FAIRMODE Technical Meeting

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FAIRMODE
Forum for air quality modelling in Europe

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$PM_{2.5}$ source apportionment in Lombardy (Italy): comparison between CAMX and CMB modelling results

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ARPA LOMBARDIA Agenzia Regionale per la Protezione dell'Ambiente **(2)**

POLITECNICO MILANO **(3)**

Questions...

- CMB and CAMx/PSAT -> two models for Source Apportionment
  ...Is it possible to validate their results?

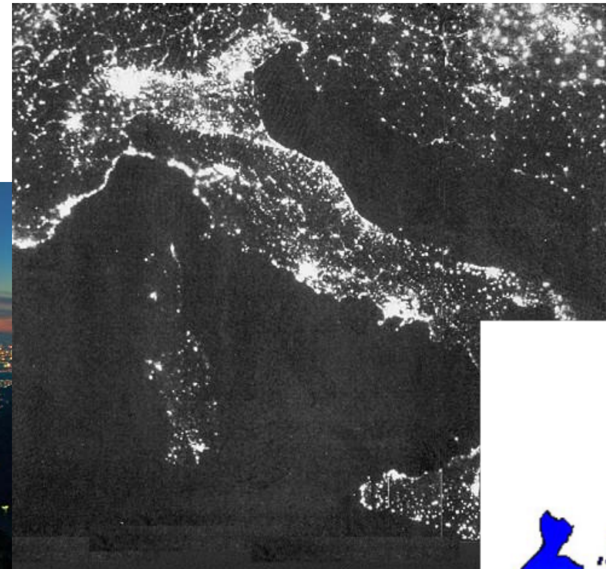
- Does the comparison of the approaches allow to point out strengths and weaknesses of both methods?

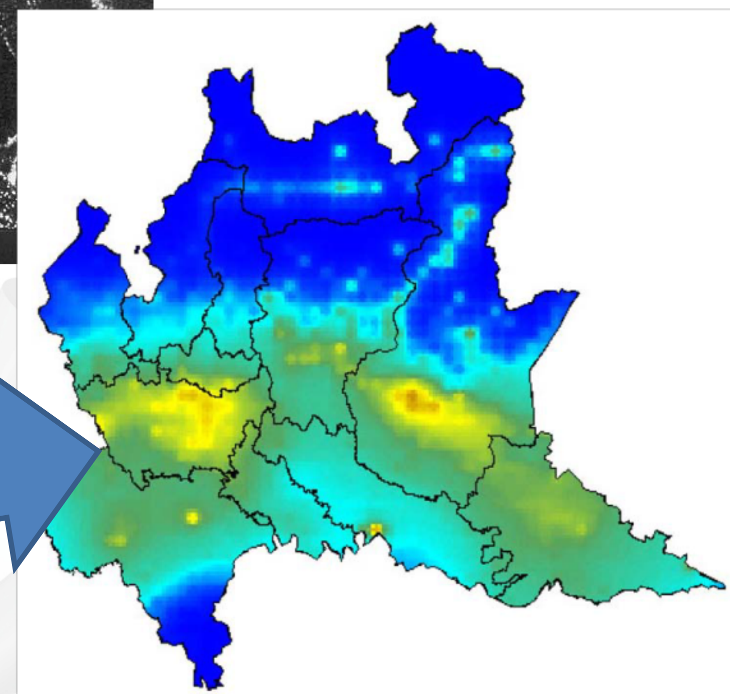
- Thanks to the combination of both approaches, is it possible to derive more accurate (quantitative) information on the role played by the different emission sources and aerosol formation processes?

The case study

on

The Po valley





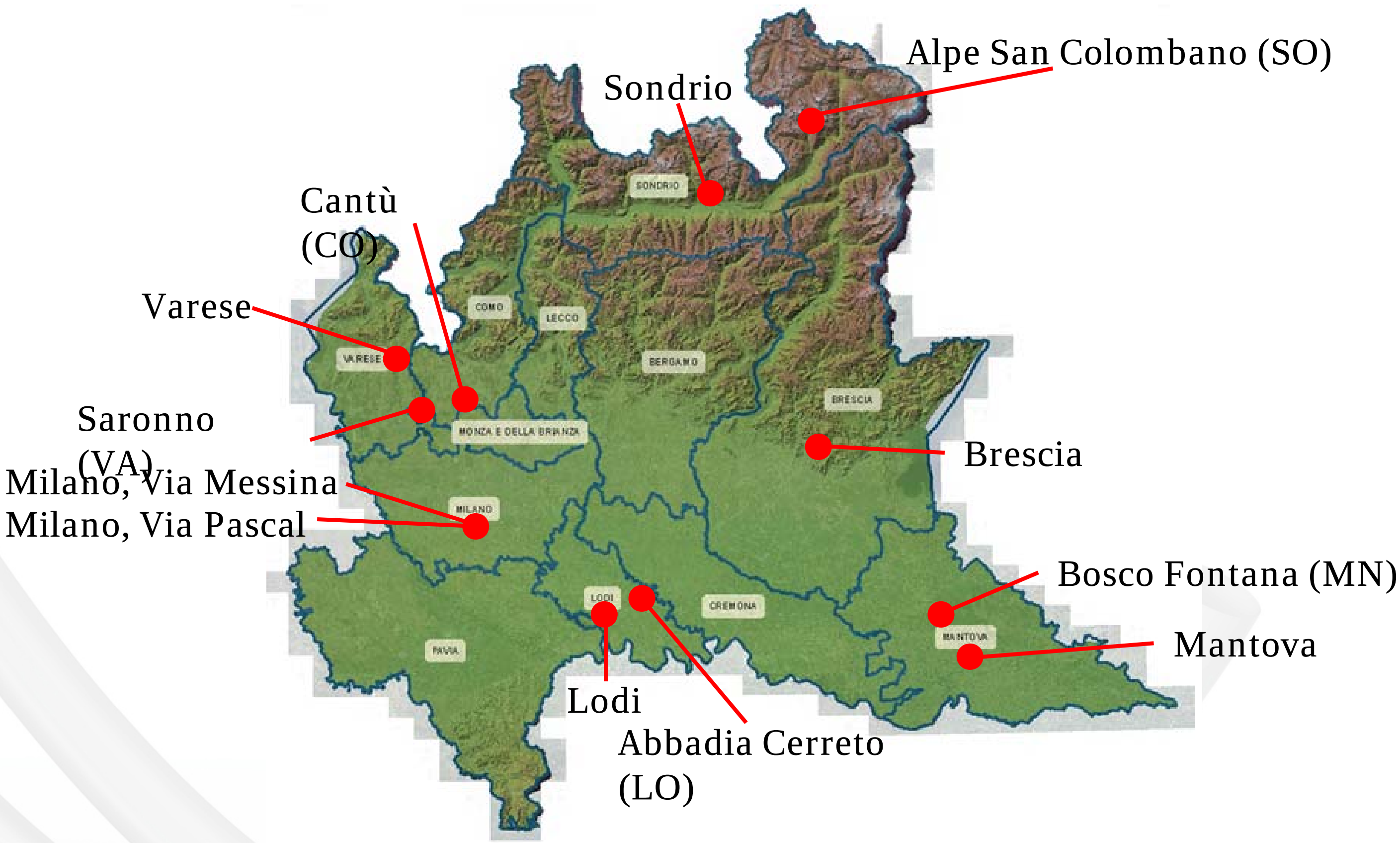
PM10 Exceedances days (2010)

