The initiative on harmonisation of source apportionment with Receptor Models in Europe
main results 2010-2013

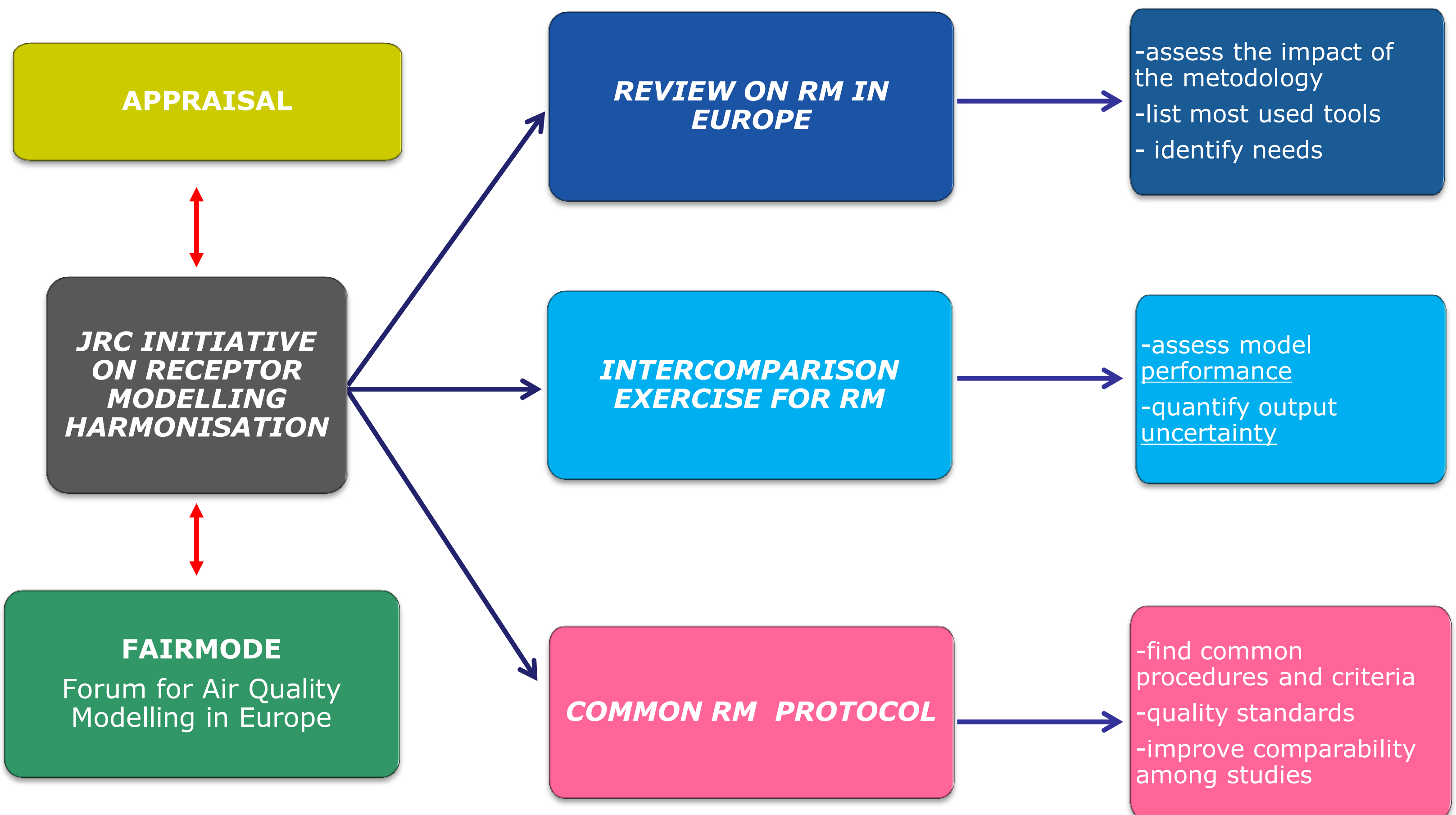
C.A. Belis
European Commission, Institute for Environment and Sustainability – JRC

and the source apportionment community

Fairmode Technical Meeting, Kjeller, 28-29/04/2014

FAIRMODE
Forum for air quality modelling in Europe

Are RMs appropriate for air quality management?



REVIEW

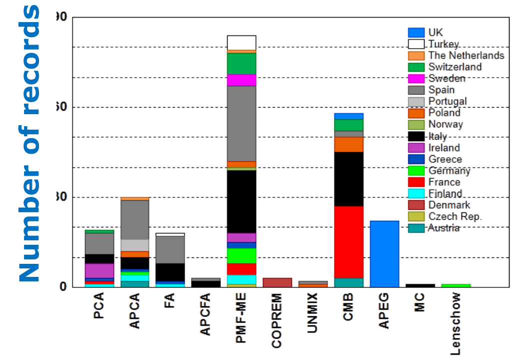
European Commission

Deliverables

Enhancing source apportionment with receptor models to foster the air quality directive implementation. Karagulian & Belis, 2012 IJEP 50

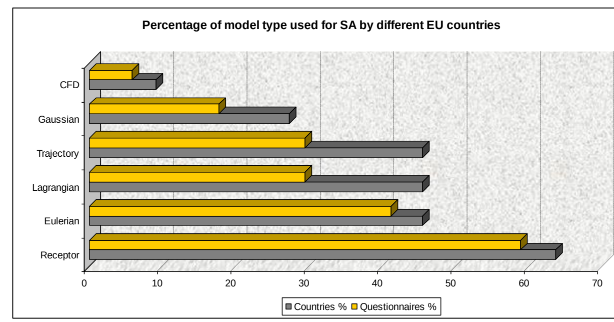
Needs for the development of RMs in Europe

80 studies 243 records



Current trends in the use of models for source apportionment of air pollutants in Europe E. Fragkou, I. Douros, N. Moussiopoulos, C. A.Belis. 2012 IJEP 50

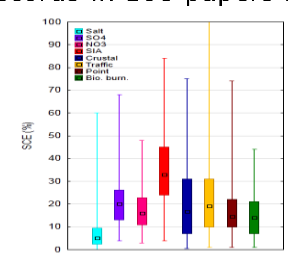
Includes all types of SA methodologies



Critical Review and meta analysis of ambient particulate matter source apportionment with receptor models. C.A. Belis. F. Karagulian, B. Larsen, P.K. Hopke. 2013 Atmospheric Environment.69,94-108

