Inter-comparison of PMF and CMB receptor models for PM_{10} source apportionment in three sites in central Italy

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Receptor models (RMs), based on chemical composition of particulate matter (PM) at specific sites, such as Chemical Mass Balance (CMB) and Positive Matrix Factorization (PMF), represent useful tools for assessing the impact of PM sources to air quality. This is a fundamental information, especially in areas influenced by anthropogenic activities, for planning mitigation strategies for environmental management. Some recent inter-comparison of source apportionment (SA) results showed that one of the drawback in the comparison of estimated source contributions is the compatibility of the sources, i.e. the chemical profiles of the factor/sources used in the different receptor models. This suggests that source apportionment analyses based on an integrated approach between several receptor models, could give more stable and reliable solutions with respect to the application of a single model. In this framework, it is clear the need of further research for evaluating receptor models performances and for standardisation of RMs applications to source apportionment (Belis et al., 2015).

The aim of this work was to perform an inter-comparison of PMF (using two EPA codes: PMF3 and PMF5) and CMB (CMB8.2 code) outputs, focusing on the discrepancies on both source chemical profiles and estimates of source contributions. The dataset used included 347 daily PM_{10} samples collected in three different sites, (a rural site, an urban background site, and an urban site) in central Italy, located near industrial emissions. The PM_{10} samples were chemically analysed to determine the concentrations of 21 chemical species (NH_4^+, Ca^{2+}, Mg^{2+}, Na^+, K^+, Mg^{2+}, SO_4^{2-}, NO_3^-, Cl^-, Si, Al, Ti, V, Mn, Fe, Ni, Cu, Zn, Br, EC, and OC). The chemical composition was used as input for the PMF3.0, PMF5.0, and CMB8.2 receptor models.

Receptor models outputs were compared in terms of chemical profiles and of estimated contributions for seven out of the nine identified sources (crustal, marine, nitrate, sulphate, traffic, resuspended dust, biomass burning, harbour-industrial, and coal-fired power plant). This because PMF3 and PMF5 were not able to directly separate crustal and coal-fired power plant contributions because of the collinearity of profiles (both loaded with Si and Al). For CMB a single profile of traffic-resuspended dust, experimentally determined, was available instead. In Figure 1 the inter-comparison of the contributions of the seven factors/sources is shown. The main differences between PMF and CMB were observed for secondary nitrate, biomass burning and harbour-industrial sources. The reason for the discrepancies is the non compatibility of these source profiles that have local specificities. These site-dependent features were taken into account, optimizing the input source profiles of CMB, basically including a mixed source of ammonium and sodium nitrate in order to account for the interaction of nitric acid and sea-spray with formation of $NaNO_3$ generally observed in Central and Southern Italy (Contini et al., 2014). The run with optimised profiles (CMB_{opt}) brought a significant improvement in the comparison of the estimated source contributions with PMF. The comparison between measured and reconstructed PM_{10} concentrations showed a negligible unexplained mass for PMF and a more relevant unexplained mass for CMB (16.8% of PM_{10} as an average for the three sites). While, using optimized source profiles in CMB reduced the unexplained mass from 16.8% of PM_{10} to 7.6%.

PMF5 was applied with some constraints on factor profiles: for NO_3^- (pull up maximally in the nitrate factor and pull down maximally in the marine factor) and for K^+ (pull up maximally in the biomass burning factor). The comparison with PMF3 results showed that the applications of constraints improved the interpretability of factors profiles and the comparability of estimated source contributions with stoichiometric calculations (based on measured concentrations of chemical species) marine and sulphate sources.

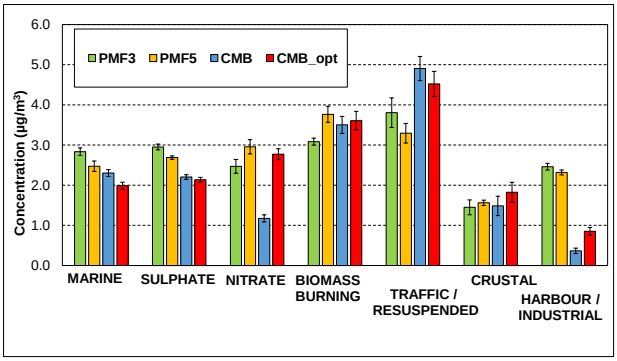

Figure 1. Inter-comparison of source contributions estimated by receptor models (average of the sites).

Belis, C., et al., (2015). *Atmos. Environ.* **123**, 240-250.
Contini, D., et al. (2014). *Sci. Total Environ.* **472**, 248–261.