PARTICULATE SOURCE APPORTIONMENT USING TWO CHEMICAL TRANSPORT MODELS OVER FRENCH SOUTH EASTERN COASTAL AREA

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Abstract: Over French south eastern Mediterranean coast, two numerical models have been used to evaluate source apportionment for particulate matter at the regional scale with two different approaches: zero-out modeling with CHIMERE model and tracer approach with CAMx model. Both winter and summer periods with distinct emissions and meteorological conditions have been investigated. In the same time, intensive air pollution monitoring campaigns have been carried on downtown in Marseille (France) to characterize sources pollution from receptor models. Between two numerical approaches, some differences have been noted due to non-linear systems, leading to an overestimation of local sources with the zero-out modeling. For the winter period, road traffic, industrial-energy and residential-tertiary sectors display significant contributions inside the large cities. During summer, natural emissions display significant contributions in a large part of the region but anthropogenic contributions with road traffic and industrial-energy sectors remain important contributions inside large cities. Long-range transport also displays an important contribution for both seasons. Comparison between numerical and receptor model results downtown in Marseille during the winter period have shown a good agreement.

Key words: *Source apportionment, particulate matter, aerosols, CHIMERE, CAMx, receptor models,*

INTRODUCTION

As several French areas display high pollution concentration above the European limit values, discussions are in progress to design efficiency action plans in aim to reduce particles concentrations. The first step of these discussions is to know the relative contributions of each emission source to determinate the major activity sectors involved in the pollution events. Currently, the contributions of activities sector are evaluated in terms of emission inventory at national and regional scales. However, due to the non-linear systems between emissions of primary particles and total concentrations, due to several factors as chemical reactions or meteorological conditions, the source contributions are not directly apportioned. Nevertheless, this estimation could be considered as a decisive element to design efficiency actions.
In the framework of the APICE project (Common Mediterranean strategy and local practical Actions for the mitigation of Port, Industries and Cities Emissions), financed by the European Program for Territorial Cooperation MED 2007/2013, an international collaboration has studied the particulate source apportionment inside five large Mediterranean cities: Barcelona, Genoa, Marseille, Thessaloniki and Venice. The scientific partnership has assessed the contribution of the whole pollutant emissions activities with a special focus on maritime activities based on monitoring campaigns and numerical approaches.
In this study, two chemical transport models (CTMs) have been updated over the French south eastern area to evaluate air quality and source apportionment for PM_{10} and $PM_{2.5}$. The first model is CHIMERE (Bessagnet et al., 2009) with an adaptation of the zero-out method and the second one is CAMx (ENVIRON, 2011) using a tracer approach.

DATA AND METHOD

Description of the modeling system

The modeling system used for this study includes three nested simulation domains (Figure 1). The first domain extends over a large part of Europe with a grid resolution of 27 km. The second domain includes the south-eastern part of France and the northern part of the Mediterranean Sea with a grid resolution of 9 km and the last one covers the PACA region with a grid resolution of 3 km. These three domains are used to compute meteorological fields, gridded emissions, boundary and initial conditions and other specific input required by chemical-transport models.
Meteorological fields are provided by the Weather Research and Forcasting (WRF) model v3.3.1 (Skamarock et al., 2008) for the three nested domains with a 1 hour temporal resolution. Initial and boundary conditions required for the first domain are provided by the National Centers for Environmental Prediction (NCEP) issue from the Global Data Assimilation System (GDAS).

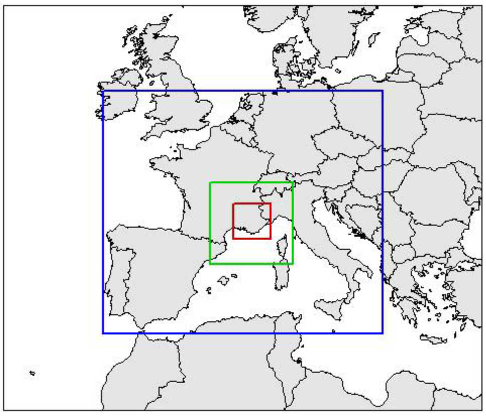

Figure 1 : Spatial extent of three domains using in the simulation chain.

