



Carbonaceous particles and aerosol mass closure in PM_{2.5} collected in a port city



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ABSTRACT

Mass concentrations of PM_{2.5}, mineral dust, organic carbon (OC) and elemental carbon (EC), water-soluble organic carbon (WSOC), sea salts and anthropogenic metals have been studied in a city-port of south Italy (Brindisi). This city is characterized by different emission sources (ship, vehicular traffic, biomass burning and industrial emissions) and it is an important port and industrial site of the Adriatic sea. Based on diagnostic ratios of carbonaceous species we assess the presence of biomass burning emissions (BBE), fossil fuel emissions (FFE) and ship emission (SE). Our proposed conversion factors from OC to OM are higher than those reported in the literature for urban site: the reason of this could be due to the existence of aged combustion aerosols during the sampling campaign (WSOC/OC = 0.6 ± 0.3).

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1. Introduction

It is known that carbonaceous species constitute a major, sometimes dominant, fraction of atmospheric particulate matter (Querol et al., 2013; Srinivas and Sarin, 2014; Pio et al., 2011). Carbonaceous aerosol is commonly divided into an organic carbon (OC) and an elemental carbon (EC) fraction. Both of these have important roles and effects on climate and health because of their physical and chemical characteristics (Pio et al., 2011; Viidanoja et al., 2002). Due to its special surface properties, EC provides a good adsorption site for many semi-volatile compounds such as the PAHs (Pio et al., 2011 and therein references). OC is an effective light scatterer and may contribute significantly to both visibility degradation and the direct aerosol climate forcing (Tegen et al., 1997; Malm and Day, 2000). A significant fraction of the particulate OC is water soluble, making it important to various aerosol–cloud interactions (Corrigan and Novakov, 1999; Decesari et al., 2000). Finally, carbonaceous species have the potential to influence many heterogeneous reactions involving atmospheric aerosols and trace gases (Grgić et al., 1998).

Despite the very large proportion of carbonaceous aerosols in particulate matter PM_x (PM₁₀, PM_{2.5} and PM₁) and the consequent relevance of this component for air quality, their origins are not fully understood and probably their contribution to PM_x load vary largely across the different regions in Europe (Querol et al., 2013 and therein references). The major sources of these species are biomass, bio-fuel

burning emission (BBE) (Gonçalves et al., 2011), and fossil-fuel combustion (FFC) (Hamad et al., 2015). The origin of OC, especially of SOA (secondary organic aerosol), is a hot topic in atmospheric sciences and a recent WHO report (REVIHAAP report, WHO, 2013) indicates that further studies are needed to characterize the health effects of SOA. Another recent WHO report (WHO, 2012) also evidences clear health consequences from the increased concentrations of EC, although this is probably not only due to EC per se but to the OC compounds adsorbed on EC particles.

During long-range atmospheric transport, oxidation of organic compounds is a prime interest in assessing the organic matter (OM) to OC ratio. OM/OC conversion factor ($f_{OM/OC}$) is not site or time dependent and in the literature, in relationship with the extent of OM oxidation and secondary organic aerosol (SOA) formation, values for $f_{OM/OC}$ from 1.2 for fresh aerosol to 2.6 for aged aerosol have been suggested (Chow et al., 2015 and therein references). Different studies have estimated the conversion factor using different methods such as: (a) measuring the distribution of functional groups by FTIR spectroscopy to estimate composite organic carbon from the number of carbon bonds present and organic mass from the molecular mass of each functional group (Russell, 2003; Gilardoni et al., 2007; Liu et al., 2009); (b) estimating aerosol organic matter as the difference of total aerosol mass and the sum of the other major species, namely mass balance approach (Sciare et al., 2005; Bae et al., 2006); and (c) examining spectral data using Aerosol Mass Spectrometer (Aiken et al., 2008; Chan et al., 2010).

In the present study, the reconstructed mass balance method for estimating the OM/OC ratio is applied to data collected at the port city of Brindisi (Italy). The estimation of $f_{OM/OC}$ could give more information

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on the aerosol ageing and formation or transport of SOA in the Mediterranean basin. In fact, the Mediterranean area is characterized by complex meteorology that favours the ageing of pollutants, thus inducing high levels of aerosols and photo oxidant gases (Rodríguez et al., 2002; Pateraki et al., 2013). Moreover, in our study we also assessed the carbonaceous species (OC and EC, water-soluble fraction of organic carbon) and their diagnostic ratios (OC/EC, $nss-K^+/EC$ and $nss-SO_4^{2-}/EC$) which provide crucial information regarding the formation and the source characteristics of carbonaceous particles. These local aerosol closure experiments, performed in the Mediterranean Sea, will bring a more comprehensive picture of the aerosol chemical composition in a region that is surrounded by strong sources of dust, sulphate and carbonaceous aerosol (Sciare et al., 2005 and therein references).

2. Experimental details

2.1. Site description

The sampling site was located in the Brindisi harbour area (Fig. 1), in the Apulia region (south-eastern part of Italy) ($40^{\circ}38'09''$ N, $17^{\circ}57'15''$ E). The sampling instruments were located on the roof of the ASI building (Consorzio per l'Area di Sviluppo Industriale) at 13 m above the ground level and, as shown in Fig. 1, it is the first building next the harbour and there are not higher buildings close to it. This allows to exclude the influence of near buildings on wind direction measured in the sampling site.

The Brindisi area is characterised by a complex scheme of emission sources including: urban emissions (88,500 inhabitants), harbour-related emissions (harbour traffic accounted yearly for >9.5 million tons, over 520,000 passengers and over 175,000 vehicles), airport emissions, petrochemical emissions, power-plant emissions and several artisan activities.

