FAIRMODE – WG2

SG2 - *Contribution of natural sources and source apportionment*

The Use Of Models For Source Apportionment And For Assessing The Contribution Of Natural Sources In Response To The Air Quality Directive

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Source apportionment in the AQD

- Source apportionment studies include assessing the contribution from **local sources** as well as from **natural sources**, **neighbouring countries** and the contribution from **resuspended road sand** and **salt**.
- **AQD**: possibility to discount natural sources and long-range transport of pollution and resuspension attributable to winter sanding-salting of roads when assessing compliance against limit values.
- Although not explicitly mentioned in the AQD, modelling is necessary for this purpose as monitoring of these contributions everywhere in a zone or agglomeration would be unrealistic.

SG2 of FAIRMODE

- The working sub-group (SG) on the "Contribution of natural sources and source apportionment" has been formed within the frame of the Forum for Air Quality Modelling in Europe (FAIRMODE).
- SG2 focuses on source apportionment and the contribution of natural sources on pollutant concentrations and aims to:
  - provide useful guidance and suggest best modelling practices and quality assurance procedures for member countries.
  - promote harmonised model use for source apportionment in the EU
- **Phase 1:** Review of the current status of modelling practices used for source attribution and quantification of contributions by member states to identify gaps and problems.

SG2 Participants

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Sources used in this review:

1. Database compiled within the frame of the COST Action 633
2. Workshop on the “Quantification of the contribution of natural sources to the ambient PM concentrations” (Ispra, JRC, October 2006)
3. Notifications submitted by member countries in support of their applications for postponement to comply with PM_{10} limit values
4. Indicative recent publications from member countries

Monitoring for source apportionment

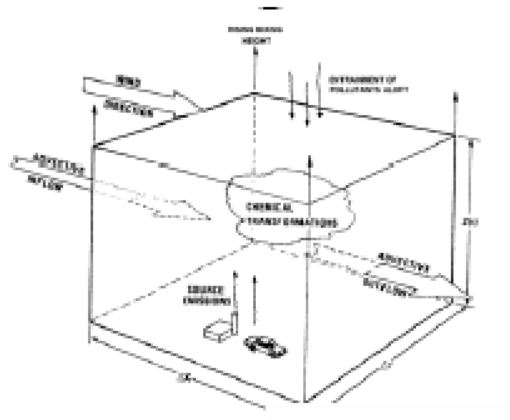
- Suggested methodologies involve:
  - Observation and analysis of monitoring data, correlation with relevant meteorological parameters.
  - Subtracting regional background levels from the urban background and hot-spot concentrations to determine the importance of local sources.
- Similar methodology used to quantify natural contributions: PM regional background levels are subtracted from those measured at the urban and traffic stations of interest for a specific period.
- The occurrence of concentration peaks of measurements simultaneously at different stations can indicate an episode due to transboundary pollutant transport or due to an accidental release.
- Limitations of monitoring (issues of spatial and temporal representativity compromised by the increased costs associated with adequate coverage and reliability).

Air Quality Modelling Techniques: Contribution & Control Assessments

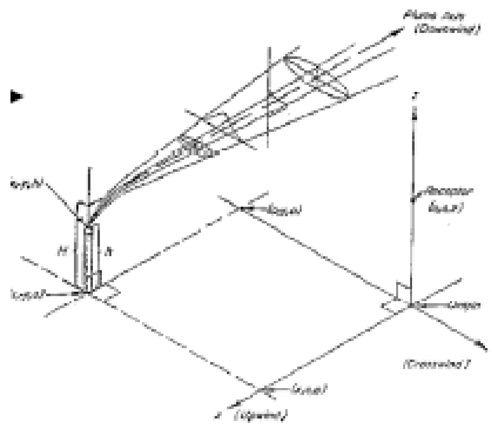
- Address source/pollutant "contribution"
  - Sector Zero-Out Modelling
    - Model simulation with "zero-out" of single or all pollutants from sector/sources of interest
  - Modelling Source Apportionment
    - Allows estimation of contributions from different source areas / categories within single runs
- Address relative efficacy of source/pollutant emissions reductions
  - Response Surface Modelling (among others)
    - A statistical "reduced-form" model of a complex air quality model

Source Models often used for regulatory purposes

- **Photochemical models**: chemical and physical atmospheric processes are described for predicting pollutant concentrations.
  - Can be applied at multiple spatial scales (local, regional/national, and global)
  - CMAQ, CAMx, MARS etc.