150 90 70 35 0

(Source: ARPA Lombardia)

- Heavily populated and industrialized (up to now...)
- Critical meteorological conditions
- Lack of specialized observations

The PARFIL data set



One of the few examples of a rather comprehensive PM composition measured data set for the Po valley…

Ambient data set

Not overlapping with the CAMx simulation

Sampling campaign design

- Period: 01.01.04-30.06.07
- Daily PM2.5 gravimetric sampling
- Sampling sites: 6 UB, 3 UT, 1 RB and 1 Alpine site.
- Analysis on collected filters (about one week each month per kind of component): about 700 daily concentration data per site and kind of component

Elements(Z>11)

Al, Si, Ca, Sr, K, Fe, Ti, Mn, V, Cr, Ni, Cu, Br, Pb, Zn, Cl, S
XRF

Carbon

OC, EC
TGA/FT-IR and ATOT

Cations and Anions

Chloride, Nitrate, Solphate, Ammonium
IC

PAH's

B(a)P, B(a)A, B(b)F, B(k)F, I(1,2,3-c,d)P, DB(a,h)A, Fluoranthene, Pyrene, Chrysene, B(g,h,i)P
HPLC

CMB8.2 Configuration & Set up

Operation

- Control file: none
- Set optios conditions: standard
- Britt and Luecke: done
- Source elimination: none
- Best fit: none
- Ambient Data Selection: Winter and Summer semestre, each site

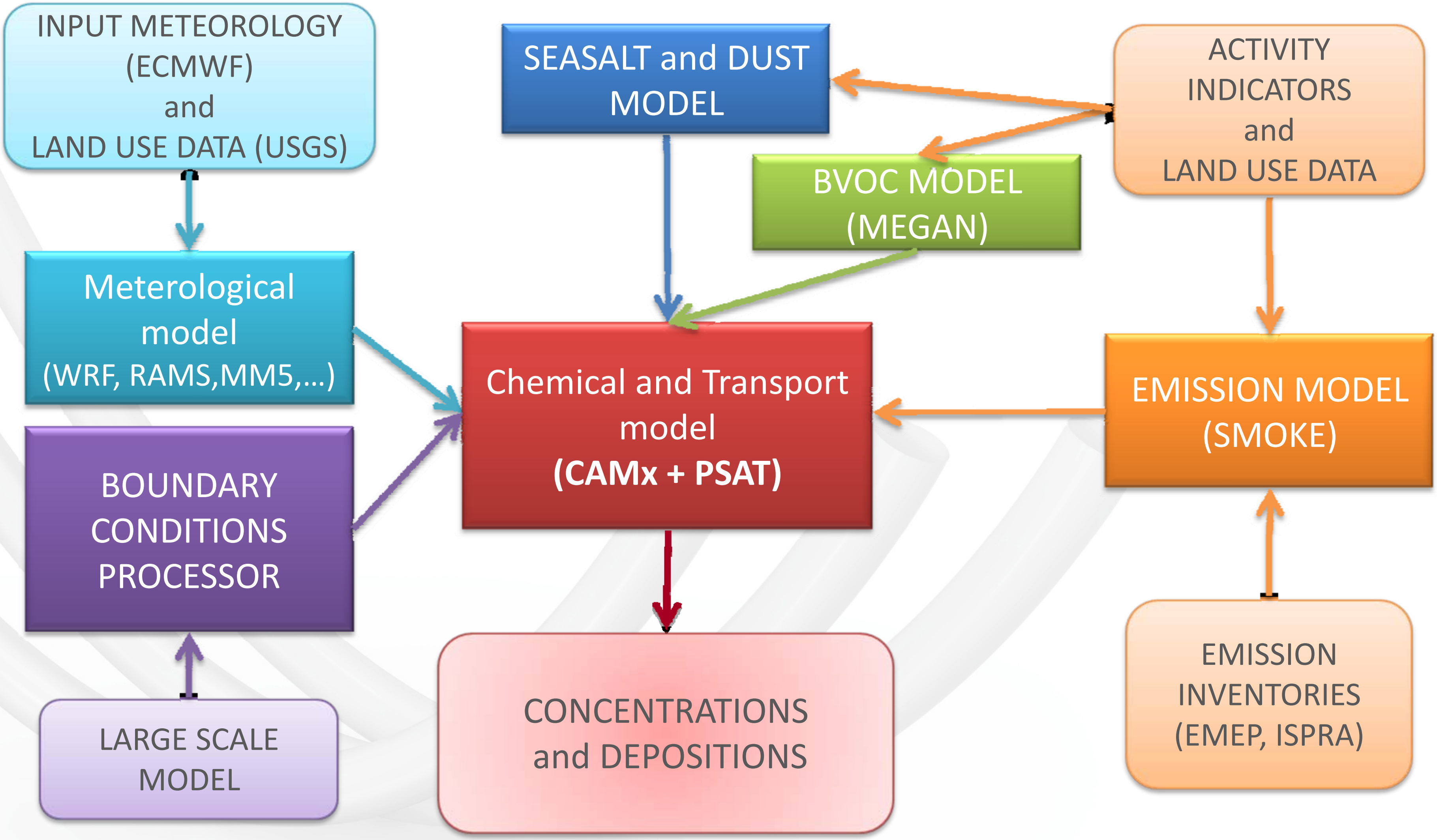
Fitting Sources Arrays:

→ Transport Light Duty Diesel Vehicles
→ Transport Heavy Duty Diesel Vehicles
→ Transport Gasoline
→ Tire and Brake Wear Erosion
→ Ammonium Sulphate
→ Ammonium Nitrate
→ Domestic Biomass Burning
→ Agricultural Burning (Open Burning)

