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## Current trends in the use of models for source apportionment of air pollutants in Europe

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**Abstract:** Forum for Air Quality Modelling in Europe (FAIRMODE) sub-group 2 (SG2) on the 'Contribution of natural sources and source apportionment' has been formed in response to the need for a harmonised European approach in the use of models for source apportionment, addressing the requirements of the current Air Quality Directive 2008/50/EU. Within SG2, a review was performed on source apportionment modelling methods used by member states for the preparation of their extension reports regarding compliance with PM<sub>10</sub> limit values. This review was extended to identify the modelling source apportionment methodologies used by member states for various pollutants. The extended study was performed by directly addressing a questionnaire to the national representatives of 38 countries of the European region and to 50 national experts. The responses revealed the widespread use of both receptor and dispersion models but also demonstrated a number of issues related to the validation of the source apportionment methodology applied.

**Keywords:** Forum for Air Quality Modelling in Europe; FAIRMODE; source apportionment; atmospheric pollutants; receptor models; dispersion models.

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## 1 Introduction

Air quality is one of the major environmental problems of European countries, particularly in urban agglomerations. Ozone and particulates are among the main substances of concern with the most frequent exceedances of limit values and subsequent public health impacts. For  $PM_{10}$ , this is also reflected in the significant number of European Union (EU) countries that have applied for a postponement of attainment of  $PM_{10}$  limits until 2011, according to Article 22 of the Air Quality Directive (AQD) 2008/50/EU. For this purpose, the European Commission (EC) requested that the main sources of pollution contributing to the observed concentrations are identified and that source apportionment (SA) should be precise enough to allow an adequate understanding of which measures should be taken to address the main sources of pollution in future efforts. Although not explicitly mentioned in the AQD, the use of modelling tools in combination with measurements is recommended for the purpose of SA, as monitoring of

the contributions from all emitting sources in an air quality management zone would be unrealistic and economically unfeasible. The Forum for Air Quality Modelling in Europe (FAIRMODE) aims to harmonise modelling approaches in Europe for regulatory purposes. Sub-group 2 (SG2) on the 'Contribution of natural sources and source apportionment' has been formed within the frame of FAIRMODE, with the scope to provide guidance and suggest best modelling practices and quality assurance procedures for member countries, in order to promote harmonised model use for SA in the EU. Within the activities of SG2, two targeted reviews have been performed in order to obtain up-to-date information on the type and frequency of modelling tools that are used by member states for SA. The first review (Review 1) is mainly focused on the use of models for preparing the postponement applications for attaining PM<sub>10</sub> limit values, and the second review (Review 2) is requesting information on model use for SA of other regulated pollutants and the relevant evaluation procedures of the methodologies applied. These two reviews are presented in the following sections of the present paper.

## **2 Review 1: the use of models in postponement applications for attaining PM<sub>10</sub> limit values**

The material used for this Review 1 included information from a number of sources apart from the analysis of the extension reports submitted by member countries in support of their applications for postponement to comply with PM<sub>10</sub> limit values. Namely, Review 1 also used information from a relevant database compiled within the frame of the COST Action 633 and from a workshop that took place at the JRC premises in Ispra in October 2006 on the 'Quantification of the contribution of natural sources to the ambient PM concentrations'. Indicative recent publications from member countries on SA with the use of models were also consulted. Review 1 focused on the use of models for SA regarding PM<sub>10</sub>.

A review of European publications, which were reported in the questionnaires submitted by EU countries within the frame of the COST 633 Action, revealed that principal component analysis – PCA was the most frequently used modelling method for SA, as it appeared in 30% of the studies, while back-trajectory analysis was represented in 11% of the studies (Viana et al., 2008). Other receptor models were also frequently used, such as positive matrix factorisation – PMF (8%), chemical mass balance – CMB (7%) and mass balance analysis (7%). In a relevant study by Samara et al. (2003), CMB modelling results from the analysis of PM<sub>10</sub> samples collected from an industrialised urban area of Northern Greece during June 1997 to June 1998 showed that the major source of PM<sub>10</sub> was diesel vehicle exhaust, but significant contribution from industrial oil burning was also evidenced at the sampling site next to the industrial area.

