



Size fractionated aerosol composition at roadside and background environments in the Madrid urban atmosphere



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ABSTRACT

The chemical composition of size-segregated particulate matter (PM) was studied during summer and winter sampling campaigns, at two different urban sites (roadside and urban background) in the city of Madrid, Spain. PM was sampled with high volume cascade impactors, in 4 size ranges: 10–2.5, 2.5–1, 1–0.5 and <0.5 μm . The carbonaceous content (OC and EC) was determined by a thermo-optical method, whilst the water soluble ionic species were measured by ion chromatography. The most common synoptic meteorological situations, including those causing the transport of African dusty air masses, were identified in both seasons. Whether the PM₁₀ mass or the highest concentrations of EC and OC were found predominantly in the ultrafine size fraction at both sites. In contrast with roadside, at the urban background, the particle mass concentrations for the different size ranges were statistically higher in summer than in winter. Observed inter-site differences suggest the existence of other sources and formation processes contributing in the summer period to the levels of PM at the urban background site apart from road traffic emissions. Secondary organic carbon (SOC) showed a clear seasonal pattern, with much higher concentrations in summer than in winter in both places, as well as higher relative contributions at the urban background than at the road traffic site. (NH₄)₂SO₄ levels were at their maximum at both sites in summer in PM_{0.5}. Differently, higher values were reached in winter for NH₄NO₃ in PM_{0.5} and for NaCl in PM_{2.5–10}. From the ion balances, it was observed that, in summer, the formation of secondary inorganic compounds included an unusual enrichment in Ca²⁺ in the submicrometre fraction, either at roadside or at urban background.

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1. Introduction

Particulate matter (PM) is a high priority atmospheric pollutant since it has strong negative impacts on human health, (Schleicher et al., 2011), climate (Das and Jayaraman, 2012), acid rain (Zhang et al., 2012), ecosystems (Katul et al., 2011), visibility (Yuan et al., 2006) and building materials (Costa et al., 2009).

A major portion of atmospheric particles is constituted by carbonaceous matter (elemental (EC), organic carbon (OC), carbonated carbon (CC)); at European urban areas, its

contribution is known to reach up to 18–32% of PM₁₀ and 25–31% of PM_{2.5} (Putaud et al., 2010). Epidemiological studies have demonstrated that cardiovascular mortality and morbidity are associated with exposure to increased levels of urban carbonaceous aerosols (Ito et al., 2011). EC is abundant in the emissions from combustion of fossil fuel (Gillies and Gertler, 2000), and biomass burning (Alves et al., 2010; Vicente et al., 2012). OC sources are poorly characterised, but include direct emission by combustion processes, soil, paved road dust, meat cooking and others, and gas to particle conversion from anthropogenic and biogenic volatile organic compounds (VOCs). OC and EC are two of the major components of atmospheric particles emitted by traffic (both diesel and gasoline) (Calvo et al., 2013). There is little information

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concerning the loading of CC in aerosols in Europe; CC results from soil or road dust emission and is usually present in coarse particles not attacked by atmospheric acidic components (Calvo et al., 2013).

Another predominant fraction of atmospheric aerosols consists in water soluble inorganic matter (WSIM). WSIM is emitted directly in the particulate form, principally as sea salt from the action of wind on the ocean surface (Donahue et al., 2009) or as secondary production from gas to particle conversion. Sulphate, nitrate and ammonia are considered the major secondary inorganic compounds in aerosols; their precursor gases (SO_2 , NO_x and NH_3) are mainly emitted to the atmosphere by combustion processes (energy production or road traffic) and agricultural activities. At European urban areas, sulphate and nitrate contributions are known to reach up to 21–28% of PM_{10} and 22–37% of $\text{PM}_{2.5}$ (Putaud et al., 2010).

Given that the presently in-force European Directive 2008/50/EC on air quality, requires the control of PM_{10} and $\text{PM}_{2.5}$, many studies have been focussed on the size integrated particulate matter, mainly in urban areas (Aldabe et al., 2011; Minguillón et al., 2012; Moreno et al., 2013; Oliveira et al., 2010; Salvador et al., 2012). However, the environmental effects, the formation processes and the source assignment depend greatly on the particle size distribution (Lighty et al., 2000). Concerning size-segregated samples, attention has been greatly focussed on polyaromatic hydrocarbons in urban areas, because of their carcinogenicity (Cancio et al., 2004; Duan et al., 2005; Wang et al., 2009). The information available on the particle size distributions for other compounds is rather scarce.

