JRC TECHNICAL REPORT

European guide on air pollution source apportionment for particulate matter with source oriented models and their combined use with receptor models

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2020

Joint Research Centre

EUR 30082 EN

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EU Science Hub
https://ec.europa.eu/jrc

JRC119067

EUR 30082 EN

PDF ISBN 978-92-76-10698-2 ISSN 1831-9424 doi:10.2760/470628

Luxembourg: Publications Office of the European Union, 2020

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How to cite this report: Mircea M., Calori G., Pirovano G., Belis C.A., *European guide on air pollution source apportionment for particulate matter with source oriented models and their combined use with receptor models*, EUR 30082 EN, Publications Office of the European Union, Luxembourg, 2020, ISBN 978-92-76-10698-2, doi:10.2760/470628, JRC119067.

Contents

Acknowledgements 1

Abstract 2

1 Introduction 3

  1.1 Scope and aims 3

  1.2 Target audience 4

  1.3 Why use receptor models (RMs) and source-oriented models (SMs) for SA...5

2 SA techniques: description and applications 7

  2.1 Techniques for SA using RMs 7

  2.2 Source oriented air quality models 8

  2.3 SM approaches for SA 11

  2.4 European SA studies with SMs and with SMs-RMs: survey results 19

3 Estimation of source contributions with SM approaches 22

  3.1 Modelling and validation of PM - base case of SM approaches 22

  3.2 Sensitivity analysis methods 24

  3.3 Tagged species methods 25

4 Combined use of SMs and RMs 26

  4.1 Source categories association and species/compound association for PM validation 26

  4.2 Comparison of source contributions from SMs and RMs 31

5 Intercomparison between different SMs and between SMs and RMs 35

References 39

List of abbreviations and definitions 47

List of figures 48

List of tables 49

Annex 1. Applications of SMs and SMs-RMs models for estimating particulate matter source contributions in Europe Annexes 50

  A1. Air quality management: 50

  The origin of ambient particulate matter concentrations in the Netherlands 50

  Establishing the contribution of agriculture to particulate matter in Flanders 51

  On the origin of particulate matter in Berlin, Germany 51

  Study 1: BelEUROS 52

  Study 2: RIO-IFDM-OSPM 53

  SA studies with air pollution models in support to AQPs in Germany 54

  SA studies with air pollution models in support to AQPs in Slovakia 56

  Source Apportionment studies in Italy at national and regional scales 56

  A2. Other studies 57

  The study performed in the frame of the MED-APICE project (www.apice-project.eu) 57

Acknowledgements

The authors express their gratitude to:

S. Banzhaf, Institut für Meteorologie, Freie Universität Berlin

L. Ciancarella, ENEA

H. Hebbinghaus, LANUV Nordrhein-Westfalen

H. Hooyberghs, VITO

J. Mateiovicova, Slovak Hydrometeorological Institute

P. Prati, University of Genoa

M. Schaap, TNO

P. Viaene, VITO, and

S. Wurzler, LANUV Nordrhein-Westfalen

for sharing their experiences on applications of SMs and SMs-RMs on particulate matter source contributions in Europe.

The authors are grateful to Jean-Luc Jaffrezo, Philippe Thunis, Alain Clappier, Stig Hellebust for their comments and suggestions on the draft version of this document. Special thanks to all the Fairmode WG3 members for their constructive criticism that led to fruitful discussions during the meetings.

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Abstract

Particulate matter is one of the most critical atmospheric pollutants, in terms of impact on human health, in Europe and worldwide. Understanding the role of the different sources and processes leading to its formation and transformation/evolution is essential to support the development of abatement strategies. The present guide, developed under FAIRMODE WG3 with the contribution of WG4, provides an overview, illustrates the methodologies and formulates recommendations for the application of air quality source-oriented models (SMs) in estimating contributions of sources to particulate matter. It also describes the potential of SM techniques and critically discusses the advantages and the limitations of applying simultaneously receptor models (RM) and source-oriented models (SM) in source apportionment (SA) studies. Among the source-oriented modelling methods, the document focuses on those using the Eulerian chemistry-transport models (CTMs) since they simulate both primary and secondary particulate matter directly emitted and formed from gas precursors in the atmosphere, respectively.

1 Introduction

Particulate matter (PM) adverse effects on human health are well documented. In the European Region, the life expectancy of population is reduced up to 8.6 months on average (WHO, 2013) due to exposure to high ambient PM concentrations.

Particulate matter is a complex mixture of aerosol particles, either emitted directly by natural or anthropogenic sources (primary) or formed in atmosphere through physical and chemical processes from gas precursors (secondary). In addition, the formation of new PM in atmosphere is influenced by the amount and the physical and chemical characteristics of the pre-existing aerosol particles. Most of the gas precursors and aerosol chemical compounds are emitted by multiple sources posing further problems to the identification and quantification of individual source contributions to atmospheric concentrations of PM. In Europe, PM_{10} and $PM_{2.5}$ limit values set by the Air Quality Directive (AQD, 2008/50/EC) are exceeded in several countries (EEA, 2018).

Source apportionment (SA) is the technique which relates a source emission (an activity sector or an area) to the ambient air concentration of a pollutant.

In order to protect human health and the environment as a whole, the reduction of pollution levels by combating the "emissions of pollutants at source (EU Air Quality Directive, 2008/50/EC) is necessary. According to the Annex XV of the AQD, information about sources is to be provided to establish the causes that determined an exceedance in order to design and evaluate the Air Quality Plans (AQPs). SA outputs are therefore among the data that Member States (MS) are requested to report to the Commission via the e-reporting scheme (Implementing Provisions for Reporting, IPR). Others applications of SA in the frame of the AQD are the estimation of the contribution of natural sources (Article 21), winter sanding and salting (Article 22) and long-range transport of pollutants in background sites (Annex IV A), among others.

The exceedances caused by natural sources and winter sanding/salting are not to be considered for the purposes of the AQD. The contributions from these sources to high PM levels (Art. 2 par 15, EU, 2008) can be estimated based on two existing EU guidelines: guidelines for demonstration and subtraction of exceedances attributable to natural sources such as atmospheric re-suspension or transport of natural particles from dry regions (http://ec.europa.eu/environment/air/quality/legislation/pdf/sec_2011_0208.pdf) and guidelines for determination of contributions from the re-suspension of particulates following winter-sanding or -salting of roads" *(http://ec.europa.eu/environment/air/quality/legislation/pdf/sec_2011_0207.pdf)*, which address them extensively.

In addition to the methods proposed in the guides mentioned above, the contribution of different emission sources to PM can be identified and quantified by two main families of modelling techniques: receptor-oriented models (RMs) and source-oriented models (SMs).

RMs identify and estimate the PM sources following the methodologies described in the EU report (Belis et al., 2014; Belis et al., 2019) from concentration measurements of different PM components at a given location, within an area representative for measured PM levels.

SMs perform grid source apportionment over a given area/domain using air quality models which have as input data the emission inventories and meteorological fields as well as air concentrations at the boundaries of the area/domain.

The air quality models are often used to evaluate the contributions of sources in the absence of measurements or as a complement to them. Therefore, the FAIRMODE (Forum for Air Quality Modelling in Europe; http://fairmode.ew.eea.europa.eu) working group 3 (WG3) has carried out activities to test and harmonise the use of both RMs and SMs and the main outcomes are synthesised in this guide while more details can be found in Belis et al. 2020.

1.1 Scope and aims

The AQD has set up an air quality management scheme under which Member States (MS) **identify the main causes** of concentrations above the AQD limit values (exceedances). The MS assess whether the exceedances are **attributable to adverse climatic conditions, transboundary contributions, site-specific dispersion characteristics, or to a combination of these factors.** Furthermore, source apportionment techniques are used to identify and quantify **the contributions from main pollution sources with the purpose to provide**

understanding on what measures should be taken to address them. Pursuant to Article 20 of the AQD regarding the exceedances, contributions from natural sources such as Saharan dust, sea salt spray are also taken into account in the evaluation of the exceedances and can be subtracted if documented.

Several guides provide support for PM modelling:

(*http://acm.eionet.europa.eu/reports/ETCACM_TP_2013_11_FAIRMODE_guide_modelling_PM*),
air quality model evaluation:

(*http://fairmode.jrc.ec.europa.eu/document/fairmode/WG1/Guidance_MQO_Bench_vs2.1.pdf*)
and for source apportionment performed with receptor models:

(*https://ec.europa.eu/jrc/en/publication/european-guide-air-pollution-source-apportionment-receptor-models*).

The present guide completes this suite and aims to provide an overview and recommendations for the application of air quality models in estimating source contributions to PM (SM techniques). It also shows the potential of these techniques and critically discusses the needs and the limitations when applying simultaneously RM and SM techniques in source apportionment studies.

Among SM methods, the document focuses on Eulerian chemistry-transport models (CTMs) since they simulate both primary and secondary particulate matter which are directly emitted and formed from gas precursors in the atmosphere, respectively. The secondary particulate matter can account up to 70% of PM_{10} in urban areas with a consequent high impact on population health.

This document does not focus on the validation of modelled concentrations addressed by WG1 (assessment) nor the emission inventory accuracy addressed by WG 2 (emissions). Nevertheless, it discusses the sensitivity analysis methodology which is commonly used for planning (WG4).

This document does not provide information on the “fitness for purpose” of the different methods because this topic is specifically addressed in a complementary FAIRMODE guidance under preparation which is focused on this issue (Thunis et al., 2019).

This document focuses on the estimation of the set of main pollution sources, in terms of activity sectors (e.g. SNAP, NFR) and geographical origin (transboundary contributions, natural contributions, etc.), affecting a specific area in a given time window. The studies oriented to characterise a single source are not primarily addressed in this document.

This document together with the guidance on the “fitness for purpose” of the different methods can also help in preparing additional documentation to support Member States in the e-reporting process of PM exceedances. In this regard, the SM approaches provide more comprehensive information on the geographical origin of pollutants than the RM methods.

1.2 Target audience

This document is intended to provide the basis for organisations, companies and institutions to carry out source apportionment studies in support of authorities responsible for air quality management according to the AQD including the development, implementation and evaluation of official air quality plans. As mentioned in the FAIRMODE recommendations (Thunis et al., 2019), source apportionment guides the choice of the most effective mitigation strategies and measures to include in air quality plans (AQPs) which generate future emission scenarios, after the identification and quantification of sources which have major contributions to high pollution levels. Source apportionment plays a key role in assessing the effectiveness of AQPs and, in combination with epidemiological studies, to assess the impacts on health and vegetation due to changes of pollution sources.

Besides, source apportionment techniques are powerful methods for understanding the way in which CTMs reproduce atmospheric processes and, consequently, provide a mean for the validation and the improvement of such models. The study of pollution sources is also relevant for the analysis of climate change strategies.

The document also provides a framework to assist users in the interpretation of modelling results for source apportionment purposes. Moreover, it also aims to help the air quality modellers, independent of their level of experience, to get introduced to the source apportionment methodologies.

1.3 Why use receptor models (RMs) and source-oriented models (SMs) for SA

The quantitative contributions of different sources to ambient air pollution cannot be measured directly. Advection processes disperse local emissions and transport emissions from regions outside the study area. In addition, the secondary processes change the physical properties of pollutants (e.g. gas to solid) and convert some of them (precursors) into others (secondary pollutants). Therefore, RM and SM models are needed to estimate the contribution from individual sources to the air concentration of pollutants. This information is fundamental for air quality management providing the basis for the air quality plans (Directive 2008/50/EC) and to link human exposure to individual sources which are influencing human health events (Laden et al., 2000; Thurston, 2017; Weidemann et al., 2016).

Both SA techniques have limitations inherent to their formulation and data availability. However, these techniques have evolved considerably in the last decade thanks to the high interest demonstrated by the scientific community.

RMs apportion the measured mass of an atmospheric pollutant at a given site to its emission sources. This technique has been used for SA at local level in many locations of the world (e.g. Zhang et al., 2007; Jimenez et al., 2009; Karagulian et al., 2015). RMs are commonly used to estimate the pollution sources attributed to the total PM, along with associated uncertainties. RMs apportion the measured mass of an atmospheric pollutant providing information derived from real-world data. The applications and the number of publications have continuously increased over the last decades and the capabilities of RMs are continuously improving (Belis et al., 2019).

The use of RMs has the following advantages:

- derives information about sources from measured data;
- estimate the contribution of sources for most of the PM chemical components;
- does not require an extensive input data set (e.g. 3D meteorological data, 3D emission data, air concentrations at boundaries);
- does not require significant computing resources and data storage is negligible;
- the uncertainty of the output is estimated.

On the other hand, RM studies provide information for specific sites and time windows. For epidemiological studies, long time series (greater than 2 years) would be necessary, however, these are not always available (Sarnat et al., 2008). Source identification with RM techniques is also limited in case of co-linearity between source profiles (i.e. sources with similar composition impacting a receptor site) and because some methods like CMB requires prior knowledge of the composition of the emission sources.

Information on the sources of air pollutants can also be obtained by applying SMs that mimic the physical and chemical processes taking place in the atmosphere in the presence of emissions of pollutants. Common SM approaches are sensitivity analysis methods (also known as brute force approach and more recently as emission reduction potential - ERP) in which separate model runs are performed, each one considering a different set of sources of interest and tagged species methods that use bookkeeping to record the different proveniences of the species masses throughout a unique model run (see section 2.2).

The use of SMs has the following advantages:

- it is not limited to sites where monitoring data are available, thus it evaluates the contributions of sources in the absence of measurements or as a complement of them;
- it can predict air quality changes in relation to emissions changes and therefore, can be used to develop effective air quality plans to reduce pollution (sensitivity analysis methods);
- the definition of the sources is directly linked to the emission inventories, so it can be detailed in terms of activity sectors and subsectors;
- estimates the contribution of different emission sectors to secondary pollutants through the air concentrations at boundaries;
- estimates the contribution of transported pollutants;
- the individual effects of meteorology can be isolated;

- it can provide output with high temporal resolution (hourly);
- it is possible to explore the variability in time and space of source contributions;

However, the results of SM approach are limited by the quality of the input data (emissions, meteorology) and by the formulation of the chemical transport model (CTM) used. The measurements available for model validation as well as the experience of the model user also impacts on the SM outcome.

As stated in the "Guidance on the use of models" (EEA, 2011), "It is the aim of FAIRMODE to promote these advantages and to address these disadvantages". In particular, activities in FAIRMODE aim to improve and harmonise the quality assurance of models to provide transparency and increased understanding of their applicability, in this case, for SA techniques addressed in WG3.

The combined use of RMs and SMs can provide a more comprehensive and reliable evaluation of source contributions given the fact that are based on different and independent input data. In recent years, some studies have combined RMs and SMs aiming to reduce uncertainty in source impact estimates (Hu et al., 2014; Ivey et al., 2015).

2 SA techniques: description and applications

2.1 Techniques for SA using RMs

Receptor-oriented models (RMs) source apportionment encompasses many tools ranging from simple techniques based on elementary mathematical calculations and basic physical assumptions (e.g. enrichment factor analysis) to complex models with pre- and post-data processing and user-friendly interfaces. Although all such tools deal with measured pollutant concentrations at the receptor site, the type of the input data may vary considerably. In general, there are three kinds of data input: ambient pollutant mass concentrations, source profiles, and meteorological data (i.e. wind speed and direction or backward trajectories). In addition, there are extended models, which can take into account other kind of information like season, weekday, precipitation, etc.

Common for all, the multivariate RMs is the basic equation that refers to the solution of the mass balance problem (Eq. (1)):

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

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

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

1. *source profiles* do not change significantly over time or do so in a reproducible manner;
2. receptor species do not react chemically or undergo phase partitioning (solid/gas or solid/ liquid) during transport from source to receptor (i.e., they add linearly).
3. data are representative of the studied geographical area and consistent with the conceptual model
4. comparable/equivalent analytical methods are used for the receptor site(s) throughout the study as well as for the characterization of the source profiles.

RM approaches

Some RM approaches are briefly described below. More details are available in the first and second edition of the guide on source apportionment with receptor models (Belis et al., 2014 and Belis et al., 2019; respectively) and in the scientific reviews on this topic (Watson et al., 2008; Viana et al., 2008; Hopke, 2010; Belis et al., 2013)

Incremental approach or Lenschow approach

This approach was applied primarily for source contribution estimations (SCEs) of traffic emissions to PM_{10} mass concentrations in urban agglomerations (Lenschow et al., 1999). Incremental methods (or "Lenschow" approaches) based on spatial gradients of concentration, calculated as the difference between concentrations at two specific locations. They depend on the assumption that source contributions can be derived from the differences between the concentrations. This approach is based on the assumptions that: a) the regional contribution is constant outside and inside the urban area, and b) the city does not contribute to the regional background. This approach is only applicable to determine contributions from different spatial sources to concentrations. For a detailed discussion of incremental approach refer to Thunis (2018). The incremental approach, in principle, could be implemented also by means of SMs, but as for the latter the application is less meaningful because SMs can investigate directly the role of a geographical source without introducing any incremental term.

Chemical Mass Balance

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

The accuracy of a CMB modelling result depends strongly on the availability of source profiles, which ideally are from the region where the receptor is located and that should be contemporary with the underpinning ambient air measurements. CMB requires a good knowledge of the emissions in the study area in order to assure that all relevant sources are included and to evaluate their uncertainty. Because of CMBs sensitivity to collinearity of the source profiles, which impedes the mathematical solution of the mass balance, it is often necessary to merge sources into groups of source types in order to produce composite profiles.

Positive Matrix Factorisation

Factor Analytical methods operate linear transformation of the original variables to create a new set of variables, which better explain cause-effect patterns. The input for this kind of method is measured receptor species concentrations associated with ambient PM (x_{ij}). Positive Matrix Factorisation (PMF) is the most commonly used factor analytical technique. It resolves a weighted factorisation problem with non-negativity constraints using known experimental uncertainties as input data thereby allowing individual treatment (scaling) of matrix. The most recent versions of the US-EPA software solve the PMF problem via the Conjugate Gradient algorithm and contain routines to estimate the optimal number of factors, to test the rotational ambiguity and to introduce constraints. The uncertainty and stability of the solution are estimated by bootstrapping and displacement methods.

2.2 Source oriented air quality models

These models simulate the distribution of concentrations of airborne pollutants over a given area starting from some knowledge of emission sources over that area and then describe with various level of detail the processes affecting the pollutants during their lifetime in the atmosphere: advection by wind, turbulent mixing, chemical and physical transformations, depletion by deposition. The linkage between sources and ambient concentrations that source oriented models reconstruct allows, among other applications, a possible quantitative assessment of the relationship between emission of a given source and its corresponding air concentration. This can be achieved in different ways, depending on the type of model employed and the technique that is then used to assess the contributions.

The source-oriented models that are most often employed for this purpose (Fragkou et al., 2012) fall in three broad categories:

- Gaussian and non-Gaussian parametrised models;
- Lagrangian puff and particle models;
- Eulerian chemical transport models.

Classical introductions and advanced references for these main modelling techniques can be found in Hanna et al. (1982), Turner (1994), Zannetti (1990), Jacobson (1998), Pielke (2001) and Seinfeld and Pandis (2006).

Although other types of models were applied for SA, in this brief overview we focus on the most commonly used ones.

Gaussian and non-Gaussian parametrised models reconstruct sequences of steady-state situations (e.g. on hourly basis), assuming for each of them either a uniform or simplified meteorology. The basic formulation describes a straight plume originating from a point source, with diffusion around plume axis parametrised according to meteorological categories and terrain type. Various formulations also allow modelling plumes of pollutants coming from sources with different geometry (i.e. line, area/volume); chemical and physical transformations are usually neglected by these types of models.

Lagrangian puff and particle models dynamically follow the pollutants released from sources through a series of computational elements moving independently in a three-dimensional flow field. Turbulent diffusion is then described with different approaches and details, depending on model type: the puff-based ones usually use Gaussian-like mass distributions around the barycentre of each puff, while the particle-based ones move the particles according to stochastic components of the motion that depend on local turbulence conditions. Lagrangian models can deal with sources with different geometry, initially shaping the puffs or releasing the particles according to sources position and form. Lagrangian models may include depositions and chemical transformations, usually in simplified way, while often they consider the pollutants as non-reactive.

