



## Road traffic impact on urban atmospheric aerosol loading at Oporto, Portugal

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### ABSTRACT

At urban areas in south Europe atmospheric aerosol levels are frequently above legislation limits as a result of road traffic and favourable climatic conditions for photochemical formation and dust suspension. Strategies for urban particulate pollution control have to take into account specific regional characteristics and need correct information concerning the sources of the aerosol.

With these objectives, the ionic and elemental composition of the fine ( $PM_{2.5}$ ) and coarse ( $PM_{2.5-10}$ ) aerosol was measured at two contrasting sites in the centre of the city of Oporto, roadside (R) and urban background (UB), during two campaigns, in winter and summer.

Application of Spatial Variability Factors, in association with Principal Component/Multilinear Regression/Inter-site Mass Balance Analysis, to aerosol data permitted to identify and quantify 5 main groups of sources, namely direct car emissions, industry, photochemical production, dust suspension and sea salt transport. Traffic strongly influenced PM mass and composition. Direct car emissions and road dust resuspension contributed with 44–66% to the fine aerosol and with 12 to 55% to the coarse particles mass at both sites, showing typically highest loads at roadside. In fine particles secondary origin was also quite important in aerosol loading, principally during summer, with 28–48% mass contribution, at R and UB sites respectively. Sea spray has an important contribution of 18–28% to coarse aerosol mass in the studied area, with a highest relative contribution at UB site.

Application of Spatial Variability/Mass Balance Analysis permitted the estimation of traffic contribution to soil dust in both size ranges, across sites and seasons, demonstrating that as much as 80% of present dust can result from road traffic resuspension.

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### 1. Introduction

Particulate matter (PM) in urban environments is a primary health issue (Katsouyanni et al., 1997; Samet et al., 2000) which has lead policy makers to legislate on exposure levels. In 1999, the European Commission (EC) Air Quality Directive 1999/30/EC established  $40 \mu\text{g m}^{-3}$  (to be decreased to 20 from 2010 on) as the limit for the annual mean of  $PM_{10}$  (PM with an aerodynamic diameter below  $10 \mu\text{m}$ ), with a daily average limit value of  $50 \mu\text{g m}^{-3}$  that cannot be exceeded more than 35 days a year (to be decreased to 7 days from 2010 on). From 1997 to 2006, 20 to 50% of the European urban population was exposed to over 35 days a year of exceedences and no improvements were observed through the decade (EEA, 2009). The Air Quality Directive 2008/50/EC added  $PM_{2.5}$  to the list of criteria pollutants, with more exigent limit

values or target values to be met for PM. PM pollution assumes a special importance in the Iberian Peninsula, including the Oporto Metropolitan Area (Sillanpaa et al., 2005; Pereira et al., 2005), due to several unfavourable climatologic factors: high temperatures and low precipitation hinder rain scavenging and promote dust resuspension (Almeida et al., 2006b) while intense insulation supports photochemistry and secondary aerosol formation (Almeida et al., 2006a). Also, the geographical proximity to the coast and northern Africa turns sea salt (Pio et al., 1996) and mineral dust (Artiñano et al., 2001; Pey et al., 2009) into frequently important contributors to the PM load. Besides the favourable conditions for PM concentrations build-up, PM health effects are worsened in the warmer climates of southern Europe (Katsouyanni et al., 2001).

Automotive emissions are one of the major contributors to atmospheric pollution in urban areas (Rodriguez et al., 2004; Almeida et al., 2005; Negral et al., 2008). Despite the constant developments on environmentally friendlier fuels and the production of less pollutant vehicles, atmospheric pollution by road

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traffic is not diminishing in several south European metropolitan areas because of continuous traffic increasing (EEA, 2009). The high traffic volumes in those areas increase the atmospheric particles concentration within the breathing zone due to both direct vehicle emissions (exhaust and wear vehicle emissions) and the resuspension of particles from the road surface (Almeida et al., 2005). Such particles were shown to be rich in heavy metals and polycyclic aromatic hydrocarbons, whose carcinogenicity induces an additional potential risk to the public health (Harrison and Yin, 2000). Thus, policy makers need to account for the traffic sector and its emissions, as a whole, in abatement strategies (Querol et al., 2004).

The national and regional authorities need a correct knowledge of source contributions to the atmospheric aerosol loading in order to develop control strategies that are effective and cost acceptable. With the implementation, by legislation enforcing, of engine control measures, direct pipe emissions are nowadays lower, and other non-exhaust sources, such as brake wear and road dust resuspension, are becoming important (Gietl et al., 2010). In southern Europe climate conditions favour re-entrainment of dust, which is a non despising contributor to PM loading. Discrimination between natural sources of dust and road dust resuspension is difficult because of the similarity of dust source profiles, although special source apportionment adaptations have been able of successfully discriminate both sources (Almeida et al., 2006b; Amato et al., 2009a,b; Pey et al., 2010). In this study, ambient particulate matter was collected at two sites within the city centre of Oporto, Portugal. Samples were chemically characterised by PIXE and ion chromatography. Factor/multi-linear regression/inter-site mass balance analysis were applied to analytical data, in order to identify association patterns between aerosol components, to quantify source contributions and to infer about the importance of aerosol sources and processes.

