



Speciation of organic fractions does matter for aerosol source apportionment. Part 2: Intensive short-term campaign in the Paris area (France)



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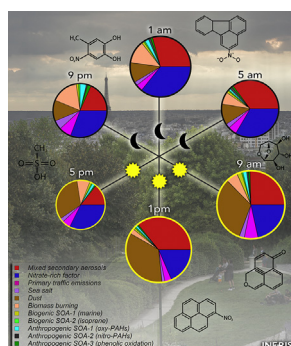
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HIGHLIGHTS

- Source apportionment using key primary and secondary organic molecular markers
- High resolution filter data allowed the understanding of the atmospheric processes.
- 7 SOA factors identified including 2 specific biogenic SOA and 3 anthropogenic SOA
- Traffic and biomass burning SOA resolved using 1-nitropyrene and methylnitrocatechols
- Role of night-time chemistry for SOA related to nitro-PAHs and phenolic compounds

GRAPHICAL ABSTRACT



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ABSTRACT

The present study aimed at performing PM₁₀ source apportionment, using positive matrix factorization (PMF), based on filter samples collected every 4 h at a sub-urban station in the Paris region (France) during a PM pollution event in March 2015 (PM₁₀ > 50 µg m⁻³ for several consecutive days). The PMF model allowed to deconvolve 11 source factors. The use of specific primary and secondary organic molecular markers favoured the determination of common sources such as biomass burning and primary traffic emissions, as well as 2 specific biogenic SOA (marine + isoprene) and 3 anthropogenic SOA (nitro-PAHs + oxy-PAHs + phenolic compounds oxidation) factors. This study is probably the first one to report the use of methylnitrocatechol isomers as well as 1-nitropyrene to apportion secondary OA linked to biomass burning emissions and primary traffic emissions, respectively. Secondary organic carbon (SOC) fractions were found to account for 47% of the total OC. The use of organic molecular markers allowed the identification of 41% of the total SOC composed of anthropogenic SOA (namely, oxy-PAHs, nitro-PAHs and phenolic compounds oxidation, representing 15%, 9%, 11% of the total OC, respectively) and biogenic SOA (marine + isoprene) (6% in total). Results obtained also showed that 35% of the total SOC originated from anthropogenic sources and especially PAH SOA (oxy-PAHs + nitro-PAHs), accounting for 24% of the total SOC, highlighting its significant contribution in urban influenced environments. Anthropogenic SOA related to

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nitro-PAHs and phenolic compounds exhibited a clear diurnal pattern with high concentrations during the night indicating the prominent role of night-time chemistry but with different chemical processes involved.

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1. Introduction

Atmospheric particulate matter (PM) plays an important role on climate and air quality (Boucher et al., 2013; Heal et al., 2012). To design effective PM concentration reduction strategies, their sources and contributions from each source need to be known thoroughly. Thus, several source apportionment methods have been developed for this purpose. Receptor-oriented modelling is one of the approaches that have been extensively used for PM source apportionment studies in the past decades (Belis et al., 2015; Hopke, 2016). The most commonly used receptor models include chemical mass balance (CMB) (Chow and Watson, 2002; Watson et al., 2002), positive matrix factorization (PMF) (Paatero and Tapper, 1994), and UNMIX (Henry, 1997). PMF is a powerful multivariate method that can resolve the dominant positive factors without prior knowledge of sources (Hopke et al., 2006; Kim et al., 2003; Shrivastava et al., 2007; Zhang et al., 2009).

Typically, PMF uses trace elements, and organic and elemental carbon (OC/EC), as well as secondary ions and metals as the input data matrix to explore the “co-variances” between species (Kim and Hopke, 2004; Kim et al., 2003). The use of tracers with high source specificity in the PMF model can enhance the interpretation of the factors. For example, levoglucosan is often used to trace the biomass burning source (Simoneit, 2002). The use of organic molecular markers in the PMF has resulted in considerable progress in the understanding of the organic aerosol (OA) fraction (Jaekels et al., 2007; Laing et al., 2015; Schembari et al., 2014; Shrivastava et al., 2007; Srimuruganandam and Shiva Nagendra, 2012; Waked et al., 2014; Wang et al., 2012; Zhang et al., 2009). However, these studies have often been based on 12 or 24 h sampling periods, making difficult to capture the information on “fast” chemical processes related to OA emissions and formation. The use of a higher time-resolution datasets (e.g. filter samplings every 6 h or less) may facilitate the understanding of the processes involved, for both primary and secondary sources, and the analysis of their diurnal cycles.