RMs description and classification + meta analysis

272 records in 108 papers 2012



INTERCOMPARISON

INTERCOMPARISON EXERCISE FOR RM

First step (real-world dataset)
16 participants

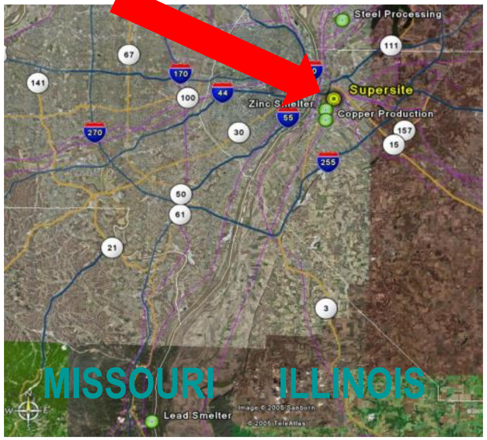
ORGANIZATION	COUNTRY
IDAEA CSIC	SPAIN
Univ. Aahrus	DENMARK
University of Genoa	ITALY
Finnish Meteorological Institute	FINLAND
INERIS/LSCE	FRANCE
University of Birmingham	UNITED KINGDOM
Norwegian Institute for Air Research (NILU)	NORWAY
Department of Physics University of Florence	ITALY
University of Milan Bicocca	ITALY
C.N.R. Institute for Atmospheric Pollution Research	ITALY
IUTA e.V.	GERMANY
NCSR Demokritos, Environmental Research Laboratory	GREECE
Dept. of Physics - University of Milan	ITALY
Paul Scherrer Institut Laboratory of Atmospheric Chemistry	SWITZERLAND
C.N.R - I.S.A.C.	ITALY
JOINT RESEARCH CENTRE	European Commission

Second step (synthetic dataset)
22 participants

ORGANIZATION	COUNTRY
IDAEA CSIC	SPAIN
Univ. Aahrus	DENMARK
University of Genoa	ITALY
Finnish Meteorological Institute	FINLAND
University College Cork	IRELAND
University of Birmingham	UNITED KINGDOM
University of Florence Department of Physics	ITALY
Faculty of Science Charles University in Prague	CZECH REPUBLIC
National Institute of Public Healt and the Environment (RIVM)	THE NETHERLANDS
C.N.R. Institute for Atmospheric Pollution Research	ITALY
Miguel Hernández University	SPAIN
NCSR Demokritos, Environmental Research Laboratory	GREECE
University of Milan Dept. of Physics	ITALY
Paul Scherrer Institute - Laboratory of Atmospheric Chemistry	SWITZERLAND
C.N.R - I.S.A.C.	ITALY
Aristotle University of Thessaloniki	GREECE
University of Milan Bicocca	ITALY
University of Aahrus	DENMARK
University of Lisbon	PORTUGAL
Pontificia Universidad Católica de Chile	CHILE
University of Sao Paulo	BRAZIL
Joint Research Centre	European Commission

European Commission

Real-world Dataset



To have inorganic and organic species two datasets collected in the St. Louis supersite were merged.

The final dataset contained 178 $PM_{2.5}$ 24 h samples with 42 chemical species.

Original publications:

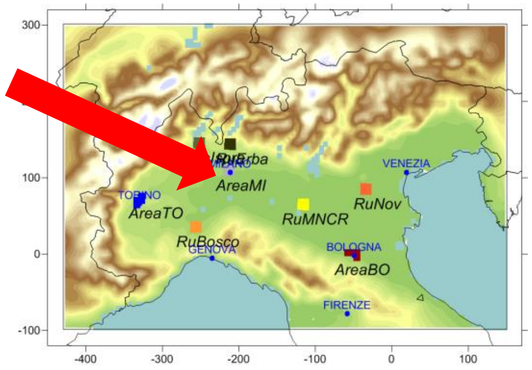
-Lee, J. H., Hopke, P. K., and Turner, J. R., 2006. Source identification of airborne $PM_{2.5}$ at the St. Louis-Midwest Supersite. Journal of Geophysical Research D: Atmospheres 111,

-Jaeckels, J. M., Bae, M. S., and Schauer, J. J., 2007. Positive matrix factorization (PMF) analysis of molecular marker measurements to quantify the sources of organic aerosols. Environmental Science and Technology 41, 5763-5769.

Synthetic Dataset

A run was executed using CAMx and PSAT over a computational domain covering the whole Po Valley. $PM_{2.5}$ sources were extracted for a cell located in the city of Milan. Noise was introduced «a posteriori».

The final input data matrices contained 364 24 h samples with 38 inorganic and organic species.



INTERCOMPARISON EXERCISE FOR RM

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MODEL	SOLUTIONS
➡ EPA PMF v3.0	8
➡ PMF-2	6
➡ EPA CMB 8.2	4
APCS	1
COPREM	1
ME-2	1
PCA	1
TOTAL	22

MODEL	SOLUTIONS
➡ EPA PMF v3.0	12
➡ PMF-2	3
EPA PMF v5.0	1
EPA PMF V4.1 (beta)	1
➡ EPA CMB 8.2	4
CMB-ROBOTIC	1
COPREM	1
ME-2	1
FA-MLRA	2
TOTAL	26

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Evaluation Methodology → by source categories (Belis et al., submission)

Complementary tests Provide ancillary information about the solutions' performance

Mass closure
Number of factor/sources

Preliminary tests Test if source/factors belong to a given source category

Fingerprints → Pearson, Pearson (log transformed), Weighted Difference

Time-trends → Pearson

Species contributions (%) → Pearson

= % of species total matrix (EPA PMF v3) = explained variation (PMF 2) = contribution by species (CMB 8.2)

$$WD_{ij} = 1/n \sum_{a=1}^{n} \frac{x_{ia} - x_{ja}}{\sqrt{s_{ia}^2 + s_{ja}^2}}$$

(Karagulian & Belis, 2012)

Source/factors accepted if pass > 50% of the tests

Performance tests evaluate if SCEs fall within an established quality objective

$$z\text{-score}(SCE) = \frac{x_i - X}{\sigma_p}$$

xi: solution i
X: reference
u: uncertainty

σ_p= uncertainty criterion 50%

$2 < |z| \leq 3$ warning area

$|z| > 3$ action area

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INTERCOMPARISON EXERCISE FOR RM

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STEP 1

Data set: mass concentrations of species and uncertainties

OCT
OC1
OC2
OC3
OC4
OP
ECT
EC1m
EC2
EC3
SO4
NO3
NH4
Al
As
Ba
Ca
Co
Cr
Cu
Fe
Hg
K
Mn
Ni
P
Pb
Rb
Se
Si
Sr
Ti
V
Zn
Zr

INORGANIC DB
From June 2001 – May 2003
24h samples collected every day

Reference:
Lee, J. H., P. K. Hopke, and J. R. Turner (2006),
Source identification of airborne PM2.5 at the St. Louis-Midwest Supersite, *J. Geophys. Res.*, 111, D10S10,

indeno(cd)pyrene
benzo(ghi)perylene
benz(a)anthracene
benzo(a)pyrene
fluoranthene
pyrene
coronene
benzo(b,k)fluoranthene
benzo(e)pyrene
benzo(j)fluoranthene
dibenz[a,h]anthracene
levoglucosan

