European Guide on Air Pollution Source Apportionment (SA) for estimating Particulate Matter (PM) source contributions with Source oriented Models (SMs) and combined use of SMs and Receptor Models (RMs)

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History of guide

- idea to make such an SA guide was launched by Claudio Belis at FAIRMODE Technical Meeting 27-29 June 2016, Zagreb, Croatia

- first outline of the SA guide presented at FAIRMODE Plenary meeting 14-15 February 2017, Utrecht, The Netherlands

-first request of contributions launched in the FAIRMODE community on 7 April 2017

-a survey about available studies on estimation of particulate matter source contributions with source oriented models (SM) and/or receptor models (RM) was administrated on 19 May 2017

-presentation of survey results at FAIRMODE Technical Meeting 19-22 June 2017, Athens, Greece ,and a new request of contributions before 15t September 2017

-distribution of an updated outline of SA guide and new request for contributions via email before 1 November 2017

-guide presentation at FAIRMODE Plenary Meeting: Baveno, 26-27 February 2018

-guide presentation and discussions at FAIRMODE Technical Meeting: Tallinn, 26-28 June 2018

-guide presentation at FAIRMODE Plenary Meeting: Warsaw, 12-14 February 2019

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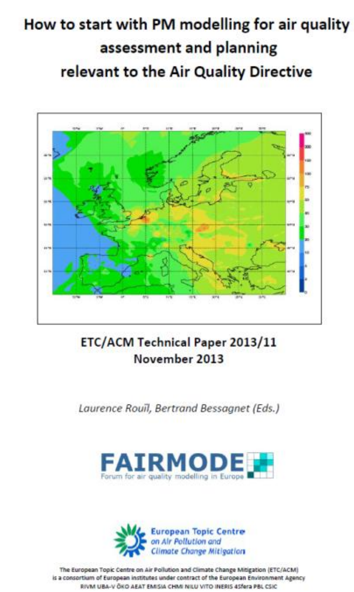
Introduction: 1.1 Scope and aims

…the document focuses on the 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…

…it also shows the potential of RM and SM techniques and critically discusses the needs and the limitations when applying simultaneously these techniques in source apportionment studies…



Introduction: 1.2 Target audience

…provide the basis to the organizations, companies, institutions which carry out source apportionment studies in support to authorities responsible for air quality management according to the AQD including the development, implementation and evaluation of official air quality plans...

….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 the users in the interpretation of the 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…

Introduction: 1.3 Why using receptor models (RMs) and source oriented models (SMs) for SA

…The quantitative impact of the sources that affect the ambient air pollution cannot be measured directly…

RMs:

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 are negligible
-the uncertainty of the output is estimated.

Disadvantages:

-provide information for specific sites and time windows(long time series for epidemiological studies not available)
-source identification is 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.

Introduction 1.3 Why using receptor models (RMs) and source oriented models (SMs) for SA

SMs:

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;

-allow to 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 could be detailed in terms of activity sectors and subsectors;

- estimate the contribution of different emission sectors to secondary pollutants through the air concentrations at boundaries;
- estimate the contribution of transported pollutants;
- the individual contribution 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;

Disadvantages:

-the results 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.

Introduction: 1.4 Techniques for SA using RMs

Incremental approach or Lenschow approach

Chemical Mass Balance

Positive Matrix Factorization

Introduction: 1.5 Source oriented air quality models

The source oriented models:

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

Model application

- 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 is well documented and validated for the specific application?

Introduction: 1.6 SM approaches for SA

Sensitivity analysis

…. can be employed to quantitatively estimate the uncertainties in the predicted concentrations when combined with estimates of emission uncertainties …, to compute incremental reactivity factors, i.e. the sensitivity of ozone concentrations to emissions of specific volatile organic compounds …, or to develop a simplified representation of a full model response over a specific range of emission levels…

Methods

…The most straightforward sensitivity method used for SA applications is brute force method (BFM) (other names: zero out method or emission reduction potential (ERP))

…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 …- requires the addition of specialised code to the host CTM

Tagged species

…this methodology labels each precursor in every time step according to its activity source and/or the geographical origin…- requires the addition of specialised code to the host CTM

-A key aspect of tagging methods is that the sum of the concentrations xi corresponding to each source i is always equal to the total concentration X due to all sources…- this is a common feature with RMs that led to quantifying the mass transferred from the source to the receptor.

Introduction

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

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 complexity of the terrain.		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

2 Estimation of source contributions with SM approaches

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

- Domain, time period and spatial resolution
- Boundary conditions: linking model outputs over different domains
- Meteorological data
- Emissions: anthropogenic and natural
- Model evaluation/validation-MQO

2.2 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 emission input….
…The difference in concentration between the base case and a perturbed simulation represents the sensitivity of the modelling system to the considered emission variations...