For the two large domains, anthropogenic emissions are supplied by the European Monitoring and Evaluation Programme (EMEP) for the reference year of 2007 with a spatial resolution of 50 km. Emissions are downscaled according to the land cover with the spatial resolution of the related domain. A local emission inventory calculated at the regional scale for the reference year of 2007 with an initial spatial resolution of 1 km is used for the finest domain. This local inventory is gridded over the grid of the last domain. For this finest domain, point emissions are provided for specific stacks of large industrial sites. Natural emissions are calculated by the Model of Emissions of Gases and Aerosols from Nature v2.04 (MEGAN) (Guenther et al., 2006). An additional module of CHIMERE, named *diagbio*, calculates dust and sea salts emissions. Both models use meteorological fields issue from WRF to force natural emissions.

Boundary and initial conditions required for the largest domain are supplied by the LMDz-INCA2 model (Hauglustaine et al., 2004). The CHIMERE v2008 model is used for the simulations of large domains. Outputs of larger domain are used to provide the finer domain with boundary and initial conditions.

Both winter and summer periods with distinct emissions and meteorological conditions have been investigated for this study. The first simulation period is the February month of 2011 and the second period is August 2011.

Source apportionment approaches have been applied to the same emission sectors, described in the Table I, and computed for both PM_{10} and $PM_{2.5}$.

Table I: Emission sectors involved in the source apportionment studies.

Name	Description
Industry - Energy	Public power, heating plants, industry, waste, …
Residential – tertiary	Biomass combustion, residential plants, commercial plants, …
Agriculture	Agriculture, forest, …
Natural	Windblown dust, sea salts, biogenic, …
Maritime transport	Shipping, loading and unloading processes, maritime activities…
Non-road transport	Inland waterways, railways, air traffic, …
Road transport	Cars, trucks, motorcycles, road abrasion, …
External	Long-range transport from outside of the domain

CHIMERE model and zero-out approach

CHIMERE v2008 is used for the finest domain with a spatial resolution of 3 km and a 1 hour output. An option allows the attachment of a tracer to a source location during the simulation. As tracers are defined as an inert species, it has been necessary to apply a sensitivity analysis method to reach the aim of this study. The initial step assumes that the total concentration of a pollutant in a cell grid (i,j) is composed of a sum of contributions of each pollutant source. The most widely use of this method considers that removing a source should directly give an estimation of its contribution by computing a simple difference with a reference run. However, an update version, given more accurate results, proposes to remove each emission source in turn to obtain a matrix system A.X=B (equation (1), where X is a concentration.

$$A = \begin{bmatrix} 0 & 1 & 1 & \dots & 1 \\ 1 & 0 & 1 & \dots & 1 \\ 1 & 1 & 0 & \dots & 1 \\ \dots & \dots & \dots & \dots & \dots \\ 1 & 1 & 1 & \dots & 0 \end{bmatrix},\ X = \begin{bmatrix} c_1 \\ c_2 \\ c_3 \\ \dots \\ c_N \end{bmatrix}\ B = \begin{bmatrix} C^*(c_1 = 0) \\ C^*(c_2 = 0) \\ C^*(c_3 = 0) \\ \dots \\ C^*(c_N = 0) \end{bmatrix}, \quad (1)$$

The relative contribution to the total concentration is computed after resolution of the matrix system. An additional run, computed with all emission sources, named as “reference run”, allows an estimation of the error due to non-linear systems. For a linear system between emissions and concentrations, this error should be null. However, for the non-linear systems, this error will be minimal if this method is applied to small sources and if the accuracy of the model is sufficient. On the other hand, due to its formulation, this method needs to compute as many runs as sources apportioned. Then, it is necessary to define a compromise between computational time and accuracy.

CAMx model and tracer approach

CAMx v5.40 is used for the finest domain with the same spatial and temporal resolution than the previous CHIMERE configuration and for the same periods. It includes a specific extension to compute source apportionment for particulate matter: the Particulate Source Apportionment Technology (PSAT) (Yarwood et al., 2004). It uses reactive tracers to track several pollutants from specific sources and to evaluate their contributions for primary particles, precursor gases and secondary aerosols. A total of 32 tracers can be associated to each source group. The fundamental advantage of this method is to be mass consistency as it is a true source apportionment approach. Also, it allows a decrease of the computational time as the source apportionment is calculated in the same run. Same input emissions are used for the CAMx model and the zero-out approach with CHIMERE.

RESULTS AND DISCUSSION

To compare results from CHIMERE and CAMx, the respective monthly contributions of activity sectors to PM_{10} and $PM_{2.5}$ concentrations are computed during both periods at a sampling site located downtown in Marseille (Figure 2).