ORGANIC DB
From May 2001 – July 2003
24h samples collected every 6th day

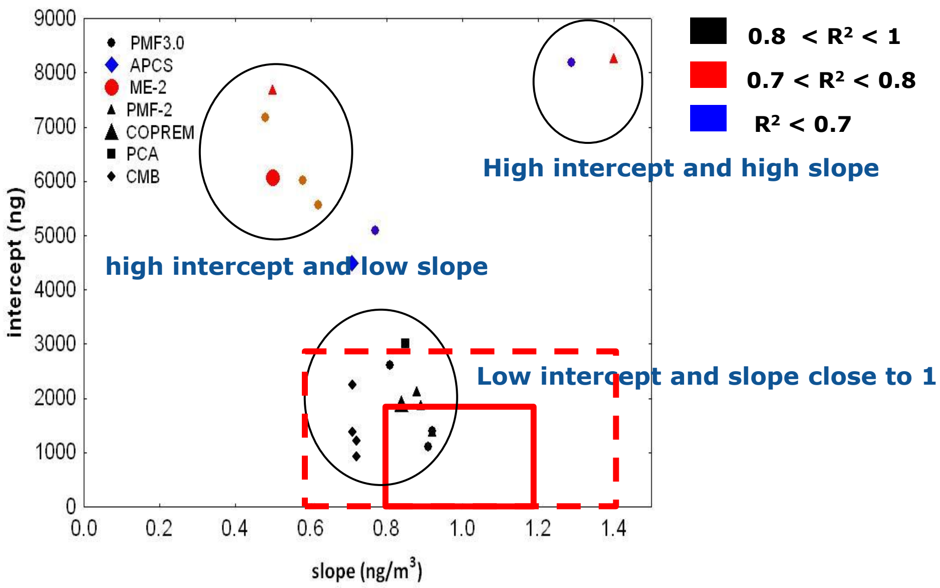
Reference:
Jaeckels JM, Bae M.S., Schauer JJ (2007) Positive matrix factorization Analysis of molecular markers measurements to quantify the sources of organic aerosols. *EST.* 41-5763

Structure of data:

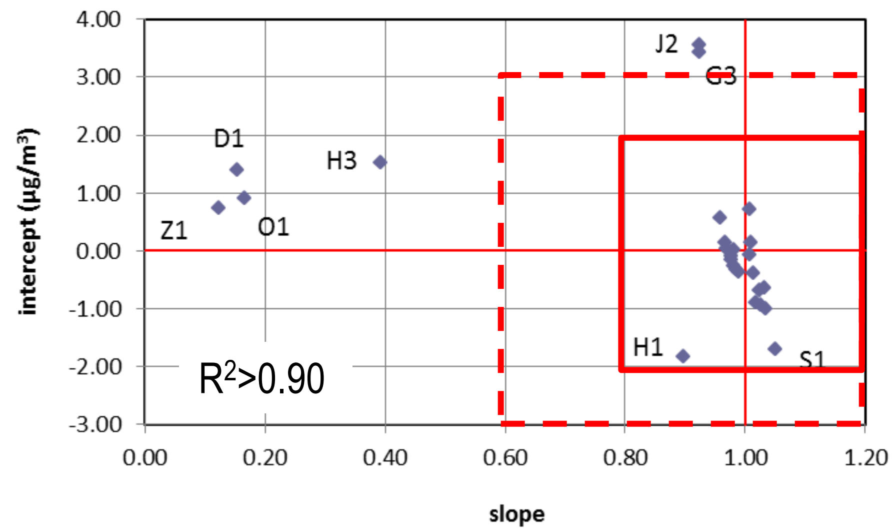
- **inorganic ions: high uncertainty**
- **Co, Cr, Hg, Ni, Rb, Ti, Va, Zr have many missing values**
- **Ca, Fe, Zn, K uncertainties below 5%**
- **there were differen MDLs, probably due to different analytical batches**
- **PAHs presented many BDL values.**

Complementary Test 1
Modelled *vs* measured mass

STEP 1



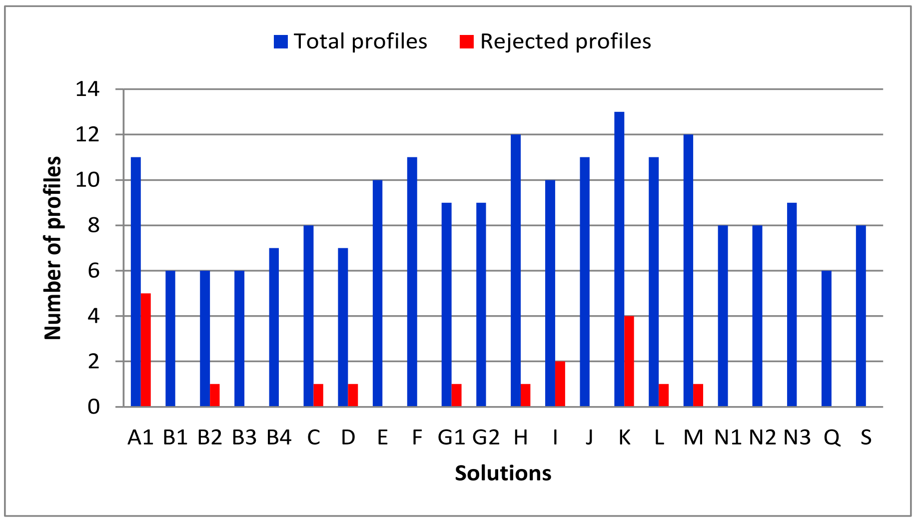
STEP 2



77% of the solutions fall close to the target (20% tolerance for the slope and 2 µg/m^3 tolerance for the intercept).

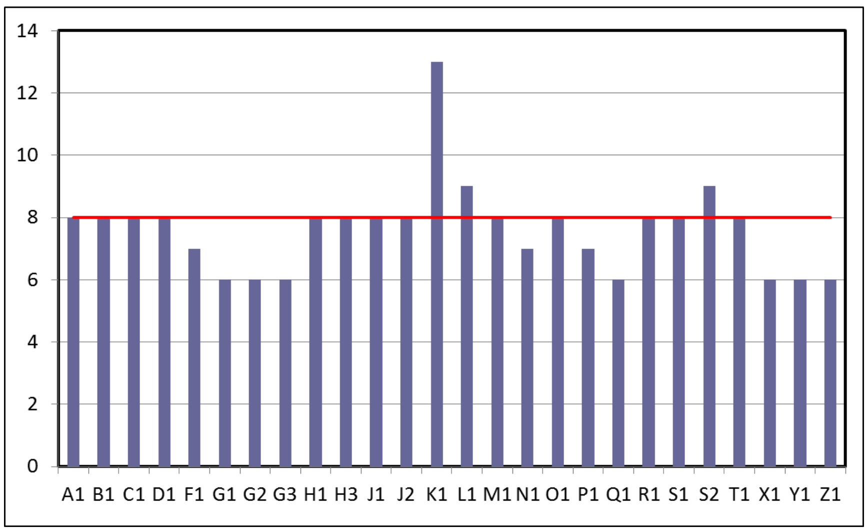
Complementary Test 2
Number of factor/source profiles