CMB Source profiles
=
CAMx emission profiles

SPECIATE (EPA)
ARPA Lombardia
AMAT-MI

The CAMx modelling system



CAMx/PSAT

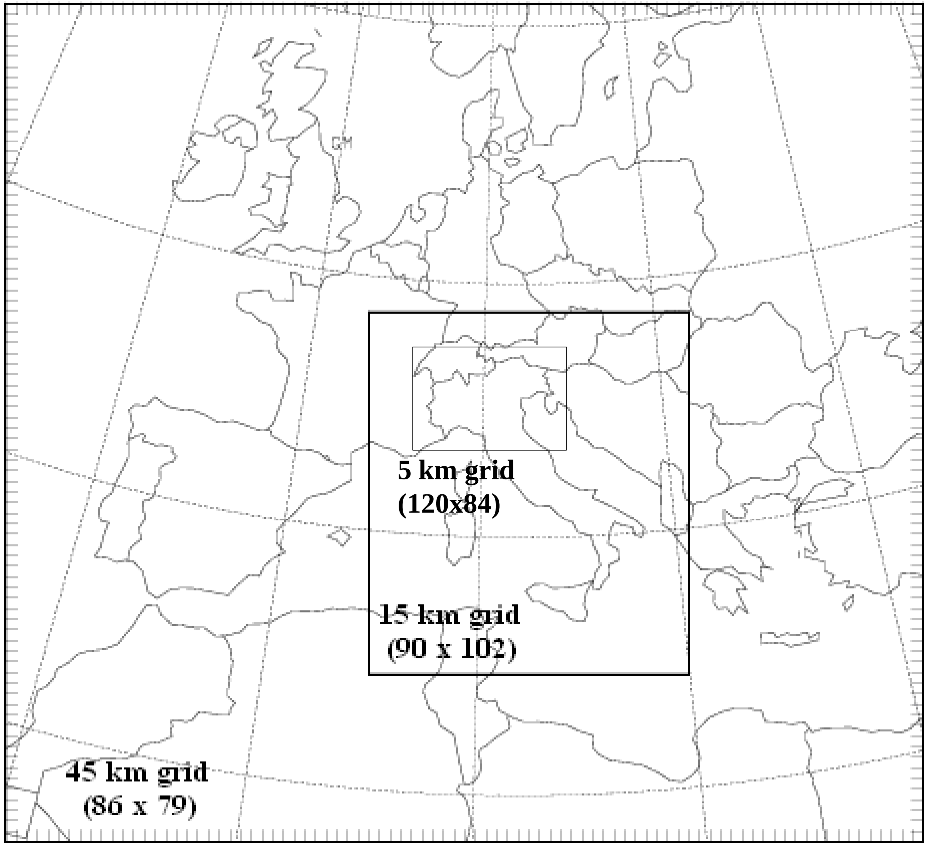
PSAT - Particulate Source Apportioning Technology (Yarwood et al., 2004)

PSAT uses reactive tracers to apportion primary PM, secondary PM ***and gaseous precursors to secondary PM*** among different source categories and source regions.

- detect the role played by the different emission sources
- investigate, source by source, the influence of non linear chemical system on the evolution of the PM concentration

A similar technology has been developed for ozone (OSAT).

Modelling system set up



WRF

WRF v.3.2.1 working over 3 domains

1) European domain - 45 Km resolution
2) Italian domain - 15 Km resolution
3) Po valley domain – 5 km resolution

ECMWF fields as initial and boundary conditions

Emissions

- Local emission inventories for Lombardy, Piemonte, Veneto and Emilia Romagna
- Italian official emission inventory for 2005
- EMEP emission inventory for 2005 outside Italy

CAMx domain e temporal coverage

CAMx 5.4 working over 2 domains

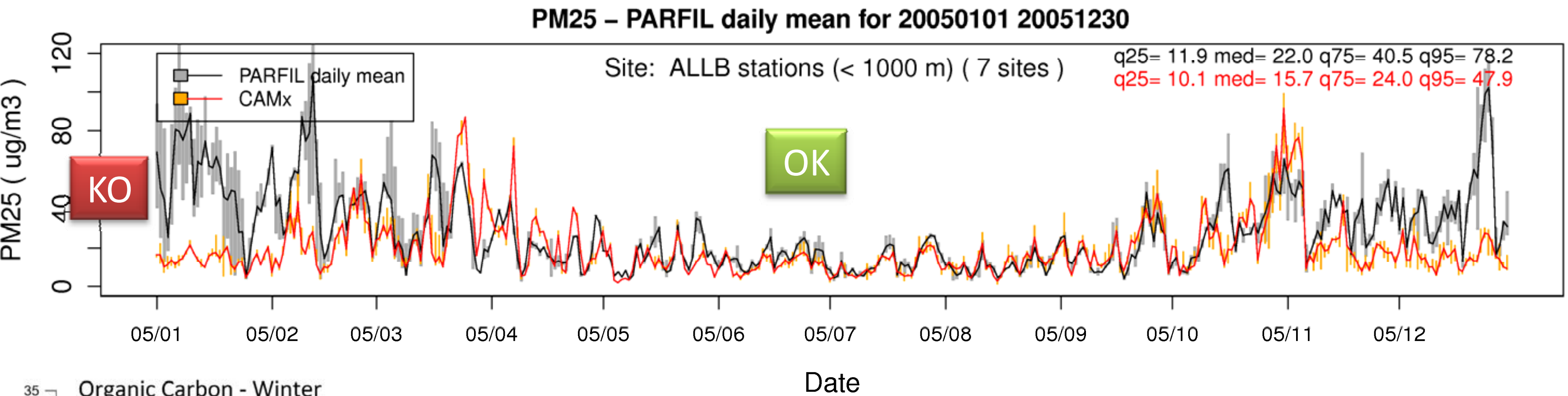
1)Italy 86 x 98 cells – 15 Km spatial resolution)
2)Po valley 116x80 cells - 5 km resolution

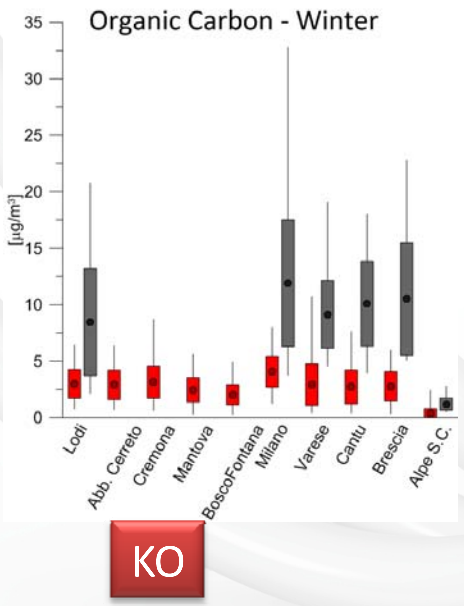
Calendar year **2005**

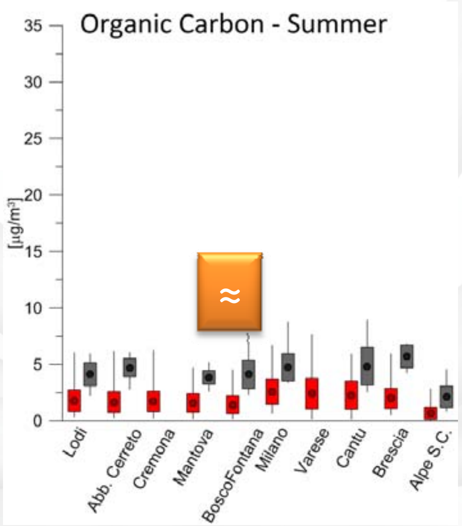
PSAT set up

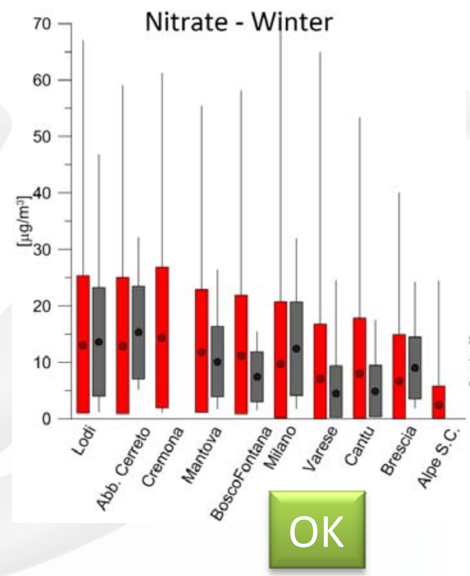
1 source area (whole domain)
28 source categories (Transport, energy, agriculture, biomass burning,...)