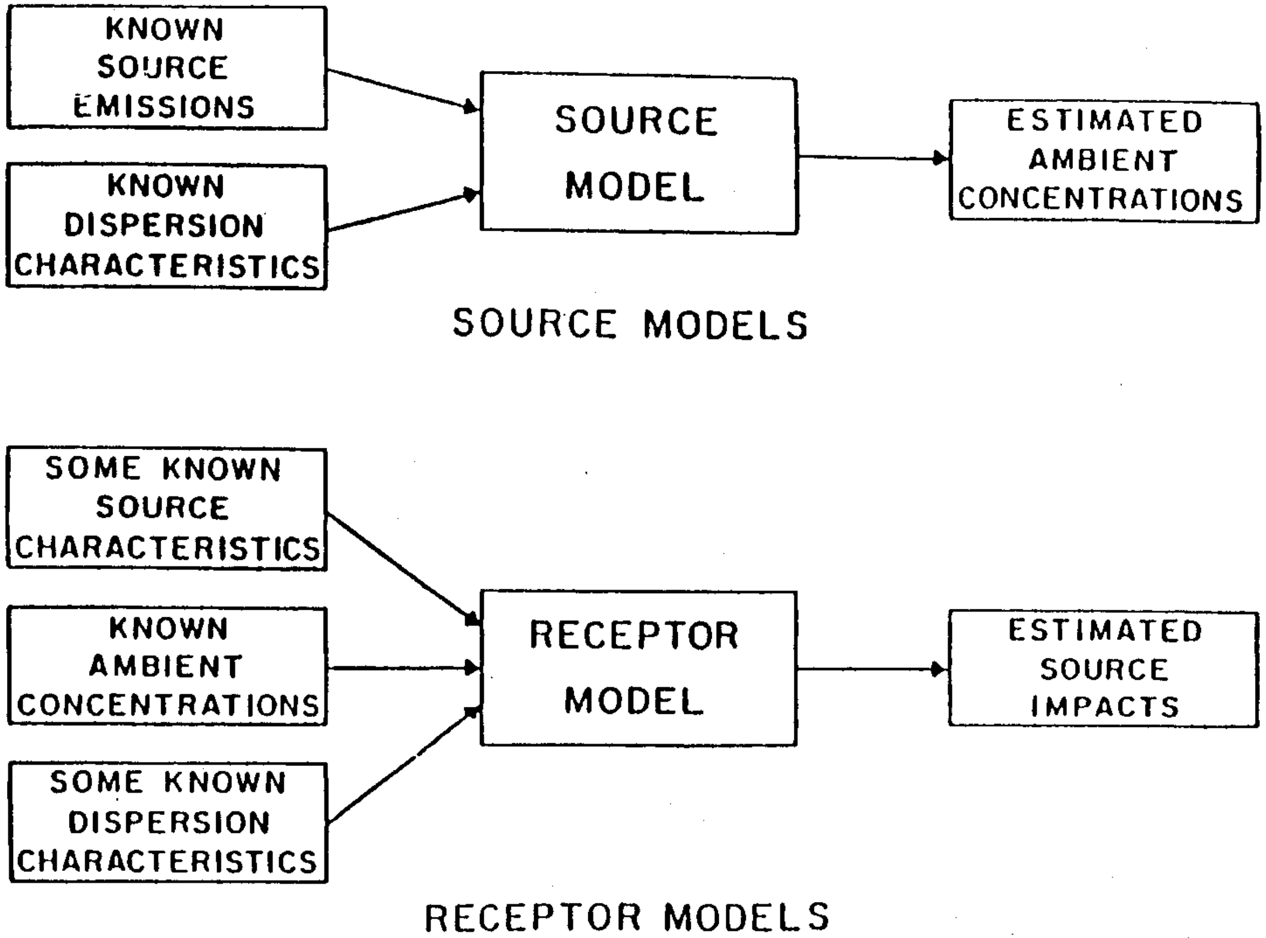
- **Dispersion models**: source-oriented models that characterise atmospheric processes by dispersing a directly emitted pollutant to predict concentrations at selected downwind receptor locations.
  - Typical of permit applications for new sources but can be run for multiple sources at once
  - AERMOD, ISC, ASPEN etc.