STEP 1



50% of the solutions report between 6 and 10 number of factor/sources.
7 solutions >10 factor/sources

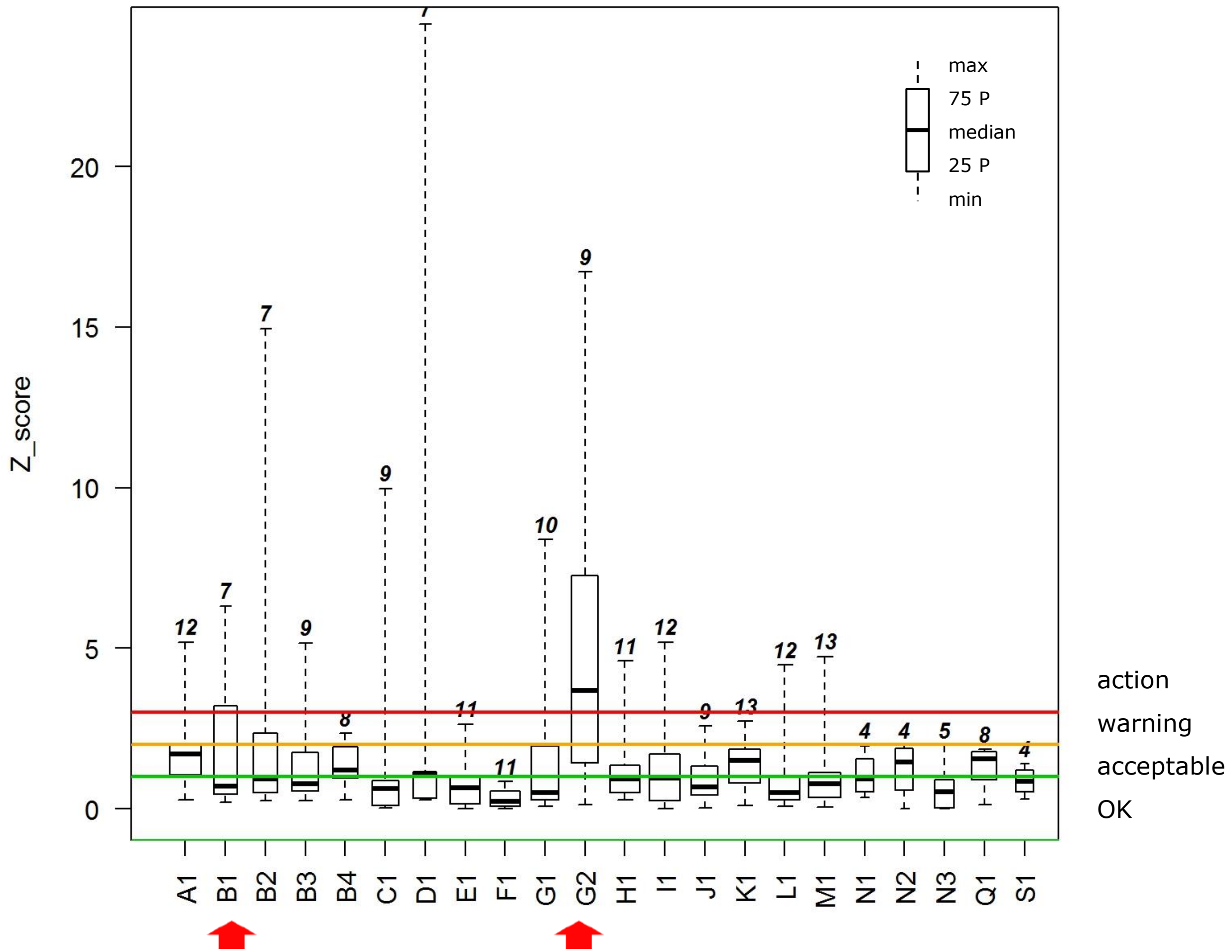
STEP 2



50% of the solutions report the correct number of sources (8).
96% of the solutions between 6 and 9 factor/sources

STEP 1

z scores grouped by solution



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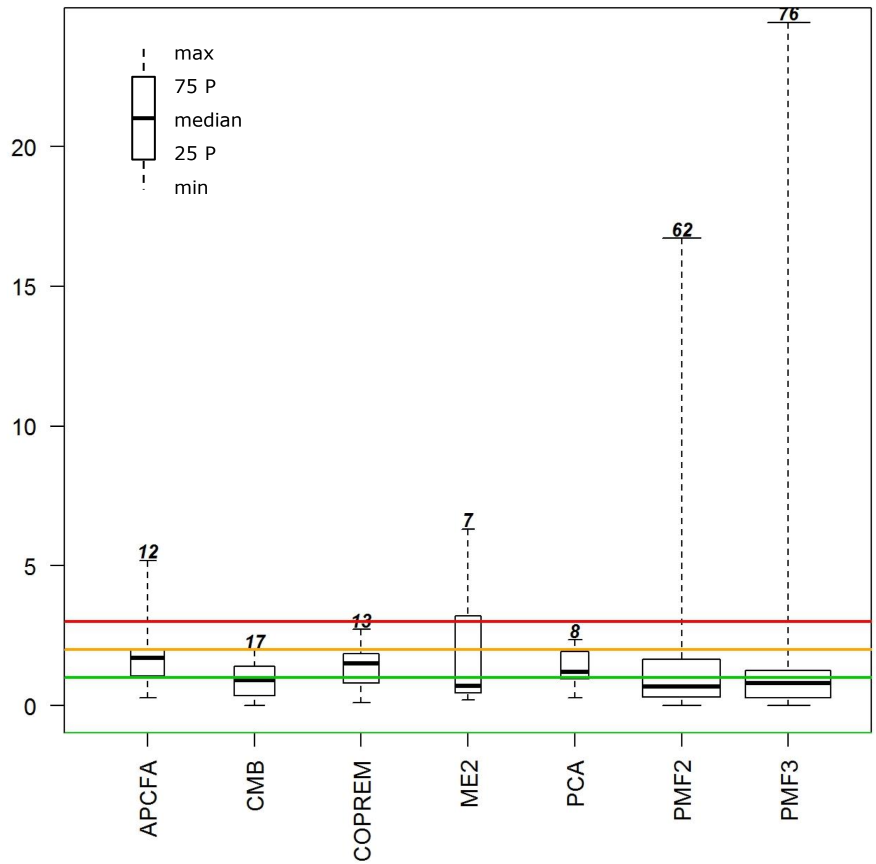
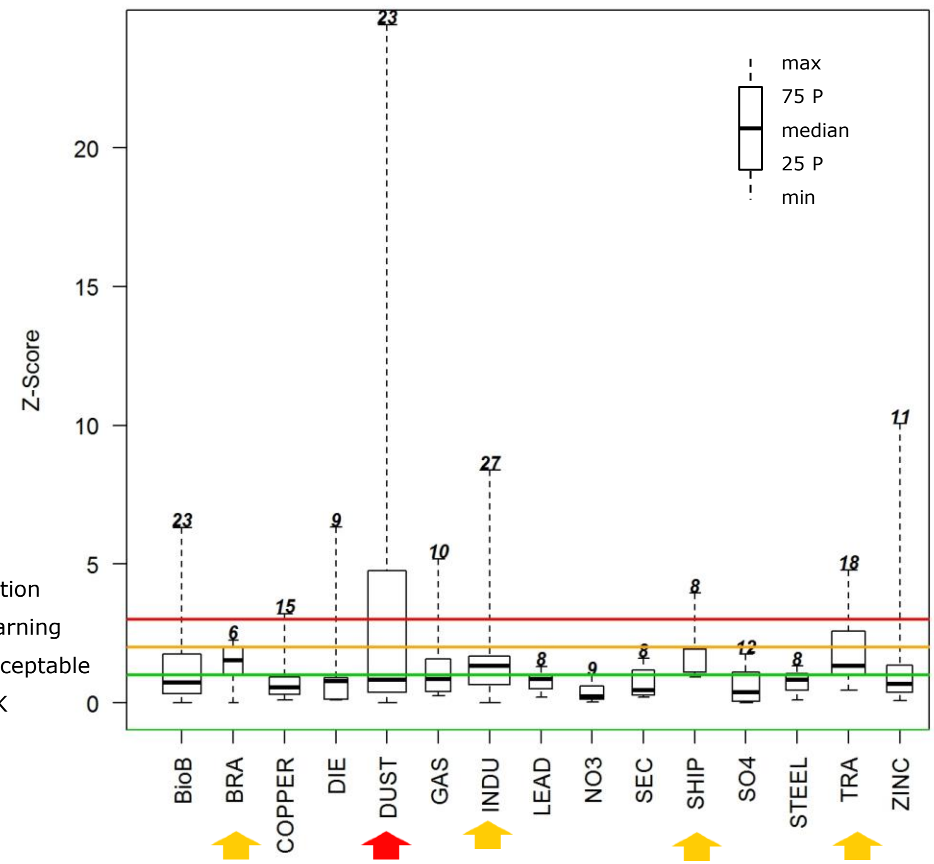
STEP 1