The use of dispersion models for source identification and quantification of corresponding contributions was found to be pronounced when natural sources of particulate matter were specifically addressed, according to the presentations from ten EU countries which participated in the workshop on 'Quantification of the contribution of natural sources to the ambient PM concentrations' in October 2006 in Ispra. Modelling was used in 90% of the cases, with the exception of the Netherlands, as the main focus of the relevant presentation was on sea-salt contribution, for which case the use of modelling tools is currently limited, but gradually growing. 50% of the countries have used dispersion models, mainly Eulerian chemical transport models (CTM), while 40%

of the countries reported the application of receptor models for SA. In order to enhance the reliability of the methodology, 30% of the countries have applied back-trajectory analysis in combination with other modelling methods.

The analysis of indicative recent publications suggested that, recently, several EU countries have relied on the combined use of available emission data and a dispersion model to estimate source contributions. For example, the hybrid Swedish AIRVIRO dispersion model has been applied in a number of European cities including Prague, Riga, Vilnius and Tallinn. The Gaussian ADMS-Urban model has been used for SA and for the evaluation of proposed emission reduction measures in Poland, within the frame of an air quality management project in Cracow between 2005 and 2006 (Adamczyk et al., 2007). Eulerian dispersion modelling systems have been used recently in Mediterranean member states to assess the Saharan dust contribution to ambient particle concentrations. In the studies by Astitha et al. (2005) and Kallos et al. (2006), the SKIRON/ETA dispersion forecasting system was applied for urban Mediterranean regions, while Rodríguez et al. (2001) have combined SKIRON results with back-trajectory analysis to determine the proportion of Sahara-induced exceedances with respect to the total annual exceedances in Southern Spain.

The increased use of dispersion models and of the combination of models for the quantification of natural contributions was also reflected in the technical reports submitted by EU countries in support of their application for postponement of attaining  $PM_{10}$  limit values. As the AQD allows for a three-year extension of the attainment deadline in case that exceedances can be explained due to contributions from natural sources, adverse climatic conditions and specific dispersion characteristics, the applicant EU countries had to develop a reliable methodology to:

- a confirm that a significant number of exceedances or high annual mean concentrations was due to natural sources
- b to quantify the proportion of these exceedances
- c to determine the extent to which the different natural sources were responsible for these exceedances by estimating the  $PM_{10}$  concentrations resulting from their relevant emissions.

At the time of preparation of Review 1, the EC has announced decisions for the time extension applications of 17 EU countries, including 289 air quality management zones. A demand for extending the period for attainment of the daily  $PM_{10}$  limit was expressed for the majority of the zones (287 zones), while a demand for extending the period for attainment of the annual limit regarded 230 zones. Nine of the applicant countries (53% of the total) considered transboundary pollution as the main cause for non-compliance, while two countries (Denmark and Austria) attributed a significant number of exceedances to winter-salting and sanding. Objections were raised by the EC for 96% of the zones applying for postponement of attaining the annual limit and for 86% regarding the daily limit. It was interesting to note that objections raised for 53% of the applicant countries (either referring to the annual or to the daily limit) were attributed to inadequate or incomplete SA.

The analysis of the reports submitted by the countries complementary to the time extension applications suggested that receptor modelling was used to a smaller extent (29%) than dispersion models (41% Lagrangian, 59% Eulerian and 35% Gaussian) and trajectory models (41%) (Table 1).

**Table 1** Modelling tools used for SA by different EU countries for the purposes of preparing the time extension reports

<i>Model type</i>	<i>Number of countries</i>	<i>%*</i>
Lagrangian	7	41
Eulerian	10	59
Trajectory	7	41
Receptor	5	29
Gaussian	6	35
CFD	1	6
Combination of models	12	71

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

This can be explained on the basis of the transboundary contributions which represented in most countries a large percentage of natural contributions. The long-distance transport of particulate matter dictates the need to account for the physical and chemical processes governing pollutant transfer, which are addressed by dispersion model simulations, while back-trajectories constitute an ideal first screening approach for identifying the origin of transported polluted air masses. Local scale modelling (computational fluid dynamics – CFD software) was only applied by one country, as usually the impact of natural and transboundary sources in street canyons is less pronounced than the impact of local anthropogenic sources.

