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APICE

Source contribution analysis by receptor models and chemical transport models

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Noemí Pérez and Jorge Pey, CSIC – IDÆA
Pedro Jiménez, Universidad de Murcia
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1. Source Apportionment analysis

The contribution of the different emission sources – both anthropogenic and natural – to the Particulate Matter concentrations – has been highlighted by two different approaches: the receptor models and the Chemical Transport Models.

The two different techniques of Source Apportionment have been applied at the same time in the five cities in order to answer to these questions:

• which pollutant emission mostly affects PM10 and PM2.5 concentrations?

• which is the weight of the presence of the port in the studied cities in terms of PM10 and PM2.5 concentrations?

The two different Source Apportionment (SA) approaches aimed at integrating the peculiar potentialities of both techniques: by one side receptor models, more suitable to pointing out specific emission sources bind to specific markers, and, on the other side, CTMs, which extend their assessment on the formation of secondary aerosols, since they apportion the gas precursor emissions, too. Moreover, while receptor models give SA outcomes on some monitoring sites only, SA by CTMs provides outcomes on the whole studied territory with a certain resolution (spatial maps).

2. Source Apportionment Outcomes by Receptor Model

Receptor Models aim to re-construct the contribution of emissions from different sources of atmospheric pollutants, e.g., particulate matter (PM), based on ambient data (i.e. PM elemental and chemical composition) registered at monitoring sites. The fundamental principle of receptor modelling is that mass and species conservation can be assumed and a mass balance analysis can be used to identify and apportion sources of airborne PM in the atmosphere. One of the main differences between models is the degree of knowledge required about the pollution sources prior to the application of receptor models. A second major difference between these different approaches is the number of observations (e.g., samples) needed to apportion sources. While Chemical Mass Balance (CMB) model assumes and needs an a-priori knowledge of the emission sources and could be used with only one sample, approaches such as Positive Matrix Factorization (PMF) need a significant number of samples (at least equal to the number of Chemicals species included in the model) to single out the emission sources active in a particular area and to provide statistically sound results. PMF (in Barcelona, Genoa, Thessaloniki and Venice) and CMB (in Marseille) are the two approaches adopted by the APICE Partners.

In Barcelona, simultaneous sampling was carried out in two sites every four days during one-year, from February 2011 to January 2012: Port of Barcelona (41°19’58”N; 2°8’27”E) and

Palau Reial (urban background site, 41°23'15"N; 2°6'56"E). A PMF analysis was performed on 295 cases, including simultaneous PM10 and PM2.5 measurements performed at both monitoring sites.

Barcelona, average results:

Sources	Port: Contribution (µg/m^3) to		Palau Reial: Contribution (µg/m^3) to	
	PM10	PM2.5	PM10	PM2.5
Industrial emissions	1.8 ± 0.1	0.3 ± 0.1	0.8 ± 0.1	0.3 ± 0.0
Mineral/road dust	9.2 ± 0.5	2.1 ± 0.2	3.3 ± 0.3	0.4 ± 0.1
Ammonium sulphate	2.9 ± 0.6	7.9 ± 0.9	4.8 ± 0.7	9.9 ± 0.9
Fuel oil combustion	3.8 ± 0.3	1.1 ± 0.2	2.8 ± 0.2	0.9 ± 0.1
Vehicle exhaust emissions	6.6 ± 0.8	5.3 ± 0.5	2.6 ± 0.4	2.9 ± 0.3
Aged sea spray + nitrate	12.1 ± 1.0	9.2 ± 0.8	2.0 ± 0.3	2.3 ± 0.4
Unaccounted	0.0	0.8	1.2	1.6

The biggest differences between the port and the urban area of Barcelona were found for the mineral dust source, attributed to the influence of dust re-suspension from the new port area under construction, but also to re-suspension of road dust from the intense truck traffic around the port area. The fuel oil combustion source was also higher for the port of Barcelona, reflecting direct emissions from shipping. The contribution of the industrial emissions was also higher at the port area. This is attributed to the transport of pollutants from the industrial area in the surroundings of Barcelona. The aged sea spray+nitrate source was also higher at the port area. This is not a single source but reflects the mixing of different emission sources and subsequent aging of pollutants during the transport of the air masses. However, the ammonium sulphate source was much higher at the urban area of Barcelona. This is attributed to the formation of secondary ammonium sulphate from SO2 shipping.

The results show that the contribution of port emissions to PM10 and PM2.5 at the port were around 40% for both PM10 and PM2.5, being mainly attributed to mineral dust (23 and 17% for PM10 and PM2.5, respectively) and fuel oil combustion (10 and 16%, respectively). Vehicle exhaust emissions accounted for 3% in both fractions, and ammonium sulphate for 2 and 6%, respectively.