z scores grouped by model and by source category



action
warning
acceptable
OK



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Conclusions STEP 1

1. The new methodology used for the evaluation of the IE appears appropriate to test the comparability between factors in terms of both fingerprint and time trend.

2. There is a reasonable quantitative agreement between SCE. 86% of the factors meet the acceptability criteria (OK or acceptable).

3. The participants' bias in the SCEs are consistent with the 50% maximum uncertainty acceptability criterion adopted in this evaluation.

4. However, there was a considerable variability in the number of factors identified by participants.

5. Some models were used by only one or two participants, therefore it is not possible to draw conclusions about the performace of these models.

6. One limitation of using real world data is that the reference SCEs are obtained as the average of participants. This may obscure a methodology bias. In our case, comparison with published solutions of the same dataset was also satisfactory.

INTERCOMPARISON EXERCISE FOR RM

STEP 2

Synthetic Dataset species

LEVOGLUCOSAN	PB
ORGANIC CARBON	NI
ELEMENTAL CARBON	SR
NO3	CR
SO4	SB
CL	SN
NH4	RB
NA	MO
K	AS
CA	CD
MG	CHRYSENE
SI	BENZO(B)FLUORANTHENE
FE	BENZO(K)FLUORANTHENE
AL	BENZO(E)PYRENE
ZN	BENZO(A)PYRENE
TI	INDENO(123,C,D)PYRENE
CU	DIBENZO(A,H)ANTHRACENE
V	BENZO(G,H,I)PERYLENE
MN	CORONENE

The final input data matrices contained 364 24 h samples with 38 chemical species including inorganic and organic components.

Synthetic Dataset sources categories

NAME	CODE	CONTRIB. ($\mu g/m^3$)
Biomass burning	BioB	4.33
Ammonium sulphate	SO4	7.12
Ammonium nitrate	NO3	12.69
Mineral dust	DUST	4.01
Road dust	ROAD	2.68
Sea salt – Road salt	SALT	0.52
Traffic exhaust	TRA	6.63
Industry	INDU	5.11

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Synthetic Dataset 2

- Real-world emission profiles for group of sources were used to estimate the concentration of species that are not calculated by the model (e.g. trace elements).
- The final SCEs were obtained by combining the time trend of the sources produced by the model and the chemical profiles of 8 source categories.
- The **noise** was introduced to each species using a *normal distributed random variable (u)* centered on zero with standard deviation equal to the species *average relative standard uncertainty* obtained from a real-world dataset (Larsen et al., 2012)

$$C_{pert,ij} = C_{ij} + (C_{ij} \times u_j)$$

- The **uncertainty** of the input species concentration was generated by fitting a curve to describe the relationship between concentration and uncertainty in the above mentioned dataset.

