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THE USE AND EVALUATION OF MULTI-POLLUTANT SOURCE APPORTIONMENT METHODOLOGIES BY EU AUTHORITIES AND RESEARCH GROUPS

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Abstract: Accurate source apportionment constitutes one of the key topics related to the use of air quality models for the purposes of the Air Quality Directive on ambient air (AQD). FAIRMODE (Forum for Air Quality Modelling in Europe) 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 the identification of source contributions that will address the requirements of the AQD. A first step in the context of SG2 activities has been a review of source apportionment methods used by different member states for the preparation of the extension reports regarding their compliance with PM_{10} limit values. The results revealed the lack of a uniform methodology and gaps in the validation of the results. In the present study, the former review is extended in an effort to identify the modelling source apportionment methodologies used by member states for various pollutants, with emphasis on evaluation and estimation of uncertainties relating to the methodologies applied. The study was performed by directly addressing a relevant questionnaire to the national representatives of 38 countries of the European region and to 50 national experts who have registered their interest for FAIRMODE SG2 activities. The responses reveal the widespread use of both receptor and dispersion models but also show that uncertainty assessment is rarely performed. The analysis of the replies is supported by a literature review of recent European source apportionment modelling applications.

***Key words:** FAIRMODE, Air Quality Directive, source apportionment validation methods, uncertainty assessment.*

INTRODUCTION

The present study was prepared within the frame of activities of FAIRMODE WG2-SG2, focusing on the *"Contribution of natural sources and source apportionment"*. This work represents an extension of a previous review on the use of models by EU member states for source apportionment (SA) and for assessing the contribution of natural sources to PM_{10} concentrations in response to the requirements of the Air Quality Directive. The results of this first review were presented in the 13th International Conference on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes, in the special FAIRMODE session. The increased interest in using modelling tools for SA in member states was revealed, 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.

METHODOLOGY

The present review was based on the compilation and analysis of meta-data (i.e. information based on previous studies and publications) received in the responses of member states to the questionnaires sent by the leading team of FAIRMODE SG2. The questionnaires were a "*Request for information concerning source apportionment methodologies using models in Europe"* and included questions on:

a) the type of models used for SA
b) the pollutants for which SA is performed by each member state
c) what SA methodology is used (short description and references)
d) if any evaluation of the SA methodology is performed (short description and references)
e) any issues, concerns related to SA using models, especially in regard to the EU Air Quality Directive (2008/50/EC)
f) 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 among **EIONET NFPs representatives** (representing **40** European countries) and **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 (1 response), Finland (1 response), Germany (1 response), Greece (2 responses), Italy (4 responses), Lithuania (1 response), The Netherlands (1 response), Slovakia (1 response), Slovenia (1 response), Spain (3 responses), United Kingdom (1 response).

RESULTS

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 (Table 1). Trajectory models are less frequently used (29% of the returned questionnaires) and always complementary to receptor or dispersion models. Dispersion models were the most commonly used tools for PM_{10} 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_{10} limit values (Fragkou et al., 2010). In the present review, 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 emissions in most EU countries. The long-distance transport of ozone and PM dictates the need to account for the physical and chemical processes governing pollutant transfer, simulated by dispersion models. Therefore, the use of dispersion models is highly associated in the questionnaire responses with source attribution of ozone emissions. Computational Fluid Dynamics (CFD) modelling was applied for SA only by a research institute in Germany, in a study to estimate non-exhaust PM emission from highway traffic.