It was of great interest to note the high percentage of member countries (71%) that have applied a combination of modelling approaches for SA. In several cases, Eulerian dispersion models were complemented by Lagrangian trajectory models to account for transboundary contributions, such as for Cyprus, Portugal and Spain (natural transboundary contributions), and Belgium and Austria (anthropogenic transboundary contributions). In Greece and Italy, Eulerian dispersion models have been used to account for transboundary transport of polluted air masses in combination with statistical receptor models for source attribution of both local/national and long-distance sources. In Slovakia a Gaussian model was used for air quality assessment complemented by a Eulerian CTM to assess transboundary contribution. Slovakia and Poland were the only countries to account for resuspension using the environmental protection agency (EPA) emissions modelling approach, which requires input information on traffic characteristics, dust load on the road and street geometry.

Several countries have verified the model results against available measurements within the frame of the application, while the majority of the models used by the member countries for SA are extensively validated in the literature. In some cases, such as for the UK, France and Portugal, model validation was explicitly described. The report submitted by the UK explained the use of a volatile correction model to calibrate and validate the model results by applying appropriate scaling factors prior to the comparison with measurements. Portugal referred to the use of the ‘Standard Guide for Statistical Evaluation of Atmospheric Dispersion Model Performance’ (ASTM Standard D6589,

2005) that has been consulted to validate the prognostic meteorological and air pollution TAPM modelling system. Finally, France applied the Eulerian CTM modelling system PREV'AIR to estimate transboundary and natural contributions, including online verification procedures.

### **3 Review 2: the use of models for SA of atmospheric pollutants in Europe**

The results of the first review revealed the increased interest in using modelling tools for SA in member states, along with the problems associated with the lack of a uniform methodology in modelling and evaluation approaches. A follow-up study was thus considered necessary to build a more complete picture on the modelling methodologies applied by member states for SA of regulated pollutants and on the evaluation procedures used to assess the confidence of SA results.

#### *3.1 Methodology*

The second review was based on the compilation and analysis of the responses of member states to the questionnaires sent by the leading team of FAIRMODE SG2. The questionnaires were a 'Request for information concerning SA methodologies using models in Europe' (see Annex) and included questions on: the type of models used for SA, the pollutants for which SA was performed, the SA methodology used, whether any evaluation of the SA methodology was performed, any other issues/concerns related to SA using models, especially in regard to the AQD, and finally affiliation and contact/personal details. Some of the questions included a number of choices to tick, while the rest of the questions were open for the stakeholder to provide a more detailed reply. The questionnaires were distributed via e-mail to National Focal Points representatives of the European Environment Information and Observation Network (representing 40 European countries) and to 49 experts and regulators who have expressed their interest on SG2 activities through FAIRMODE registration (representing 17 countries). The questionnaires were addressed to all types of stakeholders, including universities and research institutions, regulatory bodies and environmental consulting companies. Unfortunately, a limited number of responses were obtained, consisting of 17 questionnaires which represented 11 EU countries, mostly from the Mediterranean and the Balkan regions. In particular the following countries provided feedback: Cyprus (one response), Finland (one response), Germany (one response), Greece (two responses), Italy (four responses), Lithuania (one response), The Netherlands (one response), Slovakia (one response), Slovenia (one response), Spain (three responses), UK (one response).

