



## Aerosol sources in subway environments

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### ABSTRACT

Millions of people use rail subway public transport around the world, despite the relatively high particulate matter (PM) concentrations in these underground environments, requiring the identification and quantification of the aerosol source contributions to improve the air quality. An extensive aerosol monitoring campaign was carried out in eleven subway stations in the Barcelona metro system, belonging to seven subway lines. PM<sub>2.5</sub> samples were collected during the metro operating hours and chemically analysed to determine major and trace elements, inorganic ions, and total carbon. The chemical compositions of subway components such as brake pads, rail tracks and pantographs were also determined.

The mean PM<sub>2.5</sub> concentrations varied widely among stations, ranging from 26 µg m<sup>-3</sup> to 86 µg m<sup>-3</sup>. Subway PM<sub>2.5</sub> was mainly constituted by Fe<sub>2</sub>O<sub>3</sub> (30–66%), followed by carbonaceous matter (18–37%) for the old stations, while for new stations equipped with Platform Screen Doors (PSD) these percentages go down to 21–44% and 15–30%, respectively. Both the absolute concentrations and the relative abundance of key species differed for each subway station, although with common patterns within a given subway line. This is a result of the different emission chemical profiles in different subway lines (using diverse types of brakes and/or pantographs). The co-emission of different sources poses a problem for their separation by receptor models. Nevertheless, receptor modelling (Positive Matrix Factorization) was applied resulting in ten sources, five of them subway-specific: RailWheel, RailWheel + Brake, Brake\_A, Brake\_B, Pb. The sum of their contributions accounted for 43–91% of bulk PM<sub>2.5</sub> for the old stations and 21–52% for the stations with PSD. The decrease of the activity during the weekends resulted in a decrease (up to 56%) in the subway-specific sources contribution to the –already lower– bulk PM<sub>2.5</sub> concentrations compared to weekdays. The health-related elements are mainly apportioned (> 60%) by subway sources.

### 1. Introduction

People living in large urban areas spend a significant amount of time commuting. The subway system is one of the major transport modes in many metropolitan areas worldwide, due to its convenience, safety and high speed. This particular environment may present specific conditions regarding air quality, such as relatively high particulate matter (PM) concentrations, with a chemical composition clearly different from that typically present outdoors, as pointed out by several studies in subway systems worldwide (e.g. Martins et al., 2016b, 2015b and references therein). In Barcelona, currently, over 1.25 million passengers commute by subway system on a daily basis, absorbing around 50% of the urban commuting load.

It is known that the exposure to PM is linked to health effects, and

the subway emissions have been shown to contribute to personal exposure to PM (Martins et al., 2015a; Minguillón et al., 2012). However, results so far show little clarity on whether subway PM is relatively more toxic than outdoor PM dominated by road traffic-generated particles. Thus comparing subway with outdoor air, some studies found higher subway oxidative potential values (Janssen et al., 2014), reactive oxygen species activity (Kam et al., 2011), in-vitro toxicity (Steenhof et al., 2011), transient biological effects (Bachoual et al., 2007; Karlsson et al., 2005; Seaton et al., 2005), genotoxicity (Karlsson et al., 2008; Salma et al., 2009) or cancer health risks (Lovett et al., 2017), whereas other works concluded that there is no evidence for increased health risk in breathing subway air (Grass et al., 2010; Gustavsson et al., 2008; Karlsson et al., 2008; Moreno et al., 2017; Spagnolo et al., 2015). Interestingly, what evidence there is for

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enhanced oxidative stress induced by breathing subway air implicates the presence of trace elements such as Cu and Sb rather than the dominant ferruginous component (Moreno et al., 2017), so that investigating the chemistry and sources of subway PM has become more of a priority.

The concentration and chemical composition of subway airborne particles depend on various factors, such as outdoor air quality, station and tunnel design, chemical composition of subway components (wheels, rail tracks, brake pads, and current supply materials), power system, braking mechanisms, train speed and frequency, passenger influx, ventilation and air conditioning systems, cleaning frequency, and other operational conditions (Johansson and Johansson, 2003; Kwon et al., 2015; Moreno et al., 2014; Park and Ha, 2008; Ripanucci et al., 2006; Salma et al., 2007). Studies including chemical composition of subway PM, on top of bulk PM concentrations, are more scarce but demonstrate that subway PM mainly consists of Fe, accounting between 40% and 80%, and other transition metals such as Cu, Ba, Cr, Si, Mn, and Zn (Aarnio et al., 2005; Jung et al., 2010; Loxham et al., 2013; Moreno et al., 2015; Mugica-Álvarez et al., 2012; Querol et al., 2012; Salma et al., 2007). The presence of these components is attributed to the abrasion of rail tracks, wheels, catenary, brake pads and pantographs produced by the motion of the trains (Chillrud et al., 2004; Jung et al., 2012, 2010; Park et al., 2014; Querol et al., 2012). Colombi et al. (2013) identified some clusters attributed to different sources: wear of electric cables, abrasion of wheels, rails and brakes, or crustal origin. However, the contribution of the potential sources is very rarely quantified, and only four studies were found. Jung et al. (2010) used a single-particle analytical technique and identified four major particle types in subway platforms: Fe-containing (most prevalent, 29–87% in number of particles), soil-derived, carbonaceous, and secondary nitrate and/or sulfate particles. Park et al. (2012) identified and quantified PM<sub>10</sub> sources in passenger cabins using receptor modelling (Positive Matrix Factorization): soil and road dust sources (27%), railroad-related sources (48%), secondary nitrate sources (16%), and a chlorine factor mixed with a secondary sulfate source (9%). Park et al. (2014) identified and quantified PM<sub>10</sub> sources in subway tunnels: rail, wheel, and brake wear (60%), soil combustion (17%), secondary aerosols (10%), electric cable wear (8%), and soil and road dust (5%). Martins et al. (2016b) identified and quantified PM<sub>2.5</sub> sources in platforms (data also used in the present study), grouping all the subway contributions (rail tracks, wheels, brake pads, catenaries and pantographs) in a single source accounting for 11–58% of PM<sub>2.5</sub> for conventional stations.