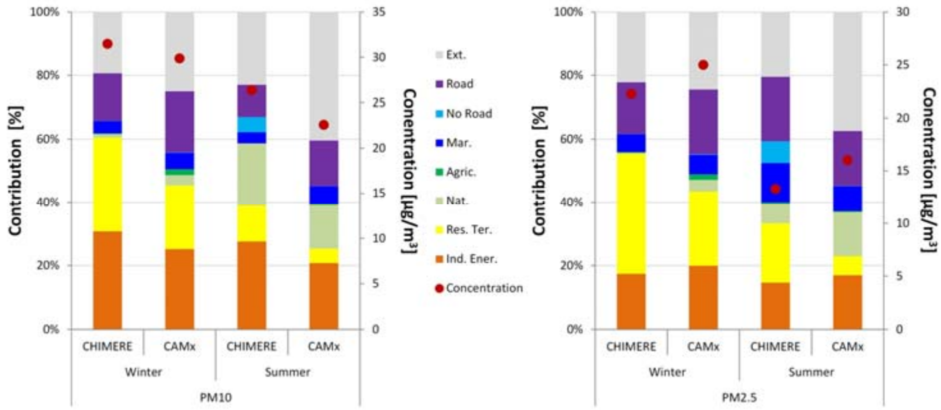

Figure 2: Concentration (red dot) and relative contribution of emission sectors to the monthly PM_{10} (left) and $PM_{2.5}$ (right) concentrations at « 5 Avenues » sampling site, located downtown in Marseille, during winter and summer periods using « zero-out modeling » from CHIMERE and « tracer » approach from CAMx by PSAT module.

Results for PM_{10} during the winter period are very similar with just a small additional contribution of both industry-energy and residential-tertiary sectors with CHIMERE model. Differences during the summer period are more important with a higher contribution of the external sector with CAMx model. Also, the summer contributions of both industry-energy and residential-tertiary sectors from CHIMERE model display a small additional contribution. During both periods, maritime activity displays a contribution around of 5% to the monthly PM_{10} concentrations from both models. In comparison with the PM_{10} concentrations, the contribution of the industry-energy sector to the monthly $PM_{2.5}$ concentration

decreases and the contribution of the residential sector increases during the winter period, in agreement with the emissions. During the winter period, the main difference is observed for the residential-tertiary sector with a higher contribution for the CHIMERE model. The same difference is also observed during the summer period. For this period, outputs of CAMx display a higher contribution of the external sector. Also, the contribution of the natural sector is more important with CAMx model for both periods. Contribution of maritime activity to the $PM_{2.5}$ concentration is 3% during the winter period and between 4% and 6% during the summer period from CAMx and CHIMERE respectively.
The application of both methodologies at the regional scale allows a spatial representation of source apportionment results and highlights significant differences (Figure 3). The main differences are observed for the residential sector during the winter period with a relative contribution exceeded 30% over the major part of the domain with CHIMERE and the zero-out approach whereas this contribution is lower than 10% over a large part of the domain with the tracer approach using CAMx, except for some grid cells, mainly located over large cities. With CHIMERE, contributions of road traffic are less dispersed and focus on the highways. For the industry-energy sector, the high contributions are located over the similar area, mainly in the south-western part close to the Berre pond (industrial area). However, output from CAMx displays a more important spatial extent for the contribution of this sector.

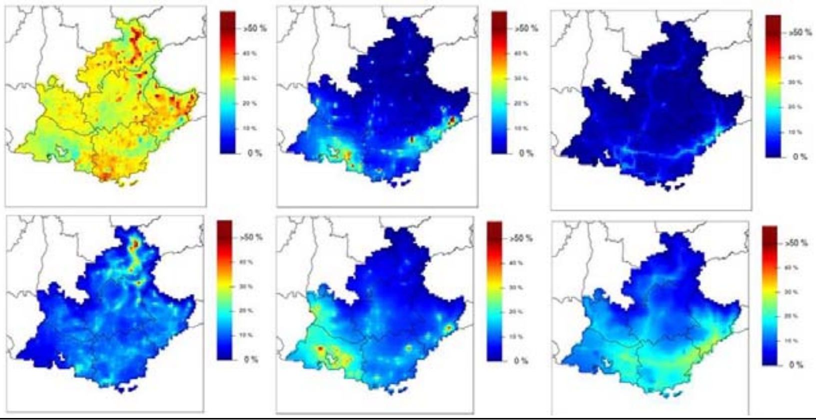

Figure 3: Monthly PM_{10} relative contributions during the winter period from CHIMERE (top) and CAMx (bottom) for Residential sector (left), Industry-Energy sector (middle) and Road Transport (right).