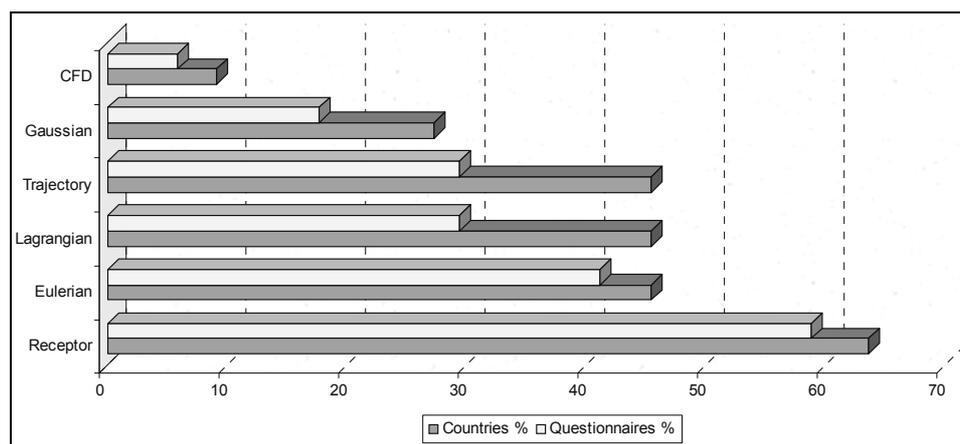
#### *3.2 Results*

The results from Review 2 are described in the following sections regarding the type of models used for SA by participants, the target metric examined and the SA evaluation methodologies applied in the studies.

### 3.3 Type of models used for SA

The information reported in the responses to the questionnaires revealed that dispersion models (59% of the reported studies) and receptor models (59% of the reported studies) are equally used by member states for SA (Figure 1).

**Figure 1** Percentage of model type used for SA by different EU countries



Trajectory models were less frequently used (29% of the returned questionnaires) and always complementary to receptor or dispersion models. As revealed from Review 1, dispersion models were the most commonly used tools for PM<sub>10</sub> SA rather than receptor models for the preparation of the reports submitted by EU countries in support of their application for postponement of attaining PM<sub>10</sub> limit values. In this extended Review 2, the need for applying dispersion models for SA can be explained on the basis of the transboundary contributions which represent a significant part of pollutant concentrations in most EU countries, particularly in the case of ozone and particles. Therefore, the use of dispersion models was highly associated in the questionnaire responses with source attribution of ozone concentrations. CFD modelling was applied for SA only by a research institute in Germany, in a study to estimate non-exhaust PM concentrations from highway traffic.

The suitability of receptor models for attribution of pollutants to their sources is recognised by experts from EU member states (Figure 1) and this was reflected in the high frequency of use reported in the returned questionnaires (59% of the questionnaires corresponding to 64% of the countries). These models are appreciated for they provide, with low computational intensity, source estimations at the urban and regional scales which are independent from emission inventories and meteorological data pre-processors (Karagulian and Belis, 2011). Regarding the frequency of use of different types of receptor models for SA, in contrast to a recent extensive literature review by Viana et al. (2008), where PCA was found to be the most frequently used receptor model, followed by the Lenschow approach, in Review 2 PMF was the preferred receptor model used for SA (45% of the countries). The second preferred receptor model was CMB (used in 36% of the countries), followed by PCA and UNMIX (both used only in 18% of the countries). Viana et al. (2008) reported an observed continued increase in the use of PMF

for SA in EU countries over the recent years, which was supported by the findings of Review 2.