Receptor models are commonly used for source apportionment

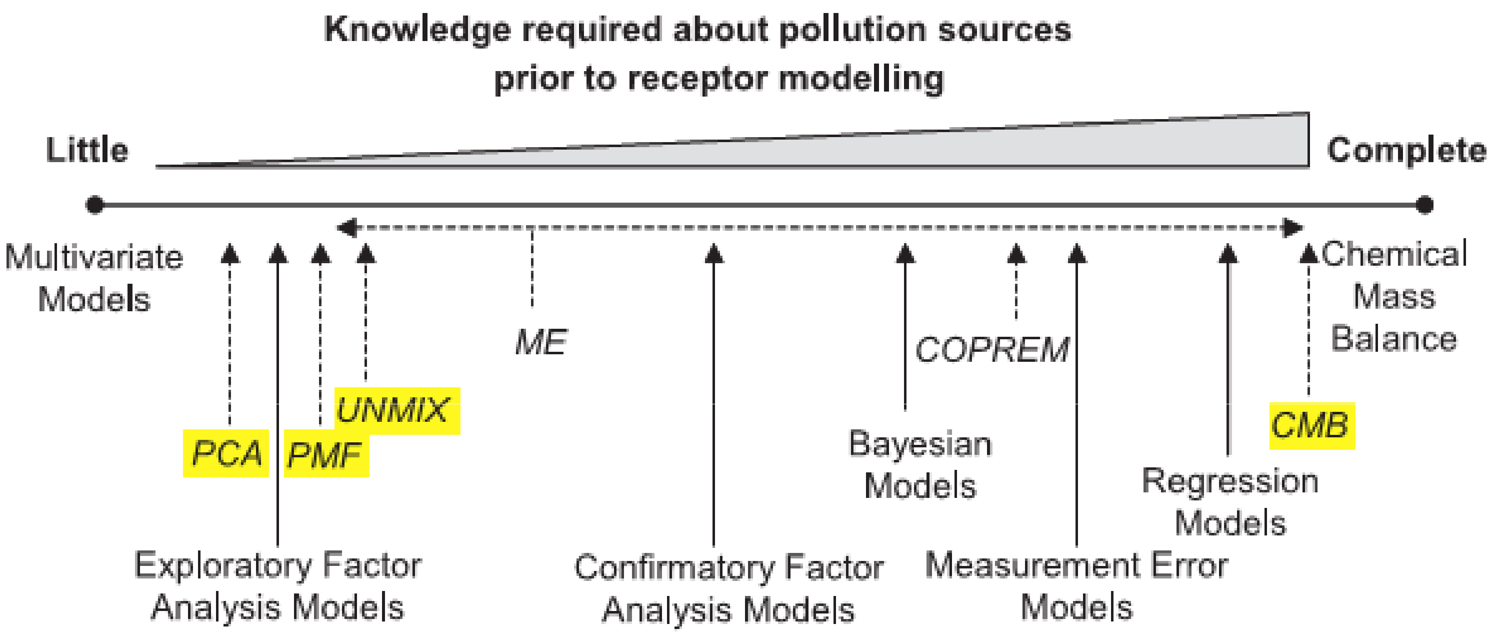
- **Receptor models** complement source models by independently identifying sources and quantifying their contributions using ambient measurements of different observables at different times and locations. Source apportionment is accomplished by solution of the **mass balance equations** that express concentrations at several measured pollutants as a linear sum of products of pollutant abundances in source emissions and source contributions. These equations can be solved by several mathematical methods.
- However, *the solution does not guarantee physical reality*, so internal and external validation measures must be evaluated. Receptor models are best used in conjunction with source models to create a “weight of evidence” for justifying emission reduction measures on different source types (Watson and Chow, 2005).

Source and Receptor Models



(From Watson, 1979.)

Receptor modelling methods



(From Viana et al., 2008)

Most commonly used methods:

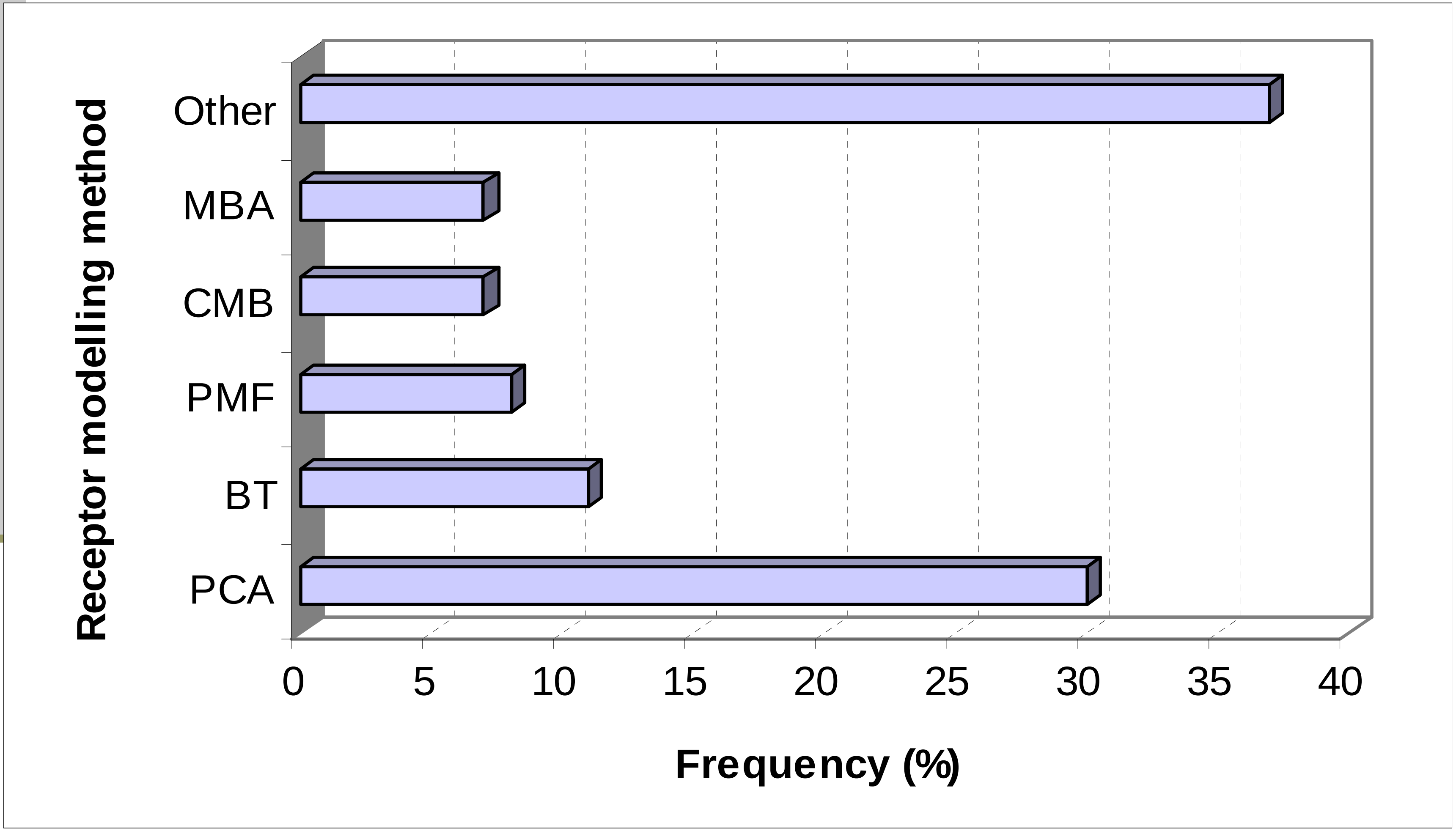
- Principal Component Analysis (**PCA**)
- Positive Matrix Factorization (**PMF**)
- Chemical Mass Balance (**CMB**)

From COST 633 Questionnaire, 2005

Cluster Analysis	CA	Multivariate statistical classification of a dataset which maximises the similarity between cases of the same cluster and minimises the similarity between clusters	Statistical packages	Worldwide	Avila & Alarcón, 1999 Rodriguez et al., 2003
Factor Analysis	FA	Multivariate technique for reducing matrices of data to their lowest dimensionality by the use of orthogonal factor space and transformations that yield predictions and/or recognisable factors	Statistical packages	Worldwide	
Principal Component Analysis	PCA	Most common form of FA. Results calculated using an eigenvector analysis of a correlation matrix	Statistical packages	Europe-USA	Thurston & Spengler, 1985 Rodriguez et al., 2003 Salvador et al., 2003
Multiple Linear Regression	MLR	Relates the aerosol mass to the composition of certain tracer elements from the sources contributing in the aerosol samples	Statistical packages	Worldwide	Groblicki et al., 1981 Chan et al., 1999a
Target Transformation Factor Analysis	TTFA	Relates the value of factor loadings derived from FA to the value of mass fraction in physical source emissions	FANTASIA (Factor Analysis To Apportion Sources In Aerosols)	USA	Hopke, 1989 Chan et al., 1999b