Table 1. Modelling tools used by different EU member states for SA

Country	Modelling Methods
Cyprus	Receptor model (PCA), Dispersion (Eulerian), Trajectory
Finland	Dispersion (Eulerian-Lagrangian)
Germany	Receptor model (Lenschow, PMF), Dispersion (Lagrangian), Trajectory, CFD
Greece	Receptor model (PMF, PCA), Disperion (Eulerian), Trajectory
Greece	Receptor model (PMF, PCA), Dispersion (Eulerian)
Italy	Dispersion (Eulerian)
Italy	Receptor model (CMB)
Italy	Receptor model (PMF)
Italy	Dispersion (Eulerian)
Lithuania	Dispersion (Gaussian)
The Netherlands	Receptor model (PMF, UNMIX), Trajectory
Slovakia	Dispersion (Lagrangian)
Slovenia	Receptor model (PCA)
Spain	Dispersion (Eulerian, Lagrangian, Gaussian)
Spain	Receptor model (PCA, PMF, ME, CMB), Trajectory
Spain	Receptor model (PCA-APCS, PMF, UNMIX)
UK	Dispersion (Gaussian-Lagrangian)

The suitability of receptor models for attribution of pollutants to their sources is recognised by experts from EU member states (Figure 1) and this is 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 preprocessors (Belis, C. and F. Karagulian, 2011).

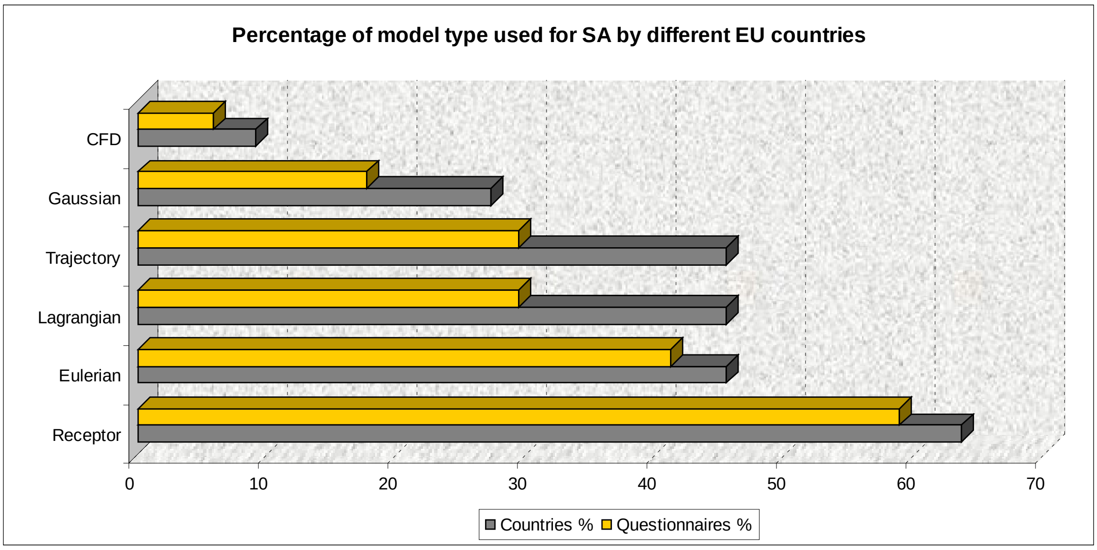

Figure 1. Percentage of model type used for SA by different EU countries

It is also interesting to note the frequency of use of different types of receptor models for source identification and attribution in the studies reported by member states in the FAIRMODE SG2 questionnaire. 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 the current study PMF is the preferred receptor model used for SA (45% of the countries corresponding to 41% of the questionnaires). The second preferred receptor model is CMB (used in 36% of the countries and 35% of the returned responses), followed by PCA and UNMIX (both used only in 18% of the countries and 12% of the questionnaires). Viana et al. (2008) reported an observed continued increase in the use of PMF for SA in EU countries over the recent years, which seems to be supported by the findings of the present study. The obvious advantage of the PMF and PCA models over CMB relates to the fact that related software is widely available and detailed information on the sources and source profiles is not required. On the other hand, CMB application requires extensive data on pollution sources prior to source apportionment and is considered as the ideal receptor model in the case of minimal changes of the source profiles between the emitter and the receptor.

