAM system is applied. One of the ways to retrieve this information is, of course, with the use of larger scale models but SA methodologies (especially those involving Lagrangian models) could help assessing this component as well.

- There could be a synergistic use of SA and IAM techniques like scenario analysis or optimization based approaches, such as cost-benefit, cost-effectiveness, multi-objective approaches. To that end, SA could drive the choice of the emission patterns to be tested through scenario analysis, to limit the number of simulations to be performed through a CTM. In alternative, it could limit the degrees of freedom of cost-effectiveness analysis, constraining the optimal solution to consider only a subset of the possible emission reductions previously identified applying SA.

Table 3. Activities explicitly mentioned in Directive 2008/50/EC for which identification of sources is relevant

Activity	Description
Reduction of emissions at source (Preamble point 16)	One of the overarching principles of the Thematic strategy on air pollution is the reduction of emission at source. Indeed, quantifying the extent to which such sources actually impact on air quality and therefore assess the effectiveness of their reduction requires source apportionment.
Assessment of air quality (article 5)	Each zone and agglomeration shall be classified in relation to the assessment thresholds. This classification shall be reviewed at least every five years or less if significant changes in activities relevant to the ambient concentrations of pollutants occur.
Natural sources, road salting and sanding (articles 20 and 21)	In order to subtract exceedances attributable to natural sources or winter sanding or salting of roads, Member States are requested to provide evidence to support their claims.
Localization of monitoring stations (Annex III B item c)	Urban background locations shall be located so that their pollution level is influenced by the integrated contribution from all sources upwind of the station.
Background measurements (Annex IV A)	Information on background levels in rural areas is essential to judge the enhanced levels in more polluted areas, assess long-range transport, support source apportionment analysis and for the understanding of specific pollutants such as particulate matter. It is also essential for the increased use of modelling also in urban areas.
Ozone precursors (Annex X A)	Measurements of ozone precursors are requested to monitor their trend, to check the efficiency of emission reduction strategies, to check the consistency of emission inventories and to help attribute emission sources to observed pollution concentrations.
Local, regional and national air quality plans (Annex XV A item 5)	Lists, maps, emitted quantities and transboundary nature of main emission sources responsible for pollution are to be provided when drafting air quality plans.
Public information (Annex XVI item 4)	Timely information about actual or predicted exceedances of alert thresholds, is to be provided to the public, including indication of main source sectors or categories and recommendations for action to reduce emissions.

Table 4. Activities explicitly mentioned in Directive 2004/107/EC for which identification of sources is relevant

Activity	Description
Target Value exceedances (Article 3 item 3)	Aiming at implementing measures to attain target values, MS are requested to specify zones and agglomerations where such values are exceeded and to indicate source contributions.
Transmission of information and reporting (Article 5 item d)	MS shall forward to the Commission information concerning the sources contributing to the exceedances.

7 Conclusions and summary

In recent years the number of studies on source apportionment in Europe has steadily increased. This is closely related to the continuous development of tools with improved functionalities and performance. Nevertheless, the lack of an appropriate European network of urban monitoring sites with enhanced chemical and physical characterization of aerosols is becoming a limiting factor for a further growth and consolidation of SA techniques for these important pollutants.

This is true for both Receptor models and Eulerian models. In the first case, speciated PM are input data essential for model execution. Moreover, detailed information on chemical and physical properties of aerosol would benefit also CTM models making it possible to test and improve their ability to reproduce not only the total mass of PM, but also important mass fractions like nitrates, sulphates, organic and elemental carbon, or toxic components like heavy metals and PAHs.

Also the partial information on European source emission factors and source fingerprints is limiting source identification. At present, there is a great deal of information on certain type of sources, like point sources or engine exhaust, for which legislation on standardization and emission control has greatly improved in recent years. Nevertheless, small diffuse sources with high variability from site to site and for which registration and standardization are not feasible, like combustion of solid fuels (wood and coal), natural sources and cooking emissions, are still poorly characterized and their contribution to atmospheric pollution is quite uncertain. More work is needed to chemically characterize and compile European databases for this type of sources.

Obtaining reliable source contribution quantification requires tools the performances of which have been tested and documented. For this purpose, validation procedures and quality assurance tests should be carried out in any study and the estimation of the output uncertainty reported.
Moreover, the definition of European methodological protocols to guarantee a minimum level of quality and to make results from different studies comparable is required. Therefore, the information obtained in intercomparison or benchmarking exercises is essential in understanding source apportionment model performances and uncertainties. On this regard, the European intercomparison exercises for Receptor models have concluded that the contribution estimations are compliant with a quality objective equal to 50% of relative uncertainty (Karagulian et al, 2012).