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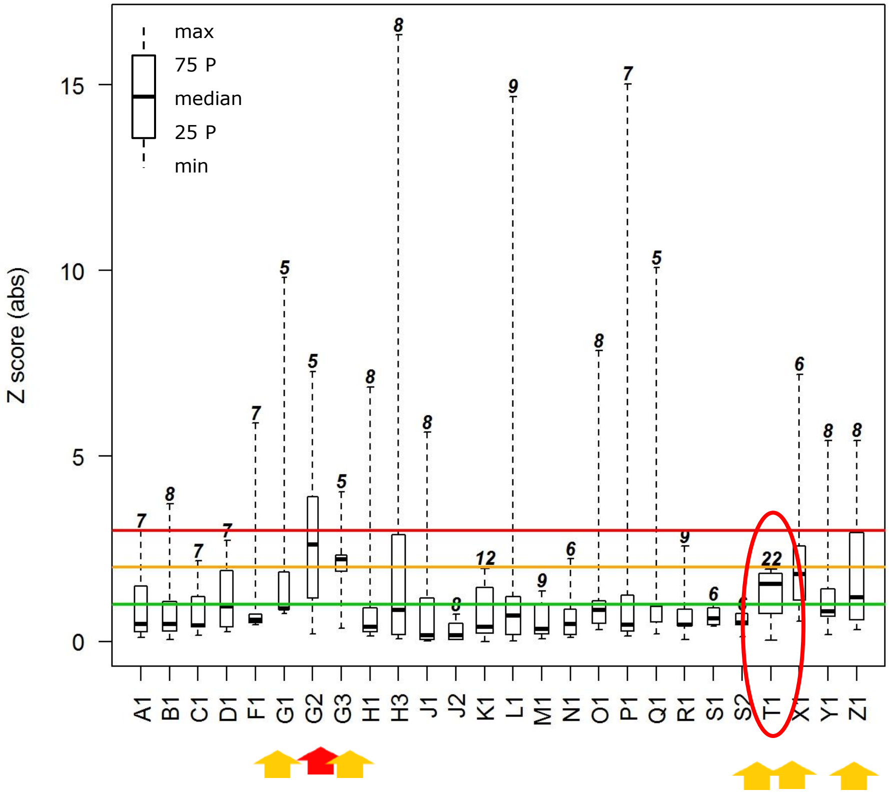
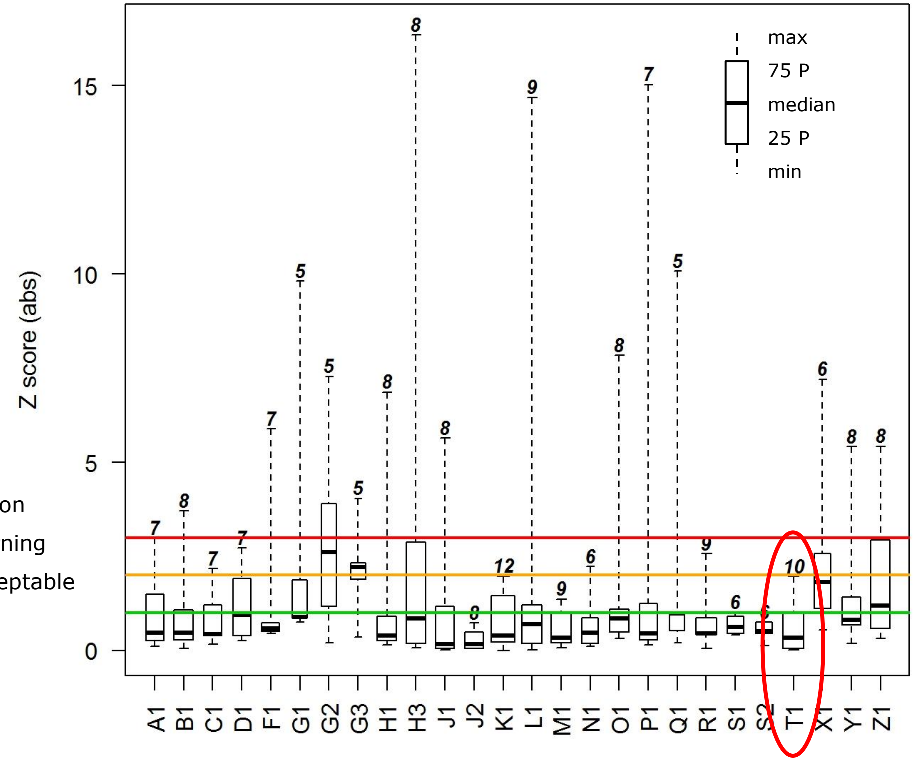
z scores grouped by solutions (original and corrected)



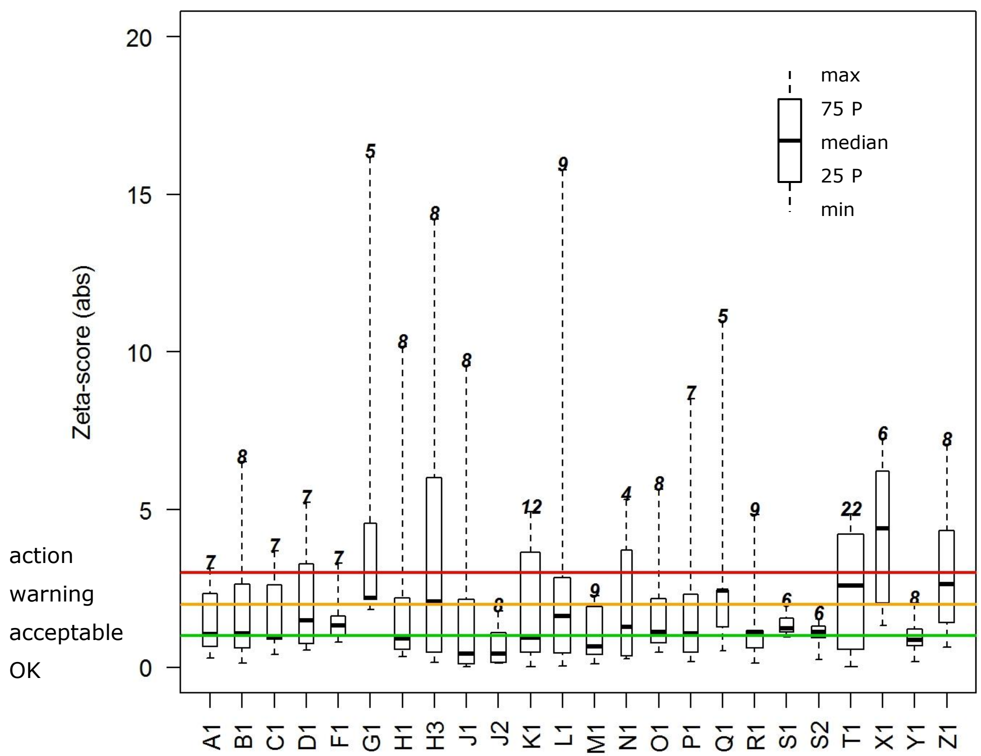
action
warning
acceptable
OK



original

corrected T1 & H1

Zeta scores


Zeta-score (abs)
20
15
10
5
0
max
75 P
median
25 P
min
action
warning
acceptable
OK
A1
B1
C1
D1
F1
G1
H1
H3
J1
J2
K1
L1
M1
N1
O1
P1
Q1
R1
S1
S2
T1
X1
Y1
Z1