Name	Abbreviation	Definition	Software	Use	Examples
Lenschow approach		Based on the assumption that the levels and chemical composition of particulate matter at a traffic site result from the addition of the local influence of traffic on the adjacent street, the sources of the agglomeration (city background) and global sources with little contribution from the agglomeration (regional background).	Mathematical packages	Europe	Lenschow et al., 2001 John et al., 2004 Querol et al., 2004
Chemical Mass Balance	CMB	Model for assessing contributions of primary particles on the basis of known source compositions	CMB7 CMB8 Mathematical packages	Worldwide	Hopke & Song, 1997 US-EPA, 1990 Abu-Allaban et al., 2002 Sun et al., 2004
Source Apportionment by Factors with Explicit Restrictions	SAFER	Multivariate receptor model based on PCA and the self-modelling curve resolution technique (SMCR). Implements explicit physical constraints in estimating source compositions		USA	Kim, 1989 Henry and Kim, 1990 Kim and Henry, 1999
Positive Matrix Factorisation	PMF	Weighted least-squares fit with the known error estimates of the elements of the data matrix used to derive the weights. Non-negativity constraint	PMF2	Europe-USA	Paatero & Tapper, 1994 Paatero, 1997 Chueinta et al., 1999 Kuhlbusch et al., 2004 Ito et al., 2004
Multilinear Engine	ME	Technique for fitting multilinear and quasi-multilinear mathematical expressions or models to two-, three-, and many-dimensional data arrays	ME2	Europe	Paatero, 1999 Rae et al, 2002 Yli-Tuomi et al., 2003

Frequency of use of different receptor models in member states (COST 633)



EEA/ETC Questionnaire

Receptor modelling: 70%, combination of receptor and source modelling: 20%, source modelling: 10 %

Country	Modelling Methods
Austria	Source modelling
Finland	Receptor modelling (PCA, MLR, MLF, SEM)
Germany	Source and Receptor modelling (PCA, MLR, PMF)
Greece	Receptor modelling (MR/APCS, CMB)
Italy	Source and Receptor modelling (PCA, PMF)
Netherlands	Receptor modelling (PCA, MLR)
Portugal	Receptor modelling (MLRA, PCA, MBA)
Spain	Receptor modelling (MLRA, PCA)
Sweden	Receptor modelling (PMF)
United Kingdom	Receptor modelling (PCA)

Workshop on the "Quantification of the contribution of natural sources to the ambient PM concentrations"

- 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 then limited, but gradually growing ever since.
- **50%** of the countries have used source models (mainly Eulerian Chemical Transport Models).
- **40%** of the countries reported the application of receptor models for source apportionment.
- In order to enhance the reliability of the methodology, a **30%** of the countries have applied back-trajectory analysis in combination with other modelling methods.

Publications

Publication	Area of application	Model type
Adamczyk, L. et al. (2007)	European cities (Prague, Riga, Vilnius, Tallinn)	Hybrid Swedish AIRVIRO Dispersion model
Adamczyk, L. et al. (2007)	Cracow, Poland	Gaussian, ADMS-urban model
Astitha, M. et al. (2005)	Urban Mediterranean	Eulerian, SKIRON/ETA
Favez, O. et al. (2010)	Grenoble, France	Receptor, CMB and PMF
Kallos, G. et al. (2006)	Urban Mediterranean	Eulerian, SKIRON/ETA
Pio, C.A. et al.(1996)	Western Portoguese coast	Receptor, PCA
Rodríguez, S. et al. (2001)	Southern Spain	Eulerian SKIRON combined with back-trajectory analysis
Simpson, D and K.E. Yttri (2009)	Switzerland, Sweden and Norway	Eulerian, EMEP SOA
Viana, M. et al. (2008)	Spain	Receptor, PCA, PMF and CMB

Notifications of time extensions (1)

In order to be eligible for the 3-year postponement of attaining PM_{10} limit values the applicant EU countries have to apply a methodology to:

(a) confirm that a significant number of exceedances or high annual mean concentrations was due to natural sources