## 2. Experimental

Oporto city (population around 250,000, density of 6300 inhabitants  $\text{km}^{-2}$ ) is located in northern Portugal ( $41^\circ 09'N$ ,  $8^\circ 37'W$ ) on the Douro river estuary, being part of Oporto Metropolitan Area (about 1.2 million inhabitants). Detailed information about Oporto characteristics and sampling sites characteristics can be found in Oliveira et al., 2007 and Oliveira et al., 2009. In order to evaluate the influence of automotive emissions on atmospheric pollutant levels, sampling was performed simultaneously at two locations in the centre of the city, one directly impacted by fresh car emissions (roadside, R) and the other, although close to the first, far enough as to be considered as urban background (UB). Two, one-month, sampling campaigns were performed (July and November/December 2003) to evaluate the different contributions of summer and winter traffic and the influence of the characteristic meteorological conditions of both seasons. The roadside station, (known as Boavista), is part of the regional air pollution monitoring network; between 2002 and 2007, yearly average  $\text{PM}_{10}$  concentrations varied from 36.6 to 52.2  $\mu\text{g m}^{-3}$  (meeting only twice the EC directive requirements) and the number of exceedences ranged from 69 to 120 a year (<http://www.qualar.org>).

Atmospheric aerosol fine ( $\text{PM}_{2.5}$ ) and coarse ( $\text{PM}_{2.5-10}$ ) fractions were collected simultaneously at both sites with Dichotomous Stacked Filter Units (DSFUs, (Luhana et al., 2001) in two parallel lines. Each DSFU unit was operated at 6  $\text{L min}^{-1}$  with two different polycarbonate membrane filters (Nuclepore), a 12  $\mu\text{m}$  pore upper filter coated with Apiezon L high-vacuum grease to avoid any bounce-off or re-entrainment of the deposited particles (Cahill et al., 1979), and a 0.4  $\mu\text{m}$  pore uncoated backup filter. Before and after sampling, filters were pre-conditioned in a box with constant

humidity (50%) for a minimum period of 24 hours, electrostatically neutralized with an ionizing bar (EXAIR<sup>®</sup>) and weighted in a micro balance to determine the deposited aerosol mass. Two intensive, one month-long, campaigns were conducted, the first from July 3rd to July 30th and the second from November 21st to December 18th 2003. Two daily, 12 hours, sampling periods were selected (starting at 7:00 a.m. or 7:00 p.m.) to account for the day/night variability of the traffic emissions.

Sampled Nuclepore filters were analysed regarding their inorganic chemical composition. The elemental content was determined at the Danish National Environmental Research Institute by PIXE for 20 elements with atomic number higher than 12 (Cl, Al, Si, S, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Br, Rb, Sr, Zr, Sn, Pb). The analytical uncertainty of measurements was estimated for each element and each sample, being the general precision, 5%, for concentrations far from the detection limit. The water-soluble ionic composition ( $\text{Na}^+$ ,  $\text{NH}_4^+$ ,  $\text{K}^+$ ,  $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$ ,  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$ ) was determined by ion chromatography at the University of Hertfordshire, U.K., after filter extraction in pure water. In addition to some typical meteorological parameters such as temperature, pressure, wind speed, wind direction, humidity and solar radiation, the concentrations of NO and NO<sub>2</sub>, CO, SO<sub>2</sub> and O<sub>3</sub> were also measured at both sites with gas analysers and  $\text{PM}_{10}$  by  $\beta$  attenuation.

This study was part of the EC funded project SAPPHIRE. In the project, besides DSFU sampling, aerosol was also collected with high-volume samplers to study the organic content of aerosols (Oliveira et al., 2007) and their particle size distributions evaluated by DMA (Oliveira et al., 2009).

## 3. Results and discussion

### 3.1. Aerosol composition and levels

Both  $\text{PM}_x$  fractions clearly exhibited higher levels at the R site than at UB (Tables 1 and 2), with the exception of the fine fraction in summer (similar values). Mass concentrations were in general agreement with other aerosol studies at Oporto (Pereira et al., 2005; Slezakova et al., 2007; Duarte et al., 2008) and in the same range of levels reported for other European urban environments (Van Dingenen et al., 2004).

The average total ionic mass was 6.4 and 4.5  $\mu\text{g m}^{-3}$  in  $\text{PM}_{2.5}$  and  $\text{PM}_{2.5-10}$ , respectively, in summer and 2.6 and 3.1  $\mu\text{g m}^{-3}$  in winter (Tables 1 and 2). Equivalent ionic balance ratio between cations and anions was usually lower than unity for fine particles and higher than unity for the coarse fraction. That is probably from the presence of non-measured  $\text{H}^+$  in the fine aerosol and non-measured carbonates in the coarse particles.

Sulphate (78%) and ammonium (89%) were overwhelmingly present in the fine fraction, whereas nitrate was prevalent in the coarse fraction (78%), as previously found by Alves et al., 2007, in a Portuguese coastal area and Nicolas et al., 2009, in an Iberian semiarid region. Photochemistry plays an important role on the formation of those species and their presence in the aerosol phase, as evidenced by the higher summer concentrations (Tables 1 and 2).

Sea-salt was the major component of coarse particles ionic fraction (55% in winter, 39% in summer). Main sea salt components ( $\text{Na}^+$  and  $\text{Cl}^-$ ) showed higher  $\text{PM}_{10}$  concentrations in winter, due to higher production of sea spray in wintery weather.  $\text{Cl}^-$  levels decreased more strongly in summer than  $\text{Na}^+$  as result of volatilization and substitution by acidic ions (sulphate and nitrate) on warmer sunnier days (Nicolas et al., 2009; Pio and Lopes, 1998).

In the following discussion, non sea salt potassium and calcium ( $\text{nss-K}^+$ ,  $\text{nss-Ca}^{2+}$ ) are defined from sea salt composition ratios, as given by Seinfeld and Pandis (1998). Calcium, commonly associated

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