At the urban area of Barcelona the contributions from the port were 11% and 18% for PM10 and PM2.5, respectively. The influence of the port in the urban background of Barcelona is mainly attributed to fuel oil combustion (4-5%) and ammonium sulphate (6 and 12%, respectively) from the formation of secondary ammonium sulphate during transport of SO2 emissions from the port to the urban background site.

It is important to highlight the formation of secondary aerosols in the urban area of Barcelona, from the gaseous precursors SO2, transported from the port, and the high levels of NH3 measured at the urban background.

3. Source Apportionment Outcomes by Chemical Transport Model

Source Apportionment analysis by CTMs has been performed using the zero-out modeling technique by the groups running CHIMERE (Barcelona and Marseille) whereas for CAMx the specific PSAT tool has been applied (Genoa, Venice, Thessaloniki and one again Marseille). The zero-out method sets to zero a specific emission on the original emission inventory and measures the change in the concentration output; a complete model run is required for each source or emission sector under investigation. Particulate Source Apportionment technology (PSAT) uses reactive tracers to apportion primary PM, secondary PM and gaseous precursors to secondary PM among different source categories and source regions.

In Barcelona, source apportionment for PM10 and PM2.5 has been evaluated by CHIMERE zero-out method for both summer (August 2011) and winter (December 2011) periods. The maritime contribution analysis has been calculated by the zero-out method applied on the Other Mobile Sources (SNAP 8) in which port emissions are included.

SA outcomes are here discussed for three sites: an urban site in Barcelona downtown and two sites near the Port: the World Trade Center, which can be considered as a port background site and a second site located at the very heart of the port of Barcelona.

All the three sites present exceedances of the daily PM10, both during summer and winter periods, with higher concentration both for PM10 and PM2.5 during summer than at wintertime, indicating the importance of secondary formation in PM levels in the city of Barcelona. The highest concentrations are recorded at the site at the very heart of the port of Barcelona (Table 1).

Table 1: PM10 and PM2.5 concentrations recorded at 3monitoring sites in Summer and winter period

Site	PM10 (µg/m3) Summer	PM10 (µg/m3) Winter	PM2.5 (µg/m3) Summer	PM2.5 (µg/m3) Winter
Barcelona downtown	40.5	24.0	19.2	14.2
World Trade Center (WTC)	52.2	40.2	24.6	24.6
Inner Port (POR)	69.4	55.2	43.2	29.4

In all the three sites, Source Apportionment outcomes for PM10 and PM2.5 are slightly different but not so much to give different ranking in the contribution analysis (Tab.2-Barcelona). At summertime the most important contributors at the various sites are the following:

• urban site: on-road transport, followed by the maritime sector (included in other mobile sources);

• World Trade Center site: maritime sector, followed by on-road transport. Here, also the boundary conditions and biogenic sources have a relevant weight on PM10 and PM2.5 concentrations;

• inner port site: the maritime sector, dominates with over 50%, followed by on-road transport.

Here, the external contribution is reduced from both the urban and the WTC sites, indicating the important local contribution of emissions to air quality.

At wintertime the most important contributors at the various sites are the following:

• urban site: the contribution from outside of the domain through the boundary conditions and combustion in manufacturing industry (SNAP 3); on road traffic loses importance, as well as the maritime sector;

• World Trade Center site: the maritime sector followed by on-road transport, with the weight of combustion in manufacturing increased conversely to biogenic contributions in respect to summertime;

• inner port site: the maritime sector still dominates but with a less important weight then summertime (38%); the second contributor is the combustion in manufacturing processes.

Table 2: SA from CHIMERE during summer (August 2011) and winter (December 2011) at 3 Barcelona sites.

PM2.5 Source Apportionment (% on total concentrations)	Urban site		World trade center		Inner port	
	summer	summer	summer	winter	summer	winter
Boundary conditions	6.3%	14.3%	8.3%	8.9%	5.5%	7.7%
Road Transport	20.5%	8.5%	18.6%	18.1%	11.7%	8.3%
Other mobile sources (including Maritime/Harbor emissions)	16.7%	4.5%	28.4%	23.2%	53.9%	38.1%
Non-industrial combustion	6.9%	3.5%	5.6%	6.5%	4.6%	3.6%
Energy production and Industries	7.4%	8.9%	7.0%	9.3%	5.8%	16.1%
Agriculture	2.5%	1.3%	2.4%	1.2%	1.4%	1.2%
Biogenic sources	5.2%	3.1%	7.2%	2.5%	4.9%	2.0%
Others	34.5%	55.9%	22.5%	30.3%	12.2%	23.0%

Focusing on the other mobile sources sector, which includes maritime emissions, the mean contribution on PM2.5 among the three sites varies between 17% at the urban sites and 54% inside the port area during summertime, whereas in winter this contribution decreases to 5% at the urban site and 38% at the port. This contribution takes into account not only the emissions from ship and vessels, but considers all the emissions coming from the SNAP 8 (other mobile sources), comprehending all on shore port activities.

