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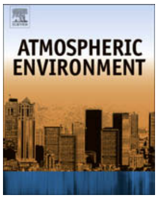

Quantifying road dust resuspension in urban environment by Multilinear Engine: A comparison with PMF2

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A B S T R A C T

Atmospheric PM pollution from traffic comprises not only direct emissions but also non-exhaust emissions because resuspension of road dust that can produce high human exposure to heavy metals, metalloids, and mineral matter. A key task for establishing mitigation or preventive measures is estimating the contribution of road dust resuspension to the atmospheric PM mixture. Several source apportionment studies, applying receptor modeling at urban background sites, have shown the difficulty in identifying a road dust source separately from other mineral sources or vehicular exhausts. The Multilinear Engine (ME-2) is a computer program that can solve the Positive Matrix Factorization (PMF) problem. ME-2 uses a programming language permitting the solution to be guided toward some possible targets that can be derived from *a priori* knowledge of sources (chemical profile, ratios, etc.). This feature makes it especially suitable for source apportionment studies where partial knowledge of the sources is available.

In the present study ME-2 was applied to data from an urban background site of Barcelona (Spain) to quantify the contribution of road dust resuspension to PM_{10} and $PM_{2.5}$ concentrations. Given that recently the emission profile of local resuspended road dust was obtained (Amato, F., Pandolfi, M., Viana, M., Querol, X., Alastuey, A., Moreno, T., 2009. Spatial and chemical patterns of PM_{10} in road dust deposited in urban environment. Atmospheric Environment 43 (9), 1650–1659), such *a priori* information was introduced in the model as auxiliary terms of the object function to be minimized by the implementation of the so-called “pulling equations”.

ME-2 permitted to enhance the basic PMF solution (obtained by PMF2) identifying, beside the seven sources of PMF2, the road dust source which accounted for 6.9 $\mu g\ m^{-3}$ (17%) in PM_{10}, 2.2 $\mu g\ m^{-3}$ (8%) of $PM_{2.5}$ and 0.3 $\mu g\ m^{-3}$ (2%) of PM_1. This reveals that resuspension was responsible of the 37%, 15% and 3% of total traffic emissions respectively in PM_{10}, $PM_{2.5}$ and PM_1. Therefore the overall traffic contribution resulted in 18 $\mu g\ m^{-3}$ (46%) in PM_{10}, 14 $\mu g\ m^{-3}$ (51%) in $PM_{2.5}$ and 8 $\mu g\ m^{-3}$ (48%) in PM_1. In PMF2 this mass explained by road dust resuspension was redistributed among the rest of sources, increasing mostly the mineral, secondary nitrate and aged sea salt contributions.

1. Introduction

Non-exhaust particulate emissions resuspended by traffic can often represent an important source of atmospheric particulate matter (PM) in urban environments (Thorpe and Harrison, 2008; Schauer et al., 2006; Norman and Johansson, 2006), especially when the scarce precipitation favors the accumulation of road dust on the pavement. Consequently, resuspension of road dust can lead to high human exposures to heavy metals, metalloids and mineral matter. Studying the contributions of road dust resuspension to urban levels of atmospheric PM is a key task for establishing eventual mitigation or preventive measures. Load and chemical properties of road dust are various and heterogeneous. Fine and coarse particles deposit daily on urban pavement from multiple sources such as wear of road pavement, brakes pads, and handling of dusty materials (demolishing, transport by uncovered trucks, etc.) among others. Through dry deposition, any source of PM is contributing to accumulation of road dust. As

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a consequence, the chemical composition of road dust is quite variable depending on the site and particle size (Amato et al., 2009; Manno et al., 2006; Han et al., 2007; Zhao et al., 2006). In southern European cities, accumulation and resuspension of road dust are favored by the scarce rainfall as compared to central and northern countries. Indeed, a comparison of several European studies shows that PM in Mediterranean countries is considerably enriched in mineral dust: soil fraction accounts for 22% in Amsterdam whilst 65% in Athens (Sillanpää et al., 2006), mineral dust achieves 10 $\mu g\ m^{-3}$ in Spain while 2–5 in central Europe and 7–9 in Sweden (Querol et al., 2004). Viana et al. (2007) registered 23–33% of mineral matter in PM_{10} in Barcelona (Spain), while only 9% in Ghent (Belgium). Moreover, even in areas with high background mineral load, Querol et al. (2004) found that the highest mineral dust concentrations were recorded at urban background sites rather than at rural or suburban background sites.

Although road dust consists mainly of mineral particles, it is also enriched in heavy metals and metalloids. Vehicle wear (brakes, rotor, muffler ablation) yields to deposition of high metal content particles. Brake pads are commonly filled with $BaSO_4$, while Sb, Sn and Mo sulfides are often added as lubricants and Cu and Zn compounds are normally used to improve friction (Garg et al., 2000; Iijima et al., 2007; Thorpe and Harrison, 2008). Querol et al. (2007) determined trace element concentrations in particles at several receptor sites in Spain, and specific tracers were identified for steel and stainless steel industries, copper and zinc metallurgies, ceramic and petrochemical estates, and traffic-related sources. In the urban atmosphere, Cu, Zn, Sb, Mo, Ba and Sr were found in similar, or even higher, concentrations than in industrial environments. Therefore, nowadays the traffic sector represents a major source of diffuse metal and metalloid emissions (EEA, 2004) that in the near past were exclusive markers of industrial emissions.