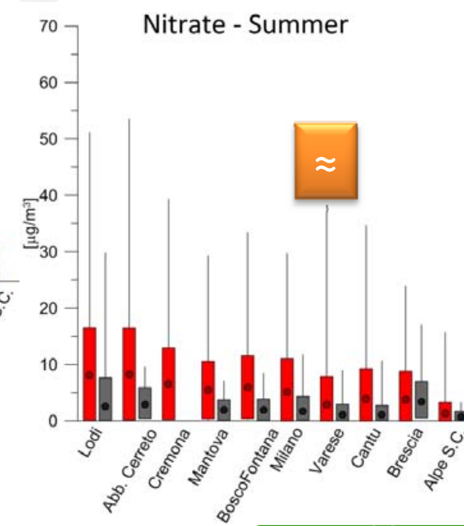
CAMx performance - $PM_{2.5}$



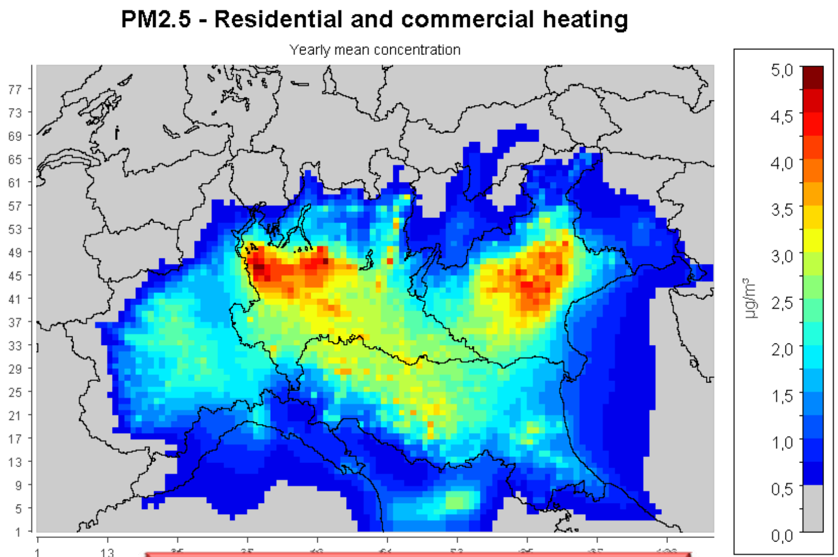


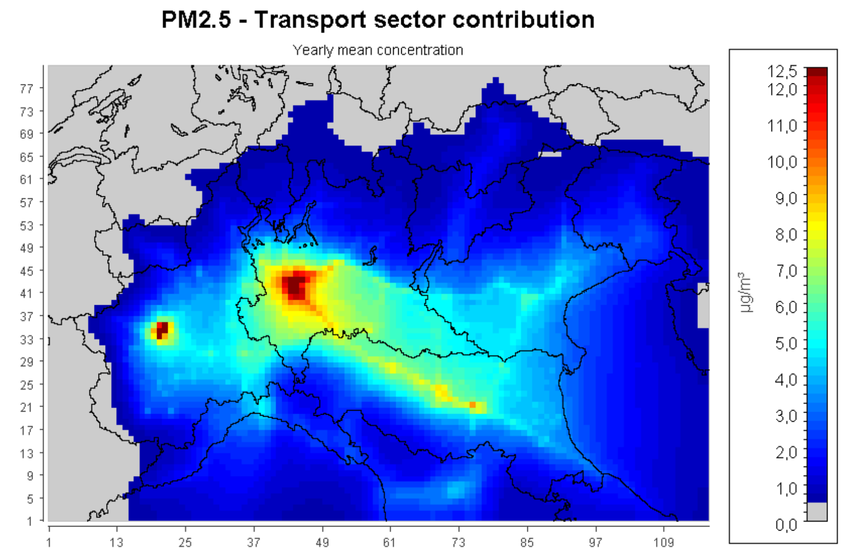






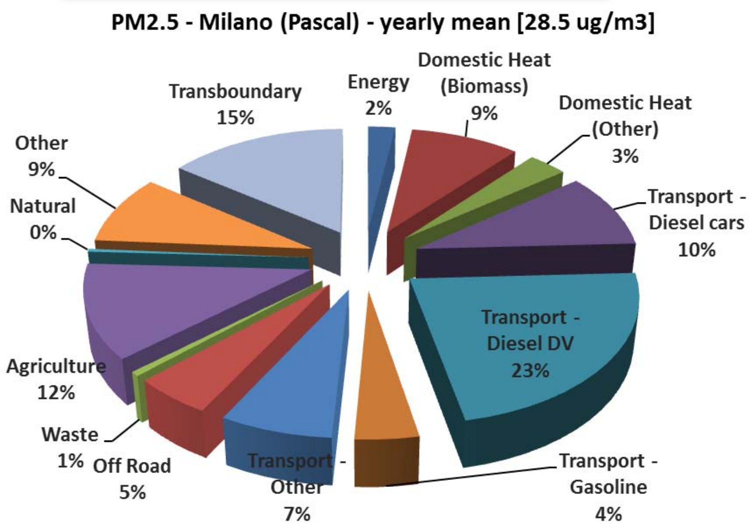
CAMx Source Apportionment – $PM_{2.5}$

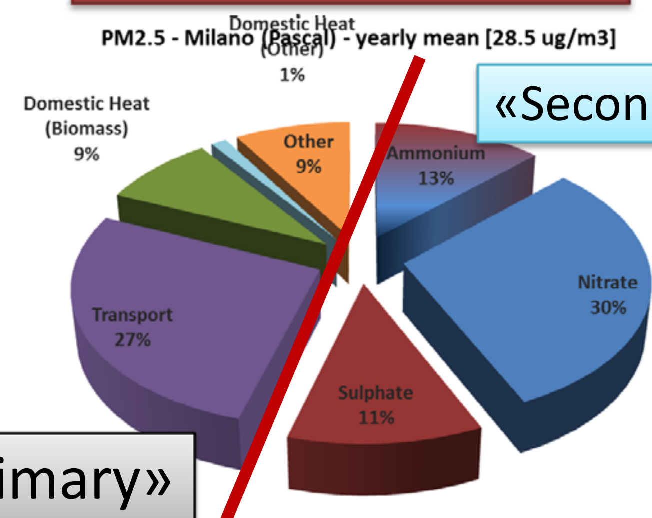




PSAT Classification