(b) quantify the proportion of these exceedances

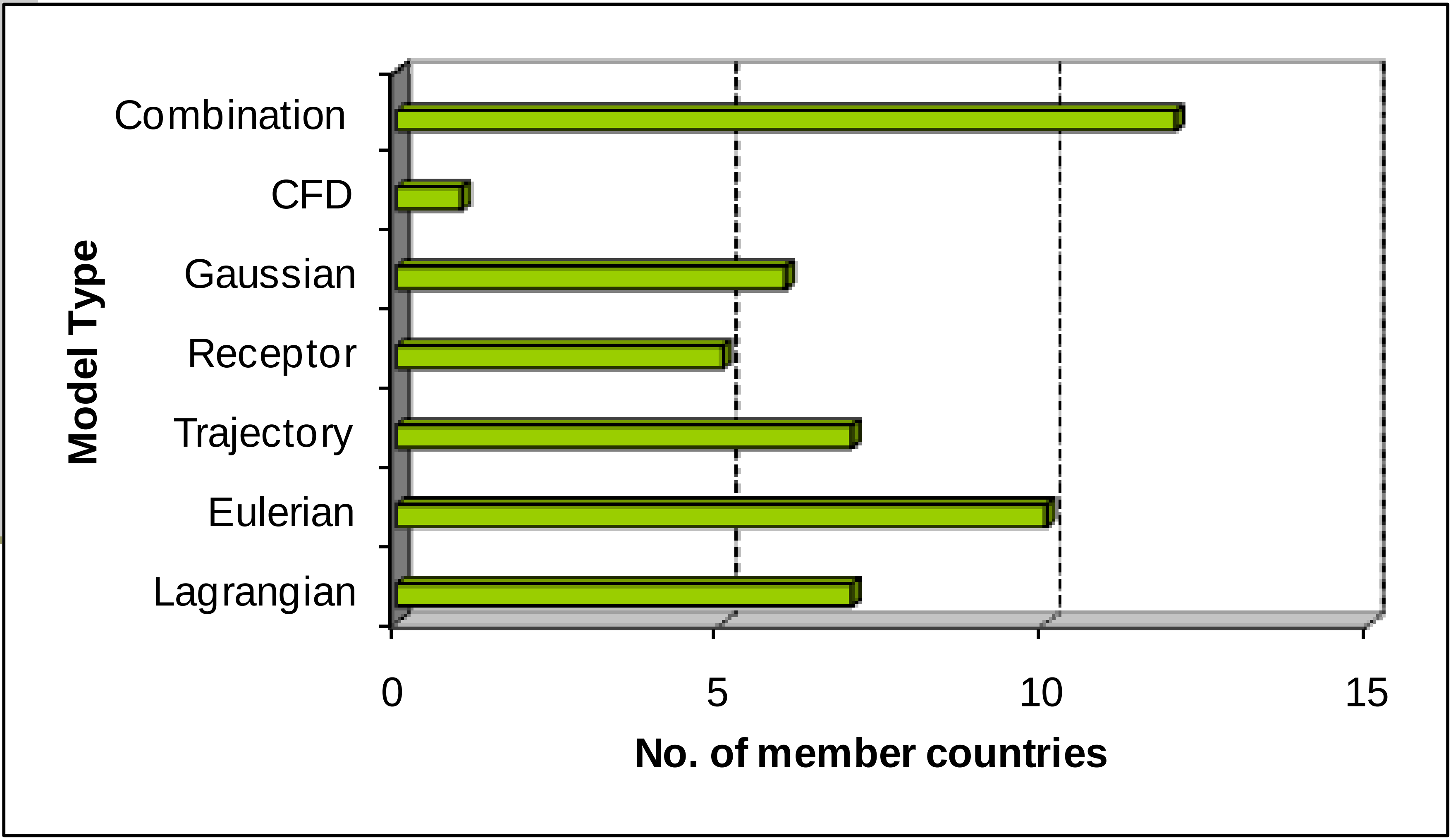
(c) determine the extent to which the different natural sources were responsible by estimating the PM_{10} concentrations resulting from their relevant emissions

Notifications of time extensions (2)

Total countries	**17**
Total zones	**289**
Zones in demand for annual limit extension	**230**
Zones in demand for daily limit extension	**287**
Zones with objections for annual limit	**221 (96%)**
Zones with objections for daily limit	**248 (86%)**