From the methodological point of view, both Receptor models and Eulerian models in combination among each other and with complementary techniques, the main of which are Lagrangian models, appear as the most dynamic areas in the evolution of source apportionment tools.

Recent studies demonstrate that, to abate exceedances of air quality limits, sources of secondary inorganic aerosol like traffic (nitric oxides) and agriculture (ammonia) are the most important source categories to target throughout the year together with biomass burning during the cold season.
Future studies on source apportionment in Europe should focus on:

a) the relevance of gas-to-particle conversion and photochemical processes as the main contributors to recalcitrant pollutants like PM and ozone,
b) sources of specific aerosol fractions (e.g. carbonaceous) or micro pollutants present in the particulate (heavy metals, PAHs) that have relevant impacts on health and/or climate,

c) identification of nitric oxides sources due to the number of exceedances and the contribution of these pollutants as precursors for both PM and ozone.

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ANNEX 1. SOURCE APPORTIONMENT CASE STUDIES

Utilization of source apportionment techniques is expanding not only in Europe but also in Africa, Asia and South America (Johnson et al., 2011). These techniques are increasingly aiding environmental compliance and answering policy relevant questions like: What sources to target for pollution abatement efforts? Where to fix the target? and How to achieve the target? (Johnson et al., 2011).

European coal combustion regions

In the European coal combustion regions – typical for many areas in the new EU member states – residential heating by coal (or coal substitutes) combustion was proved to be a major PM pollution source. In a study carried out during winter pollution episodes in Cracow (Poland) using Chemical mass balance (CMB) and constrained positive matrix factorization (CMF), residential coal combustion in small stoves and boilers was found to be a main source of PM_{10} and benzo(a)pyrene (Junninen et al. 2009). Severe winter PM episodes in five Polish cities (Szczecin, Warsaw, Cracow, Zabrze, Jelenia-Góra) were also studied by Juda-Rezler et al. (2011) using a combination of backward air mass trajectories, Lenschow (or incremental) approach and principal component analysis (PCA). The study showed that traffic as well as coal combustion in both industry and residential sector were the main sources of PM_{10}. Similarly to the Junninen et al. (2009) study, the main problem of Juda-Rezler et al. (2011) investigation was to differentiate the source profiles of coal combustion in different utilities, i.e. residential stoves/boilers, industrial high-efficiency boilers and power plants, due to the collinearity (resemblance) of these sources. PCA analysis performed in a rural background site in Diabla Góra (Poland) showed that As measured in relatively clean rural location represents a marker of industrial coal combustion (and regionally transported pollutant), while Cr results the marker of residential coal combustion (local pollution).
Also the source apportionment study conducted in Upper Silesian city of Zabrze, by PCA coupled with multi-linear regression analysis (PCA-MLRA), pointed out the residential coal combustion as the main source of $PM_{2.5}$ (Rogula-Kozlowska et al., 2013). Moreover, Trapp (2010) in his dispersion modelling study with Gaussian puff model (CALPUFF) apportioned the same sector as main source of PM_{10} concentrations in urban areas in Central Poland.

Pollution sources in Portugal

Almeida et al. (2006) carried out a chemical characterization for $PM_{2.5}$ and $PM_{2.5–10}$ samples collected in a suburban area, aiming to evaluate the performance of Multilinear Regression Analysis (MLRA) and Mass Balance Analysis (MBA) in the determination of source contribution to Particulate Matter (PM) concentrations. MLRA and MBA showed very similar results for the temporal variation of the source contributions. However, quantitatively important discrepancies were observed, principally due to the lack of mass closure in $PM_{2.5}$ and $PM_{2.5–10}$. Both methods indicated that the major $PM_{2.5}$ aerosol mass contributors included secondary aerosol and vehicle exhaust. In the coarse fraction, marine and mineral aerosol contributions were predominant.
Moreover, Almeida et al. (2005) applied PCA and MLRA to identify possible sources of PM and to determine their mass contribution at a sub-urban area located in the north outskirts of Lisbon, a Southern-European city. Seven main groups of sources were identified: soil, sea, secondary aerosols, road traffic, fuel-oil combustion, coal combustion and a Se/Hg emission sources. In $PM_{2.5}$, secondary aerosol and vehicle exhaust contributed on average, with 25% and 22% to total mass, respectively, while sea spray and soil represented, respectively, 47% and 20% of the coarse fraction mass loading. Maritime air mass transport has a significant role on air quality in the North of Lisbon. The highest PM levels were recorded during South Continental episodes. These episodes are characterized by high mineral aerosol contents, due to the transport of dust from the interior of Iberian Peninsula and the Sahara desert.

