Harmonized PM source apportionment on a large set of various French sites using constrained Positive Matrix Factorization

D. Salameh[1], O. Favez[2], B. Golly[1], J-L. Besombes[3], L. Alleman[4], A. Albinet[2], J-L. Jaffrezo[1]

[1]Université Grenoble Alpes-CNRS, LGGE UMR5183, 38402 St-Martin d'Hères, France
[2]INERIS, DRC/CARA/CIME, 60550 Verneuil-en-Halatte, France
[3]Université Savoie Mont Blanc, LCME, 73376 Le Bourget du Lac, France
[4]Mines Douai, SAGE, 59508 Douai, France
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Presenting author email: dalia.salameh@lgge.obs.ujf-grenoble.fr

Particulate matter (PM) is one of the most studied atmospheric pollutants due to their adverse effects on human health and the increased risk of morbidity and mortality (Pope et al., 2009). In order to reduce the health risks and to build effective PM abatement strategies, a detailed knowledge about the predominant sources of PM and a reliable source identification and quantification of their contribution to the PM ambient levels are strongly needed.

Multivariate receptor models such as Positive Matrix Factorization (PMF) are very useful and have long been used worldwide for PM source apportionment (Viana et al., 2008; Belis et al., 2013). PMF notably uses a weighted least-squares fit and quantitatively determine source fingerprints and their contributions to the total PM mass, providing both factor profiles and the mass contributed by the factors. However, in many cases, it happens to be tricky to separate two factors that co-vary due to similar seasonal variation, obscuring the physical sense of the extracted factors. To address such issues of source collinearities, extra specific constraints are incorporated to the model (i.e., constrained PMF, that can be performed using for instance the ME-2 software developed by Paatero (1999)), allowing for a better source separation and cleaner profiles that are more consistently interpretable.

The main objectives of the present work conducted within the framework of the SOURCES project was to perform a harmonized PM source apportionment on a large number of sites (up to 18) of different typologies (urban background, industrial, traffic, rural and/or Alpine sites) distributed all over France and previously investigated with annual or multiannual studies (2012-2015). For that purpose, and to improve the source apportionment results, a constrained version of PMF (US-EPA PMF v5.0) receptor model was applied to the PM chemical datasets in a harmonized way for all sites. PM samples collected at these sites were extensively characterized and generally analyzed for the contents of OC/EC, anions/cations, major and trace elements (such as Cu, Ni, Pb, Rb, Sb, V, Zn, Al, Ca, K, Mg, Na, Ti, etc.,), and several organic molecular markers (including oxalate, MSA, levoglucosan, polyols, etc.,).

A major outcome of the present study relies in the comparison of the chemical fingerprints of the factors identified for the source profiles commonly resolved at the different sites (Figure 1, as an example), which allowed to highlight the homogeneities and/or dissimilarities in the composition of the sources, and their spatial variabilities over different typologies of sites. Moreover, at all sites, the contributions of the different source categories to the ambient PM levels have been also compared and discussed regarding local emission sources and long-range transport processes. To do so, geographical origins of major PM sources identified through constrained PMF analysis have been investigated using potential contribution source function (PSCF), by associating the temporal contributions of the resolved factors with back trajectories (Waked et al., 2014).

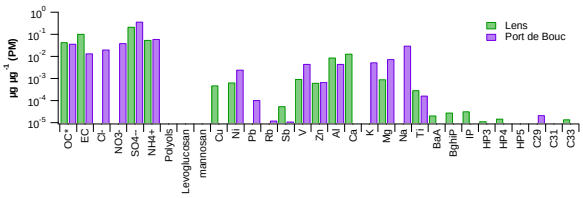

Figure 1. Example of the source profiles (µg µg^{-1} of PM) obtained at "Port de Bouc" and "Lens" for the factor "oil combustion" using constrained PMF method.

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