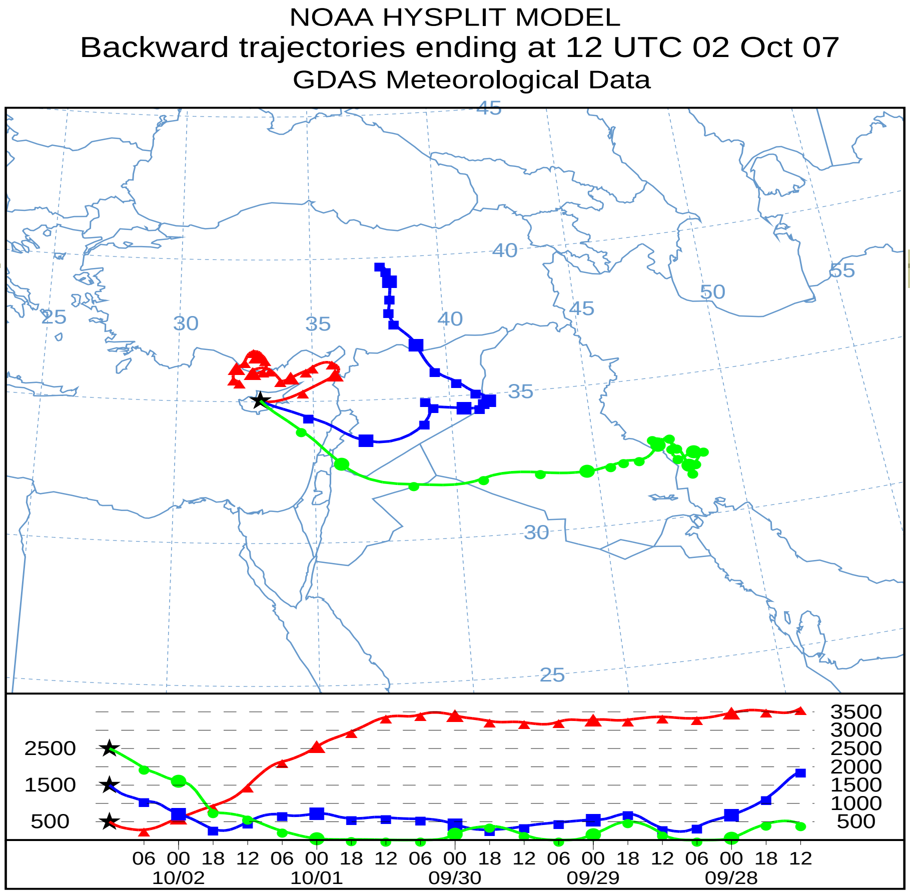
- 9 countries (53%) considered transboundary air pollution as the main factor for non-compliance
- 2 countries (12%) attributed a significant number of exceedances to winter-sanding and salting
- Objections raised for 53% of the applicant countries were attributed to *inadequate or incomplete source apportionment*!

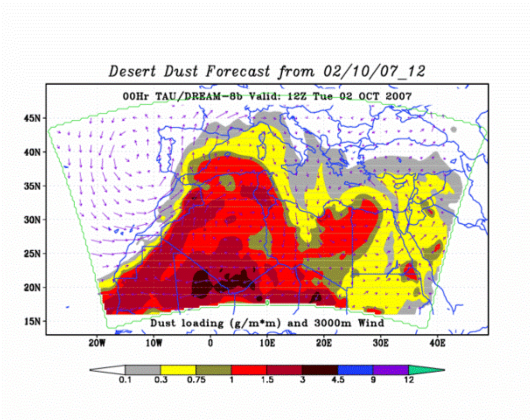
Models for source apportionment used by different EU countries according to the time extension reports

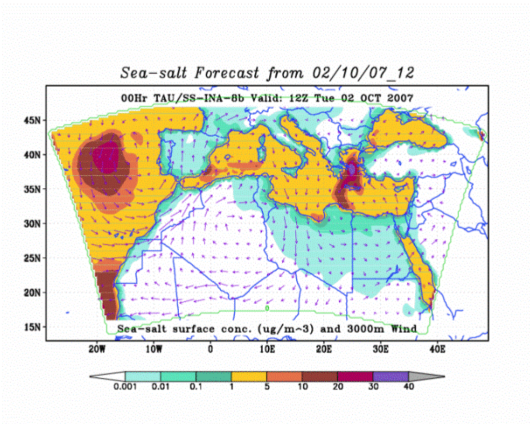


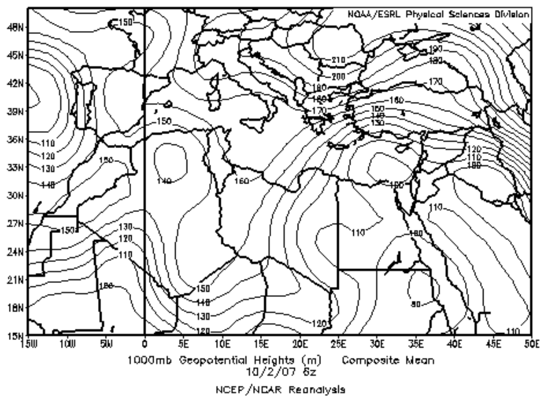
Different model combinations were used by member countries…