This paper is the second paper of a two-part series demonstrating that the speciation of the OA fraction is important for PM source apportionment. Note that, the use of organic molecular markers in source apportionment studies is based on the assumption that these compounds are chemically stable in the atmosphere (i.e. tracer compounds) (Schauer et al., 1996). Some molecules can undergo a decay in the atmosphere by photochemical processes involving sunlight and atmospheric oxidants and their use may cause a bias in the source apportionment. Nevertheless, the first paper (Srivastava et al., 2018) highlighted the advantage of using primary and secondary organic molecular markers to resolve sources rarely apportioned in the literature such as two types of primary biogenic organic aerosols (fungal spores and plant debris), as well as specific biogenic and anthropogenic secondary OA (SOA). This second paper focuses on the identification of sources during a major PM pollution event using high resolution filter data in order to apportion specific primary (POA) and secondary OA fractions using various and distinctive markers and to understand the atmospheric chemical processes involved.

2. Experimental

2.1. Monitoring site and sampling period

Measurements were conducted at the ACTRIS SIRTA atmospheric supersite (Site Instrumental de Recherche par Télédétection

Atmosphérique, 2.15° E; 48.71° N; 150 m a.s.l.; [http://sirta.ipsl.fr; Haeffelin et al. \(2005\)](http://sirta.ipsl.fr; Haeffelin et al. (2005))). This site is located approximately 25 km southwest from Paris city centre (Fig. S1), surrounded by forests, agricultural fields and small villages, and is representative of suburban background conditions of the Paris region (Crippa et al., 2013a; Petit et al., 2017a; Petit et al., 2014; Sciare et al., 2011). An intensive campaign was performed from 6 to 21, March 2015. The late winter-early spring period was chosen on purpose as intense PM pollution events are usually observed in Northern France (and Europe) during this period of the year due to the combination of significant residential emissions, manure spreading, stagnant atmospheric conditions favouring the accumulation of pollutants and possible photochemical processes enhancing the formation of secondary aerosols (Bressi et al., 2013; Crippa et al., 2013a; Dupont et al., 2016; Favez et al., 2012; Fröhlich et al., 2015; Petit et al., 2017a; Petit et al., 2014; Sciare et al., 2011; Waked et al., 2014).

2.2. Sample collection and co-located measurements

PM₁₀ samples (Tissu-quartz fibre filter, Pallflex, Ø = 150 mm) were collected every 4 h from 6 to 21, March 2015 using a high-volume sampler (DA-80, Digital; 30 m³ h⁻¹). Prior to sampling, quartz fibre filters were pre-heated at 500 °C for 12 h. After collection, samples were wrapped in aluminium foils, sealed in polyethylene bags, and stored at -20 °C until analysis. Shipping of the samples to the different laboratories for analyses have been done by express post using cool boxes (<5 °C). A total of 92 samples and 5 field blanks were collected and analysed for an extended chemical characterization following the protocols described in Section 2.3.

PM₁₀, Black Carbon (BC), NO_x and O₃ concentrations were measured using co-located online analysers: TEOM-FDMS (1405F model, Thermo), multi-wavelength aethalometer (AE33 model, Magee Scientific), T200UP and T400 monitors (Teledyne API), respectively. Moreover, assuming that biomass burning and fossil fuel combustion were the two predominant combustion sources, BC from wood burning (BC_{wb}) and fossil fuel (BC_{ff}) emissions were estimated using the so-called “aethalometer model” (Drinovec et al., 2015; Sandradewi et al., 2008). The AE33 instrument uses a dual-spot technology which provides the automatic compensation (k) of the aerosol loading effect (Drinovec et al., 2015) over the 7 wavelengths of measurements of BC (from near UV to near IR). An inaccurate automatic compensation has been observed for several days due to high scattering during the pollution episode linked to high ammonium nitrate concentrations. The associated data were thus manually corrected with fixed k values as explained by Petit et al. (2017a) to improve the separation between BC_{wb} and BC_{ff}. Finally, meteorological parameters such as temperature, relative humidity (RH), wind direction, and wind speed were obtained from nearby weather station (about 5 km).

2.3. Analytical procedure

A total number of 71 different chemical species have been quantified on filter samples. Major ions (Cl⁻, NO₃⁻, SO₄²⁻, NH₄⁺, Ca²⁺, Na⁺, Mg²⁺, K⁺), methanesulfonic acid (MSA) and oxalate (C₂O₄²⁻) were analysed using ion chromatography (Guinot et al., 2007). EC/OC was measured using a Sunset lab analyser and the EUSAAR-2 thermal protocol (Cavalli et al., 2010; CEN, 2017). Seven metal elements (namely Ca, Ti, Mn, Fe, Ni, Cu, and Pb) were quantified by PIXE (particle-induced X-ray emission) (Lucarelli et al., 2018; Lucarelli et al., 2011). Sugars, including known biomass burning markers (levoglucosan, mannosan

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