Eulerian chemical transport models dynamically simulate pollutants' dispersion in a three-dimensional framework, using a fixed grid, to which all variables are referred (emissions, meteorology and pollutants concentrations) and respect to which all physical and chemical processes are modelled. The grid resolution implies a limit to the detail in describing pollutants behaviour that can be achieved; in particular, source geometry is reduced to the grid-cell geometry, with potential loss of sub-grid detail that may exist. The identity of individual sources inside each grid cell is also lost, since, for a given cell, the emissions and the concentrations represent the total amount of the pollutant released/ present in that portion of the atmosphere. On the other hand, this allows straightforward modelling of the physical processes and chemical reactions affecting pollutants, since such processes and reactions involve in fact the total amount of the different species at a given location. In particular, in case of secondary pollutants, this may involve non-linear relationships between emission sources of primary components and resulting ambient concentrations. Eulerian models may be also employed in non-reactive mode, i.e. neglecting chemical transformations.

Model application

In general, the choice of the model to be used should be made according to the application of interest, following 'fit-for-purpose' criteria. As effectively summarised in (EEA 2011) and (ETC/ACM 2013), the following aspects should be considered in choosing an air quality model:

- Does it cover an adequate area with the necessary spatial and temporal resolution?

- Does it include the representation of the relevant physical and chemical processes at that scale?

- Is it fed by emission information concerning the most relevant sources and by suitable meteorological data?

- Is it well documented and validated for the specific application?

The spatial scale of the assessment to be performed implies a first criterion for the type of model to be used. In general, for small areas where contributions from nearby sources to the primary component of the particulate matter concentrations are relevant, high resolution models capable of accounting for source geometries may be preferred. On the other hand, for large areas with possible contributions from multiple sources at different distances, the secondary components may be important, so models allowing to account for physical and chemical processes leading to their formation are needed. Gaussian models may be used for local/hotspot and urban/agglomerate applications, while at the other end the Eulerian ones are better suited for scales ranging from urban/agglomerate to regional/continental. Lagrangian models may span different spatial scales, although the larger scale ones may need to include chemical transformations.

Another important issue is which components of particulate matter are most relevant for the application of interest. At scales from local to urban, local and low height sources are usually the most influential ones, and the primary components are the most important ones to be modelled. In such cases Gaussian and Lagrangian models, which typically neglect the transformations leading to secondary components, may be used. It is important to note that by using solely these type of models, only a part of the actual atmospheric PM can be described, since the secondary part produced from city precursors is often not treated by the model. Moreover, in most cases the computational domain of local scale models is limited to the urban area even to the city centre, therefore also the PM (primary and secondary) affecting the city concentrations, but coming from sources placed outside the computational domain, is missing. Such contributions must be described by larger scale models able to reproduce physical and chemical transformations from urban to regional scale. Eulerian chemistry-transport models (CTMs) are then the typical tools of choice, since they may incorporate formation and removal mechanisms involving secondary inorganic and organic aerosol.

Whichever model is employed, concentration contributions directly modelled are the ones coming from the sources lying inside the actual domain of analysis, or part of them. Contributions from more distant sources may be then accounted for in different ways, typically by introducing a background, superimposed on concentrations contribution coming from the "local" sources actually modelled, or by assigning boundary conditions at the boundaries of the modelling domain. Such concentrations, either as a background or at the boundaries, usually come from measurements, from models at a scale larger than the actual one, or from a combination of them.

A typical one is through boundary conditions from a coarse grid to a finer grid, which can occur either on-line, in case of an Eulerian model run on multiple nested grids, or off-line, when different models are run at the different scales or different runs of the same model are performed separately in sequence. Other techniques imply passing the information from the finer scale to the coarser one, as in the case of plume-in-grid approach where a Gaussian or Lagrangian model describe plumes at higher detail inside the framework of an Eulerian model, or the case of the feedback that occurs in two-way nested models. A further option is the superimposition of contributions, when concentrations resulting from coarser models are used as background contribution to a finer scale simulation. It follows that in some of these cases the concentration contributions at a given site or over a given domain may be the result of a “hybrid” modelling approach encompassing different modelling techniques. In all cases, when the results from multiple scales are combined it is important to avoid double counting of emissions, i.e. to be sure that the concentrations represented by the models employed at each scale carry the signals from different sources or from different components of the particulate.

Whichever is the modelling approach employed, a crucial aspect of any model application is the use of reliable input data. Concerning meteorological input, it is worth mentioning that simpler parametrised models may typically require only time series of data at a representative nearby location while Lagrangian and Eulerian models almost always imply more effort, i.e. the use of meteorological models to reconstruct the sequence of three-dimensional meteorological fields that are needed in input.

Special attention must be paid to the input emissions, since the emission inventory directly affects the uncertainties of any source-oriented models’ application. To perform a thorough source apportionment, the inventory must be as much complete as possible, i.e. it must include all the sources that may potentially affect the concentrations at the location or over the area of interest. Missing sources can significantly bias the results and should be progressively identified through validation activities, typically involving comparison against measurements and, when possible, the use of receptor models, as discussed later. Another aspect to be considered is the further downscaling or gridding of the inventory data, often performed when preparing model input starting from reference emission inventories; that phase may introduce further uncertainties, that should be also possibly assessed through validation activities and sensitivity runs.

As in all types of applications, the air quality simulations performed with SMs have to be validated. This may happen in several ways, which are summarised according to four different modes: operational, diagnostic, dynamic and probabilistic model evaluation ETC/ACM (2011). In addition to the validation of total calculated concentrations against measurements, individual source contributions should be also validated whenever possible. While this not generally possible in case of gaseous pollutants, in case of particulate matter it can be supported by chemical analysis and receptor modelling (EEA, 2011). Dynamic evaluation, i.e. the assessment of model response to particular aspects of changes in the model inputs, is also of particular interest when dealing with source contributions. Such a validation may focus on situations during which different type of sources may be active in different ways: differences can occur on the basis of seasons (e.g. building heating or road sanding during wintertime), weekends (e.g. traffic types and volumes different from weekdays) or during episodes driven by specific meteorological conditions (e.g. Saharan dust outbreaks or when relevant emission sources are upwind of the area of interest). This requires a model application running over a significant period (possibly one year or more) or during episodes driven by specific meteorological conditions. Having a source-based modelling system validated over a significant time period usually offers better chances to verify the correct representation of different sources in different situations, instead of a modelling system validated only over short time periods. In this respect, modelling systems running operationally over a given area may offer a good starting point. The wider the basis on which SMs simulations are validated, the more confidence can be put in the source influence estimates, either on long-term or on specific episodes.

As for the estimation of source contributions, various techniques are employed, depending on the different type of models.

Gaussian and Lagrangian models usually consider either primary particulate or first-order reactions, so ambient concentrations are linearly related to source strengths. Multiple emission sources are modelled independently from each other, and the ambient concentrations at each point are the result of the superposition of the contributions from all the relevant sources. Source apportionment is typically achieved with these types of models either through separate model runs, each one considering a different set of sources of interest, or by bookkeeping inside a single model run, taking account of the contributions from the different source sets. The same technique may be employed with non-reactive Eulerian models, i.e. when pollutants are modelled using a gridded framework neglecting chemical transformations leading to secondary components.

When chemical transformations cannot be neglected the evaluation of source contributions requires more sophisticated approaches. In fact, in the case of non-linear chemical transformations, the superposition principle cannot be applied due to the presence of possible interaction terms between the reactants. Consider, as a simple example, two precursors (c_a, c_b) emitted by two different sources (A, B) that can react producing a secondary compound (c_{sT}). In the case of a non-linear relationship between reactants and products, the total c_{sT} concentration produced when both sources are operating, can be different from the sum of the concentrations (c_{sA} and c_{sB}) that are produced by each source separately. In order to overcome this problem or at least to reduce errors related to non-linear processes, Eulerian CTMs able to reproduce both primary and secondary fractions of PM are required. CTMs SA applications are based on two different kinds of approaches. The "sensitivity analysis approach" (also known as brute force method (BFM) or emission reduction impacts (ERI); Thunis et al., 2019), still relies on the superposition principle, meaning the concentration associated to each source is estimated separately. The second kind of techniques is generally called "tagged species approach" (mass-transfer methods in Thunis et al., 2019), whose underlying idea is to "tag" the category/sector/activity and/or the geographical origin of the precursors and then follow their atmospheric fate. Both techniques are discussed in the next section.

An overview and guidance on how to run a CTM and on modelling techniques which must be used for meteorology and emissions is outside the scope of this report and can be found in the references given above.

2.3 SM approaches for SA

Source-oriented modelling techniques are usually based on the application of Eulerian Chemistry Transport Models (CTMs) following two main approaches: sensitivity analysis, also known as brute force method (BFM) or emission reduction potential (ERP), and tagged species.

Sensitivity analysis

Generally, sensitivity analysis measures how the concentrations predicted by an air quality model respond to variations in model parameters or input data, and is employed for many purposes (other than SA). As far as input emissions are concerned, sensitivity analysis can be employed to quantitatively estimate the uncertainties in the predicted concentrations when combined with estimates of emission uncertainties (Gao et al., 1995), to compute incremental reactivity factors, i.e. the sensitivity of ozone concentrations to emissions of specific volatile organic compounds (Carter and Atkinson, 1989; Yang et al., 1995), or also to develop a simplified representation of a full model response over a specific range of emission levels (EMEP 2015; Amann et al., 2011). This approach is used to estimate the improvement in air quality due to the measures considered in Air Quality Plans as requested by AQ Directive 2008/50/EC (Annex XV). The information produced with brute force method by CHIMERE model (Menut et al., 2014), summarised by a series of bell-shaped fitting functions, is the basis of the SHERPA tool (Thunis et al., 2018), used to derive the Urban $PM_{2.5}$ Atlas (Thunis et al., 2017) which quantifies for 150 European urban areas the contribution of both the spatial (urban, country, etc.) and sectoral (transport, residential, agriculture, etc.) sources to pollution. In the atmosphere, emissions and concentrations may be related by highly non-linear processes, consequently, sensitivities of different orders need to be evaluated. First-order sensitivities describe linearly the response of the model to a change in parameters or input data, while higher-order sensitivities describe the response according to quadratic and higher power functions. In most cases, only first-order sensitivities are computed due to the increasing complexity and computational/storage resources required by second-order CTM sensitivities. Moreover, in several cases, second order effects can be considered negligible (e.g. Koo et al., 2007). When model behaviour implies significant non-linearities, the major limitation of first-order sensitivities is that they describe the model response over a limited range of the input parameters.

The most straightforward sensitivity method used for SA applications is usually called the brute force method (BFM). The method involves running a model simulation with unperturbed emissions (baseline) and then performing a number of sensitivity simulations, each one varying emissions of all species from a set of sources, either by sector or geographic location. The difference between the results from the baseline and the sensitivity simulation for a particular source is considered as the contribution of that source. The same approach can be implemented by adding the sources one-by-one to a simulation with only the background concentrations. It is important noting that in case of non-linearities the two approaches can give rise to different results.

An important aspect to consider when dealing with PM is that most of the time the relationship between precursors emissions and PM concentrations may include non–linear effects since an important PM fraction, secondary aerosol, is produced in atmosphere by many chemical reactions and physical processes. Examples of non-linearities may be “direct”, such as the reduction of precursors emissions and their effect on corresponding secondary PM components (e.g. between SO_2 and sulphate, NOx and nitrate), or "indirect" such as the reduction in precursor emission affects other component in an indirect manner, such as via oxidant formation, aerosol thermodynamic partitioning processes and aqueous phase neutralization (Pun et al., 2008).

In non-linear situations, the sum of the concentrations of each source is different from the total PM mass obtained in the base case. This can be dealt with in different ways. A first simple method is to normalise the contributions to preserve the total mass. This goal can be accomplished by means of different techniques (see for example Belis et al., 2020). A more appropriate though resource intensive method is the application of a mathematical decomposition in which the interaction terms between all the sources are computed allowing the reconciliation with the total mass (Stein and Alpert, 1993; Clappier et al., 2017). The Stein-Alpert method cannot be considered a SA algorithm, but it can support SA analysis in order to quantify the role of non-linear interactions. This method, however, is difficult to implement in every day practice because it requires the execution of a high number of runs (two to the power of the number of sources).

The magnitude of the emission variations considered in BFM may vary. When the sensitivity simulation is performed with all emissions from a given source suppressed or added (100% of emission variation), the method is usually referred to as zero–out method (e.g. Marmur et al., 2005; Burr and Zhang, 2011a). At the opposite end, small variations of baseline emissions may be considered, although the calculation of smaller concentration changes between the simulations may be more influenced by numerical errors. A further possibility is to consider intermediate percentages of emission reductions (e.g. 20% as in Koo et al., 2009a; 15% as in EMEP, 2015; 50% as in SHERPA (Thunis et al., 2018) and then linearly scale to 100% the resulting concentrations.

The BFM calculations may be influenced by non-linearities and the indirect effects that may be present in model response, but only for the considered emission perturbations. Small perturbations are useful for observing the model response in the same chemical and physical regime as the base case (the present situation), while larger perturbations show their effects in regimes that may be substantially different from the base case. Therefore, special caution has to be paid when the emissions from the set of sources of interest are a significant portion of the emissions in the area investigated. In any case, in presence of a non-linear model response, the sum of the resulting concentration changes may not add up to the baseline values. In such cases, concentration changes may be considered only as first-order approximations of source contributions.

In a modelling application at 7 km resolution over three regional domains in Europe, Thunis et al (2015) showed that the non-linear effects for PM_{10} and $PM_{2.5}$ were relatively minor (even for emission reductions reaching 50%) for yearly and monthly averages, although they are significant for daily average values. In general, non-linearities were more important at locations where concentrations were the lowest. However, these findings may be model dependent.

The BFM can be applied with any CTM, since there is no need to modify the code. Otherwise, this method is not always practical because it involves high computational costs and storage: the computational cost increases linearly with the number of emissions perturbations to examine. This may be an important limitation in some applications, especially when large grids (large domains or high spatial resolution on small domains) are employed and many source categories or source regions need to be considered.

In an attempt to improve the efficiency and accuracy of calculating sensitivities, more complex methods have been developed and applied to CTMs.

The decoupled direct method (DDM; Dunker, 1981) directly solves inside the CTM sensitivity equations derived from the governing equations of the atmospheric processes included in the model and integrates them decoupled from concentrations.

DDM typically requires the addition of specialised code to the host CTM. A parallel set of modules operate in step with the CTM modules (i.e. advection, diffusion, gas-phase chemistry, aerosol processes, depositions) calculate the gridded sensitivity coefficients. When the governing equations and algorithm of a given module are both linear, the equations for propagating the concentrations and sensitivities are identical, however when they are non-linear the equations for the sensitivities have a different form to the equations for the concentrations. Specialised modules for the sensitivity coefficients may be also written to improve the computational efficiency, as is the case of DDM-3D, a higher efficiency variant that uses different and less rigorous numerical algorithms to solve time evolution of the chemistry sensitivity equations than those used to solve concentrations. The higher numerical efficiency and the different algorithms employed can otherwise complicate the error control and also imply potential inconsistencies between sensitivities and concentrations (Dunker et al., 2002; Koo et al., 2009b). DDM has been initially implemented for gas-phase species (Dunker et al., 2002) and later extended to particulate matter (Koo et al., 2007), as also DDM-3D (Yang et al., 1997; Napelenok et al., 2006). DDM has been also furtherly extended to calculate high-order sensitivities. The high-order DDM (HDDM) has been also implemented for gas-phase species (Hakami et al., 2003; Chohan et al., 2005; Koo et al., 2010) and then to PM (Zhang et al., 2012). For the gas phase, calculating higher-order sensitivities requires few modifications of the code implemented for first-order sensitivities and implies only a slight increase of the computational cost, but gives a higher accuracy. The method has been shown to accurately capture the non-linear response of ozone concentration to changes in precursors emissions (NOx and VOC), showing reasonable agreement with corresponding brute-force results, although higher than second order sensitivities are not necessary for the majority of potential applications (Hakami et al., 2003).

In case of PM, second-order sensitivities of secondary inorganic aerosol species with respect to emissions of corresponding gaseous precursors are also shown to agree with BFM results. In addition, it avoids the noisy behaviour that may emerge when computing differences among model results deriving from small emissions perturbations, improving the prediction of nitrate concentrations with reduction in domain-wide NO_x or SO_2 emissions rates up to 50% (Zhang et al., 2012).

With DDM methods multiple sensitivities can be calculated simultaneously, depending on the coding of the method and on the available memory. DDM methods require additional memory space and CPU time over the ones employed in standard CTM runs, depending on the number of source categories and source regions to be investigated. CPU usage for first-order DDM scales linearly with the number of sensitivity parameters of interest, but is significantly less than that required by the brute-force approach (Napelenok et al., 2006). The sensitivities obtained by BFM converge toward the DDM sensitivities, as the brute-force perturbations become smaller (Dunker et al., 2002), although DDM calculations are less affected by the numerical noise that affects BFM in some cases (Napelenok et al., 2006).

As in the case of simpler BFM, DDM first-order sensitivity is also useful for determining source contributions only if the model response to input changes is reasonably linear. As an example, Koo et al. (2009b), considering monthly averaged $PM_{2.5}$ over Central USA found that for secondary inorganic PM the response is linear for emission reductions of 20% in all cases considered and reasonably linear for reductions of 100% of sectors dominated by NOx emissions, as in the case of on-road mobile sources, while the model response for secondary organic aerosols and primary PM remains nearly linear to 100% reductions in anthropogenic emissions. As also mentioned above, linear behaviour over such ranges refer to the specific area considered, other regions may exhibit linearity over different reduction ranges.

As previously mentioned, sensitivity methods are able to simulate non-linear and indirect effects, so their results may be directly related to the development of control measures (AQ Directive 2008/50/EC, Annex XV), as it is better detailed in Thunis and Clappier (2019). On the other hand, for assessments studies, when responses are strongly non-linear the results are non-additive. In this situation, different set up of the simulations (e.g. different percentage of reduction) provide different insights of the response of the atmospheric system to emission precursor variation.

More in general as pointed out by different authors (Burr and Zhang, 2011; Koo et al., 2012), it is important to recognise that there is no unique apportionment of ambient concentrations to sources when non-linear chemistry is present. Different methods inherently give different results, and there is no “true” apportionment to which all methods can be compared. Complementary information to sensitivity methods can be given by reactive tracer methods.

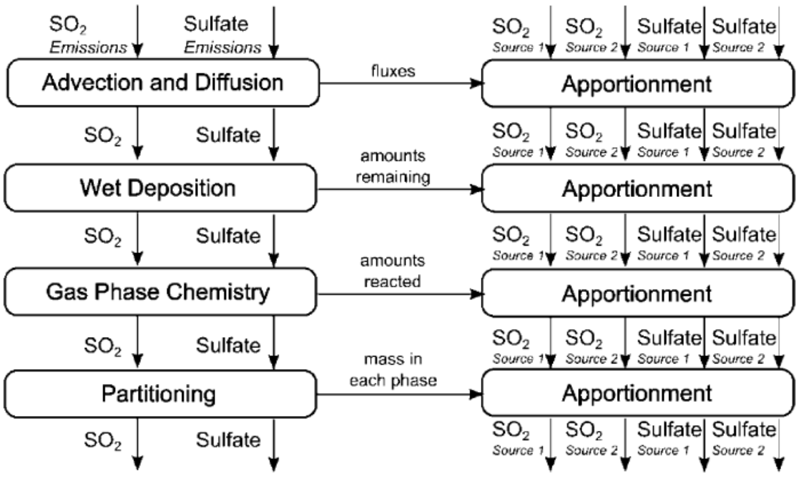
Tagged species

The tagged species methodology is designed for source apportionment purposes. This methodology labels each precursor in every time step according to its activity source and/or the geographical origin. This accountability makes it possible to quantify the mass contributed by every source/area to the pollutant concentration (e.g. PM). Tagging methods are implemented by adding extra-species to track pollutants emitted by specific sectors or areas. These species are generally known as “reactive tracers” because they can also undergo chemical transformations, being therefore able to track not only directly emitted species, but also the corresponding secondary products as well as intermediate species useful to track the key steps of the chemical pathways linking emitted precursors to secondary compounds. A key aspect in developing tagging methods in the CTM models is to fulfil the assumption that the sum of the concentrations x_i corresponding to each source *i* is always equal to the total concentration X due to all sources (Yarwood et al., 2004). This common feature of the tagged species approach techniques with RMs that led to quantifying the actual mass transferred from the source to the receptor. For that reason, these two methodologies can be grouped under the category mass-transfer source apportionment (Thunis et al., 2018).