The Madrid metropolitan area is located in the central part of the Iberian Peninsula, where extreme weather conditions associated with an inland climate are usual. The region has no relevant nearby heavy industrial activity, being the traffic sector the dominant emission source of carbonaceous matter (Salvador et al., 2004, 2012). Moreover, residential wood burning activities are rather inexistent in this urban area. Domestic heating devices in Madrid City in 2009–2010 were preferably fed by natural gas (74%), fuel-oil (19%), coal (4%), butane (2%) and propane (1%) (Ayuntamiento, 2010). Over the last years atmospheric pollution levels have been decreasing in the metropolitan area due to a reduction in residential coal burning emissions and to changes in the vehicle fleet composition (Salvador et al., 2012). However, long-range transport episodes of mineral dust, usually related to the occurrence of African dust outbreaks, significantly affect aerosol concentrations in the Madrid air basin (Salvador et al., 2004). The concentration of inhalable mineral dust in Madrid air is also strongly influenced by traffic resuspension (Moreno et al., 2013).

This study reports, for the first time, data from four size fractions in two distinct places in the metropolitan area of Madrid (Spain), one with fresh traffic influence and the other in a suburban area, for two periods of time during the summer of 2009 and the winter of 2010. The innovative character of this study consists in obtaining time series with a size-segregated detailed chemical composition of PM for differently polluted urban sites, which may be further used for a more accurate assessment of emission sources and atmospheric processes (Lighty et al., 2000). It would be important to understand how the above mentioned components behave in the small size fractions, helping to fulfil the task of reducing human exposure to PM.

2. Methods

2.1. Sampling site

The Madrid metropolitan area is located in the Madrid air basin, at the centre of the Iberian Peninsula. This basin is bordered to the north–northwest by the Sierra de Guadarrama range, located 40 km from the metropolitan area, to the northeast–east by lower mountainous terrain and to the South by the Toledo Mountains (Fig. 1). Population is about 6.6 million inhabitants, involving a car fleet of almost 4 million vehicles with very intense traffic on weekdays on the two existing ring roads and the roads connecting Madrid with the surrounding towns, where more than 2.5 million residents live (Gómez-Moreno et al., 2007).

One sampling site was located next to an automatic monitoring station (Escuelas Aguirre) which belongs to the Madrid municipality Air Quality Network. It is located at a heavy traffic street intersection in Madrid downtown (Fig. 1). The distance to the kerbside was approximately 5 m. Escuelas Aguirre provides a classical example of a city centre roadside traffic polluted site, dominated by the daily double peak in PM, NO_x , and CO concentrations, associated with morning and evening journeys across the city (Moreno et al., 2013).

The other sampling point was positioned within a non-residential, non-traffic area on the NW outskirts of Madrid (CIEMAT, Fig. 1). It is located downwind of the city centre, between the main university campus, “Ciudad Universitaria” and “La Dehesa de la Villa” park. The nearest road (Complutense Avenue) is separated from the site by more than 300 m and no industrial or domestic heating sources are present in the vicinity. Thus, according to the criteria established by the E.E.A. (European Environment Agency) (1999) this station can be classified as an urban background site. From now on, the Escuelas Aguirre traffic site and the CIEMAT urban background site will be designated as EA and CIE, respectively.

2.2. Sampling

Sampling campaigns were carried out in June of 2009 and January–February of 2010. About 30 days of sampling was done for each summer and winter campaigns. Samples were collected in parallel at both sites during 24 h, starting at 08:00–09:00, local time. High volume samplers, operating at a flow of $1.13 \text{ m}^3 \text{ min}^{-1}$, were equipped with a PM_{10} size selective inlet and a cascade impactor, from Tisch Environmental, Inc., which consists in three stages and a back-up filter ($20.3 \times 25.4 \text{ cm}$). Particles were collected in four size fractions: <0.49 , 0.49 – 0.95 , 0.95 – 2.5 , and 2.5 – $10 \mu\text{m}$. For easiness, cutting size values will be expressed throughout the paper, in an approximated way, as <0.5 , 0.5 – 1 , 1 – 2.5 , and 2.5 – $10 \mu\text{m}$. For comparison with the standard parameters more frequently used in air quality requirements, concentration values are also reported as PM_{10} , $\text{PM}_{2.5}$ and PM_{10} .

Particle matter collection was carried out onto quartz fibre filters from Whatman (QM-A) pre-fired ($500 \text{ }^\circ\text{C}$ for 6 h). At the EA site NO_2 , NO_x and CO hourly concentration levels were obtained from the local automatic station. At the CIE site, gaseous species NO, NO_2 and O_3 were measured using a calibrated DOAS spectrometer (OPSI, model AR-500) along a

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