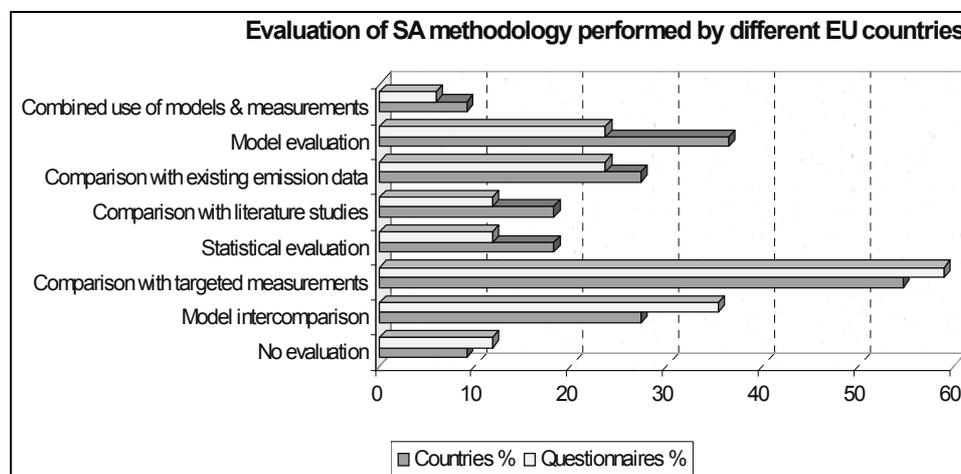
### 3.4 *Target metric*

PM was the pollutant targeted by all SA studies reported in the FAIRMODE SG2 questionnaires. In particular, PM<sub>10</sub> was the preferred target metric in 55% of the EU member states and 47% of the reported studies, while only 18% of the countries focused on fine fractions, such as PM<sub>2.5</sub>. This low number of European SA studies on fine PM fractions was not expected, considering the recent evidence on the adverse effects of fine particulates on health. Limited availability of necessary data for source attribution of fine particles, including the characterisation of specific tracers and chemical profiles for these smaller fractions, may also have contributed to the low number of SA studies targeted at this pollutant. A significant number of studies reported on the questionnaires (35% corresponding to 45% of the countries) have performed SA for NO<sub>x</sub> and NO<sub>2</sub>, while O<sub>3</sub>, SO<sub>2</sub>, CO were the target metrics in 27% of the countries. Source attribution of VOCs, metals and dioxins was examined in a very small percentage of the reported studies.

### 3.5 *SA evaluation methodologies*

Previous reviews have revealed that in most EU SA studies reported in the literature, evaluation of results is indirectly accounted for, and efforts to systematically evaluate the performance of alternative methodologies and estimate their intrinsic uncertainties have been scarce (Favez et al., 2010; Viana et al., 2008). In the extended Review 2, it was encouraging to note that a high percentage (88%) of reported SA studies have evaluated their results. The most frequently used SA evaluation method was by comparing model results to data obtained from dedicated measurement campaigns (59% of reported studies corresponding to 55% of EU countries). For specific pollutants, such as Polycyclic Aromatic Hydrocarbons, correlating calculated levels with other pollutants measured at the receptor site during sampling campaigns can be used for evaluating SA results. This method is only feasible if the ratio between the pollutant of interest and the measured pollutant is characteristic for a specific source (Larsen and Baker, 2003).

The next most common SA evaluation method used by EU member countries participating in Review 2 was model validation (36% of countries, 24% of questionnaires). This approach can involve the comparison and statistical evaluation of calculated pollutant concentrations against measured values, in order to test the performance of the dispersion model, as in the case of Finland, Slovakia, Lithuania and UK. Another SA evaluation method related to model validation is the sensitivity method, which is highly represented in the literature, particularly when dispersion models are applied for the identification and attribution of sources. SA modules incorporated into dispersion models can be evaluated by comparing SA results with results from model runs in which emissions from a particular source are greatly reduced (brute force method – BFM) or set to zero (zero-out method). In Review 2, information reported in a questionnaire returned by researchers from Spain, indicates the use of the zero-out sensitivity method to evaluate SA results for NO<sub>x</sub> and O<sub>3</sub>, based on dispersion model calculations. The use of the brute force sensitivity method has been reported by researchers in Italy for SA evaluation of NO<sub>x</sub>.

**Figure 2** Evaluation method of SA in the responses of SG2 Review 2

An added advantage of the sensitivity methods for SA evaluation, especially in terms of regulatory needs, is that the relative importance of each source category and the potential implications on source-oriented emission control strategies can be examined. Also, they can be applied with a limited computational cost as the runs for SA evaluation need only cover limited time periods, ideally for which measurement data are also available. However, the applicability of this method is pollutant-specific and depends on the linearity of the chemical reactions of the examined pollutant (Yarwood et al., 2005). For example, due to non-linearity of nitrate chemistry reactions, zero-out results have potential deficiencies as SAs for the case of SOA and NO<sub>x</sub>.