This study aims to identify and quantify the sources of PM<sub>2.5</sub> within the subway microenvironment, and to relate those to the wide variety of conditions and the different materials used within the subway network of Barcelona, with the final purpose of serving as a guide for possible abatement measures and health risk assessments applicable not only to the studied subway network but also transferable to other underground systems.

## 2. Methodology

### 2.1. Sampling campaigns

The subway network in Barcelona (managed by Transports Metropolitans de Barcelona, TMB) comprises 8 lines stretching 123.2 km and including 156 stations. A total of 11 subway stations with highly contrasting designs and belonging to 7 different lines were selected for the present study (Table 1, Fig. S1). Stations from lines L1 to L5 have an old design (built < 1980): one wide tunnel with two rail tracks, with or without middle wall; a single narrow tunnel with one rail track; or two wide tunnels with one rail track separated by a middle platform; whereas stations from lines L9 and L10 have a new design: a single tunnel with one rail track separated from the platform by a glass wall with platform screen doors (PSD). The stations from these lines have advanced platform ventilation systems and driverless trains. A

newly built platform from L9 not in use yet was also studied here (Collblanc2, Table 1). Both the old and the new trains use an electrical brake when approaching the station followed by mechanical brake. The different types of brake pads in use are described later.

The measurements were conducted continuously at each station during varying periods of time. The ventilation settings in the Barcelona subway system are different for the warmer (April to September) and colder (October to March) periods, with more intense ventilation in the tunnel sections during the warmer period with the aim to achieve temperature comfort. The stations were monitored during different campaigns, each of them falling in a different season period. See the details in Table 1.

Aerosol monitoring was daily carried out during train operational hours, hence including weekdays and weekends, which have a different train frequency (Table 1). The aerosol instrumentation was placed at the end of the platform corresponding to the train entry point, far from the commuters' access-to-platform point whenever possible, and protected by a light fence. The location was a compromise between meeting conditions for undisturbed measurement and minimizing the annoyance to commuters.

### 2.2. Instrumentation and chemical analyses

PM<sub>2.5</sub> samples were collected on quartz microfiber filters by a high volume sampler (HVS, Model CAV-A/MSb, MCV) at a sampling flow rate of 30 m<sup>3</sup> h<sup>-1</sup> over a 19 h period (from 5 a.m. to 12 p.m., subway operating hours) on a daily basis. A field filter blank per period was taken at each station.

PM<sub>2.5</sub> concentrations were determined gravimetrically weighing the filters before and after sampling after being stabilized for at least 48 h in a conditioned room (20 °C and 50% relative humidity). A microbalance (Model XP105DR, Mettler Toledo) with a sensitivity of ± 10 µg was used.

One quarter of each filter sample was acid digested with HNO<sub>3</sub>:HF:HClO<sub>4</sub> and subsequently analysed by Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES) and Mass Spectrometry (ICP-MS) to determine major and trace elements, respectively. A few milligrams of a standard reference material (NIST 1633b) were added to a fraction of a blank filter to check the accuracy of the analysis of the acid digestions. Another quarter of each filter sample was water leached with de-ionized water to extract the soluble fraction and analysed by ion chromatography for determination of soluble anions (Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, and NO<sub>3</sub><sup>-</sup>), and by selective electrode for ammonium (NH<sub>4</sub><sup>+</sup>). Soluble anions analyses are available for 408 samples but are missing for 273 samples due to laboratory analytical problems. A third portion of the filter sample (1.5 cm<sup>2</sup>) was used to measure total carbon (TC) using the Thermal – Optical Transmittance (TOT) method by means of a laboratory OC-EC Sunset instrument or an elemental C analyzer. Blank filters were analysed in the batches of their respective filter samples and the corresponding blank concentrations were subtracted from each sample in order to calculate the ambient concentrations. Uncertainties were calculated as described by Escrig et al. (2009).

For simplicity, chemical components were grouped into seven categories: iron oxide (Fe<sub>2</sub>O<sub>3</sub>), calculated from Fe concentrations assuming that all the Fe is in this oxidized form (Lu et al., 2015; Querol et al., 2012), although magnetite and metal Fe can be still present (Moreno et al., 2015); crustal matter, sum of Ca, Mg, Al<sub>2</sub>O<sub>3</sub> (assuming all Al is in this form), SiO<sub>2</sub> (estimated as 3\*Al<sub>2</sub>O<sub>3</sub>), CO<sub>3</sub><sup>2-</sup> (estimated as 1.5\*Ca), K<sub>2</sub>O (assuming all K is in this form), TiO<sub>2</sub> (assuming all Ti is in this form), and P<sub>2</sub>O<sub>5</sub> (assuming all P is in this form); carbonaceous matter, calculated from the TC concentrations, assuming an elemental to organic carbon (EC/OC) ratio of 0.5 (Querol et al., 2013), and an organic matter to organic carbon (OM/OC) ratio of 1.6 for Barcelona (Minguillón et al., 2011); insoluble sulfate, calculated as the difference between total sulfate (calculated from total S) and soluble sulfate;

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