CMB Classification

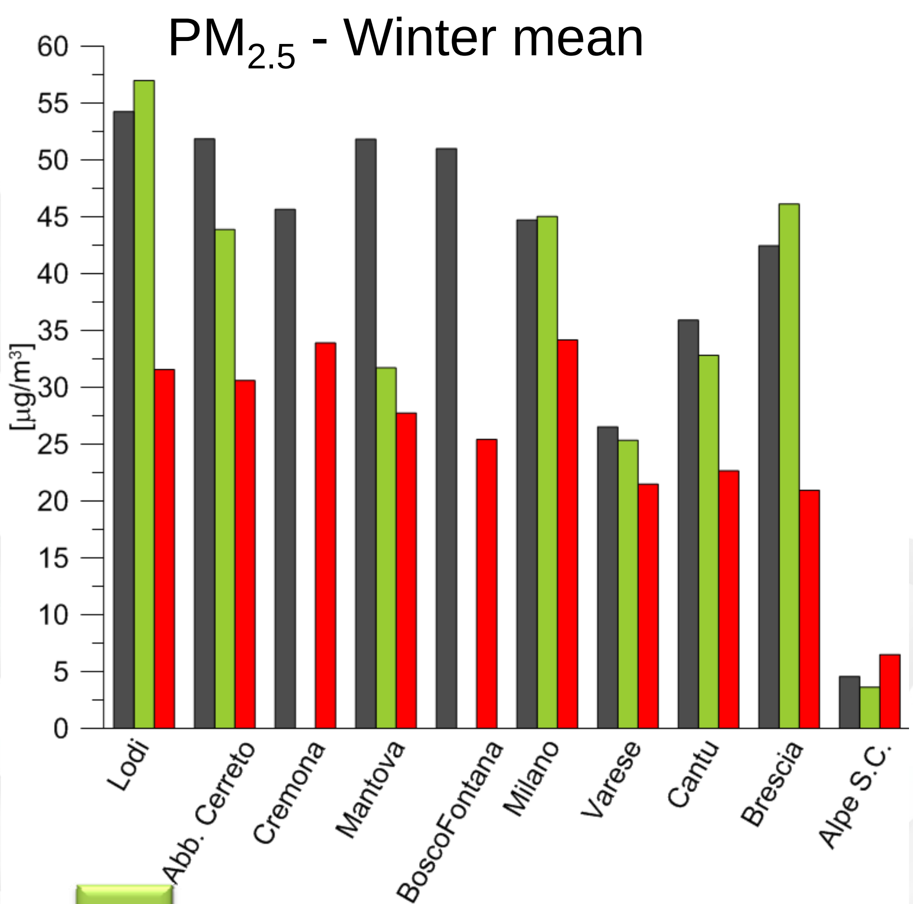


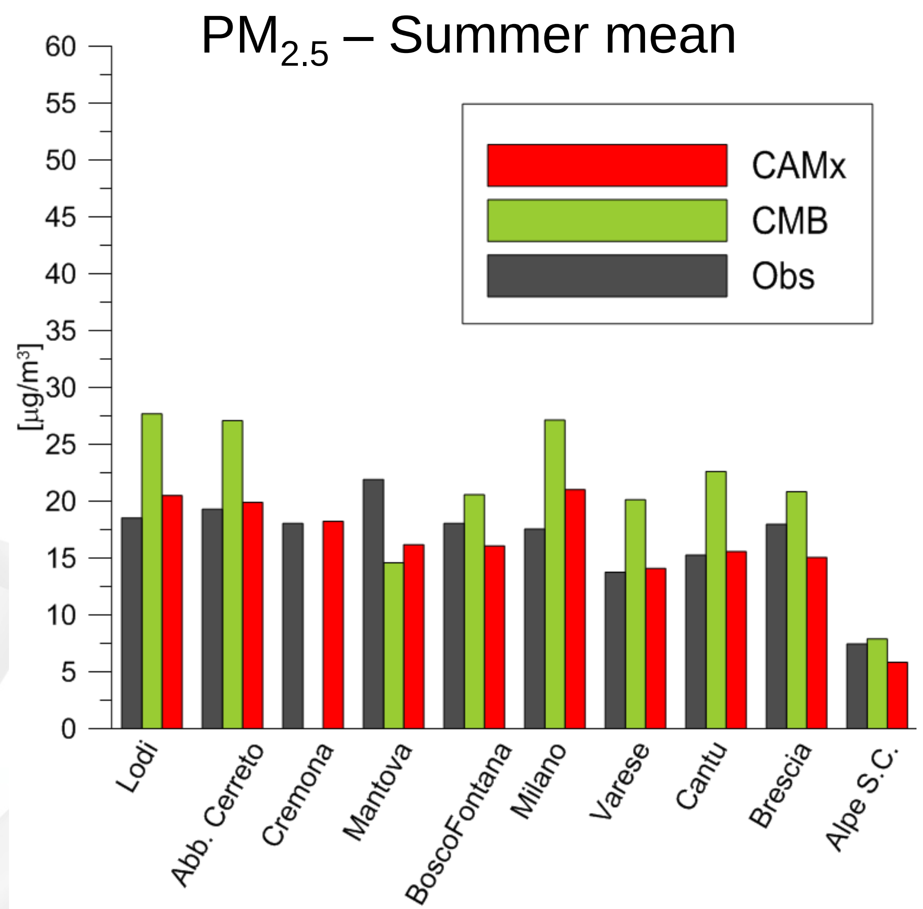


«Secondary»

«Primary»

Mass Closure





OK CAMx in summer – CMB in winter

KO CAMx in winter

≈ CMB in summer

Seasonal analysis

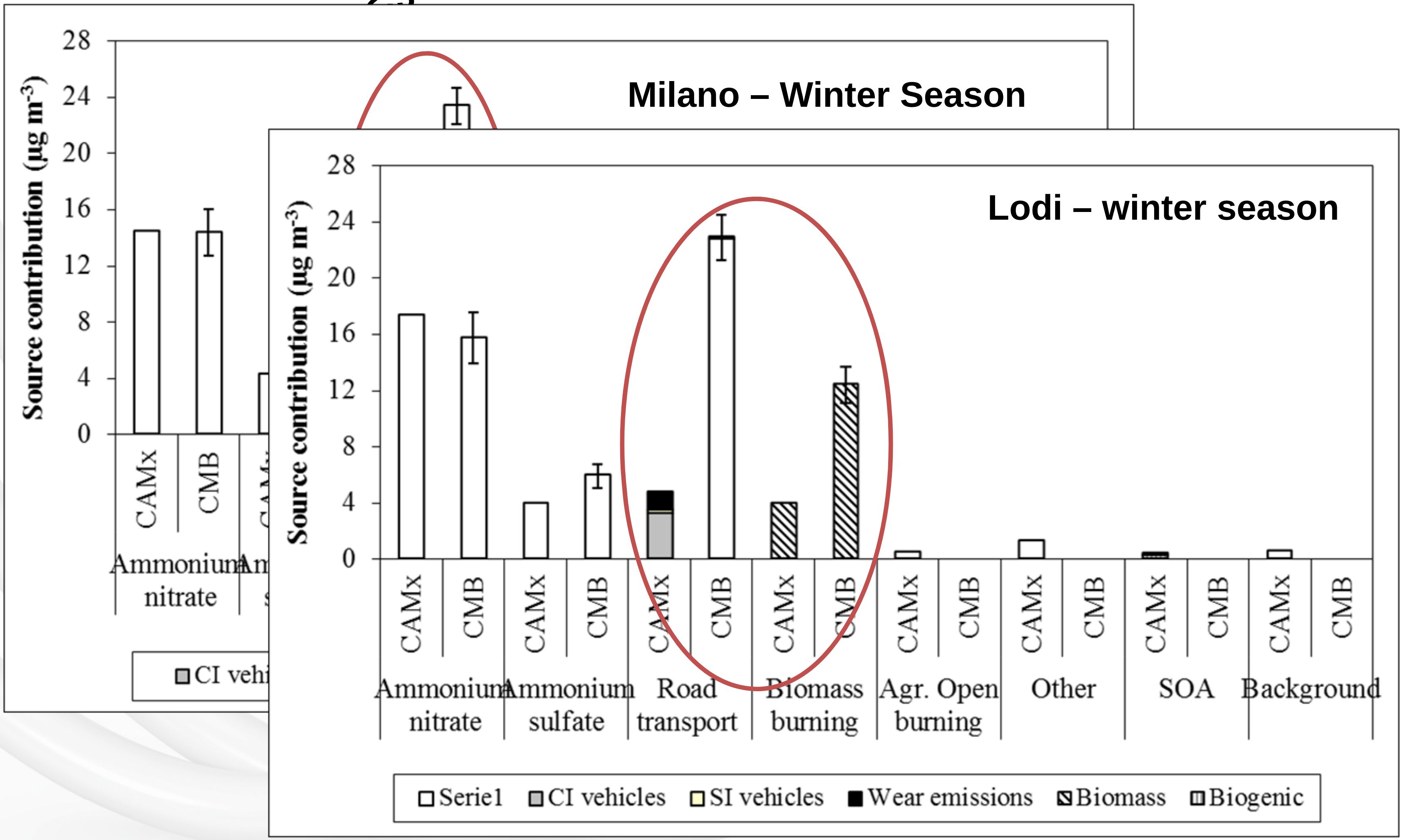
$PM_{2.5}$ - Seasonal mean concentration