Developing computational algorithms to solve the reactive tracer concentrations to ensure the maintenance of equality is non-trivial. However, tagged algorithms are implemented in several chemical transport model systems. The particle source apportionment technology (PSAT) within the Comprehensive Air Quality Model with Extensions (CAMx) (Yarwood et al., 2004; Wagstrom et al., 2008, ENVIRON, 2014) the tagged species source apportionment algorithms within CMAQ, such as (TSSA) within CMAQ (Bhave et al., 2007; Wang et al., 2009), the SIA source apportionment algorithm (Zhang et al., 2012) and, more recently, the Integrated Source Apportionment Method (ISAM, Kwok et al., 2013). A source apportionment module is available also within LOTOS-EUROS (Kranenburg et al., 2013) and a tagging method has been implemented in the DEHM model (Brandt et al., 2013a) too.

The CAMx model implements the PM Source Apportionment Technology (PSAT), a SA algorithm embedded into the code that uses reactive tracers to apportion primary and secondary PM compounds and corresponding precursors among different source categories and source regions (Yarwood et al., 2004; 2005b; Wangstrom et al., 2008). PSAT adds reactive tracers in the CAMx model to apportion PM in every single grid cell. More precisely, PSAT apportions among the different sources the variation in total PM concentration, taking place after each physical and chemical process reproduced by the CAMx host model. The key assumption in PSAT approach is that the variation in the total concentration due to a specific process is apportioned among the different sources proportional to the distribution of the tracers’ concentration at the beginning of the integration step. A schematic representation of PSAT algorithm is shown in Figure 1 for the simple case of SO_2-Sulfate system.

Figure 1. Information flow in PSAT for an example of two source classes and the SO_2–sulphate system.



Source: Wangstrom et al., 2008

Let’s consider a chemical reaction where reactants A are transformed in products B (Yarwood et al., 2004):

$$A \rightarrow B \tag{1.2}$$

Where A and B are the total concentrations apportioned to the different sources, in compliance with the following constraints:

$$A = \sum_{k=1}^{n} a_k \qquad B = \sum_{k=1}^{n} b_k \qquad (1.3)$$

Where n is the number of sources and a_k and b_k are the tracers concentration corresponding to each source. The equation for the reactant species (A) is:

$$a_k(t+\Delta t)=a_k(t)+\Delta A\times[a_k/A] \qquad (1.4)$$

Where ΔA is the variation (decrease) of A concentration due to the chemical reaction and A is total concentration before the chemical step. The equation for product species (B) is defined in a similar way:

$$b_k(t+\Delta t)=b_k(t)+\Delta B\times[a_k/A] \qquad (1.5)$$

Where ΔB is the variation (increase) of B concentration due to the chemical reaction. It is worth noting that the repartition coefficient for ΔB is the same of ΔA and proportional to the concentration of the precursor A. Similar equations can be written for other chemical and physical processes. As it can be easily verified, the approach adopted in PSAT ensures that the total concentration is preserved, as requested in (1.3). The main limitation of PSAT derives from the assumption that the apportionment of each secondary PM compound is related only to one precursor species. This implies, for example, that PSAT cannot take into account the indirect effect on nitrate concentration of a source that emits, both NOx, and a species able to produce oxidants.

PSAT is designed to source apportion most of the PM species modelled in CAMx including: sulphate, particulate nitrate, ammonium, particulate mercury, secondary organic aerosol (SOA) and six categories of primary PM. The number of tracers required by each PM species depends on the different reaction pathways. For example, primary PM species require only a single tracer, corresponding to the species itself, whereas secondary PM species require several tracers to track the relationship between gaseous precursors and the resulting PM. For example in case of nitrate PSAT introduces 5 gas phase tracer and 1 particulate phase tracer in order to keep track of the complex chemical pathways that start from NO and NO_2 to give rise to different oxidised nitrogen species, including HNO_3.

PSAT has been extensively tested against potentially more accurate methods such as SOME (Yarwood et al., 2004) and OPSA (Wangstrom et al., 2008) and has proven to be robust and computationally efficient. PSAT has been applied in several source apportionment studies in USA, Asia and Europe. As for the latter, Karamchandani et al. (2017) performed a source apportionment study at European scale aiming to reconstruct the contribution of the main emission sectors to $PM_{2.5}$ and ozone concentration in 16 cities. The analysis concerned two months (February and August) of 2010. Skyllakou et al. (2014) used the CAMx PSAT approach to distinguish the contributions of local and regional sources to fine PM in Paris. Bove et al., (2014) compared the results of a PSAT application over the city of Genoa against the results of a PMF analysis of an observed dataset. A similar application was carried out by Pirovano et al. (2015) that compared the results of a PSAT application over the Po valley against the results of a CMB application at several receptor sites placed in the Lombardy region.

The CMAQ model included a first PM source apportionment algorithm called Tagged Species Source Apportionment (TSSA, Wang et al., 2009). The algorithm adds approximately 20 new tracers or ‘’tagged species’’ for each emissions source to be evaluated. Like PSAT, TSSA has been implemented in order to track the main particulate species such as sulphate, nitrate, ammonium, elemental carbon and other primary PM species (e.g. dust). TSSA included also a preliminary treatment of secondary organic species. The results of the TSSA algorithm are three-dimensional concentration fields showing the transport of mass of both primary emissions and secondary products from the selected emissions sources. The algorithm is designed to track the emissions either of a single source or of selected classes of sources, grouped either by source category and/or by geographical region.

Like PSAT, the TSSA treatment of linear processes such as emission, advection, diffusion and deposition is the same as in the bulk model, meaning that TSSA applies the same algorithms of the host model also to the tracers. TSSA performs also additional mass conservation checks (i.e. verifying that the sum of the tracer concentrations correspond to the bulk concentration in the host model) adjusting the mass (renormalisation), if needed.

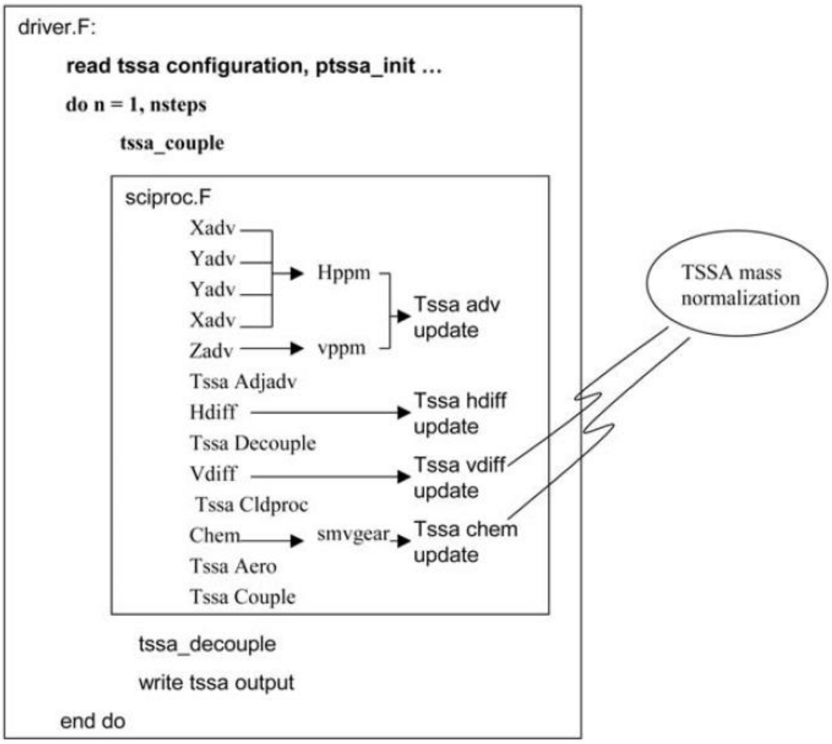
The treatment of chemistry required the development of specific algorithms able to capture the non-linear interactions that take place among the tracers. The TSSA approach, described by Wang et al. (2009), is rather different from PSAT particularly concerning the gas phase chemistry. TSSA assumes that the concentration variation of a generic tracer can be expressed as:

$$dC_{j,i}/dt=P_{j,i}-L_j C_{j,i} \qquad (1.6)$$

Where Cj,i is the concentration of the tracer species j, accounting for source i, $P_{j,i}$ is the production rate of species j from source i and L_j is the loss frequency of species j. The production and loss terms of equation 1.6 are defined according to the reaction rates of the reactions involving each tracer. Equation 1.6 admits an analytical solution that can be expressed in the following form:

$$C_{j,i}^{t_{n+1}} = \frac{P_{j,i}}{L_j}\left(1 - e^{-L_j \Delta t}\right) + C_{j,i}^{t_n} e^{-L_j \Delta t} \qquad (1.7)$$

Figure 2. Flow chart of the TSSA algorithm



Source: Wang et al., 2009.

Where the first term on the right side of the equation represents the production during period ∆t of species j due to source i while the second term is the amount of C^{tn} remaining at t_{n+1}.

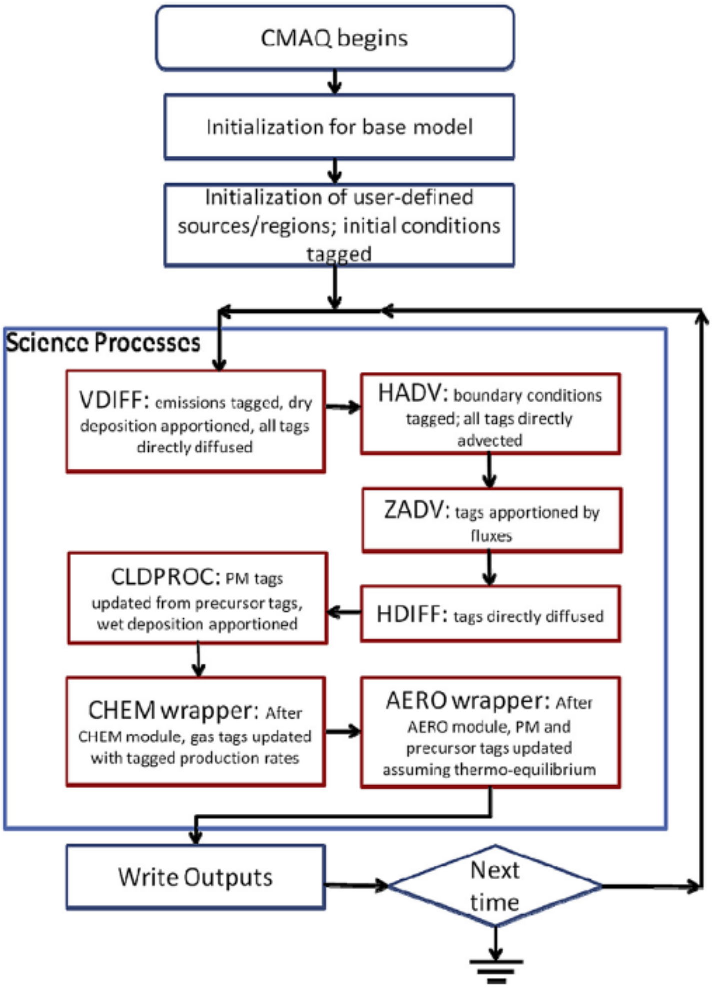
A similar equation is written for each tagging species giving rise to a set of equations that are then solved with the same chemistry solvers implemented in the host CTM model.

Aerosol chemistry is solved through a simpler approach than gas chemistry. Indeed, TSSA assumes that aerosol and gas species are at the equilibrium and computes the total mass of each tagging species based on this assumption. Then the total mass is shared into gas and PM phases proportionally to change in the corresponding bulk species.

A simplified flow chart of the TSSA module depicting the different steps involved in PM apportionment is shown in Figure 2.

CMAQ/TSSA module has been reviewed and updated in recent years introducing several improvements (Kwok et al., 2013). The new module is called ISAM (integrated Source Apportionment Method) and relies mainly on TSSA but including among others a faster and more consistent link with the base model approach for advection of tagged tracers, improved tracking of precursor species through gas phase chemical reactions using an efficiently formulated solver for determining incremental precursor reactivity. The main features of ISAM are described in Figure 3.

Figure 3. ISAM flow chart



Source: Kwok et al., 2013

As shown in Figure 3 the updated algorithm introduces an explicit treatment of most of dispersion processes for tracer species (tags), with the exception of vertical advection whose effect on tracers is still computed through apportionment of the bulk concentration. Relevant changes have been introduced also in the treatment on the gas phase chemistry of tracer species. Particularly the computation of the concentration variation is performed taking advantage of the same Jacobian matrix that describe the composed reaction rates of each chemical species, due to the changes in all other species (Kwok et al., 2013). This approach allows to reconstruct the concentration variation of each tracer based exactly on the same set of reaction rates adopted by the corresponding bulk species.

An example of application of the new CMAQ SA module is described in Napelenok et al. (2014) where CMAQ-ISAM was used to compute source contributions from ten categories of biomass combustion, including Residential Wood Combustion (RWC). CTM results were then compared and combined with the results of the receptor model, UNMIX, that was used to subdivide the observed concentrations of levoglucosan, a unique tracer of biomass combustion.

The DEHM tagging method, described in Figure 4 is based on the concepts of background and tagging concentration. The former accounts for all sources except for the "tagged" one which is traced by the tagging species. Then, as shown in Figure 4, all linear processes such as emissions (Emis), advection (Adv), atmospheric diffusion (Diff), wet deposition (Wet), and dry deposition (Dry) are integrated in parallel with the same algorithms and techniques.

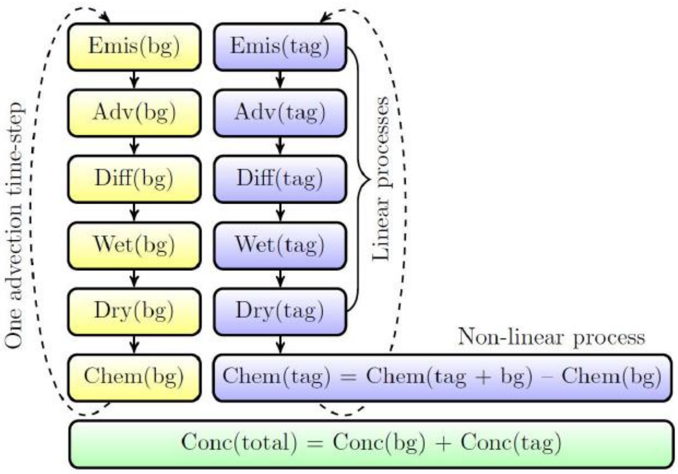
For the chemistry, the tagged concentration fields are estimated through a sequence of steps:

- the background and tag concentration fields are added to produce total concentration fields;
- the chemistry operator is applied to both total concentration field and background field alone;
- the concentration field obtained by applying the non-linear operator to the background field alone is then subtracted.

This approach, is simpler than the other tagging methods described above and is fast and efficient, but has the disadvantage that it can handle only one "source" at a time.

The EVA system, including DEHM-Tagging method was applied to identify the anthropogenic emission sources in Europe and Denmark that contribute the most to human health impacts. This work was based on the assessment of the role of ten main source categories. Another EVA application (Brandt et al., 2013b) performed a detailed assessment of past, present and future health cost externalities of total air pollution levels in Europe for the years 2000, 2007, 2011, and 2020. The study also assessed the health-related external costs specifically related to international ship traffic.

Figure 4. Overview of the DEHM tagging method.



Source: Brandt et al., 2012

LOTOS-EUROS calculates source contributions using a tagged method, based on PSAT where both primary and secondary tracers are attributed to their source (Kranenburg et al, 2013). For secondary tracers, source contributions are considered to be divided among the different 'sub-tracers'. For example, in $NH_4^+NO_3^-$ ammonium is attributed to the NH_3 sources and nitrate to the NO_x sources. The distribution of the combined tracer can be calculated by either mass or molecular basis. This method has been used in various studies in the

Netherlands (Hendriks et al, 2013) and China, in the megacities of Beijing and Shanghai (Timmermans et al, 2017).

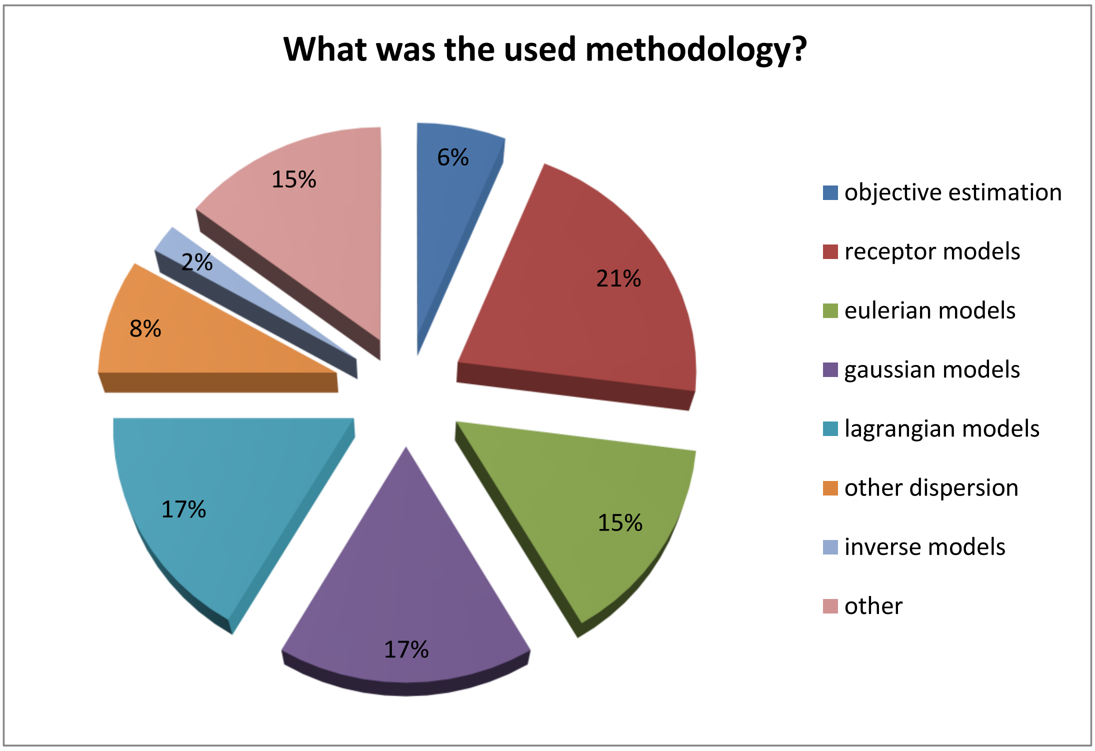
2.4 European SA studies with SMs and with SMs-RMs: survey results

Fragkou et al. (2012) published a review of the SA methodologies for PM used by EU Member States in preparing their extension reports regarding compliance with PM_{10} limit values at 1st January 2005. Based on the information from 18 respondents from 12 EU countries, source-oriented models and receptor models were equally used by member states for SA studies (61% of the reported studies).

According with a more recent review (48 answers) on the use of models for source apportionment carried out in the frame of the project APPRAISAL (Belis et al., 2017), 49% of the source apportionment studies to support air quality plans at local and regional levels were carried out using SMs (15% Eulerian, 17% each Lagrangian and Gaussian) (Figure 5).

According to the survey carried out in 2017, in the framework of FAIRMODE WG3, regarding the SA methodologies used in the latest years, the number of SM studies was 11 while RM studies were more than 150. The SA studies using RMs-SMs and SMs techniques are shown in Table 1 and 2, respectively. SMs were used in support of air quality management in Belgium, Germany, Italy, Netherlands, and Slovakia. RMs-SMs studies were only performed in Italy and were for research purposes. Most of the SM studies used well-known CTMs (BelEUROS, CAMx, EURAD, LOTOS-EUROS, AMS-MINNI) and were performed by institutions/agencies with experience in the field of air quality. One study was performed with an integrated Lagrangian puff modelling system (CALPUFF) (Table 2, No. 7) and other two used a combination of models to perform local-scale source apportionment (Table 2, No. 2 and 6). Half of the reported studies used the brute force method/zero-out approach for SA while the other half the tagged species approach (labelling subroutine, PSAT technique). More details about these studies can be found in the Appendix 1.

Figure 5. Distribution of SA studies based on project APPRAISAL.



Source: Belis et al., 2017

The relatively small number of SM studies recorded in the FAIRMODE WG3 review may be explained by the complexity of setting up and carried out CTM simulations of air quality. Where a combination of models is used, a special coupling procedure between them is necessary to avoid double-counting of the sources when estimating the background contribution to air pollution levels for local scale simulations.