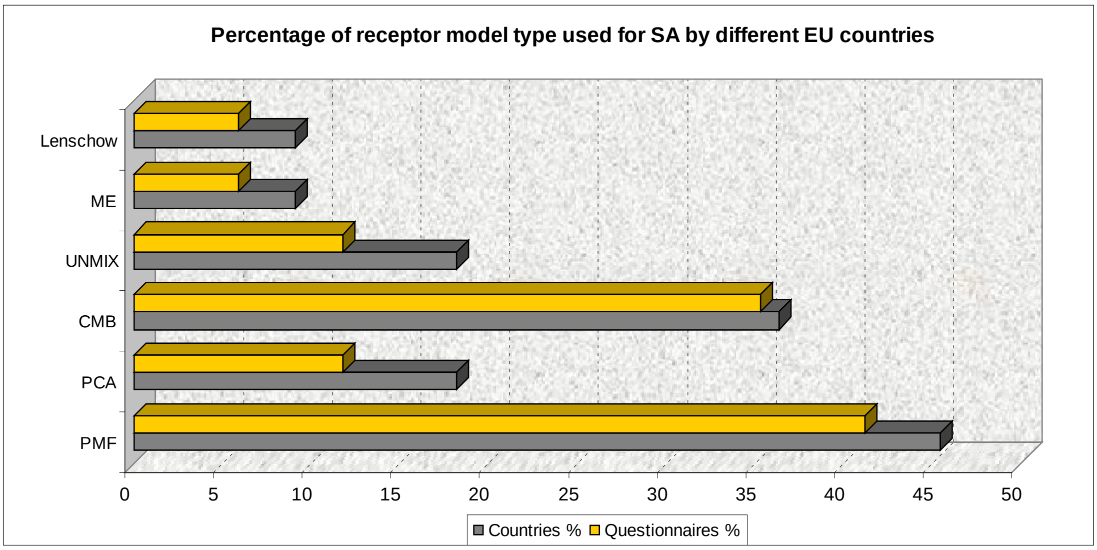

Figure 2. Percentage of model type used for SA by different EU countries

*ME: Multilinear Engine, CMB: Chemical Mass Balance, PCA: Principal Component Analysis, PMF: Positive Matrix Factorisation

Target metric

As shown in Figure 3, PM was the pollutant targeted by all SA studies reported in the FAIRMODE SG2 questionnaires. In particular, PM_{10} was the preferred target metric in 55% of the EU member states and 47% of the reported studies, while only 18% of the countries (represented by 12% of the questionnaires) focused on fine fractions, such as $PM_{2.5}$. This low number of European SA studies on fine PM fractions is 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 contribute 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_x and NO_2, while O_3, SO_2, 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.