Emission Category	Seasonal mean at site ($\mu g\ m^{-3}$)		
	CMB		PSAT
Ammonium Sulphate	3.4 ± 1.8 3.2 ± 2.1	OK	4.0 ± 0.5 3.6 ± 0.6
Ammonium Nitrate	2.3 ± 1.0 10.2 ± 6.2		6.7 ± 2.8 12.4 ± 4.6 ≈
Road Transport	9.4 ± 9.2 10.7 ± 10.1		2.4 ± 1.6 3.9 ± 2.7
Biomass Burning in D.H.	4.2 ± 4.5 10.1 ± 10.6	KO	0.8 ± 0.3 4.2 ± 1.2
Combustion in Agriculture	1.6 ± 2.3 1.4 ± 3.9		0.2 ± 0.1 0.3 ± 0.2

Summer **Winter**

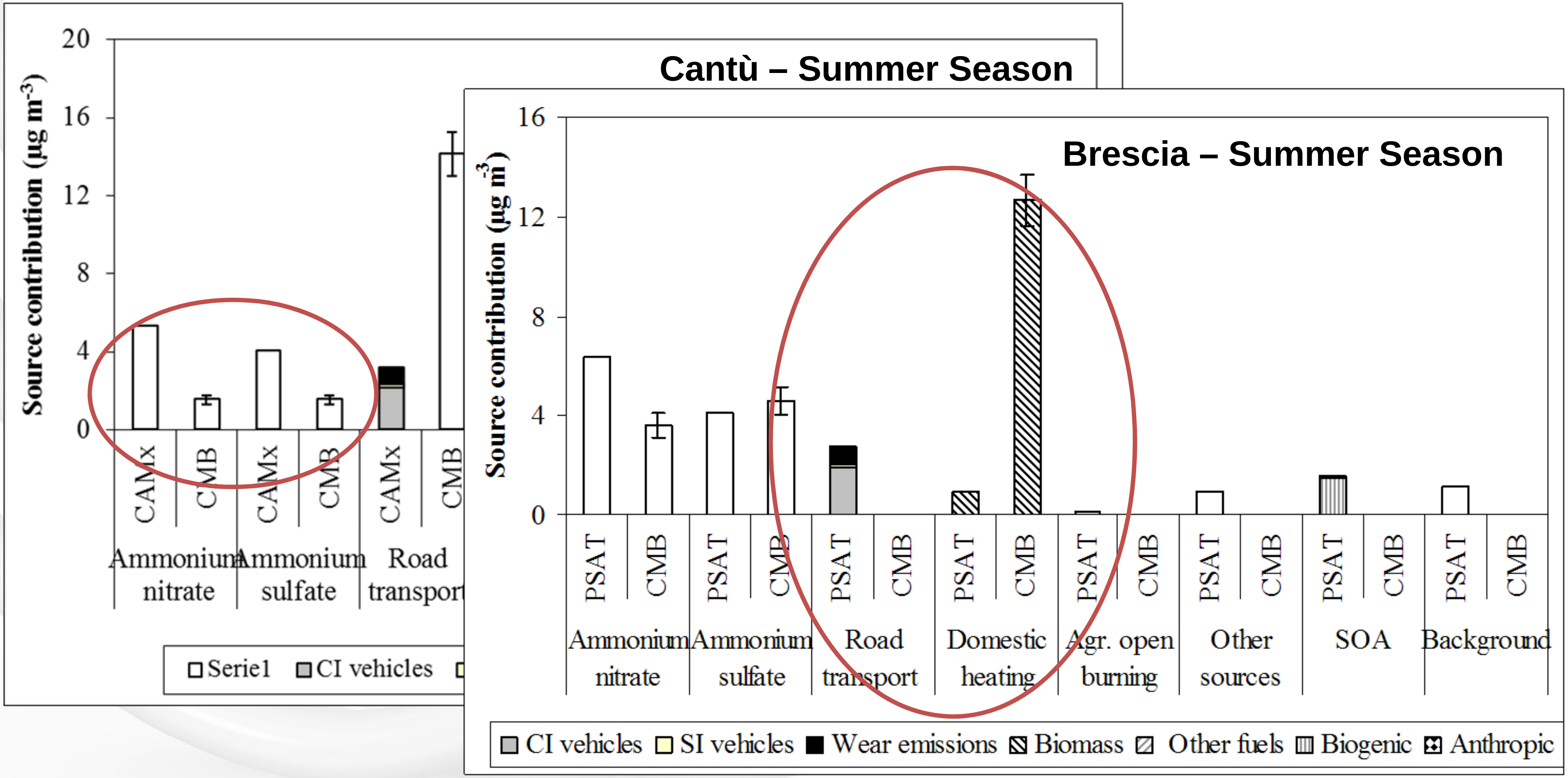
Receptor analysis

$PM_{2.5}$ - Winter mean concentration



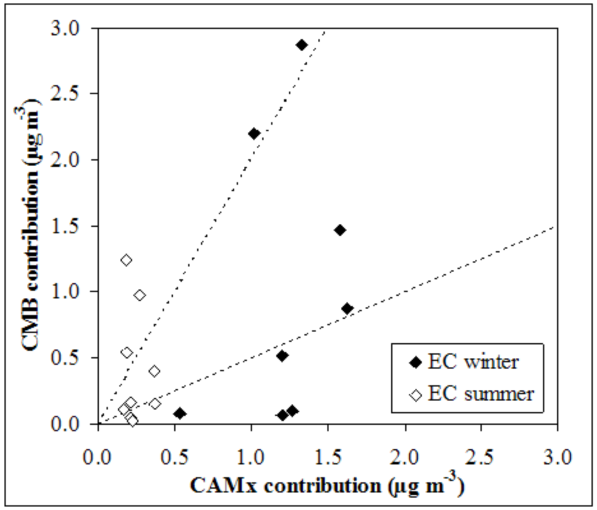
Receptor analysis

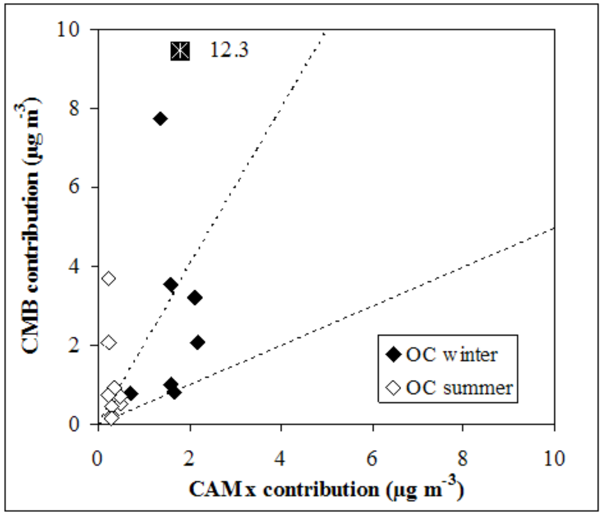
$PM_{2.5}$ - Summer mean concentration



Comparison of CAMx and CMB Source Contribution Estimates

Domestic Heating

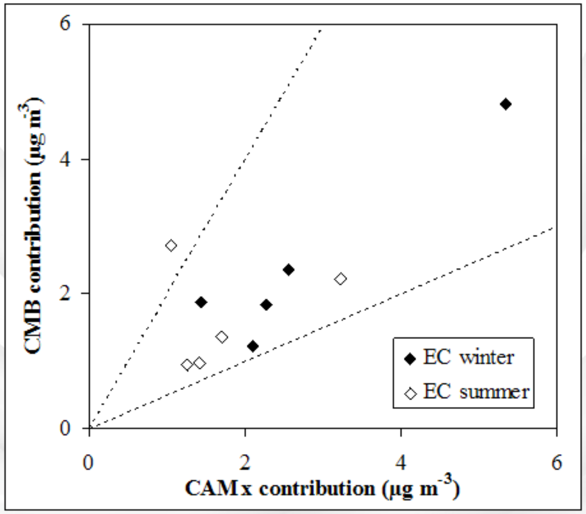


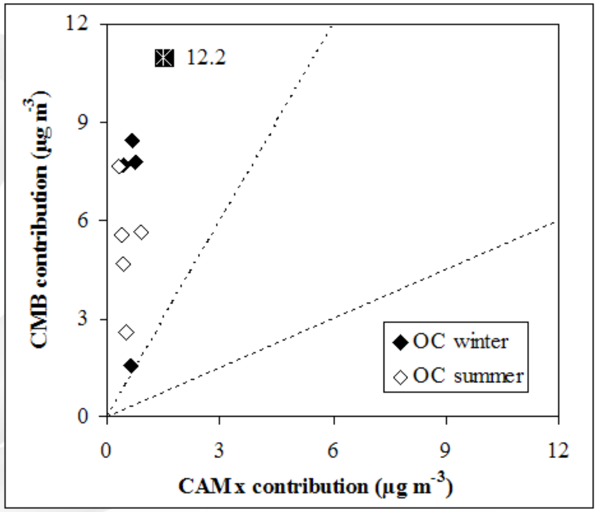


Winter ◆

Summer ◇

Transport





EC **OC**

Conclusions (1)

- ***Emission speciation profile*** (CTM) vs ***Source profile*** (RM): the two information overlap in case of non-reactive species, while they can differ a lot in case of secondary organic species

- ***Mass*** oriented approach (CTM) vs ***Marker*** oriented approach (RM): CTMs are more skilful in reproducing sources providing a strong signal in term of mass than low emitting sources, whereas RMs can capture also the contribution of low emitting sources, if they show a distinct marker pattern.