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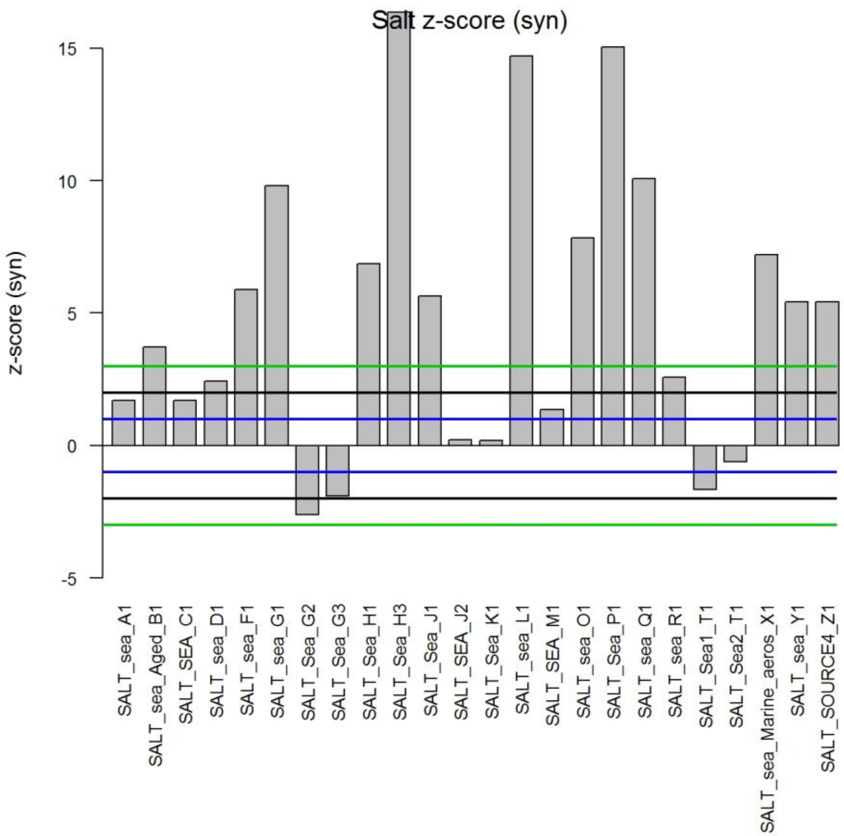
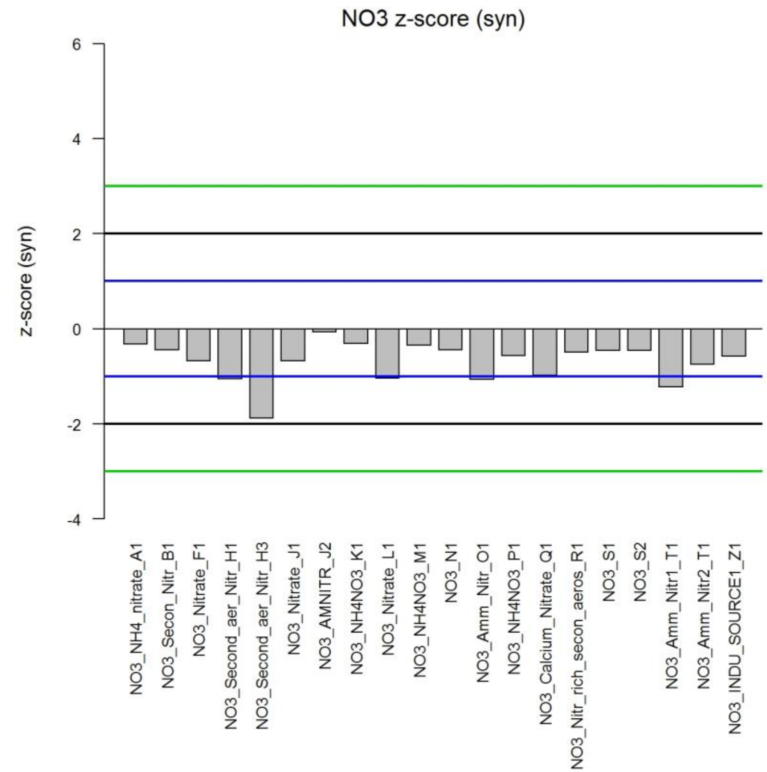
z-scores in SALT and NO3





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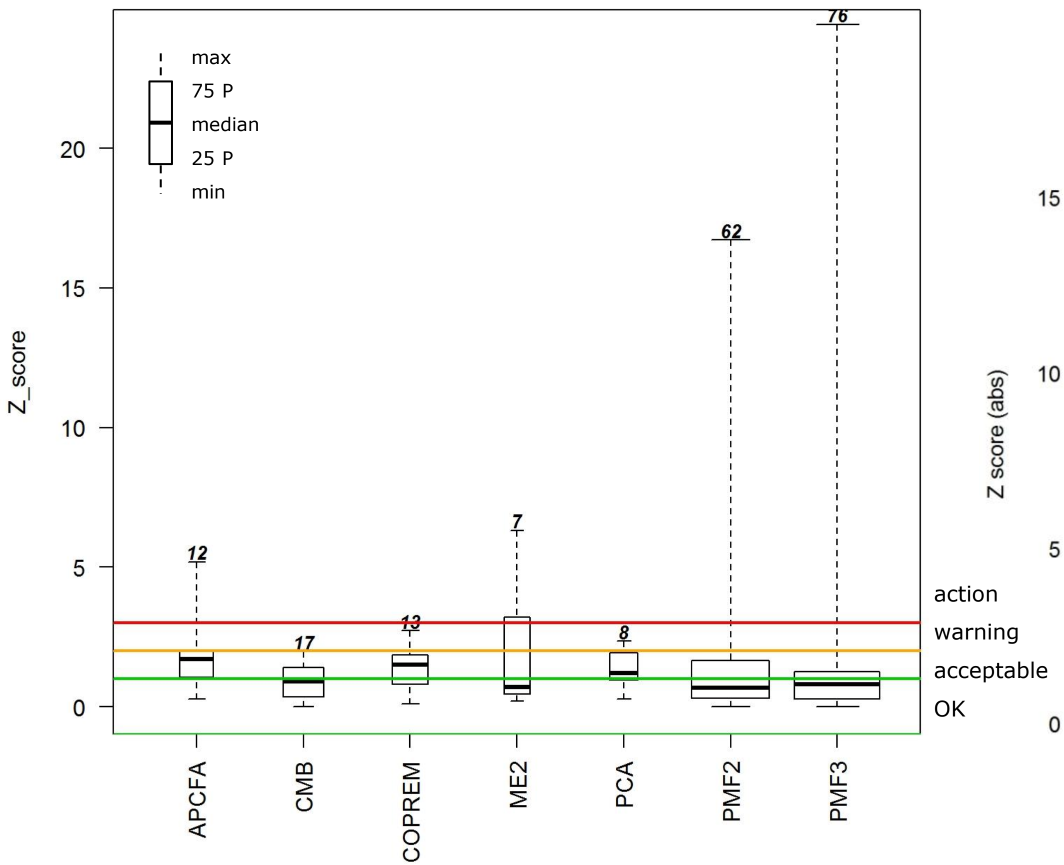
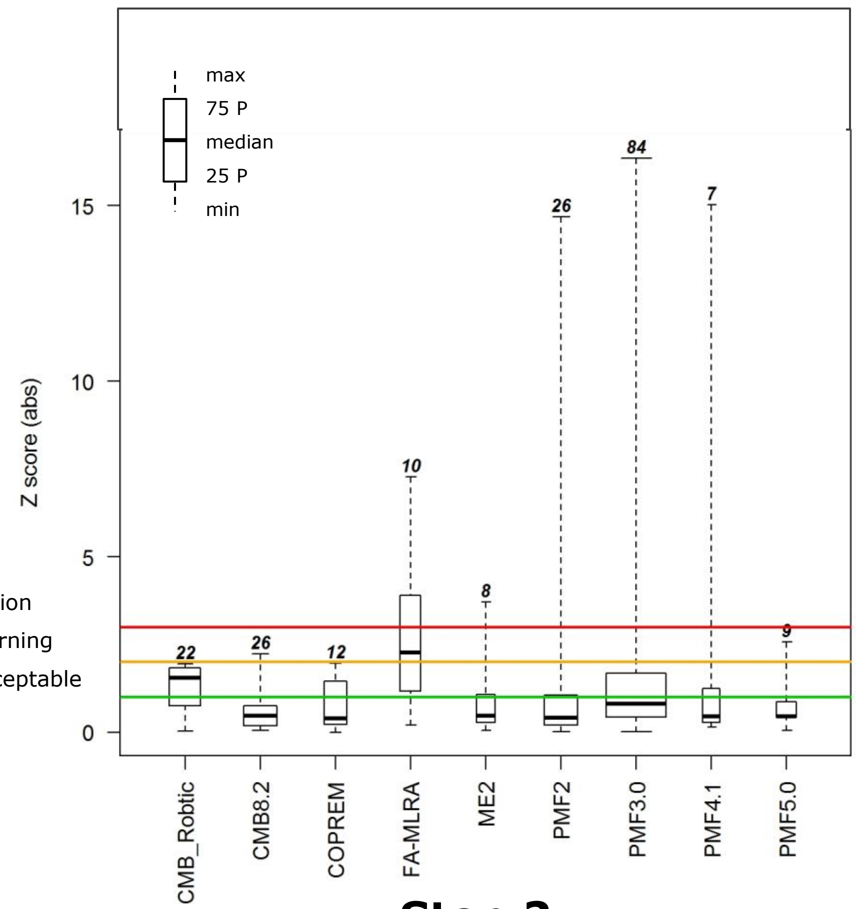
z scores grouped by model