None of the studies presented in Table 2 used assimilation techniques, apart from study No. 2 in which such techniques were used for calculating the regional background contribution with EURAD-IM and EURAD-ladis. All the studies, with the exception of AMS-MINNI, addressed source categories and did not estimate the influence of source regions. The survey showed that the model setup varies with the cases (agriculture, traffic, domestic heating) and the years. The time window covered in the studies ranged from annual average (No 1, 2, 5) and multi-annual evaluations (No. 3) to seasonal evaluations (No. 4, contribution of agriculture to PM_{10}). The RMs-SMs studies used CAMx/PSAT combined with PMF (Bove et al., 2014) and with CMB (Pirovano et al., 2015). Both studies considered several sites and investigated the differences in performance due to seasonal variations.

Table 1. RMs-SMs applications in Europe.

Country	Pollutant	RM	CTM	SA approach	Resolution	Year	Reference
Italy	$PM_{2.5}$	CMB	CAMx	tagged species	5 km	2005	Pirovano et al, Atmos. Environ, 2015
Italy	$PM_{2.5}$ - PM_{10}	PMF	CAMx	tagged species	1.1 km	June - August 2011 Nov. – Dec. 2011	Bove et al., Atmos.Environ., 2014

Source: Fairmode WG3

Table 2. SM applications in Europe.

Study area	Pollutant	CTM	SA approach	Resolution	Year	Application type	Reference	Contact person
1. Germany (Berlin)	PM_{10}	LOTOS-EUROS	tagged species	7 x 7 km	2015	Air quality management		Sabine Banzhaf, Freie Univ. Berlin, Institute of Meteorology, DE
2. Germany	PM_{10}	EURAD, LASAT, Miskam, EURAD-IM, EURAD-Fladis	combination of models, brute force method	from single meters to 250 m, in some cases up to 1 km	2005-2017	Air quality management	http://gis.uba.de/website/umweltzonen/lrp.php	Heike Hebbinghaus; Sabine Wurzler, LANUV NRW, DE
3. The Netherlands	PM_{10}, $PM_{2.5}$	LOTOS-EUROS	tagged species	7 x 7 km	2007-2009	Air quality management	Hendriks et al., Atmos. Environ. 2013	Martijn Schaap, TNO,Netherlands
4. Belgium (Flanders)	PM_{10}, $PM_{2.5}$	LOTOS-EUROS	tagged species	7 x 7 km	2007-2011	Air quality management	Hendriks et al., Atmos. Environ. 2016	Martijn Schaap, TNO, NL
5. Belgium (Flanders)	PM_{10}, $PM_{2.5}$	BelEUROS	brute force method	60km, 15 km	2007, scenarios 2020	Air quality management	Deutsch et al., Applied Mathematical Modelling, 2008	Peter Viaene, VITO, BE
6. Belgium (port of Ghent)	PM_{10}, $PM_{2.5}$	RIO-IFDM-OSPM	combination of models, zero out approach			Local air quality management		Hans Hooyberghs, VITO, BE
7. Slovakia	PM_{10}	CALPUFF	brute force method	horizontal resolution of 200–500 m, depending on the terrain complexity.		Air quality management	Krajčovičová et al., Int. J. Environment and Pollution, 2014	Jana Matejovičová, Slovak Hydro-meteorological Institute, SK
8. Europe AQMEII	$PM_{2.5}$	CAMx	tagged species	23 km	2013	Research study	Karamchandani et al., Atmos.Chem.Phys., 2017	
9. Italy	PM_{10}	AMS-MINNI	brute force method	20 km	scenarios 2011	Notification for time extension		Luisella Ciancarella, ENEA, IT

Source: Fairmode WG3

3 Estimation of source contributions with SM approaches

3.1 Modelling and validation of PM - base case of SM approaches

This chapter looks at specific issues related to the first step of a SA study: modelling PM concentrations over the investigated area with CTMs. PM (PM_{10}, $PM_{2.5}$, PM_1) is a mixture of primary and secondary particulate matter emitted or formed locally and/or transported from very large distances. Therefore, the modelling domain has to be large enough to include the major emission sources which contribute to air pollution over the investigated area and also to consider the contribution from various spatial scales to PM through the concentrations at the boundaries. The CTM has to be able to reproduce the main production paths of PM from pre-existing PM and gaseous emissions in various meteorological conditions and land cover type. The meteorology, in addition to influencing the chemical processes leading to PM formation, is responsible for PM accumulation in low dispersive conditions and for the re-suspension of deposited particles.

The air quality modelling system (CTM, the meteorology and the emissions) used for SA applications has to have adequate peer-review and performance evaluation history in PM applications.

All these PM modelling aspects were extensively addressed by ETC/ACM (Technical Paper 2013) and issues related to validation can be found in air quality modelling guide (EEA, 2011).

A CTM run including data assimilation technique produces the most probable representation of the state of the chemical variables assimilated using a combination of modelling and observational data. However, since no observational data are available when the emissions are reduced, BFM cannot be performed when data assimilation is used and the differences in simulated concentrations due to a reduction in emissions cannot be evaluated. Similarly tagged methods have no recipes to apportion the contribution of observed assimilated concentrations. The base case simulation should therefore be performed without using any data assimilation technique.

Domain, time window and spatial resolution

The configuration of the modelling domain has to take into account several aspects:
- to ensure the maximum possible spatial resolution in the area with PM exceedances;
- to have as much as possible measurements in the domain;
- to have details on anthropogenic emissions at source category level.

When choosing the size of modelling domain, the potential inflow of dust, sulphate and other chemicals from agricultural and biomass fires outside the domain needs to be taken into account (more details in Section 2.4.2 of ETC/ACM (2013).

The choice of the modelling time period for SA purposes also depends on the nature of exceedances: systematic or episodic, dependent on season or on specific atmospheric circulation patterns, etc. Therefore, the modelled period may be:
- from days to weeks, for episode days in correspondence of exceedances;
- a season, since the chemical composition of PM vary from winter to summer;
- a year, since AQ Directive (EU, 2008) set limits to number of exceedances on annual basis (PM_{10} limit value must not be exceeded more than 35 times a year).

The Spatial resolution of the simulations has to be as high as possible and should be defined taking into account the characteristics of the model used and the spatial representatively of available measurements. As indicated in Table 3.2 of EEA Technical Report (2011), for urban stations the typical CTM resolution can vary from 1 to 5 km and for rural stations from 10 to 50 km. Higher spatial resolutions over the domain of interest are advised in order to have the best representation of local phenomena. By applying the simulation results of a larger model domain with lower spatial resolution to the boundaries of smaller simulation domains with higher spatial resolution, non-linearities of atmospheric chemistry can be captured while conserving computational resources.

Multiscale-nested modelling is the current state-of the-science for air quality modelling over various scales: from regional/continental scale (few thousands km with a spatial resolution of a few tens of km) to urban scale (domain size from 100 km to few tens of km, with spatial resolution of few to 1 km) (more details in Section 2.3.1 of ETC/ACM (2013)). Various nested-grid modelling techniques are available. The static grid nesting is the most used technique to increase the resolution over a specific area. The fine grid domain is placed or “nested” inside the coarse grid domain and the domains and their spatial resolutions are fixed throughout the simulation. This technique allows one-way or two-way exchange of information among finer and coarser grids. In the one-way nesting, there is no feedback from finer grid simulation to coarser grid simulation; the coarser grid simulation is used as input for finer grid simulation. In this case, the primary concern is the mass conservation at the interface between the finer and coarser grids which is ensured by setting the flux at the finer grid inflow boundaries equal to the flux passing through this interface according to the coarser grid simulation. In principle, simulations made with different models can be nested but this introduces other uncertainties in modelling the finer grid due to the fact that the models may not use the same chemical species, same chemical and physical representation of atmospheric transport, same meteorological parameters (almost all air quality models use their own meteorological pre-processor) or the same emissions. It is therefore highly recommended to perform multiscale-nested modelling with the same model over the regional and local scales.

Boundary conditions: linking model outputs over different domains

All simulations require as input concentrations of chemical compounds at the top and lateral boundaries of the modelling domains, so-called boundary conditions. Thus regional simulations would use global model outputs or measurements-derived profiles while urban scale simulations use the outputs of regional simulations. The contribution of particulate matter from outside the domain over the domain of interest varies with the aerosol compound and with the length of investigated period. For example, according to Liu et al. (2009), particulate matter originating from outside Europe contributes to ca. 30% of $PM_{2.5}$ concentrations, which is due to ca. 17.4% of dust, 8.4% of sulphate, 1.5% of organic matter (more details in Section 2.4 of ETC/ACM (2013)).

In order to capture the intensity and the occurrence of high PM concentrations due to transport through the boundaries, boundary conditions have to be provided with a relative high time frequency, at least every 3 hours at regional scale. For these reasons, dynamic boundary conditions from other simulations are preferable to static ones, derived from long term series of measurements.

Meteorological data

The impact of meteorology on air quality is well recognised. Wind, temperature, humidity, precipitation and solar radiation are controlling the transport, formation, destruction and removal of pollutants in atmosphere. Therefore, the choice of the meteorological data to be used is fundamental for the PM modelling (Section 4 of ETC/ACM (2013)).

It is highly recommended to use meteorological data provided by meteorological services, reanalyses when possible, or to perform a validation of the output of the meteorological model before performing air quality simulations.

It is also recommended to use meteorological data produced on a grid resolution close to the one used for air quality simulation, on the same projection if possible, in order to avoid uncertainties introduced by interpolation methods.

Emissions: anthropogenic and natural

The anthropogenic emissions are usually available as emission inventories as total mass of pollutant emitted in a given period (“static” emission data; ETC/ACM, 2013) by a number of activities. At present, in Europe, the activities are usually classified using three sets for source categories: SNAP97, EMEP/NFR and UNFCCC/CRF. The correspondence between the categories may be found in Table III, Annex II at the following link:

https://www.unece.org/fileadmin/DAM/env/documents/2002/eb/ge1/eb.air.ge.1.2002.7.e.pdf

Emissions inventories with enhanced source classification detailing some macrosectors are required. The maximum number of source categories that can be tracked by the SMs depends on the detail available in the emission inventory.

These national emissions have to be distributed over the domain grid cells, on hourly basis according to spatial proxies and temporal profiles, respectively. Generally, both proxy and temporal profiles vary with source category. The most used proxies are population, land-use and, more recently, bottom-up emission inventories. More discussion about proxies can be found in Trombetti et al. (2018) and about temporal profiles in Menut et al. (2012).

The spatial resolution of proxy data has to be higher or equal to that of the grid used for SA simulations.

Natural emissions of dust, sea-salt and biogenic volatile organic carbon (BVOC) precursors of organic aerosol, an important fraction of PM, are often included in CTM formulation or calculated by other software in relation to meteorological conditions and land cover data. In the last decade, emission models for ammonia as a function of soil moisture, temperature, wind speed and agricultural practices have been also developed.

Forest fires may have also an important impact on air quality in some specific periods. These emissions are available from Copernicus/MACC-II project (EU FP7) which estimate them based on satellite observations *(http://www.gmes-atmosphere.eu/about/project_structure/input_data/d_fire/)* and a fire emissions model.

More information about anthropogenic emissions inventory available in Europe and their treatment as well as on modelling natural emissions and forest fires can be found in ETC/ACM (2013).

Model evaluation/validation-MQO

The simulated PM concentrations have to be evaluated against measured ambient concentrations using statistical model performance measures which will be compared against established model performance goals and criteria (EEA, 2011 – Annex 2; ETC/ACM, 2013 – Section 5). They can be also evaluated using the FAIRMODE model quality objectives (MQO). This kind of analysis can be performed, by, for example using the DELTA software, which is a diagnostic tool for assessing the model performances in terms of various statistical indicators and diagrams, produced in the framework of FAIRMODE. The software is available at https://ec.europa.eu/jrc/en/scientific-tool/fairmode-delta-tool. The DELTA tool diagnostics are mainly related to implementation of AQD (EC, 2008) and focus on PM_{10} and $PM_{2.5}$ rather than individual PM components.

In SA applications, the evaluation of PM components (chemical species or groups of compounds) is highly recommended since their concentrations are sensitive to source distribution and time trends. Therefore, a good reproduction of individual species is generally associated with the ability of the model to identify and quantify source contributions.

3.2 Sensitivity analysis methods

After the application of the CTM model for the base case described above, sensitivity modelling involves multiple runs of the CTM with perturbations in emissions input.

Due to its simplicity, BFM (or ERI) is the easiest approach to implement and it has been frequently used to estimate the concentrations associated with changes in source emissions by sectors. In order to identify *N* source sectors, *N* simulations have to be performed with the respective source sector emission reduced by a given percentage x%. The concentration fields obtained from each run (the base case simulation and the *N* altered runs) are then post-processed to obtain the desired statistics over the modelled periods (typically average values), over the simulation domain and/or on a set of chosen receptor points. The difference in concentration statistics between the base case and a perturbed simulation represents the sensitivity of the modelling system to the considered emission variations. For all pollutants, the difference of a given source, rescaled to the full extent of the emissions (i.e. from x% to 100%), are interpreted as the contribution of that source sector.

Substantial changes in the emissions may lead to significant changes in the chemistry of the simulated airshed, thereby altering the subsequent production/destruction of PM secondary component. Therefore, the sum of concentration variations corresponding to all perturbed runs, rescaled, may not equal the concentration of the base case simulation in a simulation with reactive species as some PM components. When reducing precursors emissions, the CTM chemistry has direct non–linear effects on the secondary PM components (i.e., sulphate, nitrate, and organics), and non–linear indirect effects on the other components that result from interactions between secondary PM species and their gaseous precursors through processes such as aerosol thermodynamic partitioning processes, gas phase oxidation, etc. (Burr et al.., 2011a). It is therefore advisable to test for each application the extent of any non-linear effects of reductions in emissions for the source sectors of interest. Ideally, the sum of contributions from all sources has to equal the concentration simulated in the base case. When this does not happen, an evaluation of the acceptability of the actual results should be carried out. To date acceptability criteria have not been agreed by the scientific community, therefore each modelling application should undergo a critical analysis of the extent of the deviation of the sum of contributions from base case concentrations as well as a clear identification of the range of applicability of the obtained results. A very first guess criterion could be to compare the deviation, expressed in relative terms, from the computed contribution, still expressed in relative terms, of each source. This comparison could provide a preliminary evaluation of the reliability of the SA results. Other tests can be derived from available literature, after a careful evaluation of the modelling context. For example Kiesewetter et al., (2015) showed that the error in reproducing the yearly mean background concentration of PM_{10} at the European scale was lower than 2 $\mu g/m^3$. Of course the same criterion should not be used, in a different context (e.g. traffic site) or for a different time scale (e.g. daily mean concentrations). A second example is reported in Thunis et al, 2018, showing that the reconstruction of the delta concentration of the $PM_{2.5}$ yearly mean concentration due a 50% variation of total emissions can be reproduced as a sum of single source contributions with an error lower than 10%. Also in this case, similar criteria should only be applied in the same modelling context.

Sensitivity methods used to estimate source contributions in terms of sectors may be applied to estimate source contributions in terms of geographic area, by reducing the emissions of that area and comparing the concentrations obtained against the base case. The SAL technique was used to quantify the origin of pollution, both spatial (urban, country, etc.) and sectoral (transport, residential, agriculture, etc.) in 150 European Cities (Thunis et al., 2018). By contrast, DDM is less common since it requires the addition of specialised code to the host CTM. To our knowledge, there are no DDM applications in Europe.

3.3 Tagged species methods

Tagged species maintains a mass balance when estimating the mass transferred from the sources to the receptors. Therefore, the sum of all source contributions equals the simulated baseline concentrations. The tagged species approach is able to detect and allocate pollutants affected by direct non-linear chemical regimes, however this information is not explicitly available for assessing chemical regimes different to those represented in the modelled area and time period investigated. In addition, according to Burr et al. (2011b) this approach is not able to simulate indirect effects (e.g. oxidant–limiting effects like the formation of secondary PM species limited by availability of oxidants) because of assumptions made in the source apportionment for secondary PM species.

All tagged species methods run in parallel to the CTM host model to determine the contributions of sources sector to PM, consequently, only one run is necessary. The introduction of additional chemical species (reactive tracers) and additional algorithms performing concentration apportionment generally increase the computational time with respect to the standard simulation, but such a computational burden is not normally substantial (generally lower than 100% of the base case run when estimating the contributions of about 10 sectors over 5 regions). Of course, any additional computational burden is less and less relevant with respect to the total BFM runs time as the number of categories/regions increase. Conversely, increasing the number of tracers can give rise to memory issues, because unlike the BFM approach, all sources are tracked simultaneously. The number of tracers needed to track a source emission depends on the complexity of the chemical mechanism and on the number of PM species involved. For example, the sulphur compounds require only two reactive tracer species (SO_2 and SO_4) to track the formation of particulate sulphate from SO_2 emission source contributions for each source sector. SOA components require more tracers. For example, in case of PSAT, 18 reactive tracers are used to track the four VOC precursors (aromatics, isoprene, terpenes and sesquiterpenes) and the 7 condensable gas (CG) and SOA pairs.

As in BFM methods, the main limitation in designing a SA study with a tagged species approach stems from the sectorial detail available in the emission inventory. Conversely, if the tagging algorithm is robust, there are no limitations with respect to the strength of the source: tagging methods can be applied to track also a single point source.

The tagged species approach can also be applied simultaneously to evaluate source contributions according to their geographical origin. The evaluation of source regions contribution requires exactly the same approach and algorithms and in many cases tagging methods are implemented in order to track sources according to a combination of regions and categories. In the case of geographical source apportionment, the only limitation derives from the spatial resolution of the emissions, meaning the single computational cell for diffuse emissions and the single source for point emissions.

Examples of application of spatial source apportionment, also in combination with source categories can be found in Kranenburg et al., (2013), Wu et al., (2013), Skyllakou et al., (2014), Wang et al., (2017).

4 Combined use of SMs and RMs

SA evaluations can be performed using both receptor-oriented (RMs) and source-oriented models (SMs). Both approaches try to reconstruct the same kind of information, namely the atmospheric concentration of pollutants associated to the different emission sources. The former is based on inferential methods, whilst the latter is predictive of source contributions. The comparison between SMs and RMs is generally focused on source categories, because the latter are more appropriate to detect the source contributions related to different kind of activities more than different emission regions. It is also important to recall that the different SA techniques (e.g. sensitivity analysis or tagging methods) that SMs can implement strongly influence the resulting source contribution estimates as well as the comparability against RMs results. For this reason, source contribution estimates obtained by means of tagging methods are considered the most suitable to be compared with RM results, being more related to the role of the source precursors than sensitivity methods.

As introduced in Section 2.2, single sample models (e.g.: Chemical Balance Model, CMB), provide the source contribution estimates (SCEs) separately for each single PM sample. They also require as input information the emission profiles of the sources that are expected to influence air quality at the receptor sites. Multivariate models (e.g.: Principal Component Analysis, PCA; Positive Matrix Factorization, PMF) identify both the number of sources, each associated to a composition profile of the emitted PM, and their SCEs by processing the entire set of PM samples altogether. Composition profiles are then interpreted, usually on the basis of marker species or specific features of the chemical composition profile (e.g. ratio or relative weight of pairs of chemical species). Such analysis allows the different factors to be associated with the different sources (e.g.: road traffic, combustion processes, soil resuspension).

The comparison of the two families of models, that are inherently different and based on different input data, improves both the evaluation as well as the interpretation of the SA results. For example, RMs outputs are usually 'tuned' to reconstruct the measured PM concentrations through a variable number of source categories; while CTMs calculate the PM concentration based on known source categories derived from the emission inventories, therefore both source categories and PM concentrations can be compared. The comparison can concern the total observed and modelled mass, similar to the usual model performance evaluation or, more interestingly, it can provide information about the reliability of the source contribution estimated with the two approaches. Indeed, validation of the SA results is a critical issue because the true contribution of sources to the levels of one pollutant cannot be measured directly, therefore, the comparison of SA results obtained from different methods, such as SMs and RMs can help to overcome this limitation (Lane et al, 2007; Park et al., 2013). CTMs are designed to reconstruct a few main PM compounds, contributing to the major part of the bulk aerosol mass. Differently, RMs are designed to identify sources according to their chemical fingerprint. As a consequence, CTMs are generally skilful in reproducing sources providing a strong signal in term of mass, even if they are co-linear. Conversely, RMs can also capture the contribution from minor sources that may not be captured by CTMs, if they show a distinct marker pattern. For instance, RMs have been useful in identifying the role of sources like biomass burning, sea salt, primary biogenic aerosol or soil resuspension that have unique chemical markers, but are not well represented in the emission inventories. Despite the potential of combining RMs and SMs, this approach for SA evaluation is not commonly applied yet, as reported by Fragkou et al. (2012), since the simultaneous application of both SM and RM methods requires availability of tools, data, expertise as well as a substantial financial support and a considerable period of time.