Two sequential low-volume samplers ($2.3 \text{ m}^3 \text{ h}^{-1}$) were used for PM_{2.5} collection: a Thermo ESM Andersen (FH95SEQ) sampler was used with Teflon filters and an Explorer Plus (Zambelli) sampler was used with quartz fibre filters. The two samplers were operated for 24 h according to the European standard (EN12341) with sampling starting at midnight. Teflon samples were used for the determination of the concentrations of water-soluble species and quartz samples for the determination of metal concentrations. In total 100 samples were collected in the period between June and October 2012.

Meteorological conditions such as wind, temperature, relative humidity and atmospheric pressure were obtained from two independent meteorological stations, one located inside the harbour area and the other located in the urban area of Brindisi at approximately 2.5 km from the sampling site.

2.2. Sample collection and analysis

All filters were weighed before and after sampling with an analytical balance (Sartorius CP2/F, reading precision $1 \mu\text{g}$) after stabilizing for 48 h. Temperature differences between pre-sampling and post-sampling weighing were usually within 1°C and relative humidity differences within 10%. The measurements on Teflon substrates were taken as reference for PM_{2.5} because of the lower uncertainty with respect to the concentrations on quartz substrates. The daily concentrations of quartz and Teflon substrates were similar, with a Pearson correlation coefficient of 0.87 and an average value of $15.1 \mu\text{g m}^{-3}$ for Teflon substrates and of $14.2 \mu\text{g m}^{-3}$ for quartz substrates.

The Teflon filters were analyzed for the evaluation of concentration of soluble ionic species (Cl^- , SO_4^{2-} , NO_3^- , $\text{C}_2\text{O}_4^{2-}$, NH_4^+ , Na^+ , K^+ , Mg^{2+} and Ca^{2+}) via High Performance Ion Chromatography (HPIC, Dionex DX-500 System). Each Teflon filter has been extracted in Milli-Q water: two successive extractions of 20 min in an ultrasonic bath were

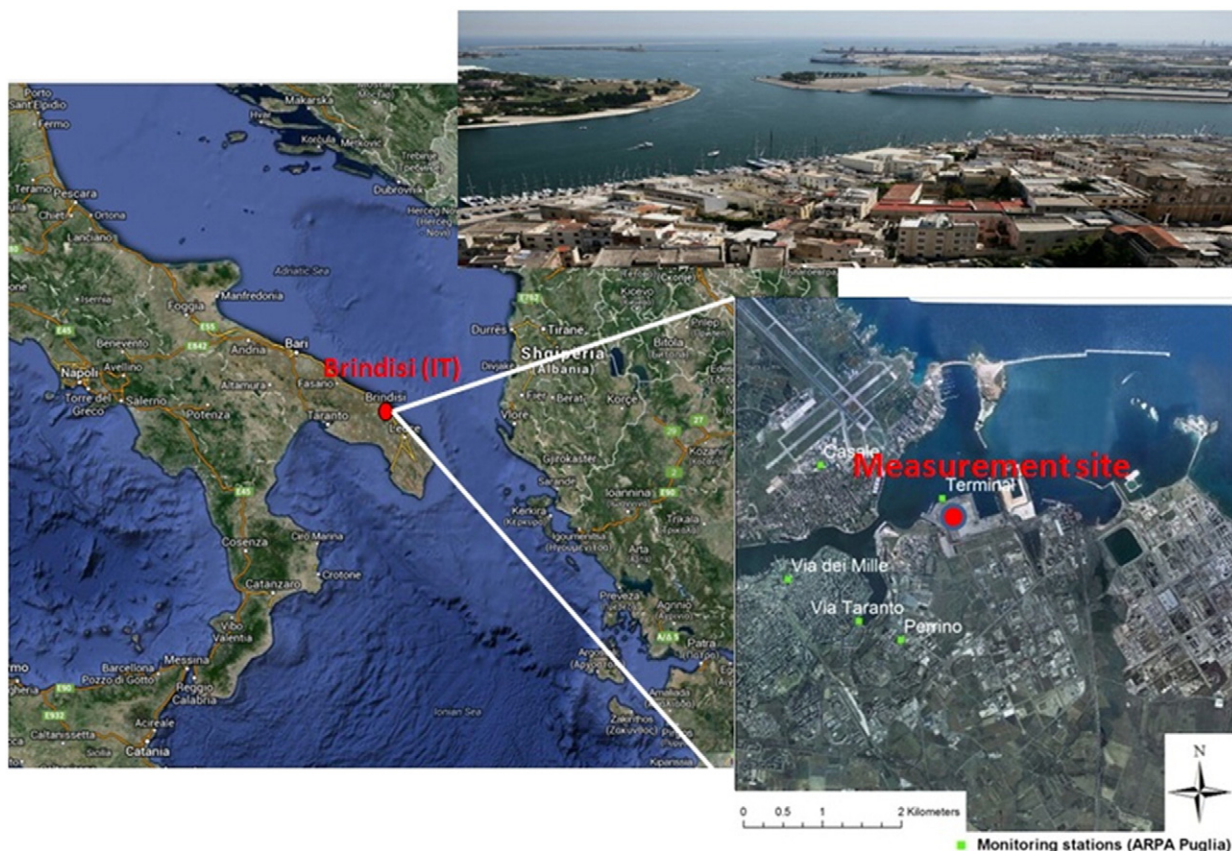


Fig. 1. Map of the southern part of Italy showing the position of Brindisi and satellite image showing the measurement site.

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