Very similar results are recorded for the PM10 source apportionment (between 16% and 52% in summer and between 7% and 41% in winter). The mean contribution is rather constant throughout the year in the entire domain (approx. 7-9%), but a strong seasonality

can be found at the urban site (16-17% in summer vs. 5% in winter). The minimal contributions are lower than 1% for both summer and winter period at the scale of the APICE domain.

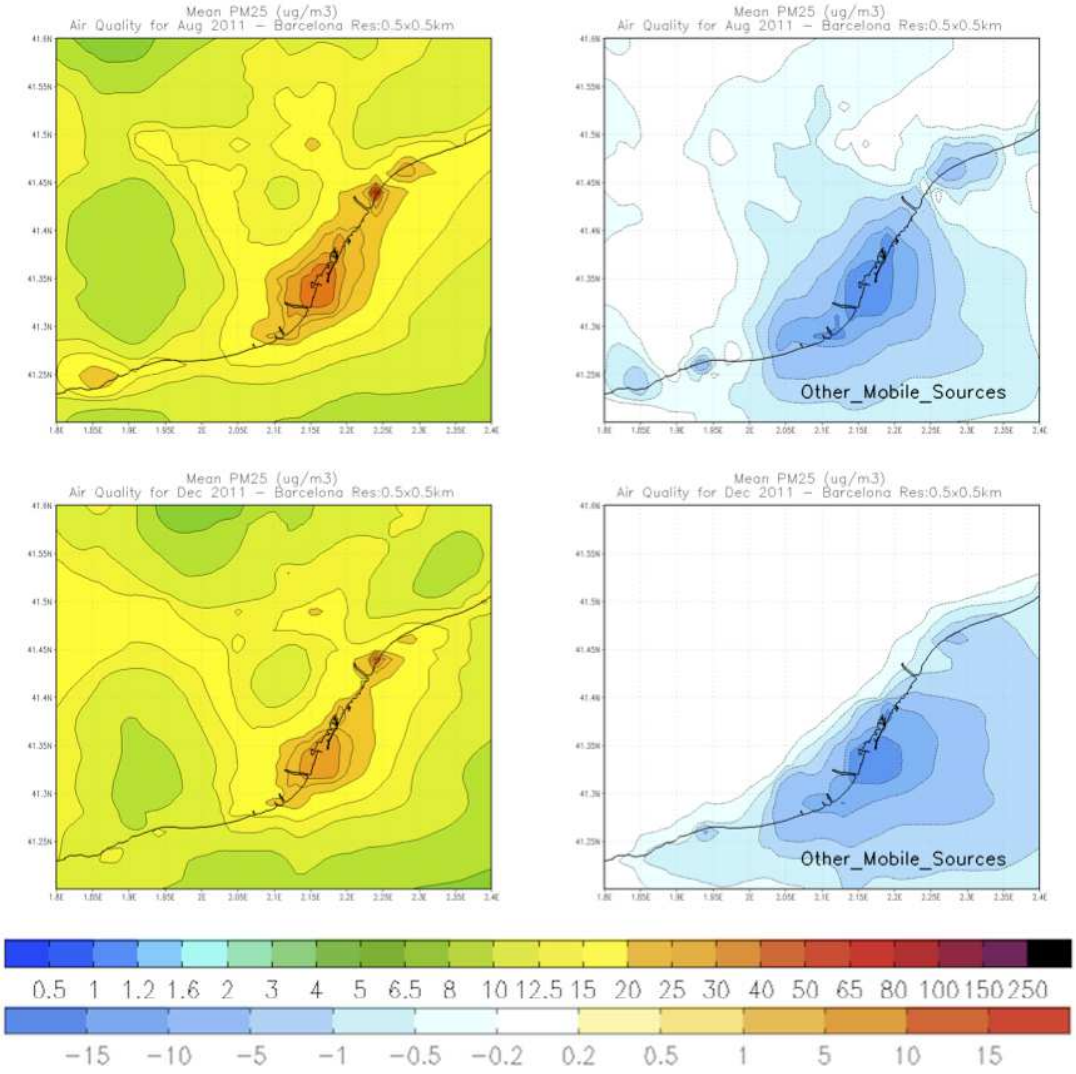

Figure 1: Monthly PM2.5 concentrations (µg m-3) (left) Turing the summer (top) and winter (bottom) periods at the Barcelona APICE domain scale from CHIMERE model and contributions for maritime sector (right) (obtained by zero-ing out the SNAP8 sector).