The enrichment of such trace elements in road dust can be exploited in order to quantify the contribution of road dust resuspension to atmospheric PM concentrations. Source apportionment studies are generally performed by receptor models that are based on the mass conservation principle:

$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{jk} \quad i = 1,2,\ldots,m \quad j = 1,2,\ldots,n \qquad (1)$$

where x_{ij} is the jth species concentration measured in the ith sample, g_{ik} is the contribution of the kth source to ith sample and f_{jk} is the concentration of the jth species in kth source. Equation (1) can be also expressed in matrix form as $\mathbf{X} = \mathbf{GF}^T$. If f_{jk} are known for all the sources then the Chemical Mass Balance (CMB) can be applied (Watson et al., 1984). For this model the experimental profiles of all major sources are needed. When both g_{ik} and f_{jk} are unknown, factor analysis (FA) techniques such as *Principal Components Analysis* (PCA) (Thurston and Spengler, 1985; Henry and Hidy, 1979) and *Positive Matrix Factorization* (PMF) (Paatero and Tapper, 1994) are used for solving (1). PMF can be either solved with the Multilinear Engine (ME-2) developed by Paatero (1999). ME-2 is a more flexible program than the earlier PMF2, and permits the incorporation of any *a priori* information such as chemical properties or linear constraints into the model as a target to be fit to some specified precision. Therefore, ME-2 is especially suitable for source apportionment studies where some knowledge (chemical ratios, profiles, mass conservation, etc.) of involved sources is available.

Heavy metals can be used as useful markers and at the same time, their high content in road dust makes it especially relevant in the health burden of urban PM because of their potential for catalyzing the formation of reactive oxygen species. Adverse health effects of PM in urban areas have been observed in several epidemiological studies (US EPA, 2004, 2006; Brunekreef and Forsberg, 2005; Kan et al., 2007; Peng et al., 2008). Schlesinger et al. (2006) indicated that transition metals such as Cu, Zn, Fe, Ni, Cr and Mn, which may act as redox compounds, are likely related to PM toxicity. Several toxicological studies also indicate that coarse particles can elicit inflammatory effects (Schins et al., 2004; Schwarze et al., 2007).

Comparisons of six European cities (Jalava et al., 2007, 2008; Happo et al., 2007) evaluated the cytotoxic and inflammatory activities of atmospheric PM in contrasting air pollution scenarios. Coarse particles showed higher inflammatory effect than the other PM size fractions. The highest inflammatory activities were induced by the Athens summer and Barcelona spring samples rather consistently in all the measured parameters. This high activity for these samples was attributed to the lack of rain, which may account for the poor washout of road dust and the consequent accumulation of coarse PM (with high levels of brake pads metals) on the road pavement. The city of Barcelona represents a typical Mediterranean scenario of urban pollution strongly influenced by road dust resuspension.

The Barcelona metropolitan area is characterized by high road traffic density and by a wide range of industrial activities. Facilities comprise ferrous and non-ferrous smelters, cement and asphalt production industries, which are spread between the two river basins in the North and South of the Metropolitan area. Furthermore, two power stations and two city waste incinerators are also based in the considered area. Traffic density is a consequence of the high population concentration in the city of Barcelona (101 km^2 with 1.6 million inhabitants, 4.5 million in the greater metropolitan area) leading to one of the highest car densities in Europe (http://w3.bcn.es/fitxers/mobilitat/dadesbasiques2006.222.pdf). Jointly, the urban architecture, characterized by square-blocks with narrow streets, reduces the dispersion of pollutants and the scarce precipitation favors the accumulation and resuspension of particulate matter deposited at ground. In addition to local emissions, African dust outbreaks reach Barcelona in the order of 7–10 events per year with the highest frequency in the summer and winter–spring periods (Rodríguez et al., 2001). PM levels in the urban background of Barcelona reflect the critical scenario of pollution, exceeding the daily limit value from the European Air Quality Directive 1999/30/CE (50 $\mu g\ PM_{10}\ m^{-3}$) 97 times per year. Approximately 80% of such exceedances are due to anthropogenic PM contribution with high proportion of mineral dust (Perez et al., 2008; Querol et al., 2001a). Previous studies explored the origin of such mineral matter in the specified area applied PCA and demonstrated the difficulty in separating resuspended road dust as an independent factor. Its profile was nearly a linear combination of other sources, mainly mineral, vehicle exhaust and wear (Querol et al., 2001a, 2004; Rodríguez et al., 2004; Viana et al., 2007). CMB has not yet been applied in this area.

The aim of this study was double: firstly to quantify the contribution of road dust resuspension in PM_{10} and $PM_{2.5}$ separately from other mineral sources and the vehicular exhaust in the urban background of Barcelona (Spain), secondly to introduce new techniques for PMF like the implementation of experimental emission profiles in the ME-2 PMF model. Thus, in order to show how the basic PMF can be enhanced, both ME-2 and PMF2 were applied and compared.