Step 1



Step 2

Intercomparison Final Remarks

- An 86% and 85 %, of the factors/sources met the acceptability criteria in the first and second step, respectively, indicating that the 50% uncertainty target is substantially observed.
- The overall assessment is mainly valid for models with high number of solutions: EPA-PMF3, PMF2, and EPA-CMB 8.2 (and to a lesser extent ME-2 and COPREM).
- The combination of real-world and synthetic datasets made it possible to assess both models' performance with respect to an unbiased reference and their ability to deal with data noise.
- A tendency to slightly underestimate the relative contribution of dominant sources and to overestimate the relative contribution of small sources (< 5%) was observed.

INTERCOMPARISON EXERCISE FOR RM

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Intercomparison deliverables

- EUR Report 2012 (STEP1)
- Oral presentation at EAC 2012 (STEP1)
- Oral presentation at EAC 2013 (STEP2)
- Oral presentation AAAR 2013 (both steps)
- Scientific paper on methodology (submission)
- Scientific paper on results (in preparation)

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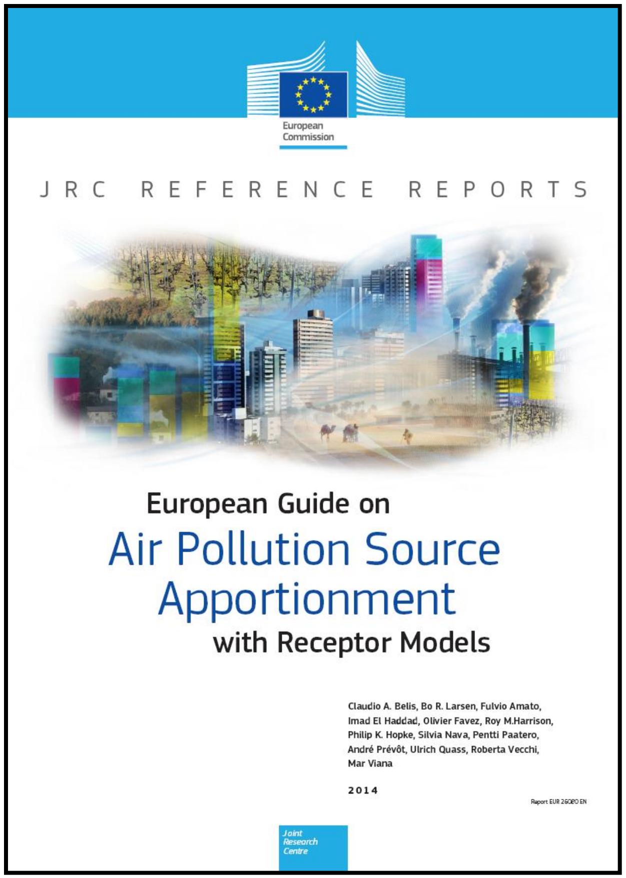
Common Protocol: Driving elements



- The main objective is to promote the best available operating procedures and to harmonize their application across Europe.
- Promote implementation of the protocol in new studies
- Establish a feed-back mechanism from users in Ms
- Schedule dissemination and capacity building activities

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COMMON RM PROTOCOL

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TABLE OF CONTENTS

Common Protocol: structure

SA studies can be considered as being consistent with the present protocol if :

1. The results are described according to the steps proposed in sections B1- B12.
2. Expert decisions are described and evidence of the objective information that support them is provided. (essential for critical steps).
3. The documentation includes the references of the source profiles used as input or to validate factor assignment.
4. The model and version used are clearly reported.
5. The quantitative uncertainty of the output is estimated and reported.
6. Estimation of overall uncertainty and validation is achieved by comparing outputs from independent models on the same dataset and/or using Monte Carlo permutation and/or displacement analysis techniques.
7. Sensitivity analysis is performed to demonstrate that there are no substantial deviations from the mass conservation assumption.
8. Only solutions that implement the quality assurance steps described in this guide can claim state-of-the-art performance supported by community-wide intercomparison exercises.

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Concluding Remarks

-Most used models and needs were identified in the review process.

Needs:

1. Quantification of model performances
2. Harmonisation of methodologies
3. Network of permanent monitoring sites with speciated PM in urban areas
4. Creation of source profile repositories
5. Mutual validation and integration among different SA techniques

-The intercomparison exercises demonstrated that RM outputs are consistent with a 50% uncertainty criterion (bullet 1).

-The common protocol provides harmonized procedures and criteria for most common RM (bullet 2). Continuous update is required to catch up with new and continuosly evolving techniques.

-More work is neeeded to deal with points 3, 4, and 5.

THANKS TO ALL THE COLLEAGUES WHO CONTRIBUTED TO THIS INTIATIVE

Intercomparison exercises :

F. Karagulian, M. Almeida, F. Amato, G. Argyropoulos, P. Artaxo, D.C.S. Beddows, V. Bernardoni, M.C. Bove, S. Carbone, D. Cesari, D. Contini, E. Cuccia, D. Contini, E. Diapouli, K. Eleftheriadis, I. El Haddad, O. Favez, R.M. Harrison, S. Hellebust, I. Hovorka, E. Jang, H. Jorquera,T. Kammermeier, M.Karl, F. Lucarelli, D. Mooibroek, S.Nava, J. K. Nøjgaard, M. Pandolfi, M.G. Perrone, J.E. Petit, A. Pietrodangelo, G. Pirovano, P. Pokorná, P. Paatero, P. Prati, A.S.H. Prévôt, U. Quass, X. Querol, C. Samara, D. Saraga, J. Sciare, A. Sfetsos, G. Valli, R. Vecchi, M. Vestenius, J.J. Schauer, J.R. Turner, E. Yubero

European common protocol for receptor models:

B. R. Larsen, F. Amato, O. Favez, I. El Haddad, R.M. Harrison, A.S.H. Prévôt, S. Nava, U. Quass, R. Vecchi, M. Viana, P. Paatero