Thus, contributions of each activity sector are more dispersed with the tracer approach of CAMx, in comparison with the “zero-out methodology” where contributions are more similar to the emission grid. This difference is explained by the calculation method of each approach. With the “zero-out methodology”, contributions of activity sectors are mainly associated with the emissions of primary particles and do not reproduce their contributions to the secondary particles. So, this approach overestimates the main emission sources of primary particles as the residential sector by way of biomass burning and underestimates the contributions of precursors as the natural sector during the summer period. With both approaches and during both periods, the external sector associated with the long range transport of pollutants, displays high contributions over large areas. However, these high contributions are mainly related with low $PM_{2.5}$ concentrations. The external contribution computed with CAMx displays a typical long range transport condition over Marseille area. Air masses coming from northern part are canalized by the Rhone Valley, which is a heavy urban and industrial area and turn with North-western winds to reach Marseille. In the case of moderate winds, like for the beginning of the winter period, these air masses bring to Marseille significant loads of particles with an additional load due to the industrial area located at the north-western part of Marseille.
Results issue from receptor models and CTMs are compared at a sampling site downtown in Marseille named “5 Avenues”. The methodologies used for the receptor models are described in Detournay et al (2012). Two different approaches from receptor models are compared to numerical approaches. As the receptor models are able to apportion only the primary fraction of particles, the same restriction is applied to numerical models. During the winter season, results from CTMs and receptor models display a major fraction of secondary aerosols, between 34% and 51% of $PM_{2.5}$ concentration (Figure 4). Outputs of CTMs are in the range of receptor models results. The main anthropogenic contributor to the primary fraction of $PM_{2.5}$ is the residential sector with a good agreement between receptor models and CTMs outputs. This

sector is mainly related to biomass burning emission during the winter time. Road traffic contributions also display a good agreement between receptor models and CTMs. The main differences are computed for the natural and industrial-maritime contributions. Between receptor models, the industrial contribution ranges from 1% to 14% whereas it ranges between 15% and 22% for the CTMs. However, a good agreement is found between PMF and CAMx. At last, CTMs underestimate the natural contributions in comparison with receptor models outputs due to a lack of emissions.

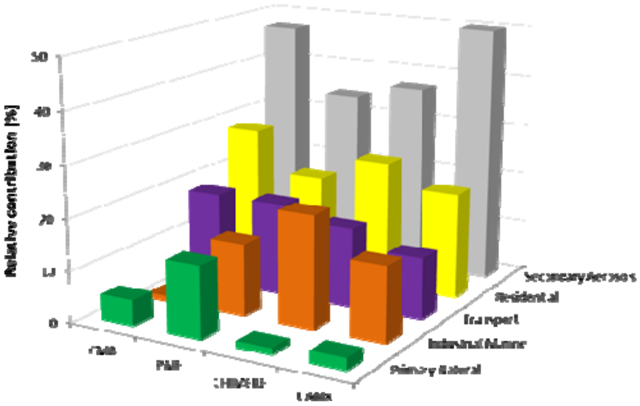

Figure 4 : Relative contribution of emission sectors to primary $PM_{2.5}$ concentration downtown in Marseille during winter period using CMB and PMF methodology and from CHIMERE and CAMx.

CONCLUSION

Between numerical approaches, some differences have been noted due to non-linear systems, leading to an overestimation of local sources with the zero-out modeling. During the winter period, road traffic, industrial-energy and residential-tertiary sectors, mainly associated to biomass burning emission, display significant contributions to PM concentrations for large cities of the South-eastern part of France. During the summer period, the sector of natural emissions displays significant contributions in a large part of the region but anthropogenic contributions with road traffic and industrial-energy sectors remain important inside large cities. Long-range transport from areas outside of the studied domain also displays an important contribution at the regional scale for both seasons. A first comparison between numerical and receptor model results downtown in Marseille during the winter period have shown a good agreement. It confirms the important contribution of biomass burning to the PM concentration for the cold season.

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