Model intercomparison as the preferred SA evaluation method was reported in a considerable number of responses (27% of countries). In some cases (Greece and Spain) different receptor models were applied for SA and their results were compared. The combined use of different types of receptor models could solve the limitations of the individual models (Viana et al., 2008) and is therefore a method used frequently for SA evaluation. In SA studies reported in responses by other countries (e.g., Spain) results from different dispersion model types were compared to evaluate NO<sub>2</sub> and O<sub>3</sub> SA results. In the questionnaires reported by researchers from Germany and the Netherlands, HYSPLIT back-trajectories were performed to evaluate receptor model SA results.

Existing emission data and emission inventories were used for SA evaluation in questionnaires returned by Italy, Finland and Spain, representing 24% of the questionnaires and 27% of the countries. Other SA evaluation methods that were less frequently reported by Review 2 participants include statistical evaluation (18% of the countries and 12% of the questionnaires), comparison with literature studies for the area of interest (18% of the countries and 12% of the questionnaires) and the combined use of model results and meteorological observations to verify the validity of the SA results (9% of the countries corresponding to 6% of the questionnaires). Meteorological variables such as wind direction and seasonal circulation phenomena can be correlated to pollutant transport, thus indicating possible pollutant sources (Chakraborty and Gupta, 2010).

Regarding the estimation of uncertainties, no particular approach for calculating uncertainties was reported in the questionnaires returned by the member states in

Review 2. Receptor modelling tools, such as CMB, provide uncertainty estimates corresponding to the calculated values for contributions from each source as their standard output. However, source profile species and receptor concentrations, each with uncertainty estimates, should be provided as input data to the CMB model in order to calculate uncertainties of SA results (Fujita et al., 2007). Routine PMF analysis provides output uncertainty estimations based on input data uncertainty and bootstrapping.

#### 4 Conclusions

Two targeted reviews have been performed by SG2 of FAIRMODE to gain an insight into the current use of models used for SA of regulated atmospheric pollutants by EU countries. The results confirm the simultaneous use of different modelling tools and methods for SA, as the appropriate methodology to combine the advantages and reduce the limitations of the individual model components. The majority of the reported studies have applied some SA evaluation methodology but no particular approach for calculating uncertainties was reported. Several researchers have commented on the need for a guidance including common criteria, indicators and performance measures to facilitate the SA evaluation procedure, which will form the basis for future FAIRMODE SG2 activities.

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## Annex

### *Request for information concerning SA methodologies using models in Europe: a task of SG2 (SA) WG2 FAIRMODE*

<i>Question 1 What type of models do you use for source apportionment (tick one or more)?</i>		
<i>Type of model</i>	<i>Tick</i>	<i>Model name/method</i>
Receptor model		
Eulerian model		
Lagrangian model		
Trajectory model		
Gaussian model		
CFD model		
Combination of models		
Other		

*Question 2* For which *pollutants* are you performing source apportionment with the use of models?

<i>Pollutant</i>	<i>Model name/method</i>
Particulate matter	
NO <sub>2</sub>	
O <sub>3</sub>	
Other (please specify)	

*Question 3* Are you using any method to *evaluate* your model results for source apportionment (e.g., intercomparison with other modelling results, comparison with measurements from sampling campaigns, other)? *If yes*, please provide a short description below.

*Question 4* Please provide a short (1 paragraph) description of your methodology for *source apportionment* (including references).

*Question 5* Please provide a short (1 paragraph) description of your methodology for *evaluating* the source apportionment results from the model, if any (including references).

*Question 6* What are your main *issues, concerns* related to source apportionment using models, especially in regard to the EU Air Quality Directive (2008/50/EC)?

*Question 7* Would you like to suggest *other colleagues or contacts* who you think would be interested and could be approached by FAIRMODE SG2?

<i>Name</i>	<i>Organisation</i>	<i>Contact details</i>

*Question 8* Any other comments

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*Personal details*

Name and Title

Affiliation

Address

Country

Telephone

E-mail

Function

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