- The **effectiveness** of RMs approach is strongly dependent on the **uncertainty** associated to the observed dataset that in some cases can drive the RMs toward unexpected and questionable answers, that can be easily pointed out through the **comparison** against CTMs results.

Conclusions (2)

- In this study **CMB** showed a **better** reconstruction of the **PM2.5 mass** closure, while **CAMx** systematically **underestimated** cold season concentrations.

- Nevertheless both models provided the **same ranking** for SCEs at several receptors.

- The most relevant **discrepancies** were related to **road transport and domestic heating**, whereas models generally **agreed** in the reconstruction of **secondary inorganic aerosol** contributions.

- **CAMx/PSAT** approach can provide **additional** information on source apportionment:
  - Correlated sources (Diesel and gasoline...) and mixed sources
  - SOA
  - Source regions (e.g transboundary)

- Deeper investigation of the possible causes of CAMx failures (e.g. **organic carbon from transport** during winter)

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Authors are grateful to M. Bedogni (AMAT-MI) for having kindly shared information about road transport emission profiles.

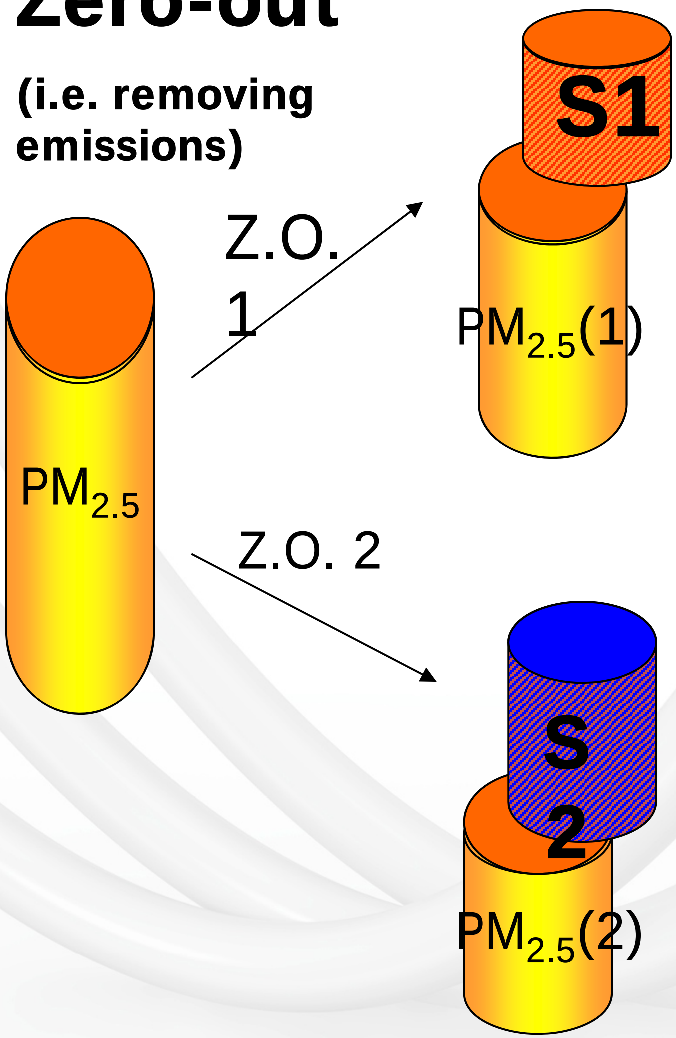
The authors are thankful to all people that performed the PARFIL campaign.