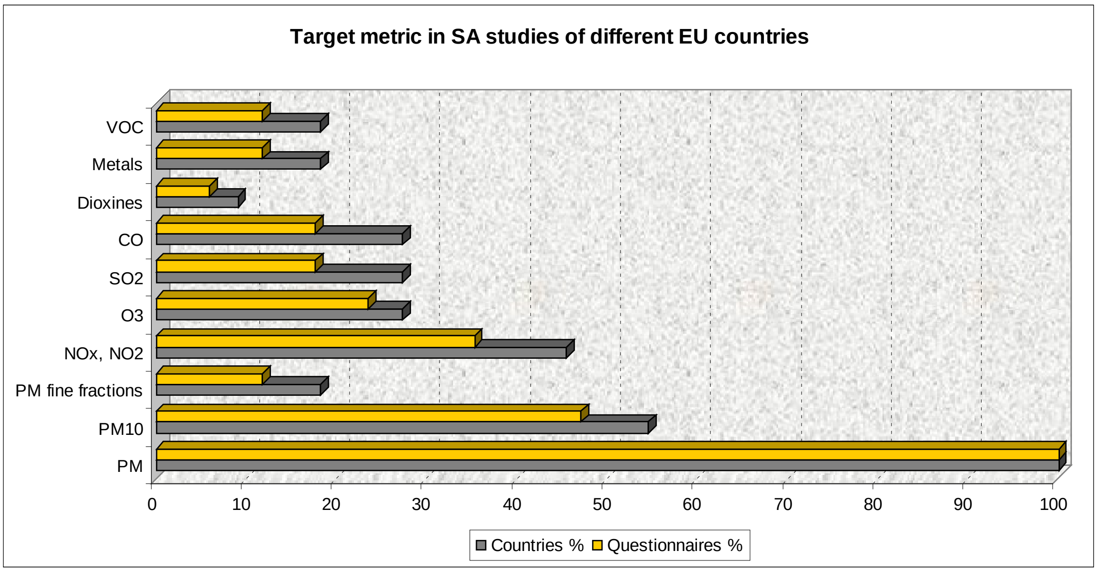

Figure 3. Target metric of SA studies reported in the FAIRMODE WG2 SG2 questionnaires

SA evaluation methodologies

Previous reviews have revealed that in most EU SA studies reported in the literature, evaluation of results is indirectly accounted for, usually by validation of the dispersion model used against measured concentrations (Fragkou et al., 2010). In the present review it was encouraging to note that a high percentage (88%) of reported SA studies by EU member states 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, R. K. III and J. E. Baker, 2003).

Model intercomparison as the preferred SA evaluation method was reported in a considerable number of responses (27% of countries, 35% of questionnaires) received within the frame of the current review. In some cases, such as in questionnaires returned by 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 questionnaire responses by other research institutes, such as from Spain, results from different dispersion model types were compared to evaluate NO_2 and O_3 SA results. In the questionnaires reported by researchers from Germany and the Netherlands, HYSPLIT back-trajectories were performed to evaluate receptor model SA results.

The next most common SA evaluation method used by EU member countries participating in the present review 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 that have recently emerged and 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 the present review, 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_x and O_3, 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_x.

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. The sensitivity method is commonly used for SA evaluation of dispersion models. In a relevant study by Koo et al. (2009), the Particulate Source Apportionment Technology (PSAT), implemented in the Comprehensive Air-quality Model with extensions (CAMx), was tested against the Decoupled Direct Method (DDM) and the BFM. SO_2 was the target metric for SA, and it was concluded that good agreement between sensitivity methods and direct SA can be expected only in the case of substantial emission reduction (almost by 100%), while noticeable difference between PSAT and BMF results was observed for smaller (by 20%) reduction in point source SO_2 emissions. Sensitivity methods are useful to ascertain the attribution of sources suggested by dispersion models and to provide a quantification of the contributions. 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 (Moussiopoulos et al., 2010). 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, the sum of nitrate contributions from zeroing-out all NO sources may differ substantially from the total nitrate concentration. In this case, the use of sensitivity methods for evaluating SA may not provide scientifically acceptable results. For the same reason, namely due to the non-linear relationship between VOC precursor emissions and SOA formation, zero-out results have potential deficiencies as source apportionments for the case of SOA.