Developing countries

Johnson et al. (2011) in their review of source apportionment techniques in developing countries identified 11 common PM source categories, which they grouped into 4 main types: (1) dust (road dust, other dust including: soil dust, re-suspension, fugitive dust and construction), (2) transport (gasoline, diesel), (3) industry (industry & commercial, including: oil burning & brick kilns, coal burning, power plants), and (4) non-urban (biomass & open burning, long range transport, marine, others). The case studies were conducted in 18 developing country cities over the last decade. The receptor model applied most frequently was CMB, followed by PMF, APFA, APCA, and PSCF. The following source apportionment case studies were performed:

- by CMB: in Shanghai and Beijing (China); Mumbai, Delhi, Kolkata and Chandigarh (India); Cairo (Egypt); Qalabotjha (South Africa); Bangkok (Thailand) and Addis Ababa (Ethiopia);
- by PMF: in Beijing (China); Dhaka and Rajshahi (Bangladesh); Hanoi (Vietnam) and Mexico City (Mexico);
- by APFA: in Sao Paulo (Brazil) and Santiago (Chile);
- by APCA: in Xi'an (China);
- by PSCF: in Hanoi (Vietnam).

The most common PM sources identified in these cities were dust emissions, followed by urban clusters of small-scale manufacturers. In the rural areas and in secondary cities, biomass burning was one of the major sources of pollution.

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ANNEX 2. TOPIC 4 QUESTIONNAIRE

(note: not all the choices of each question are visible in this facsimile)

TOPIC 4: Source apportionment

1. **What was the purpose of the source apportionment study?**

Identify causes of exceedances
Detract natural sources or road salting and sanding from PM (Dir. 2008/50/EC art. 21)
Apply for postponement of attainment (Dir. 2008/50/EC art. 22)
Design air quality plans/ action plans (Dir. 2008/50/EC arts. 23 and 24)
Identify the contribution from different geographic areas within a country
Assess remediation measures effectiveness
Refine emission inventories
Identify the contribution from other countries (transboundary pollution - Dir. 2008/50/EC art. 25 and 24)
Other

2. **What was the used methodology?**

☐ receptor modelling (select type)

tracer method
PCA
CMB
PMF
Hybrid models
Incremental approach (Lenschow)
Other — please, specify

☐ dispersion modelling (select type)

lagrangian model
gaussian model
Other — please, specify

☐ inverse modelling

☐ objective estimation techniques (e.g. statistical models, spatial interpolation of measured data, statistical relationship between emission density/traffic data/meteorology fields and air pollution levels etc.)

☐ other

3. **What were the considered source categories**

SNAP1-combustion in energy and transformation industries
SNAP2-non-industrial combustion plants
SNAP3-combustion in manufacturing industry
SNAP4-production processes
SNAP5-extraction and distribution of fossil fuels and geothermal energy
SNAP6-solvent and other product use
SNAP7-road transport
SNAP8-other mobile sources and machinery
SNAP9-waste treatment and disposal
SNAP10-agriculture
SNAP11-other sources and sinks
Other — please, specify

4. What were the pollutants considered

PM25
Lead
Nitrogen dioxide (NO2)
Ozone (O3)
Sulphur dioxide (SO2)
carbon monoxide (CO)
Nitrogen oxides (NOx)
Arsenic
Cadmium
Nickel
benzo(a)pyrene
Other — please, specify

5. Study Area

☐ Local

Urban roadside
Industrial

☐ City
☐ Region
☐ Country
☐ Other

6. Types of input data used (depending on type of SA methodology)

Field campaign results
Routine ambient air quality measurements (e.g. from monitoring network)
Source profiles
Meteorological fields
Backward trajectories
Other — please, specify

7. Sampling design (only for receptor modelling methodology for SA)

Sampling site number and type	please, specify
Number of samples	please, specify
Sampling period(s) (to include seasonal variation within a year)	please, specify
Sampling analysis (equipment used)	please, specify
Sample chemical (or physical) analyses (used technique)	please, specify

8. Open questions:

8.1. Explain to what extent was it possible to achieve the objectives of your source apportionment study and what were the limitations of the used methodology?

8.2. Are source apportionment capacities in the institutions of your country/region/city enough to achieve reliable source identification and support air quality management?

8.3. Is data collection in your country/region/city appropriate for source identification?