Thanks for your attention

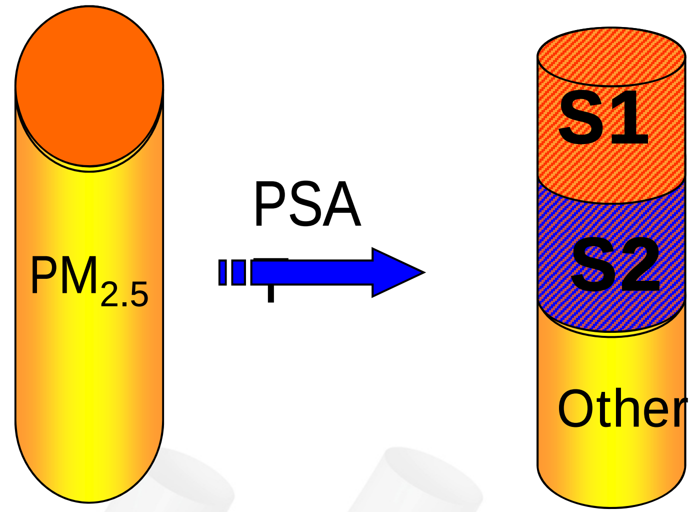
CAMx/PSAT

Zero-out

(i.e. removing emissions)



PSAT



PSAT Advantages

- Time saving (one simulation)
- Mass consistency
- Fully traceable

CAMx/PSAT

Reactive tracers approach: sulphur *source apportionment*

Tracer concentration in each cell = Emission+Transport+ chemical transformation...

CAMx
Total rate

TRACERS

WEIGHTS

1. **Emission**

$$[TSO_{2i}(t+\Delta t)]_{em} = TSO_{2i}(t) + \frac{E_{SO_2}(t)}{V} \cdot \frac{e_{SO_{2i}}(t)}{\sum_i e_{SO_{2i}}(t)}$$

2. **Transport**

$$[TSO_{2i}(t+\Delta t)]_{tr} = [TSO_{2i}(t)]_{em} + [\Delta SO_2]_{tr} \cdot \frac{[TSO_{2i}^{in}(t)]_{em}}{\sum_i [TSO_{2i}^{in}(t)]_{em}}$$

3. **Chemistry**

$$[TSO_{2i}(t+\Delta t)]_{chem} = [TSO_{2i}(t)]_{tr} + [\Delta SO_2]_{chem} \cdot \frac{[TSO_{2i}(t)]_{tr}}{\sum_i [TSO_{2i}(t)]_{tr}}$$

CAMx/PSAT

Reactive tracers approach - **Sulphate**

Sulphate formation
(chemistry step in cell I,J,K)

$SO_2 \rightarrow PSO_4$

CAMx
Total rate

- SO_2 removal

$$SO_{2i}(t+\Delta t) = SO_{2i}(t) + \Delta SO_2 \frac{SO_{2i}(t)}{\sum SO_{2i}(t)}$$

TRACERS

WEIGHTS

- Sulphate production

$$PS_{4i}(t+\Delta t) = PS_{4i}(t) + \Delta PSO_4 \frac{SO_{2i}(t)}{\sum SO_{2i}(t)}$$

Mass Balance

$$SO_2 = \sum_i SO_{2i}$$

$$PSO_4 = \sum_i PS_{4i}$$

CAMx/PSAT - Emission categories

CAMx emission category	SNAP category	Fuel	PSAT classification	CMB classification
Power Plants	01	Mix	Other sources	
Power Plants	01	Biomass	Other sources	
Power Plants	01	Other	Other sources	
Energy production in industrial activity (except power plants)	01	Mix	Other sources	
Energy production in industrial activity (except power plants)	01	Biomass	Other sources	
Energy production in industrial activity (except power plants)	01	Other	Other sources	
Domestic and commerical heating	02	Other	Domestic heating - Other fuels	
Domestic and commercial heating	02	Biomass	Domestic heating -Biomass burning	Domestic heating - Biomass burning
Industrial combustion	03	Other	Other sources	
Industrial combustion	03	Biomass	Other sources	
Road transport - cars	07-01	Mix	Road transport - SI vehicles	Road transport
Road transport - cars	07-01	Gasoline	Road transport - SI vehicles	Road transport
Road transport - cars	07-01	Diesel oil	Road transport - CI vehicles	Road transport
Road transport - cars	07-01	LPG-Natural gas	Road transport - SI vehicles	Road transport
Road transport - cars (tire and break wear)	07-01		Road transport - Wear emissions	Road transport - Wear emissions
Road transport (except 0701)	07	Mix	Road transport - SI vehicles	Road transport
Road transport (except 0701)	07	Gasoline	Road transport - SI vehicles	Road transport
Road transport (except 0701)	07	Diesel oil	Road transport - CI vehicles	Road transport
Road transport (except 0701) (tire and break wear)	07		Road transport - Wear emissions	Road transport - Wear emissions
Evaporation	07		Transport - SI vehicles	Road transport

CAMx/PSAT - Emission categories

CAMx emission category	SNAP category	Fuel	PSAT classification	CMB classification
Off road	08	Mix	Road transport - CI vehicles	Road transport
Off road	08	Gasoline	Transport - CI vehicles	Road transport
Off road	08	Diesel oil	Transport - CI vehicles	Road transport
Waste treatment	09		Other sources	
Agriculture	10		Agricultural open burning	Agricultural open burning
Natural emissions (without Sea Salt and Biogenic VOCs)	11		Background	
Sea Salt and Biogenic VOCs	11		Background	
Long range transport	-		Background	
Anthropogenic SOA	-		SOA - Anthropic	
Biogenic SOA	-		SOA - Biogenic	
Other	-		Other sources	

CMB8.2 - Performance

Diagnostic parameter	Cold season	Warm season	Target values
R^2	0.21-0.82 (0.58 ± 0.22)	0.23-0.83 (0.58 ± 0.20)	> 0.8
X^2	15-73 (36 ± 20)	14-77 (38 ± 19)	<1 (good fit) 1 – 2 (acceptable fit)
Percent mass	70-128 (97 ± 19)	72-149 (125 ± 26)	80% - 120%
SCEs t-statistics	always > 2	always > 2	> 2

Milan area and Lombardy region

ARPA Lombardia dataset available for this exercise

Site: Milano-Pascal, urban background

Sampling period: 01.01-31.12.2012

Daily PM10 concentrations (359 data) and composition:

- Elements (246 data), Z>11: Al, Si, S, Cl, K, Ca, Ti, Cr, Mn, Fe, Ni, Cu, Zn, Br, Pb, Rb
- OC-EC-TC-BC (303 data): NIOSH-Like protocol applied
- PAH's (122 data, 60 data >LOD): B(a)P, B(a)A, B(b)F, B(j)F, B(k)F, I(1,2,3,c,d)P, dB(a,h)A
- Potential availability of Ions and Levoglucosan
- Shorter campaigns in other Lombardy region sites might be available too
- Possible comparison against observation at Ispra EMEP site