4.1 Source categories association and species/compound association for PM validation

RMs and SMs reconstruct the relationship between sources and concentrations adopting similar but not completely overlapping concepts. Therefore it is particularly important to clarify some conceptual differences between the two approaches as well as to define some methodological steps that should be implemented in order to correctly compare their results.

Emission profiles (SMs) and emission source profiles (RMs)

In principle, each emission source is characterised by a specific emission profile that represents the relative chemical composition observed at the release point. Such profiles can be derived from literature or repositories such as SPECIEUROPE (Pernigotti et al., 2016) and SPECIATE (Hsu et al., 2014), or defined locally by means of specific experimental activities. Emission profiles are used by SMs to split emissions of aggregated compounds, such as PM and VOC, usually provided by emission inventories, into the model chemical species. The same kind of information is used to drive RMs based on chemical balance approach (e.g. CMB) in order to provide the model information about the “fingerprint” of the different sources. Therefore, **when comparing SMs with RMs like CMB is extremely important to the check that both models are fed with the same kind of emission profiles, at least for the main emission sources**. This practice helps the user in reducing the a-priori sources of discrepancies between the two approaches.

Table 3. Example of link between some emission categories tracked by the CAMx chemistry and transport model and the corresponding CMB profiles.

Emission category	*SNAP category*	*Fuel*	*Tagged categories*	*CMB classification*
Domestic and commercial heating	02	Biomass	Domestic heating - Biomass burning	Domestic heating - Biomass burning
Road transport - cars	07-01	Gasoline	Road transport - SI vehicles	Road Transport - SI vehicles
Road transport - cars	07-01	Diesel oil	Road transport - CI vehicles*	Road Transport - CI light duty vehicles*
Road transport - cars	07-01	LPG-Natural gas	Road transport - SI vehicles*	Road Transport - SI vehicles*
Road transport - cars **(tire and break wear)**	07-01		Road transport - Wear emissions	Road transport - Wear emissions
Road transport (except 0701)	07	Gasoline	Road transport - SI vehicles	Road Transport - SI vehicles
Road transport (except 0701)	07	Diesel oil	Road transport - CI vehicles*	Road Transport - CI heavy duty vehicles*
Road transport (except 0701) **(tire and break wear)**	07		Road transport - Wear emissions	Road transport - Wear emissions
Off road	08	Diesel oil	Transport - CI vehicles	Road Transport - CI heavy duty vehicles
Agriculture	10		Agricultural open burning	Agricultural open burning

* source categories where tagged species approach and CMB have different chemical profiles

Source: adapted from Pirovano et al., 2015

As an example, Table 3 shows the setup of a comparison between the CAMx source-oriented model and the CMB receptor model. The table shows the link between a subset of emission categories tagged in CAMx (Tagged categories column) and CMB available profiles (CMB classification). SNAP category and Fuel columns describe the sources included in emission inventory. CMB profiles have been used to create chemical profiles of the PM bulk emissions available from emission inventories. As can be inferred from the table, CMB profiles were not available with the same details of the source sectors in the emission inventory, for example “Road transport cars” using gasoline and natural gas (indicated with asterisk), partially limiting the detail in the emission chemical speciation. It is also worth noting that the source categories tracked by the CTM are less than the emission sectors available in emission inventories, due to a limit in computational resources. Moreover, in case of using factor analytical RMs (e.g. PMF), linking “factors” to emission inventory categories is not always straightforward. All these aspects have to be correctly taken into account when comparing SM and RM results, as discussed below.

An appropriate correspondence between source emission categories in the emission inventories used as input for CTM and the RM classification is essential when comparing RM and SM outputs.

Link between factors in RMs and source categories in SMs

As previously mentioned, the comparison between RM and SM results is generally not straightforward and it needs to be carefully designed.

The first and simplest case refers to the comparison of SM source categories and RM factors sharing exactly the same "emission fingerprint".

Referring to the previous example (Table 3), PM concentrations produced by the SM (CAMx) and RM (CMB) results could be directly compared, in terms of PM bulk mass and chemical composition, for all RM profiles except for diesel vehicles. For the latter, the SM tracks only one source category including emissions related to a pair of RM profiles (light and duty CI vehicles). This issue partially hampers a direct comparison between SM and RM results for diesel vehicles.

It is important to point out that a direct comparison can involve only the primary fraction of PM of the tracked emission category and it can be performed only if the RM profile describes the source fingerprint at the release point or, eventually, if such profile refers only to non-reactive compounds. Otherwise, in case of secondary compounds or RM profiles evaluated at the receptor, the consistency between the emission speciation profile in the SM source category and the corresponding RM profile is not guaranteed.

This is not an issue in case of RMs based on multivariate analysis techniques, such as PMF, which do not make use of emission profiles as input data. They identify a set of factors each representing a single source or an aggregation of sources. Factors still represent a relative distribution of chemical species, therefore identifying a chemical profile. But, such profiles describe the fingerprint of a source at the receptor site (source profile) not at the release point (emission profile) and in case of reactive or decaying compounds, the two profiles can differ. However, SM and RM results can be still compared; differences between the two approaches can be ascribed to the combination of three different aspects:

(a) differences in the PM bulk mass reconstructed by SMs for a source category with respect to the mass associated to the factor by RMs

(b) emission speciation profile adopted by SMs with respect to the chemical fingerprint of primary (e.g. emitted) compounds identified by RMs in the different factors

(c) contribution of secondary compounds reconstructed by SMs starting from precursor emissions with respect to the fraction of observed secondary species associated by RMs to the different factors

For all these reasons, in case of RMs based on multivariate analysis techniques, the comparison of chemical profiles derived from the SM results and the "factors" delivered by RMs can provide interesting information. Unfortunately, such a comparison is generally weakened by the limited chemical speciation of the PM concentration computed by SMs. In fact, SMs usually implement aerosol chemical modules dealing with less than 10 species, including a few primary compounds and the main secondary inorganic and organic species.

From a practical point of view, the definition of the source categories adopted in both RMs and SMs need to be carefully checked in order to correctly compare "apples and apples". There several cases that can be considered:

- SMs can handle fuel-resolved results for the same anthropogenic activity that could require to re-aggregated into the main source categories because RMs are not always able to capture them as distinct sources, due to their collinearity**. The re-aggregation has to be performed only for the comparison against RMs;**

- RMs can identify all kind of resuspension contribution (long range, natural dust, road dust, etc.) if enough species are measured while SMs can simulate different processes if they include algorithms to realistic represent them. For example, in most SMs applications, emission models are able to take into account natural dust resuspension, while they do not include road dust; such discrepancies must be taken into account when comparing the results of the two SA modelling approaches;

- As already mentioned most of secondary compounds accounted for SMs need to be re-associated to more generic "secondary" sources.

An example of the linking processes between RMs factors and SMs source categories is summarised in Tables 4 and 5 from the design of an Inter-comparison Exercise (IE) carried out in the framework of FAIRMODE WG3.

Table 4 shows the different source categories tracked by participating SMs defined according to the available emission categories with two different levels of details: mandatory including 8 source categories and optional including 14 source categories. **The maximum number of source categories that can be tracked by SMs depends on the detail available in the emission inventory.** In this IE, all emission categories were described in terms of SNAP sector, with the exception of a few key sectors (residential heating, road transport and international shipping) whose emissions were split also according the adopted fuel. But, as already mentioned, the number of source categories tracked by the SMs can represent just a subset of the emission categories, due to limitation in computational resources. Indeed, in the case of SA analysis based on BFM approach, each tracked category requires a whole model run. In order to take into account this computational limitation/problem, two category subsets were defined: mandatory and optional. The first one included just 8 source categories, while the second one was extended to 14 categories in order to give more details of the contributions from road transport, domestic heating and natural sources. On the basis of the two subsets, SMs reconstructed the contribution of the different source categories. Then, SA results produced by SMs were linked, whenever possible, to a set of RM factors. As shown in Table 5, the link between SM source categories and RM factors is not obvious, Indeed, there are source categories such as biomass burning for domestic heating, sea salt and dust than can be easily related to a corresponding source profile ("factor" for factor analytical RMs), while there are other categories that can be linked only to more general factor, such as industry, exhaust and shipping.

Primary and secondary species

Performing the link between SM source categories and RM factors is important to recall that many RMs do not perform a source apportionment of secondary pollutants. The most critical situation is generally related to secondary inorganic aerosol (SIA) which are usually handled by RMs as aggregated class called "SIA" unrelated to the source categories emitting the corresponding precursors (e.g Transport, heating, agriculture). In case of secondary organic aerosol (SOA), RMs can provide more detailed information about the origin of the precursors, for example being able to distinguish the role of natural biogenic sources and anthropogenic sources, where the latter can be also distinguished with respect to the fuel (e.g. biomass vs fossil fuel). SMs, on the other hand, can always tag both primary and secondary compounds to a corresponding emission source because, in case of secondary pollutants, they can tag the corresponding precursors. As a result of these inherent differences between RMs and SMs, additional assumptions are required when comparing the results of the two approaches. In particular, it is necessary to reshape the contribution computed by SMs for both primary and secondary compounds of a specific source, assigning, when necessary, the secondary fraction to more general and aggregated "secondary sources" identified by RMs. As mentioned before, a typical example concerns SIA. Indeed, while SMs are able to track the contribution of each emission category to Nitrate (e.g. Transport, Heating, Energy production), RMs usually define either a generic "Nitrate" or "Ammonium Nitrate" source including all contributions. In this case each Nitrate contribution computed by SMs for the different emission sources must be merged into a generic "Nitrate" source. One of the few exceptions to this approach is agriculture whose contribution is largely due to ammonium (more than 90%). Therefore, in this case, being the factor simply identified by a secondary species, ammonium was not removed from the corresponding source category.

Table 4. Set of source categories (mandatory and optional), adopted in the FAIRMODE IE

SNAP (and fuel)	***Description***	***Mandatory 8 sources***	***Optional 14 sources***
1	Energy industry	01_ENI	01_ENI
2-1	R & C combustion, coal	99_OTH	02_OTH
2-2	R & C combustion, light liquid fuel	99_OTH	02_OTH
2-3	R & C combustion, medium liquid fuel	99_OTH	02_OTH
2-4	R & C combustion, heavy liquid fuel	99_OTH	02_OTH
2-5	R & C combustion, gas	99_OTH	02_OTH
2-6	R & C combustion, solid biomass (wood)	02_BIO	02_BIO
34	Industry (combustion & processes)	34_IND	34_IND
5	Fugitive emissions from fuels	99_OTH	99_OTH
6	Product use including solvents	99_OTH	99_OTH
7-1	Road transport, exhaust, gasoline	07_RTR	71_RTG
7-2	Road transport, exhaust, diesel	07_RTR	72_RTD
7-3	Road transport, exhaust, LPG/natural gas	07_RTR	07_RTR
7-4	Road transport, non-exhaust, evaporation	07_RTR	07_RTR
7-5	Road transport, non-exhaust, wear	07_RTR	75_RTW
8	Non-road transport	99_OTH	99_OTH
8-1	International shipping, marine diesel oil	08_SHP	08_SHP
8-2	International shipping, heavy fuel oil	08_SHP	08_SHP
9	Waste treatment	99_OTH	99_OTH
10	Agriculture	10_AGR	10_AGR
11P	Dust	11_DST	11_DST
11	Sea Salt	99_OTH	11_SLT
11	Biogenic SOA	99_OTH	11_BSO

Source: Belis et al., 2020

Table 5. Correspondence between SM sources (SNAP) and RM sources (SPECIEUROPE) defined in the FAIRMODE for mandatory and optional subsets of the IE.

SM sources Mandatory	*RM corresp. Mandatory*	*SM sources Optional*	*RM corresp. Optional*
01_ENI	30 fuel oil combustion or 28 power plant	01_ENI	30 fuel oil combustion or 28 power plant
99_OTH		02_OTH	
02_BIO	40 biomass burn.	02_BIO	40 biomass burn.
34_IND	20 industry	34_IND	20 industry
07_RTR	1 traffic	71_RTG	2 exhaust
		72_RTD	
		07_RTR (OTH)	
		07_RTR (OTH)	
		75_RTW	5 road dust
08_SHP	37 ship	08_SHP	37 ship
99_OTH		99_OTH	
10_AGR	NH4 sum	10_AGR	NH4 sum
11_DST	10 dust	11_DST	10 dust
99_OTH		11_SLT	12 marine, 71 aged sea salt
99_OTH		11_BSO	
99_OTH		99_OTH	

Source: Belis et al., 2020

4.2 Comparison of source contributions from SMs and RMs

This section summarises the main findings of several studies that provide a comparison between SMs and RMs.

A first example is provided by Bove et al., (2014) who compared CAMx and PMF source apportionment results for $PM_{2.5}$ over the area of Genoa (Italy). PMF was applied over a set of daily samples collected at three sites for six months in 2011. The CAMx chemistry and transport model was applied for the same period over the Genoa area through two nested domains using 10 and 1.1 km grid resolutions. CAMx was driven by the WRF meteorological model, fed by GFS analysis fields. Emissions for the coarse domain were derived from the TNO emissions inventory, while the high resolution emissions inventory from Liguria region was used for the inner domain. Natural emissions (including biogenic VOC, sea salt and dust) were computed by NEMO model (Liora et al., 2015; 2016).

CAMx/PSAT was setup in order to track five source categories: Maritime and Harbour activities (SNAP code 0804), Road Transport (SNAP code 07), Energy production and Industry (SNAP codes 01, 03, 04), Non-industrial combustion plants (SNAP code 02, labelled as "Residential") and "Other sources" (including boundary conditions). CAMx and PMF results were compared over June-August 2011.

Observed $PM_{2.5}$ concentrations ranged between 12.6 and 14.0 μg/m^3 at the three sites. CAMx was able to capture the overall evolution of bulk $PM_{2.5}$, showing a good performance for sulphate, partial underestimation for OC, and clear difficulty in reproducing EC and nitrate.

PMF results identified 5 factors labelled as: traffic (Cu, Zn, Pb), Soil Dust (Al, Ti), Heavy Oil Combustion (V, Ni), Secondary Sulphates ($SO4_2^{=}$, NH_4^+), and Secondary Nitrates (NO_3^-). In order to compare CAMx and PMF results, Bove et al. split the contribution of the two "secondary factors" (Sulphate and Nitrate, respectively), into SNAP categories and additional factors. The split of the secondary factors was based on several assumptions mainly derived from the analysis of the chemical profile of the "primary factors (i.e. traffic, soil dust and heavy oil combustion) and the relative weight of each factor. Nitrate mass was shared between the Traffic (6:10), Heavy Oil combustion (3:10) and Natural sources (1:10, sea spray). The OM mass fraction of secondary factors was assigned entirely to Traffic, thereby neglecting any regional OC contribution. Furthermore, approximately 10% of the Secondary Sulphates were attributed to the biogenic emissions and then accounted for in Other Sources. The remaining concentration of sulphate was then attributed to an additional factor representing Energy Production and Industry sectors.

The reallocation of the PMF factors thus permitted more coherent comparison of CAMx SA results and PMF outcomes. The comparison showed some qualitative agreement between SM and RM results, particularly concerning the relative ranking of the three main sources: road transportation, energy production/industry and maritime emissions, accounting for 40% - 50%, 20% - 30% and 10% - 15%, of $PM_{2.5}$, respectively.

On the other hand, to make SM and RM results comparable, the authors introduced several assumptions about the origin of the secondary compounds that increased the uncertainty associated to the PMF results and that would require additional investigations in other contexts with different groups of prevailing sources.

A second interesting study is described in Napelenok et al. (2014) that aimed to verify and improve model capabilities in predicting PM concentration due to Residential Wood Combustion (RWC). The study was based on CMAQ/ISAM chemistry and transport model and UNMIX receptor oriented model (Henry et al., 1989, 1990).

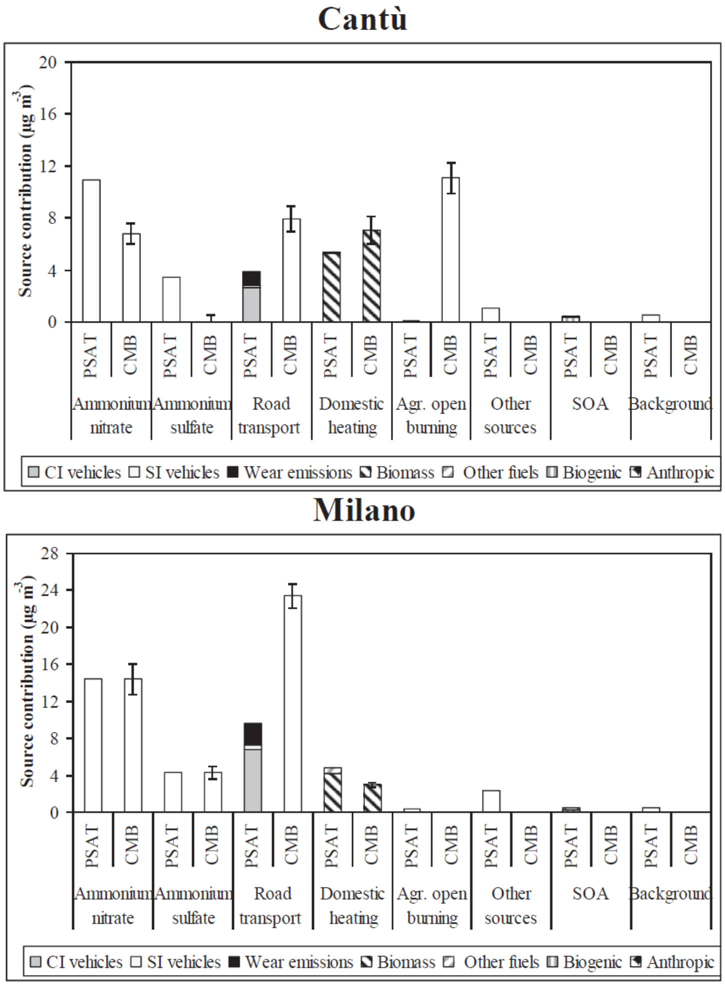
The observed dataset was based on $PM_{2.5}$ composition data collected in 15 sites located in the South East of U.S.A. during 2007. Observed data included levoglucosan. CMAQ simulations were fed with the U.S.A. National Emission Inventory (NEI) for year 2007/2008. The model was applied over the whole U.S.A. at 12 km grid resolution. Meteorological fields were provided by WRF meteorological model. The embedded SA module (ISAM) was then setup in order to track the contribution of 10 biomass-combustion source categories including, among others; Fire (prescribed and wild), Woodstoves, Household waste burning, Open burning for land clearing, Burning of crop residue and Fireplaces. According to NEI data, the largest contribution to $PM_{2.5}$ emissions of the ten biomass-burning categories comes from prescribed wildfires, whereas woodstoves and fireplaces (RWC) contribute just a minor fraction. As a first step, OC concentrations computed by CMAQ for each tracked category were transformed into corresponding levoglucosan concentrations and then summed up in order to be evaluated against available observations. As a first guess, the UNMIX receptor model was applied to the observed dataset, including levoglucosan, mannosan and galactosan plus 8 other species were available at the 15 sites. UNMIX detected three factors associated with biomass combustion, but only one of them showed a clear fingerprint for “fire emissions”. In order to augment the information retrievable from the observed dataset, a further step, combining the SM results and RM features was carried out. The SA results produced by CMAQ/ISAM for the 10 categories were used as additional concentration of fictitious “chemical species” together with the observed levoglucosan concentrations, to produce a hybrid synthetic and observed dataset. Once fed with this new dataset UNIMIX detected 7 factors, each clearly associated to one or just a few CMAQ source categories, also including a corresponding repartition of total observed levoglucosan concentration. One of these factors was clearly associated to RWC, thus allowing a comparison with the corresponding contribution of levoglucosan concentrations simulated by CMAQ/ISAM. The comparison clearly pointed out an underestimation of the modelled contribution that could be ascribed to the underlying emission inventory. The results were then used to explore possible refining of the RWC emission estimates as well as of the temporal allocation methodology. As for the latter, some improvements in the yearly-to-daily and diurnal profiles gave rise to improvements in model predictions of carbonaceous $PM_{2.5}$.