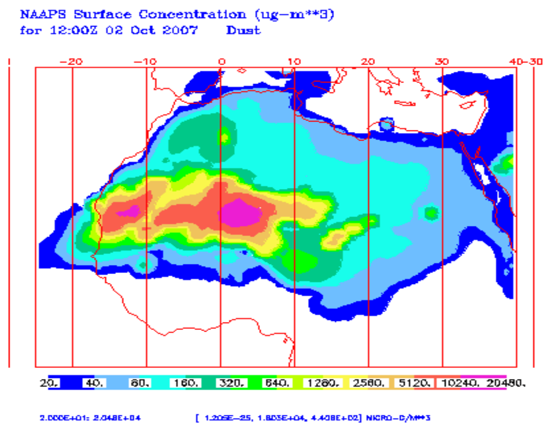
- Eulerian dispersion models were complemented by Lagrangian trajectory models to account for transboundary contributions:
  - Cyprus, Portugal and Spain (natural transboundary contributions)
  - Belgium and Austria (anthropogenic transboundary contributions)
- Eulerian dispersion models have been used in combination with statistical receptor models for source attribution of both local/national and long-distance sources (Greece, Italy).
- A Gaussian model was used for air quality assessment complemented by a Eulerian Chemical Transport Model to assess transboundary contribution (Slovakia).
- Slovakia and Poland were the only countries to account for resuspension using the EPA emissions modelling approach, which requires input information on traffic characteristics, dust load on the road and road type.


NOAA HYSPLIT MODEL
Backward trajectories ending at 12 UTC 02 Oct 07
GDAS Meteorological Data
2500
1500
500
3500
3000
2500
2000
1500
1000
500
06 00 18 12 06 00 18 12 06 00 18 12 06 00 18 12 06 00 18 12
10/02
10/01
09/30
09/29
09/28


Desert Dust Forecast from 02/10/07_12
00Hr TAU/DREAM-8b Valid: 12Z Tue 02 OCT 2007
Dust loading (g/m*m) and 3000m Wind


Sea-salt Forecast from 02/10/07_12
00Hr TAU/SS-INA-8b Valid: 12Z Tue 02 OCT 2007
Sea-salt surface conc. (ug/m-3) and 3000m Wind


NOAA/ESRL Physical Sciences Division
1000mb Geopotential Heights (m) Composite Mean
10/2/07 6z
NCEP/NCAR Reanalysis


NAAPS Surface Concentration (ug-m**3)
for 12:00Z 02 Oct 2007 Dust
20. 40. 80. 160. 320. 640. 1280. 2560. 5120. 10240. 20480.

The need for model validation

- Uncertainties in input data and model processes (e.g. Emissions, secondary organics, nitrate partitioning, meteo variability etc.)
- Models have to be assessed to ensure that they meet certain quality objectives recommended for regulatory use
- Common methodologies for model validation and evaluation:
  1. Comparison with data from dedicated monitoring campaigns to test model accuracy and representativity (monitoring data accuracy and coverage is essential)
  2. Model intercomparison studies:
     - provide useful information on model accuracy and reliability
     - reveal model limitations for specific pollutants, spatial scales and applications
     - through similar exercises, hybrid models or combined model application may emerge as innovative solutions to reduce uncertainty

Model validation in extension reports

- Several countries verified the model results against available measurements within the frame of the application.
- The majority of the models used by the member countries for source apportionment are extensively validated in the literature.
- In some cases (United Kingdom, Portugal and France) model validation was explicitly described:
  - **United Kingdom**: use of a Volatile Correction Model to calibrate model results prior to comparison with measurements
  - **Portugal**: “Standard Guide for Statistical Evaluation of Atmospheric Dispersion Model Performance” (ASTM, 2005) was used to validate the TAPM modelling system
  - **France**: the Eulerian CTM modelling system PREV’AIR was used to estimate transboundary and natural contributions, including on-line verification procedures

Conclusions

- This review confirms the **increased use of modelling** tools for source apportionment by member states and researchers
- The analysis of the time extension reports revealed (as expected!) the **lack of a uniform methodology** for source apportionment
- A standardised methodological framework and guidance would be useful, stressing on issues of QA and uncertainty estimation
- Still many limitations regarding:
  - certain compounds not adequately quantified (e.g. biogenic secondary organics, nitrate components etc.)
  - specific anthropogenic emission sources not sufficiently discriminated in many source apportionment studies (e.g. shipping emissions)
  - the identification of biomass combustion sources

Thanks for your attention!