2.3 Tagged species methods

…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 … in the modelled area and time period under investigation…
…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 some assumptions made in source apportionment for secondary PM species.

3 Combined use of SMs and RMs

3.1 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...

…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…

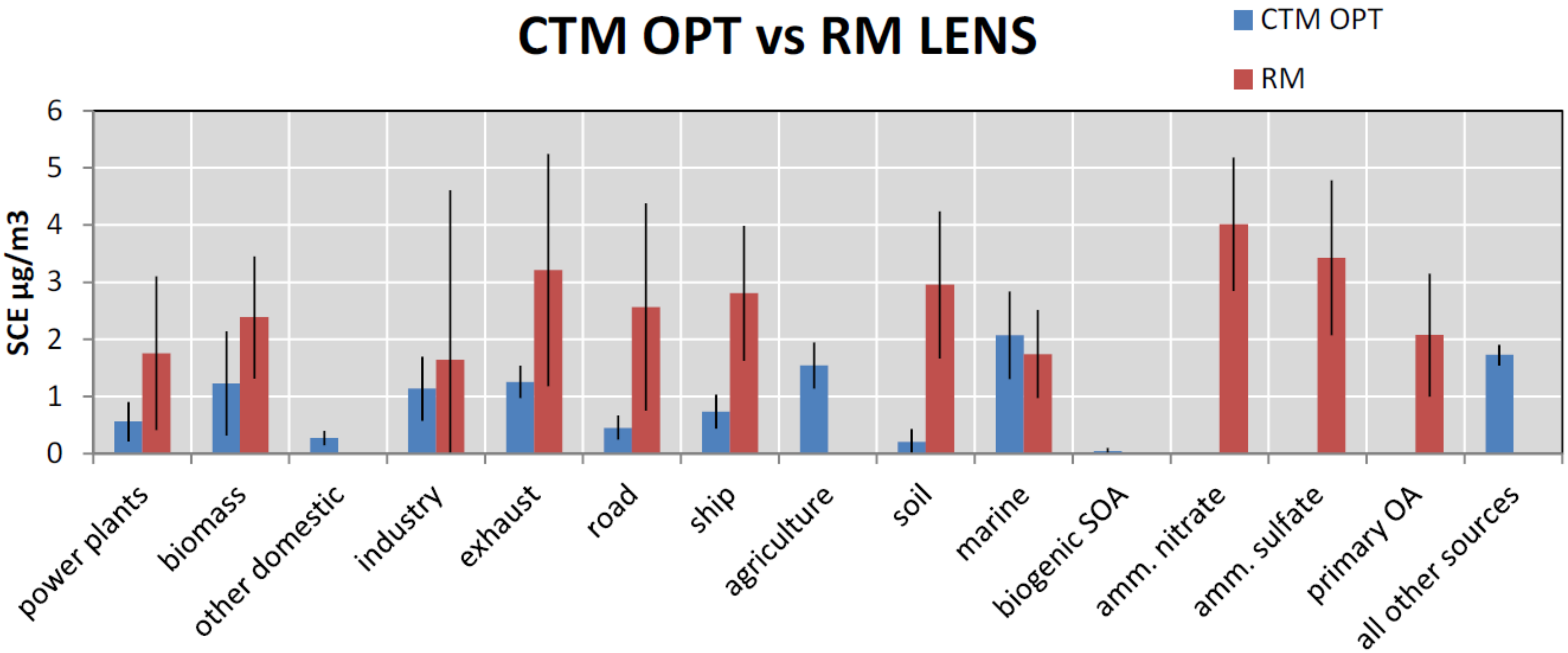
3.2 Comparison of source contributions from SMs and RMs

Bove et al., (2014): …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 PM2.5, respectively…

Pirovano et al., (2015):…CMB showed a better reconstruction of the PM2.5 mass closure; conversely, CAMx systematically underestimated PM concentrations especially during the severe episodes in the cold season, due to difficulties in reproducing very stable meteorological conditions and inaccuracies in emission inventories. Nevertheless, both models provided the same ranking for source contribution estimates (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….

4 Intercomparison between SMs and between SMs and RMs

...the most comprehensive attempt to compare SM and RM... FAIRMODE intercomparison exercise (IE, Belis et al., 2019b). The exercise involved more than 40 modelling teams, mostly applying RMs...



...SCEs deriving from RMs are always higher than the corresponding value reported with CTMs... The most important primary sources in RMs are: exhaust, soil, ship, road dust and biomass burning and in CTM mandatory set are :traffic, agriculture, industry and biomass burning...
... verify whether SM underestimation can be ascribed to factors not depending from the sources (e.g. meteorology and long range transport) or can be related to deficiencies in emission inventory instead a poor representation of some emission processes.


The End