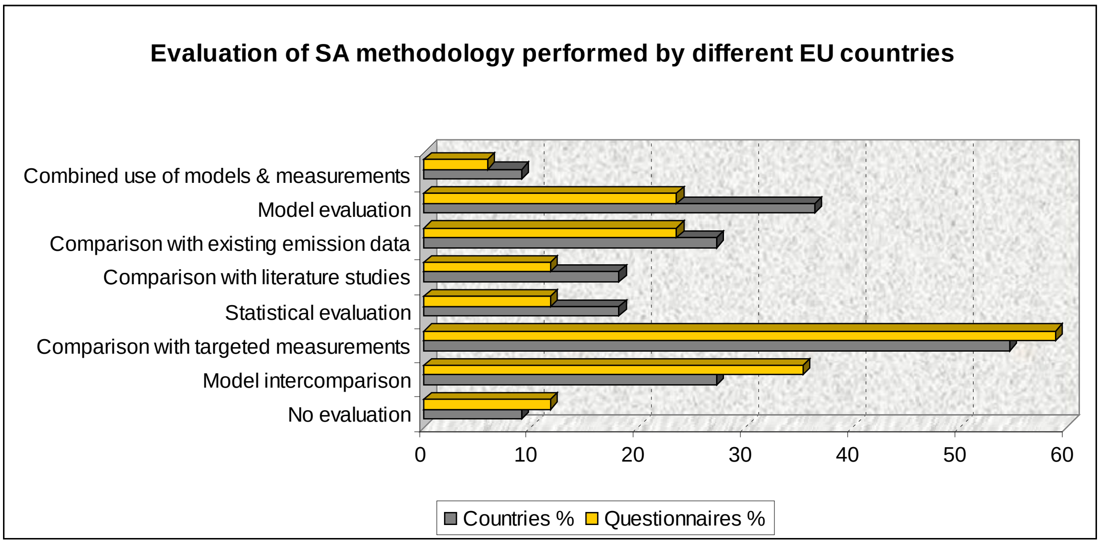

Figure 4. Evaluation methodology of SA performed in studies reported in the FAIRMODE WG2 SG2 questionnaires

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. In a relevant study by Manoli et al. (2004), SA of PAHs for northern Greece was performed with PCA receptor modelling and the results were evaluated against known emission profiles of certain sources. Other SA evaluation methods that were less frequently reported by EU member states participating in the present review 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). It should be mentioned at this point that, in regard to the use of meteorological data to complement source attribution performed by modelling tools, the simplicity of this method does not reduce its usefulness and efficiency. Meteorological variables such as wind direction and seasonal circulation phenomena can be correlated to pollutant transport, thus indicating possible pollutant sources (Chakraborty, A and T. Gupta, 2010). Statistical evaluation of SA models can be applied using the same performance measures for model validation, in particular percent mass, degrees of freedom, r-square, chi-square, fit measure and difference between calculated and measured concentrations (Watson et al., 2001). These are the preferred performance measures for the least squares calculation methodology of SA.

Regarding the estimation of uncertainties, no particular approach for calculating uncertainties was reported in the questionnaires returned by the member states in the current review. At this point it should be mentioned that 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.

CONCLUSIONS

The present review on modelling tools used for SA by EU member states and the evaluation methods applied to validate the SA results was performed within the framework of scheduled activities of the WG2-WG2 of FAIRMODE. The findings of the review were based on a less-than-satisfactory number of returned questionnaires; however some general conclusions can be drawn to guide further study. The results confirm the simultaneous use of different modelling tools and methods for identification and attribution of sources at examined receptor sites, 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. However, limited information was contained in the returned questionnaires on the estimation of uncertainty and several researchers have commented on the need for a guidance including common criteria, indicators and performance measures to facilitate the SA evaluation procedure. Uncertainty is in a large degree linked to the uncertainty related to input data used for the SA calculations. Some issues raised that need to be considered in the preparation of a guidance document for harmonisation in model use for SA are:

1. The rough/incomplete uncertainty information related to the lack of uncertainty data in the input data as well as in the final SA results
2. The absence of appropriate tracer species and the lack of harmonisation between studies regarding the marker elements used for specific sources
3. The small size of examined data sets or the inappropriate time coverage of measurements from sampling campaigns, particularly for determining particle size distributions and chemical characterisation
4. The difficulty in source classification of specific pollutants such as mineral dust, due to the overlap between the elemental composition of natural and anthropogenic crustal dust
5. The need for a multi-scale approach in the use of models for SA, particularly for secondary pollutants such as secondary aerosols

The above points, along with general issues related to harmonisation, including treatment of the heterogeneity in source categories and source classifications reported by different researchers and best complementary model methods for specific pollutants, should be further examined in order to provide a useful guidance document for SA and SA evaluation.

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