A third study concerning the comparison of RM and SM results was carried out by Pirovano et al., (2015) based on CAMx chemistry transport model and CMB8.2 (US EPA, 2004; Watson et al., 1984). An ambient daily $PM_{2.5}$ dataset based on a multi-year campaign (PARFIL project 2004-2007) was collected at 9 sites in Lombardy representative of different environmental conditions. Local source profiles were used for domestic biomass burning, agricultural open burning, and tyre and brake wear (Colombi et al., 2006), while literature profiles were used for other sectors such as road transport. CAMx version 5.41 was applied over a modelling domain of 580 x 400 km^2) including the whole Po Valley at 5 km horizontal resolution. Input meteorological data for 2005 reference year were generated by the WRF model, while gridded and point source emissions were based on three different kinds of inventories: regional based (for Lombardy, Veneto, Piedmont, and Emilia-Romagna regions); the ISPRA national based inventory (for the remaining Italian regions) and the EMEP database (http://www.emep.int/) for the non-Italian fraction of the domain. The source profiles used for the CMB analysis, were also adapted to “speciate” the bulk PM emissions for the corresponding categories, namely road transport, domestic heating and agricultural open burning, in order to reduce as much as possible the sources of difference between the two SA approaches, as discussed in §3.1.

The PSAT tool was configured to track several emission categories including transport sector, residential heating, energy production and agriculture. Where relevant, emission categories were also defined according to the fuel used. CAMx/PSAT source categories results for primary PM were re-aggregated as domestic heating (split by fuel), transport (split by engine type and wear source), agriculture, background (including natural sources and long-range transport), and other sources (essentially including stationary industrial sources), as indicated in Table 5. The secondary compounds were re-associated as: ammonium nitrate, ammonium sulphate and SOA.

The results showed that CMB showed a better reconstruction of the $PM_{2.5}$ mass closure while CAMx systematically underestimated PM concentrations, particularly during severe pollution episodes in the cold season, due both to difficulties in reproducing very stable meteorological conditions and inaccuracies in the emission inventories. Nevertheless, both models provided the same ranking for SCEs at several receptors, with a general agreement in the reconstruction of secondary inorganic aerosol contributions and the most relevant discrepancies related to road transport and domestic heating.

Figure 6. Example of site by site comparison of $PM_{2.5}$ SCEs at PARFIL sites of Cantù and Milan in the cold season.



Source: adapted from Pirovano et al., 2015

CMB had difficulty with the apportionment of emission from biomass combustion processes and provided rather questionable results at two urban receptors for the cold season, not identifying the road transport contribution that, conversely, was clearly pointed out by CAMx/PSAT. However, CAMx/PSAT and CMB provided similar results at some receptors as those shown in Figure 6, concerning the comparison of winter season mean SA concentrations: Milano and Cantù (North of Milan) receptors.

Additionally, CMB proved to be unable to resolve the traffic source contributions by engine and fuel type, simply recognising the total traffic contribution. These limitations of the CMB approach may be either due to peculiarities in the environmental dataset of aggregated seasonal data, which reduced the relative variability between the two kind of sources, or to the source profiles, despite the use of local profiles for some sources, or to the combination of both these reasons.

On the other hand, simulating the dispersion from the various sources, CAMx/PSAT could distinguish the contribution of sources sharing similar profiles and strongly correlated, such as spark and compression ignition vehicles. CAMx/PSAT could also identify the contribution of secondary PM deriving from complex chemical transformation, such as anthropogenic and biogenic SOA or related to different emission areas, such as the long-range transport burden.

CAMx/PSAT experienced difficulty in reconstructing carbonaceous species, with OC systematically underestimated at all sites, particularly during the winter season. By comparing apportioned OC against CMB results, the authors were able to show that missing OC was mainly related to road transport, mostly as a consequence of deficiencies in the emission inventories concerning the primary OC from traffic in the cold season and of the concurrent underestimation of both primary emission, including the emission of semi-volatile organic carbon, and secondary OC formation.

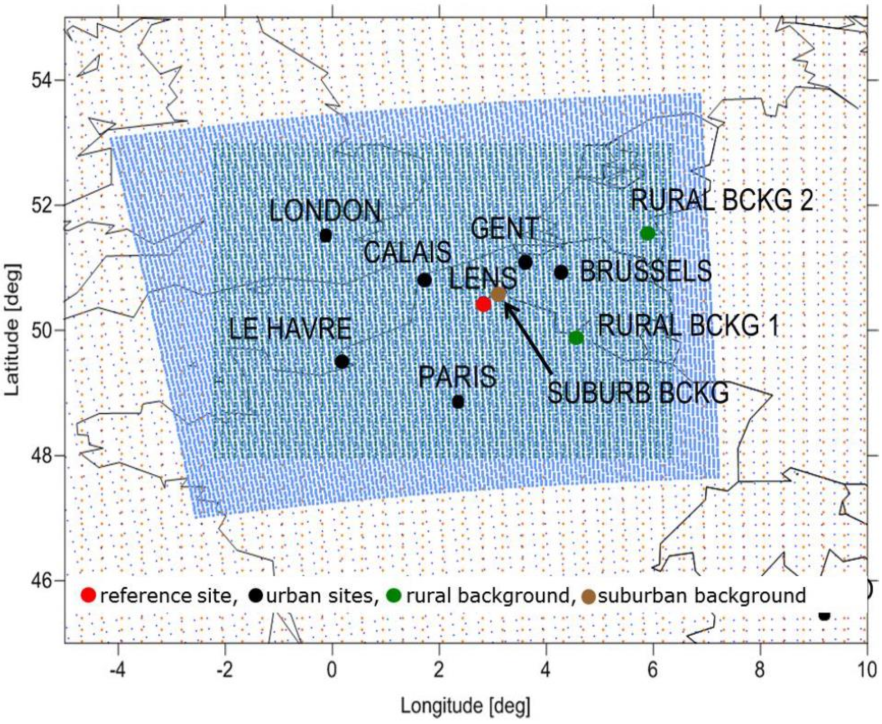
5 Intercomparison between different SMs and between SMs and RMs

Finally, the most comprehensive attempt to compare SM and RM results is the FAIRMODE intercomparison exercise (IE, Belis et al., 2020). The exercise involved more than 40 modelling teams, mostly applying RMs.

One of the distinctive features of this inter-comparison is that both RMs and SMs were applied on the same study area so called “reference site”. This approach allowed the creation of an unprecedented dataset of independent and comparable SM and RM source contribution or impact estimates (SCIEs). One of the key features of the exercise was the evaluation of the performance of the SA models, by comparing the model results with reference values established by means of a well-defined methodology (Belis et al., 2015a, b).

The IE focused on the area of Lens, in the North of France, where a comprehensive dataset of measurements was available for PM_{10} daily 24h concentrations collected every third day between March 2011 and March 2012 (116 samples) (Waked et al., 2014). Observed data included PM_{10} mass concentrations and concentrations of many species to give a detailed PM chemical composition. Participating SMs were applied over a computational domains based on two nested grids covering the whole Europe as well as the area of Lens.

Figure 7. Map zoomed on Lens domain with indication of the receptors selected for reporting results. FAIRMODE-LENS domain (green dots); WRF_FAIRMODE domains (blue and light blue dots).



Source: Belis et al., 2020

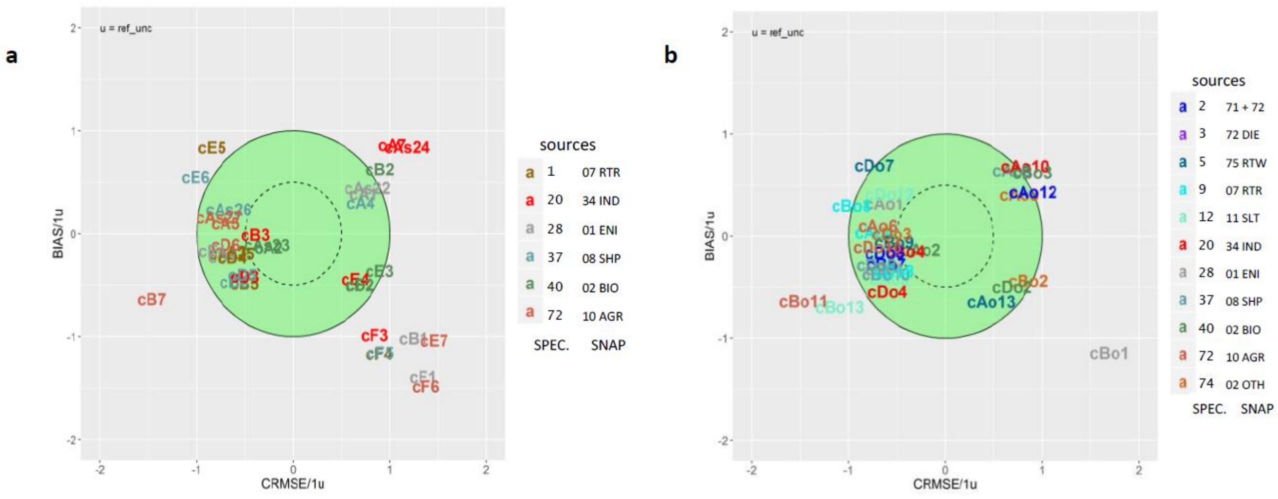
The inner domain used a grid resolution corresponding to the one of the emission inventory (around 7 km). SMs were applied over two periods: Summer from 1/6/2011 to 31/08/2011 and Winter from 15/11/2011 to 15/2/2012. These two periods were selected to be representative of both “hot” and “cold” seasons in France, as well as being long enough to include high and low levels of air pollution. Moreover, observed data being available every third day, three months of simulation were needed to pair at least 30 daily source apportionment results of RMs and CTMs. SMs results were delivered and analysed at Lens receptor as well as at a set of additional sites being representative of different emissive and meteorological features.

Meteorological fields were provided by WRF model fed with GFS analysis fields, while the emission inventory was provided by TNO over a geographical grid covering the whole Europe with a horizontal resolution of 0.125x0.0625deg, corresponding to approximately 7 km x 7km. Emissions were classified according to the SNAP sector, except for a few key source categories (Domestic and Commercial Heating, Road Transport and Shipping) that included also details about the main fuels (see Table 3).

As already mentioned, the SA exercise for SMs was designed in order to track two possible sets of source categories with increasing detail, mandatory and optional (Table 4). The results produced by SMs were firstly validated according to the usual Model Performance Evaluation (MPE) techniques (see Belis et al., 2020 for additional details) and evaluated according to the SA performance evaluation methodology (Belis et al., 2015a) suitably adapted to SMs features and results.

The evaluation was performed in two steps: the first one involved only SMs and therefore the reference profiles used for SA validation were based only on SM results. The second step involved also the RM results that were used as a reference for the evaluation of SMs.

Figure 8. Target plot for the mandatory (a) and optional (b) sets in the site of Lens. SMs are tested against the ensemble average of all models. The alphanumeric codes indicate the SMs and sources (see explanation in the text).



Source: Belis et al., 2020

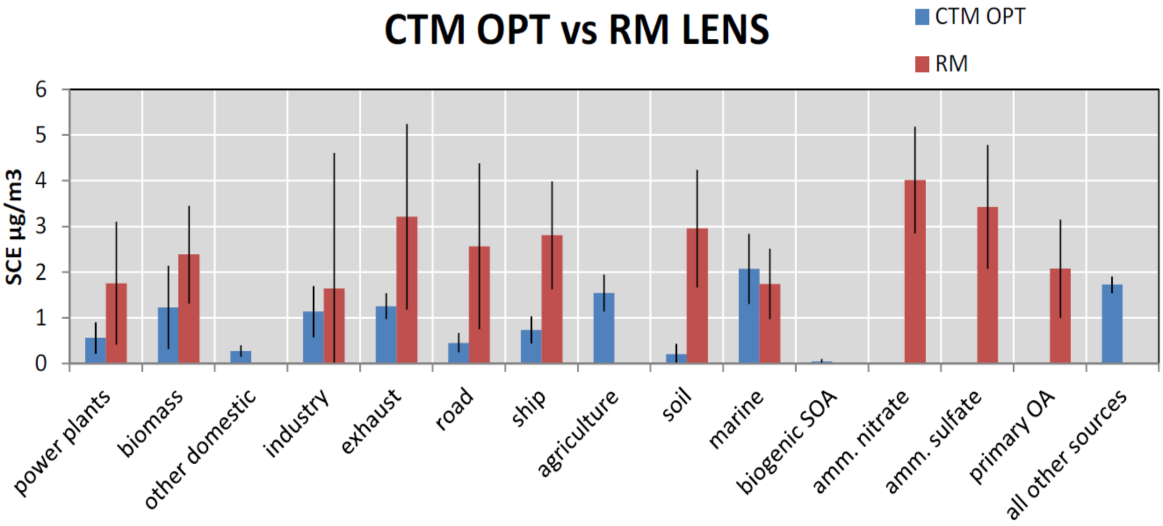
Figure 8 shows the evaluation of the first step, particularly the target plot for the different models and categories for optional and mandatory datasets, respectively. The target plots are used to display the result of the evaluation because provide a clear and prompt information about the "dispersion" of the SA results produced by SMs, when they are compared with a reference value obtained as the average of all the results. The scores of the models for the single sources are represented in the target plot by alpha numeric codes and colours. The prefix "c" indicates it is the result of a CTM run while the capital letters indicate the used model: CAMX-PSAT (A), FARM (B), LOTOS-EUROS (D), EURAD (E) and CHIMERE (F). In Figure 8a, are shown the results for the mandatory set of sources (Table 4), while Figure 8b displays the results for the optional set of sources (Table 4), denoted with the suffix "o". The last number of the code denotes the sources of each model result in sequential order. The legend to the right of each plot indicates the colour corresponding to each source category (Table 5).

In Lens, the share of results meeting the performance criteria of the testing methodology (inside the green circle of Figure 8) ranges between 60% (mandatory) and 73% (optional). In the mandatory set, industry is often out of the acceptability area. The contributions from agriculture are underestimated in three results cB (FARM), cE (EURAD) and cF (CHIMERE). With the exception of biomass burning, the contributions from cF are always underestimated with a prevailing amplitude problem. In cB energy is underestimated and biomass burning overestimated while cE overestimates ship and traffic contributions.

The performance evaluation methodology also supports additional investigations, such as comparing subsets of SMs results (e.g. SMs adopting tagging methods vs SMs adopting BFM techniques).

Then, as mentioned before, the IE allowed –for the first time- a comprehensive and coherent comparison of RM and SM results. Indeed, both kind of models referred to the same receptor and the same temporal coverage, making thus possible the analysis of only those differences related to the modelling approaches. A preliminary step of such comparison was to define a link between SM source categories and RM factors, which is shown in Table 5. As already mentioned one of the critical issues when matching the two series of sources is the comparability of the inorganic ions ammonium, sulphate and nitrate, that in the RMs are attributed by definition to one or two secondary sources (ammonium nitrate, ammonium sulphate) while the CTMs allocate these chemical species to the corresponding precursor sources.

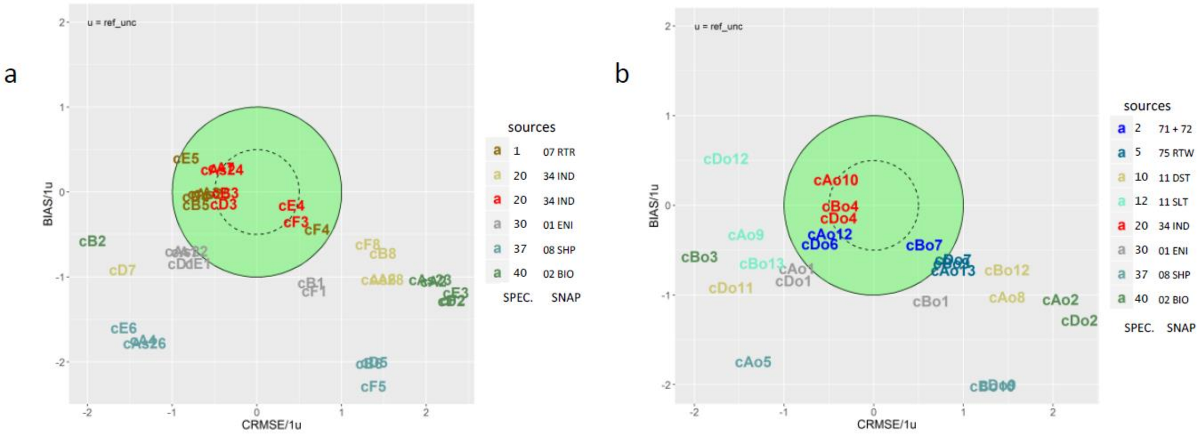
Figure 9. Mean and standard deviation of the SCIE reported in the present intercomparison obtained with RMs and CTMs for the optional set of sources.



Source: adapted from Belis et al., 2020

As an example of the obtained results, Figure 9 shows the comparison between the mean source contributions or impact estimates (SCIEs) reconstructed by SMs for the optional set of sources (CTM OPT) and RMs at Lens receptor. Such a comparison represents the most comprehensive cross-validation of the two families of models (RMs and SMs) based on a multi-model and multi-user database. It is very important to keep in mind that the evaluation was not based on direct observation of the different source contributions (because they cannot be measured), it is based instead on modelling results. Therefore, it is extremely important to carefully check the assumptions and the limit of validity that is behind such comparison.

Figure 10. Target plots for the CTM mandatory (a) and optional (b) sets against a reference profile derived from RM results at the site of Lens. The meaning of the alphanumeric codes is the same as Figure 8.



Source: adapted from Belis et al., 2020

With specific reference to Figure 9, the comparison pointed out that SCIEs deriving from RMs are always higher than the corresponding value reported with CTMs. The only exception is marine aerosol where the two families of models are very close (less than 10% difference). The most important primary sources in RMs are: exhaust, soil, ship, road dust and biomass burning. Traffic, agriculture, industry and biomass burning are the most important sources in the CTM mandatory set. The different level of performance shown by the source categories can help in designing steps to improve the overall performance of the SMs. For instance, verify whether SM underestimation can be ascribed to factors not depending from the sources (e.g. meteorology and long range transport) or can instead be related to deficiencies in emission inventory due to a poor representation of some emission processes.

The evaluation of the SM performance involved also other steps, based on the methodology developed by Belis et al. (2015). A second analysis of the obtained results is shown in Figure 10, where the SCIEs produced by SMs are compared against a reference profile derived from RMs results using pre-established acceptability criteria. Unlike Figure 9, the target plot evaluates the modelled SCEs on daily basis and not only as a seasonal average. Moreover, in this case, the performances are displayed separately for each model. This comparison therefore shows how far each source category and model was from the reference profiles.

Traffic (including both exhaust and resuspension components) and industry were the most comparable sources between CTMs and RMs. The marine source presented little bias, however, showed differences in the amplitude of the time series between the two families of models in the target plot. On the contrary, the most critical sources in the comparison between CTMs and RMs are dust and road dust, likely due to the different way in which the former was reconstructed by the different models and a possible underestimation of the latter in the emission inventory. For a more detailed explanation of the SA model evaluation see Belis et al. (2020).

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List of abbreviations and definitions

AQD	air quality directives
BFM	brute force method
CMB	chemical mass balance
CTM	chemical transport models
ERI	emission reduction impact
IE	intercomparison exercise
MS	member state
OA	organic aerosol
OPT	optional set of sources
PM	particulate matter
PMF	positive matrix factorization
RM	receptor oriented models / receptor models
SA	source apportionment
SCE	source contribution estimations
SCIE	source contribution or impact estimations
SM	source oriented models
SNAP	Selected nomenclature for air pollution

List of figures

Figure 1. Information flow in PSAT for an example of two source classes and the SO_2–sulphate system. 14

Figure 2. Flow chart of the TSSA algorithm.......16

Figure 3. ISAM flow chart 17

Figure 4. Overview of the DEHM tagging method. 18

Figure 5. Distribution of SA studies based on project APPRAISAL. 19

Figure 6. Example of site by site comparison of $PM_{2.5}$ SCEs at PARFIL sites of Cantù and Milan in the cold season. 33

Figure 7. Map zoomed on Lens domain with indication of the receptors selected for reporting results. FAIRMODE-LENS domain (green dots); WRF_FAIRMODE domains (blue and light blue dots). 35

Figure 8. Target plot for the mandatory (a) and optional (b) sets in the site of Lens. SMs are tested against the ensemble average of all models. The alphanumeric codes indicate the SMs and sources (see explanation in the text). 36

Figure 9. Mean and standard deviation of the SCIE reported in the present intercomparison obtained with RMs and CTMs for the optional set of sources. 37

Figure 10. Target plots for the CTM mandatory (a) and optional (b) sets against a reference profile derived from RM results at the site of Lens. The meaning of the alphanumeric codes is the same as Figure 8. 37

List of tables

Table 1. RMs-SMs applications in Europe. 20

Table 2. SM applications in Europe. 21

Table 3. Example of link between some emission categories tracked by the CAMx chemistry and transport model and the corresponding CMB profiles. 27

Table 4. Set of source categories (mandatory and optional), adopted in the FAIRMODE IE 30

Table 5. Correspondence between SM sources (SNAP) and RM sources (SPECIEUROPE) defined in the FAIRMODE for mandatory and optional subsets of the IE. 31

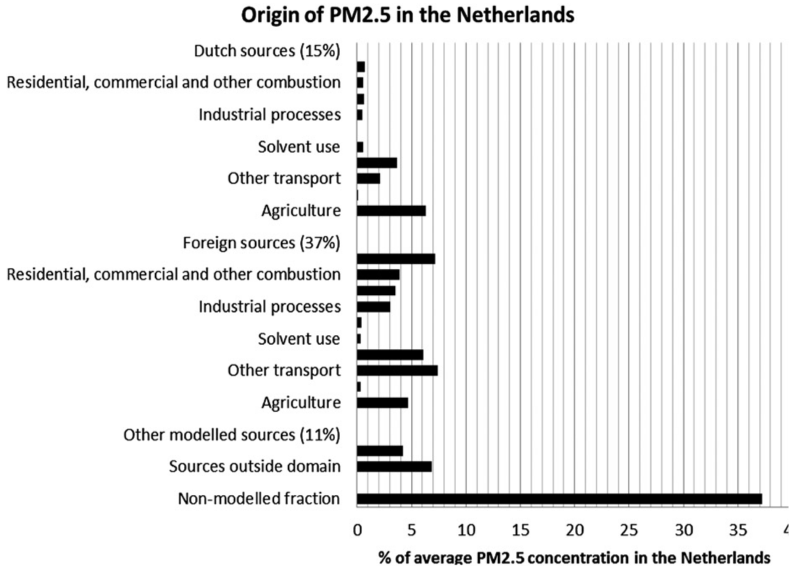
Annex 1. Applications of SMs and SMs-RMs models for estimating particulate matter source contributions in Europe Annexes

A1. Air quality management:

The origin of ambient particulate matter concentrations in the Netherlands

To be able to develop effective mitigation strategies, the origin of particulate matter needs to be established. The regional air quality model LOTOS-EUROS, equipped with a labelling routine, was used to establish the origin of PM_{10} and $PM_{2.5}$ in the Netherlands for 2007-2009 at the source sector level, distinguishing between national and foreign sources. A simulation across Europe at 0.5° longitude x 0.25° latitude (about 28 x 28 km) resolution was performed, the results of which were used as boundary conditions for a simulation at a resolution of 0.125° longitude x 0.0625° latitude (7 x 7 km) over the Netherlands. For anthropogenic PM and trace gas emissions we used the European TNO-MACC emission database for 2007 and replaced the data for the Netherlands with the emissions reported by the official Dutch Pollutant Release & Transfer Register. Emissions of sea salt, dust and biogenic NMVOC emissions were calculated online. The model results were evaluated against PM and PM component concentrations from rural and suburban stations of the Dutch National Air Quality Monitoring Network. In addition, data from a dedicated measurement campaign (August 2007-October 2008) were used for a more detailed comparison including carbonaceous aerosol and mineral dust. There were no assimilation techniques used.

Figure 1. Average $PM_{2.5}$ source apportionment to national and foreign sectors for the Dutch territory.



Source: TNO

The results suggest that 70-80% of modelled PM_{10} and 80-95% of $PM_{2.5}$ in the Netherlands is of anthropogenic origin. About 1/3 of anthropogenic PM_{10} is of Dutch origin and 2/3 originates in foreign countries. Agriculture and transport are the Dutch sectors with the largest contribution to PM_{10} mass in the Netherlands, whereas the foreign contribution is more equally apportioned to road transport, other transport, industry, power generation and agriculture. For the $PM_{2.5}$ fraction, a larger share is apportioned to foreign and anthropogenic origin than for PM_{10}, but the same source sectors are dominant. The national contribution to PM levels is significantly higher in the densely populated Randstad area than for the country on average and areas close to the borders. In general, the Dutch contribution to the concentration of primary aerosol is larger than for secondary species. The sectoral origin varies per component and is location and time dependent. During peak episodes, natural sources are less important than under normal conditions, whereas especially road transport and agriculture become more important.

Source oriented models for PM underestimate the observed particulate matter concentrations systematically. Here, the non-modelled fraction (mostly carbonaceous material) was quantified and potential contributions discussed. In addition, the apportionment of ammonium nitrate to the sectors emitting ammonia and NOx is not straightforward.

Reference: Hendriks et al., Atmos. Environ., 69, 289-303, 2013.

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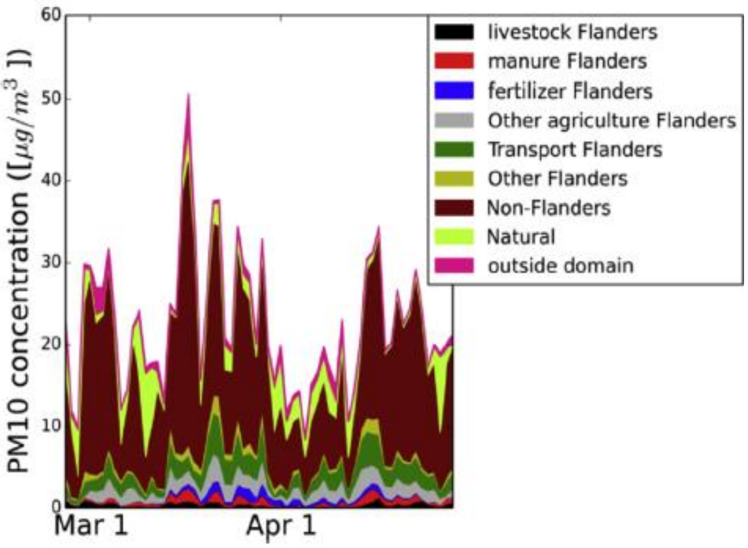
Establishing the contribution of agriculture to particulate matter in Flanders

Agriculture is one of the dominant source sectors for particulate matter in Europe. Agricultural activities directly emit particles, but the largest contribution is made though the formation of secondary inorganic aerosol (SIA). In this study the contribution of agriculture to annual average and episodic levels was quantified.

The regional air quality model LOTOS-EUROS, equipped with a labeling routine, was used to establish the agricultural contribution to PM_{10} and $PM_{2.5}$ in Flanders for 2007-2011. A simulation across Europe at 0.5° longitude x 0.25° latitude (about 28 x 28 km) resolution was performed, the results of which were used as boundary conditions for a simulation at a resolution of 0.125° longitude x 0.0625° latitude (7 x 7 km) over Belgium and the Netherlands. For anthropogenic PM and trace gas emissions we used the European TNO-MACC-II emission database for 2011 and replaced the data for Flanders and the Netherlands with the emissions reported by the national inventories. In this study the meteorological influence on the temporal variability of ammonia emissions from livestock housing and application of manure and fertilizer are included in the chemistry transport model LOTOS-EUROS. For manure application, manure transport data from Flanders (Belgium) were used as a proxy to derive the emission variability. The model results were evaluated against ammonia and PM component concentrations from rural and suburban stations. There were no assimilation techniques used.

Figure 2. Source attribution of PM10 in Flanders during spring, 2011



Source: TNO

Using improved ammonia emission variability strongly improves model performance for ammonia, mainly by a better representation of the spring maximum. The impact on model performance for SIA was negligible as explained by the limited, ammonia rich region in which the emission variability was updated. The contribution of Flemish agriculture to modelled annual mean SIA concentrations in Flanders were quantified at 1-2 µg/m^3. During spring time about half of the PM apportioned to Flanders derived from its agricultural sector (See Figure 1).

The main challenge of the study was the update of the temporal emission variability consistently across the different emission inventories.

***Reference**:* Hendriks et al., Atmos. Environ., 131, 83-96, 2016.

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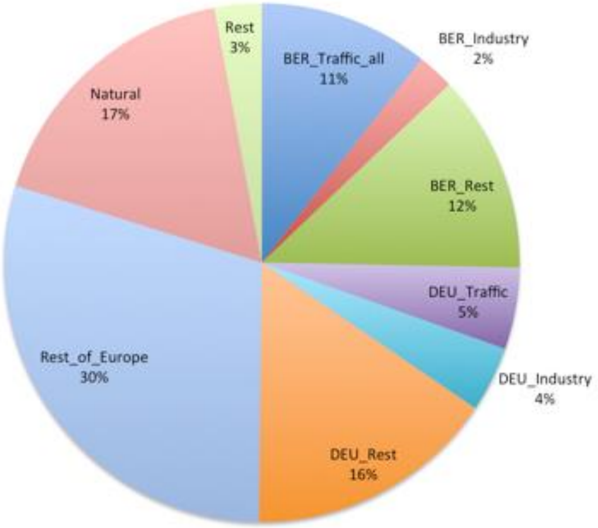
On the origin of particulate matter in Berlin, Germany

Although particulate matter levels are slowly decreasing, Berlin regularly observes particulate matter concentrations above the daily limit value. In this study the LOTOS-EUROS model has been set-up at the Freie Universitaet Berlin for source apportionment purposes.

The regional air quality model LOTOS-EUROS was used to quantify the contributions of long range transboundary transport as well as German and urban contributions to urban background levels in Berlin. A simulation across Europe at 0.5° longitude x 0.25° latitude (about 28 x 28 km) resolution was performed, the results of which were used as boundary conditions for a simulation at a resolution of 0.125° longitude x 0.0625° latitude (7 x 7 km) over eastern Germany. For anthropogenic PM and trace gas emissions we used the European TNO-MACC-III emission database updated with reported national totals in 2016. The model results were evaluated against PM component concentrations from the urban background station Neukölln. There was no assimilation techniques used.

Figure 3. Source attribution of PM10 in Berlin for 2015.



Source: Freie Universität Berlin

The simulations showed that on average the modelled PM_{10} concentration derives for about one quarter from local sources, one quarter from German sources and one third from anthropogenic activities abroad. The remainder is explained by natural sources. To illustrate that the tracing of sector dependent regional categories is possible the contributions of traffic and industry were separated out. As expected, traffic has a larger share in the local contribution than in the long range transport. The German industrial contribution is about similar as the German traffic contribution in Berlin.

The evaluation shows that the organic aerosol mass is underestimated. In addition, few SIA episodes occur which are largely underestimated by the model although the concentrations in general are reflected very well. Hence, future work will focus on the analysis of these particular episodes and how to improve the modelling for these.

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Study 1: BelEUROS

In this study the CTM BelEUROS (Deutsch et al., 2008) was used to assess the contribution of 6 different emission sectors to the PM_{10}, $PM_{2.5}$, and O_3 concentrations in Flanders. Both the contribution of the Flemish and the emissions outside Flanders on the air quality in Flanders was quantified. The model was also run for both the reference year 2007 and 3 future scenarios representing possible emission scenarios for the year 2020. The BelEUROS model area extends from -8.25° to 20.35° East and from -36.9° to 67.15° North and effectively covers a large part of Europe. The model resolution is 60 km over the whole domain and encompasses a grid refinement area where the resolution is enhanced to 15 km for the part covering Belgium. The model uses 6-hourly reanalysed ECMWF meteorological data for 2007. The emission data is based on the officially reported data and were gridded using the EMAP model (Maes et al., 2009). The emission projections for 2020 were provided by IIASA. The model does not use data assimilation

The calculation procedure consisted of doing calculations in which the emissions of individual emissions sectors or of all sectors were reduced by 20%. This reduction was selected as a compromise to assure on the one hand significant concentration changes in BelEUROS concentrations while on the other hand avoiding that the chemical regime would be totally different. For PM_{10} and $PM_{2.5}$ the results for the 20% reduction were then extrapolated to a result for 100% reduction by multiplying the concentration changes with a factor 5. For O_3 the results for the 20% decrease were considered as such. Emission reductions were applied both for the Flemish emissions and the emissions outside Flanders.

The study highlights the importance of reducing both emissions inside and outside Flanders to improve air quality. To improve the air quality with respect to PM the two most important abatement measures for emissions in Flanders are the reduction of primary PM emissions and the reduction of ammonium emissions from agriculture. According to the model results, the benefits of Flemish emission reductions will also increase in the future as Europe wide emission reductions will result in a decrease of the pollutant surplus in the air that is responsible for the significant contribution of transboundary pollutant transport. In other words, air quality will in the future be more dependent on Flemish emission reductions than on the reduction of emissions outside Flanders.

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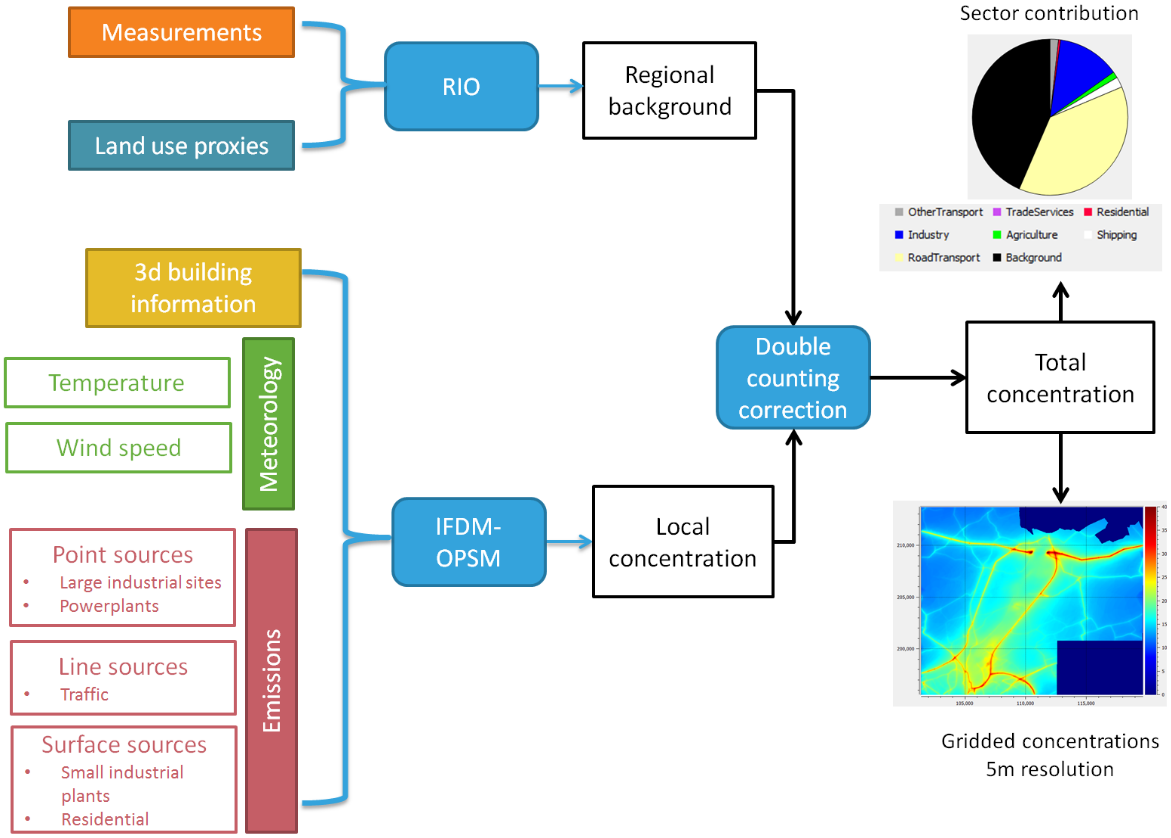
Study 2: RIO-IFDM-OSPM

In this study, VITO has used the RIO-IFDM-OSPM model chain to perform *local-scale* source apportionment. Only the pollution originating locally (i.e. in a radius of 20km from the location of interest) is characterised, all the other pollution is labelled as 'background concentration'. For the local concentration, we determine which sector is emitting the pollution. The list of sectors is depend on the details of the emission input information, but usually we at least include transport (road, shipping and rail), residential, industrial, and agricultural emissions.

Figure 4: Methodology used in the RIO-IFDM-OSPM source apportionment studies.

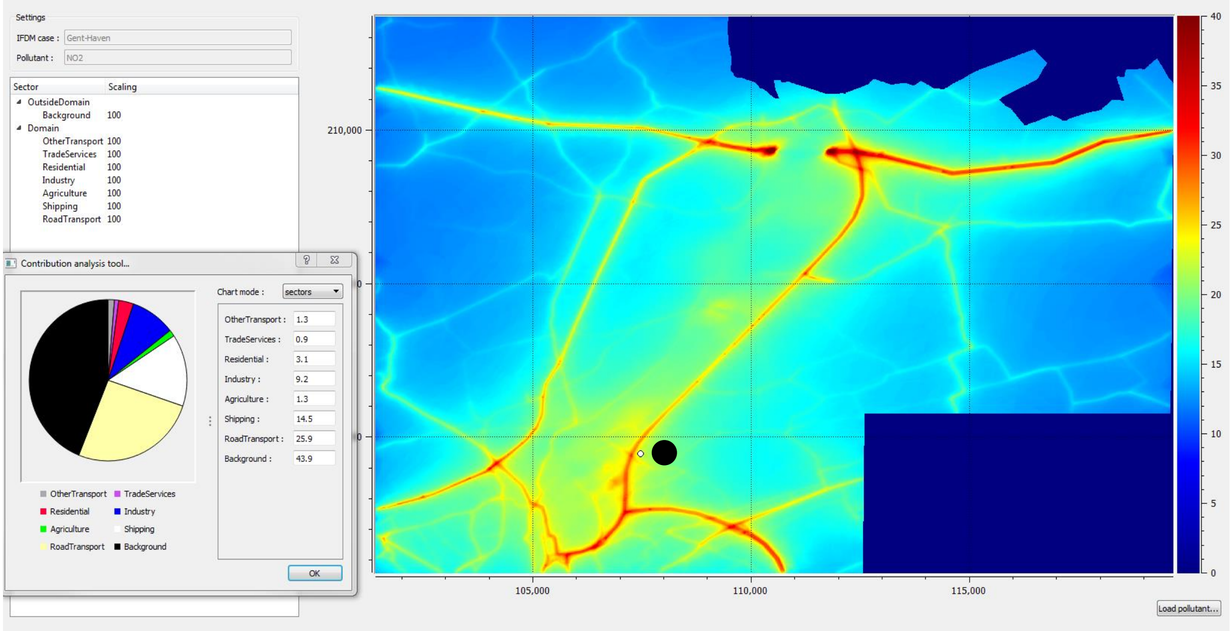


Source: VITO

The methodology relies on three models, each modelling the pollutant concentration for NO_2, PM_{10}, $PM_{2.5}$ and Black Carbon (BC) at a different scale. The RIO-model, a land-use regression model based on the data of the official monitoring network in Belgium, provides the regional background concentration. The local increment due to, amongst other, traffic and industrial emissions is calculated using IFDM, a bi-Gaussian plume model designed to simulate non-reactive pollutant dispersion at a local scale. To avoid double-counting of the sources, a special coupling procedure between the regional model (RIO) and the urban-scale model (IFDM) is applied. Finally, to calculate street level concentrations in street canyons, the RIO-IFDM chain is coupled to the OSPM box model. The methodology is illustrated in Figure 4.

To determine the contribution of each sector to the pollution at a certain location, we perform several model runs with the model chain, where, in each run, the emissions of only one of the sectors have been retained. The unexplained fraction is labelled as 'background'. This fraction contains not only the pollution emitted outside the domain, but also secondary pollution and pollution caused by unknown sources in the domain. The results are visualised in the IFDM Analyzer tool, as shown in Figure 5.

Figure 5: Visualisation of the results. The large map shows the yearly mean NO2-concentration in the port of Ghent, the small inset shows the contribution of the different sectors at the marked location.



Settings

IFDM case : Gent-Haven

Pollutant : NO2

Sector	Scaling
OutsideDomain	
Background	100
Domain	
OtherTransport	100
TradeServices	100
Residential	100
Industry	100
Agriculture	100
Shipping	100
RoadTransport	100

Contribution analysis tool...

Chart mode : sectors

OtherTransport : 1.3
TradeServices : 0.9
Residential : 3.1
Industry : 9.2
Agriculture : 1.3
Shipping : 14.5
RoadTransport : 25.9
Background : 43.9

Source: VITO

References

Lefebvre, W., Vercauteren, J., Schrooten, L., Janssen, S., Degraeuwe, B., Maenhaut, W., de Vlieger, I., Vankerkom, J., Cosemans, G., Mensink, C., Veldeman, N., Deutsch, F., Van Looy, S., Peelaerts, W., Lefebre, F. Validation of the MIMOSA-AURORA-IFDM model chain for policy support: modeling concentrations of elemental carbon in Flanders, Atm. Env., 45/37, 6705-6713., 2011b. doi: 10.1016/j.atmosenv.2011.08.033

Lefebvre W., Van Poppel M., Maiheu B., Janssen S., Dons E. Evaluation of the RIO-IFDM-street canyon model chain, Atmospheric Environment 77, 325-337, 2013

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SA studies with air pollution models in support to AQPs in Germany

We use SM in the framework of air quality planning for a considerable number of cases with limit value exceedances in NRW. See *http://gis.uba.de/website/umweltzonen/lrp.php* for a list of current air quality plans in NRW (46 at present, number still growing). Due to the number of cases (much higher than 46, as there are often several street canyons with exceedance situations to be considered in each plan, number still growing), we cannot fill in the questionnaire for each case study separately. The setup and the models used are not identical for all cases and throughout the years. In the following we are trying to give a general description of our proceeding, focusing on our current approach.

Usually we focus on annual average concentration of NO_2 and PM_{10}, even though in this survey only PM_{10} is looked at.

Please note that our main problem in current air quality planning is NO_2, not PM_{10}. NRW for example had no PM_{10} limit value exceedances for the last 3 years. Therefore, we are somewhat surprised that NO_2 is not target of this survey. When handling NO_2 there are additional challenges and problems to be tackled.

None of our models used for local contribution includes a complex air chemistry module, radiation or a gas to particle conversion module. For PM_{10} it is not the annual average concentration of PM_{10}, but the allowed number of exceedance days that poses a challenge to air quality planning. The number of exceedance days is generally not calculated explicitly by our models. We use a parameterization based on an empirical relationship between annual average PM_{10} concentration and number of exceedance days.

We address source categories, not source regions. The area investigated ranges from a street canyon to a whole city/several cities. The horizontal resolution ranges from single meters to 250 m and in some cases up to 1 km.

The regional background concentration is determined either by field observations or by EURAD-model simulations. For the urban background and the contributions of local sources we use several models for different purposes. LASAT (*http://www.janicke.de/de/lasat.html)* is used for all urban source groups (e.g., industry, heating, off road and so on). For road traffic contribution we use, depending on the situation, LASAT (mainly for urban traffic, seldom for local traffic), Miskam (mainly for complex building situations, *http://www.lohmeyer.de/de/node/134*), Immis^{Luft} (used in most of our cases of street canyons, *http://www.ivu-umwelt.de/front_content.php?idcat=94*), and/or RLuS (mainly for autobahn, *http://www.lohmeyer.de/de/node/493*). Meteorological data (wind speed, wind direction, stability of the atmosphere) stems from measurements, in most cases from the German Weather Service (DWD). The emissions are taken from the emission inventory NRW (see *http://www.lanuv.nrw.de/emikat97/startfr2.htm*). The emissions from road traffic are calculated according to the current HBEFA at the time of the study.

LASAT and Miskam use open boundaries for the outflow. Immis^{Luft} and RLuS are simple screening models. All models take into account the roughness length, though usually set as a constant value for the whole model area. When necessary we include building data and/or orography in the models.

The models have been validated by their developers in various studies. Additionally, we usually compare the results with field measurements using the data quality goals of EC/2008/50.

We are not using data assimilation methods, apart from those applied for the calculation of the regional background (EURAD-IM, EURAD-Fladis).

The model results give us information on source apportionment and in some respect on air pollution levels and can also be used to estimate the effectiveness of measures.

The number of exceedance days is not calculated explicitly by our models. We use a parameterization based on an empirical relationship between annual average PM_{10} concentration and number of exceedance days. Using a much more sophisticated method to calculate the daily average PM_{10} concentration does not necessarily lead to better results. Most of the time PM_{10} concentrations are ruled by long range transport. For PM_{10} we often overestimate the measured values when diffusive sources near ground level have to be considered. Some known problems with the modelling approach are: Input data often has uncertainties, e.g., emission data are being updated in a four year turn, and may additionally contain uncertainties that are not always quantifiable, but not negligible, e.g., with regard to diesel vehicle emissions. Model uncertainties may also sum up owing to the use of different models with different scales and resolutions. When calculating the urban road traffic contribution with LASAT for the whole city and then calculating the local road traffic contribution for a street with a high resolution, it is necessary to ensure that the local road traffic is not taken into account twice (from the city-wide and the local simulation run). Some physical and chemical processes are implemented by relatively simple parameterizations in the models. The high number of cases to be considered rules out the usage of more complex models that may or may not give more accurate results.

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SA studies with air pollution models in support to AQPs in Slovakia

Lagrangian air quality model CALPUFF has been used for the PM_{10} source apportionment in air quality management areas (AQMAs) in Slovakia.

Local PM_{10} emissions include traffic emissions, domestic heating, industrial and point sources, and fugitive sources. CALPUFF has been driven by meteorological fields created by CALMET meteorological model. Domains ranged between 20–400 km^2 in size, with the horizontal resolution of 200–500 m, depending on the complexity of the terrain. The results were post processed using multi linear statistical model, in order to account for the emission input uncertainties. In most AQMAs the major contributors are local heating using biomass burning (in winter) and road transport in both seasons, with high contribution from regional and transboundary transfer.

Krajčovičová, J. Matejovičová, J. Kremler, M. Nemček, V., 2016: Air quality modeling of non-attainment areas as a basis for air quality plans. 17th Conference on Harmonization Within Atmospheric Dispersion Modeling, Budapest, Hungary, 9-12 May 2016

Krajčovičová, J. - Kremler, M. - Matejovičová, J.: Local PM_{10} source apportionment for non-attainment areas in Slovakia. In: Int. J. Environment and Pollution, 2014. *https://doi.org/10.1504/IJEP.2014.065117*

Krajčovičová, J. Matejovičová, J. Szabó, G. (2010). 'PM_{10} source apportionment for non-attainment areas based on routinely available data'. 13th Conference on Harmonization within Atmospheric Dispersion Modeling, 1–4 June, Paris, France

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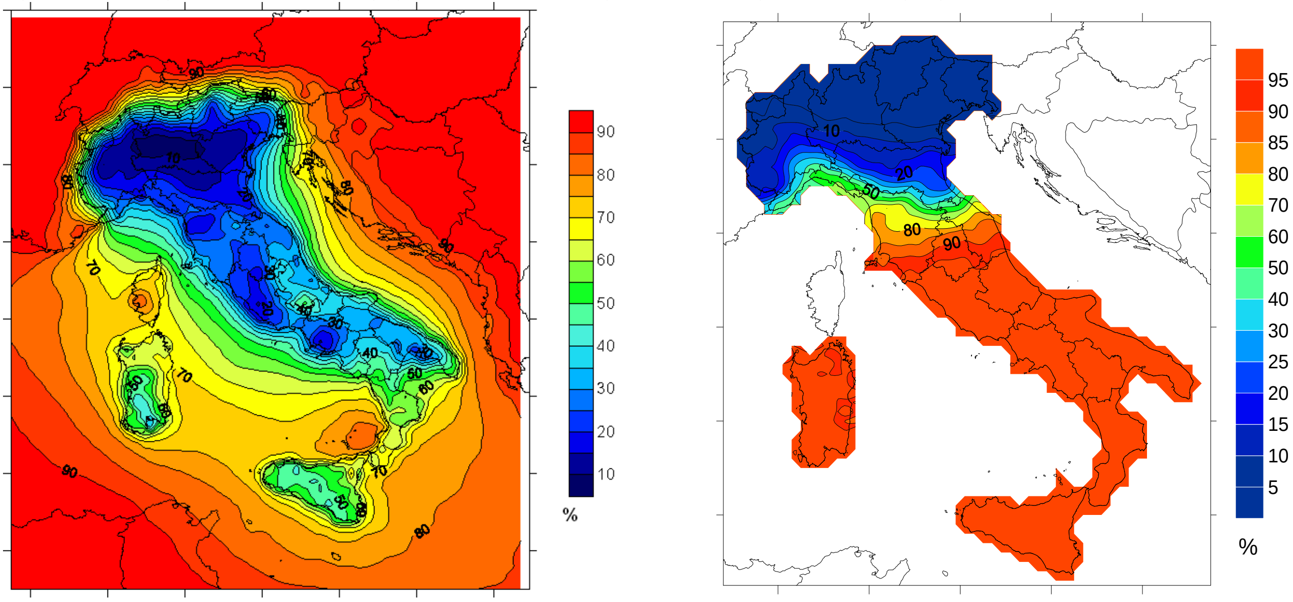
Source Apportionment studies in Italy at national and regional scales

ENEA (Italian National Agency for New Technologies, Energy and Sustainable Development) supported the Italian Ministry for Environment and Territory and Sea in preparation of the reports for postponement of attainment of PM_{10} limit values required by Air Quality Directive 2008/50/EC (27 January 2009). The study investigated 67 air quality zones located in 14 regions in order to support also the regions in designing their air quality plans for the zones with exceedances.

The methodology used in all the studies was Brute Force Method (BFM) and the simulations were performed with MINNI modelling system which is composed of an atmospheric modelling system (AMS-MINNI) (Mircea et al., 2016) and an Integrated Assessment Model (IAM) derived from GAINS-Italy (Greenhouse Gas - Air Pollution Interactions and Synergies, D'Elia et al., 2018). The two components complement each other, offering a powerful tool that provides policy makers with detailed analyses on emission and concentrations spatial and temporal variability which can be tailored to the specific national, regional and local needs.

Source apportionment estimates have been performed to separate the transboundary contribution to PM_{10} concentrations and to understand the relative contribution of the rest of Italian territory on each Italian administrative region for the year 2005. Such examples are shown in Figure 6 where the percentage contribution of foreign pollution to PM_{10} concentrations over Italy and the contribution of the rest of Italy on Po Valley pollution to PM_{10} in, respectively.

Figure 6. Percentage contribution of foreign pollution to PM$_{10}$ in Italy (left) and of Italy to the Po Valley (right)



Source: ENEA

References:

D'Elia I., Piersanti A., Briganti G., Cappelletti A., Ciancarella L., Peschi E., 2018. Evaluation of mitigation measures for air quality in Italy in 2020 and 2030. Atmospheric Pollution Research, 9: (2018), 977–988.

Mircea, M., Grigoras, G. D'Isidoro, M., Righini, G., Adani, M., Briganti, G., Ciancarella, L., Cappelletti, A., Calori, G., Cionni, I., Cremona, G., Finardi, S., Larsen, B.R., Pace, G., Perrino, C., Piersanti, A., Silibello, C., Vitali, L., Zanini, G. (2015) Impact of grid resolution on aerosol predictions: a case study over Italy, Aerosol and Air Quality Research, 16: 1253–1267, 2016.

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A2. Other studies

The study performed in the frame of the MED-APICE project (www.apice-project.eu)

PM$_{2.5}$ samples were collected by low-volume sequential samplers placed in the urban area of Genoa in three sites. Two out of the three sites are key nodes of the municipal air quality monitoring network and are located immediately outside the harbour area: Corso Firenze (44º25'5.71"N; 8º55'38.98"E) and Multedo (44°25'37.18"N; 8°49'49.21"E). The third site was located in a northern district, namely Bolzaneto (44°27'45.92"N; 8°54'4.40"E), about 7 km inland. Very briefly, Corso Firenze is located in a residential area nearby a road with moderate traffic, the Multedo station lies along a private road with a very limited traffic, and the Bolzaneto station is located in a suburb with some industrial activities, at about 400 m from an important highway. The samplers were operated almost continuously and simultaneously from May 2011 to October 2011 with Teflon and Quartz fibre filter membranes. In each site, PTFE and Quartz filters were alternated (i.e. one day PTFE and the other day Quartz) every 24 hours.

A PMF analysis was carried out separately with the data sets of each sampling site, using the time series of PM concentration values including single elements, ionic species, OC and EC. Variables were selected according to the signal-to-noise criterion (Paatero and Hopke, 2003) and 18 variables were finally retained in the analysis: Al, Si, K, Ca, Ti, V, Mn, Fe, Ni, Cu, Zn, Pb, SO_4^{2-}, NO_3^-, NH_4^+, Na^+, OC and EC.. All the concentration values and their associated errors were treated according to Polissar et al. (1998). Since PMF is affected by the rotational ambiguity, it directly implements rotations in the minimization algorithm which is controlled using the FPEAK parameter (Paatero et al., 1997). In this work, the best rotation for each factor was chosen in the FPEAK range from -2 to +2 discarding the rotations producing profiles without physical meaning (i.e. sum of elemental concentrations greater than 100%) and selecting rotations giving concentration ratios between elements typically considered tracers of natural sources (e.g. crustal matter) comparable to literature values. The PMF analysis was performed separately for the three data sets (Corso Firenze, Multedo, Bolzaneto); however, five factors were resolved everywhere and labelled, according to their characteristic tracers, reported in parenthesis, as follows: *Traffic* (Cu, Zn, Pb), *Soil dust* (Al, Ti), *Heavy Oil Combustion* (V, Ni), *Secondary Sulphates* (SO_4^{2-}, NH_4^+), *Secondary Nitrates* (NO_3

Secondary inorganic compounds turned out to be the major components of $PM_{2.5}$ (on average: *Sulphates* ~ 50%, *Nitrates* ~ 7% of $PM_{2.5}$, respectively) with percentages very similar in the three sites. *Traffic* resulted everywhere the major primary source of $PM_{2.5}$ with a variability related to the specific site location. The *Heavy Oil Combustion* impact on $PM_{2.5}$ was higher in the sites closer to the harbour even if significant at the inland site too (~ 13%, 11% and 9%, respectively in Corso Firenze, Multedo and Bolzaneto). Actually, no major resident source (i.e. oil-fuelled power plants) was and still is active in the area and ships emissions were very likely the predominant source of *Heavy Oil Combustion*.

Source apportionment was obtained through an integrated air quality forecasting system too. Meteorological fields were obtained by the state-of-the-art non-hydrostatic mesoscale model WRF-ARW, version 3.2.1., whereas air quality simulations were performed using the photochemical dispersion model CAMx. Three nested computational domains in a Lambert Conic Conformal projection were defined for the WRF model, covering Western and Central Europe with horizontal resolution of 10 km, Northern Italy with horizontal resolution of 3.3 km and the urban area of Genoa with grid spacing of 1.1 km. The number of terrain-following vertical levels adopted was 35, with higher resolution close to the surface. Initial and boundary conditions for atmospheric simulations with the WRF model were generated from the operational global model GFS outputs (0.5°x0.5° resolution). Particulate matter Source Apportionment Technology (PSAT) has been implemented into the CAMx model and is publicly available. PSAT estimates the contribution from specific emissions source groups, emissions source regions, initial conditions, and boundary conditions to PM using reactive tracers. In this study we used CAMx version 5.2 with Carbon Bond 2005 (CB05) gas phase chemistry mechanism (Yarwood et al., 2005). PPM advection solver by Colella and Woodward was used for horizontal advection (Odman et al., 1993), while a simple K-theory approach was used to describe turbulent vertical diffusion. Hertel's EBI solver was adopted to solve gas-phase time evolution (Hertel et al., 1993), while aerosol chemistry was described by the coarse-fine (CF) scheme. To limit the computing time and due to the lack of adequate emission data on the intermediate WRF domain, just two 2-way nested CAMx modelling domains were defined: an outer domain with horizontal resolution of 10 km and an inner one coincident with the 1.1-km resolution WRF finest domain. Sixteen vertical levels were defined and the ten lowest CAMx layers exactly matched the WRF layers, the surface layer corresponding to about 30 m above ground level. Large-scale anthropogenic emissions data for gaseous (NO_x, SO_x, NH_3, CO, NMVOCs) and particulate (PM_{10}, $PM_{2.5}$) pollutants were calculated processing the 2005 European dataset by TNO with the MOSESS code (Markakis et al., 2013). High-resolution (1 km) gridded emission data were obtained instead from the Liguria Region inventory. Finally, biogenic and natural emissions (Isoprene, Monoterpenes, biogenic other NMVOCs, sea salt and wind-blown dust) were computed from WRF outputs using the NEMO model (Poupkou et al., 2010).

Source apportionment for PM_{10} and $PM_{2.5}$ have been evaluated by CAMx in two periods: Summer (June - August 2011) and late Autumn (November, 15 - December, 15 2011). Categories to be apportioned have been defined following the declaration of SNAP and have been chosen according to the analysis of local emission inventory, where sources expected to have high impact on Genoa air quality can be identified. In particular, five PM source categories were considered: Maritime and Harbour activities (SNAP code 0804), Road Transport (SNAP code 07), Energy production - Industry (SNAP codes 01, 03, 04), Non-industrial combustion plants (SNAP code 02, in Genoa area mainly residential sources, hence labelled as "Residential" hereafter), Other sources (including boundary conditions). Even if the PSAT results are available for the whole domain, source apportionment outcomes are here focused for the three sites where the $PM_{2.5}$ monitoring campaign was performed: Corso Firenze, Multedo and Bolzaneto. The main impact is related to road traffic, and minor contributions are given by industries and by maritime activities. A seasonal trend can be identified in both coastal and inland sites. In fact, during winter period a strong increase in the contribution of "Residential" sources is observed, which can be ascribed indeed to the presence of residential heating emissions. Moreover, in the coastal sites, which lie near the harbour (almost inside when considering the spatial resolution of simulation domain), a strong reduction of maritime activities contribution is observed during winter period, when passengers ship traffic in the harbour is lower (effect mainly related to the seasonal tourism trend). The maritime contribution to $PM_{2.5}$ concentrations varies among the three sites between 4% and 11% in summer, whereas in winter decreases to 3% - 5%. Finally, the comparison between coastal and inland sites is consistent with what expected, in particular considering that lower contribution of harbour activities to $PM_{2.5}$ is observed for the inland site.

The $PM_{2.5}$ apportionment obtained with the two approaches (i.e. PMF and CAMx-PSAT) was compared considering the time period common to the two data sets, i.e. June - August 2011. Actually, with PMF we extracted the factor profiles processing the entire data sets while we calculated the average $PM_{2.5}$ apportionment for the quoted three months only. The comparison presented two main difficulties:

The way to single out PM sources is completely different: while with the receptor model approach a "source" (or "factor", following the PMF terminology) is basically a group of PM components which concentration values remain constant in time (or, from another point of view, show correlated time trends), the identification of sources by CTMs is based upon the emission inventory categories structure, that is defined by the user via a bottom-up approach with each activity associated to a specific emission pattern.

Secondary components of PM are resolved and treated as the other primary PM sources in the receptor model approach (i.e. they have a specific profile which corresponds to a certain fraction of the whole PM), while with CTMs the impact of each source/activity to the PM level is calculated including both the primary and the secondary components of the emission pattern, therefore the CTM does not (and actually cannot) resolve any kind of "secondary" source to be compared with a PMF factor.

Trying to overcome these two problems, we rearranged the $PM_{2.5}$ sources singled out by PMF and considered the factor profiles to redistribute the PM associated to *Secondary Sulphates* and *Nitrates* to the other primary sources. Moreover, the source names were uniformed to the PSAT classification, in particular: *Traffic* was just renamed as *Road Transport*, *Heavy Oil Combustion* was considered as entirely related to the *Maritime* sector, the remained *Sulphates* (i.e. after the corrections above described) was included in the *Energy Production - Industry* sector (according to the emission inventory, a 340 MW coal-fuelled power plant located in the harbour area is largely the main source of SO_x and sulphates in the study area). This is a quite crude approximation since part of the *Secondary Sulphates* concentration could be possibly due to other source categories (in particular: the SO_4^{2-} relative concentration in the *Heavy Oil Combustion* PMF profiles could not incorporate the total of secondary $PM_{2.5}$ due to ships emissions even considering the 24 hours sampling time of each sample): in this respect the figure obtained for *Energy production – Industry* (see below) should be considered more likely as an upper limit. *Soil dust* and the "local" sources resolved in some sites were added and labelled, together with the natural component of *Sulphates* and *Nitrates*, as *Other sources*.

Despite the several approximations introduced in the comparison, the overall picture shows a fair agreement with the apportionment by CAMx-PSAT. The global picture obtained with the repartition of the secondary $PM_{2.5}$ quantified by the field campaigns and associated by PMF to two main secondary *sources* was basically confirmed by the CAMx-PSAT analysis and showed that the emissions related to traffic, energy production and maritime activities contributed for about 40% - 50%, 20% - 30% and 15% of the total $PM_{2.5}$ in summer 2011, respectively.

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KJ-NA-30082-EN-N

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doi:10.2760/470628

ISBN 978-92-76-10698-2