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Source apportionment of size-segregated atmospheric particles based on the major water-soluble components in Lecce (Italy)

D. Contini [a,*], D. Cesari [a], A. Genga [b], M. Siciliano [b], P. Ielpo [a,c], M.R. Guascito [b], M. Conte [a]

[a] Istituto di Scienze dell'Atmosfera e del Clima, ISAC-CNR, Lecce, Italy
[b] Dipartimento di Scienze e Tecnologie Biologiche e Ambientali, Università del Salento, Lecce, Italy
[c] Istituto di Ricerca Sulle Acque, IRSA-CNR, Bari, Italy

HIGHLIGHTS

- Size-segregated source apportionment of aerosol was performed using PMF.
- Limits in identification of traffic and nitrate aerosol sources were discussed.
- Losses of chemicals in the multi-stage impactor were estimated as 19% of PM10.
- The effects of impactor losses on source apportionment were assessed.
- The method used allowed reasonable apportionment of accumulation and coarse mode.

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ABSTRACT

Atmospheric aerosols have potential effects on human health, on the radiation balance, on climate, and on visibility. The understanding of these effects requires detailed knowledge of aerosol composition and size distributions and of how the different sources contribute to particles of different sizes. In this work, aerosol samples were collected using a 10-stage Micro-Orifice Uniform Deposit Impactor (MOUDI). Measurements were taken between February and October 2011 in an urban background site near Lecce (Apulia region, southeast of Italy). Samples were analysed to evaluate the concentrations of water-soluble ions (SO_4^{2-}, NO_3^-, NH_4^+, Cl^-, Na^+, K^+, Mg^{2+} and Ca^{2+}) and of water-soluble organic and inorganic carbon. The aerosols were characterised by two modes, an accumulation mode having a mass median diameter (MMD) of 0.35 ± 0.02 μm, representing 51 ± 4% of the aerosols and a coarse mode (MMD = 4.5 ± 0.4 μm), representing 49 ± 4% of the aerosols. The data were used to estimate the losses in the impactor by comparison with a low-volume sampler. The average loss in the MOUDI-collected aerosol was 19 ± 2%, and the largest loss was observed for NO_3^- (35 ± 10%). Significant losses were observed for Ca^{2+} (16 ± 5%), SO_4^{2-} (19 ± 5%) and K^+ (10 ± 4%), whereas the losses for Na^+ and Mg^{2+} were negligible. Size-segregated source apportionment was performed using Positive Matrix Factorization (PMF), which was applied separately to the coarse (size interval 1–18 μm) and accumulation (size interval 0.056–1 μm) modes. The PMF model was able to reasonably reconstruct the concentration in each size-range. The uncertainties in the source apportionment due to impactor losses were evaluated. In the accumulation mode, it was not possible to distinguish the traffic contribution from other combustion sources. In the coarse mode, it was not possible to efficiently separate nitrate from the contribution of crustal/resuspension origin.

1. Introduction

Atmospheric particles are an important component of the earth's atmosphere, playing an important role in determining visibility (Watson, 2002), in the deterioration of air quality and human health (Pope and Dockery, 2006), in the pollution transfer to ecosystems (through deposition), in the direct and indirect effects on the radiative balance, and in the climate system (Seinfeld and Pandis, 1988). All of these effects and phenomena are strongly dependent on the size distributions of aerosols and on the distribution of the chemical species in the different size ranges. For example, exposure to elevated concentrations of fine particulate matter is associated with adverse health effects in epidemiological studies (Laden et al., 2000; Dominici et al., 2005; Ostro et al., 2006). The smallest particles (ultrafine particles with a diameter less than 100 nm) showed the ability to cross cell membranes (Li et al., 2003), which may greatly influence their health effects. Organic aerosols have the potential to scatter solar radiation and to reduce the

* Corresponding author at: Istituto di Scienze dell'Atmosfera e del Clima, ISAC-CNR, Str. Prv. Lecce-Monteroni km 1.2, 73100 Lecce, Italy. Tel.: +39 0832 298919; fax: +39 0832 298716.
E-mail address: d.contini@isac.cnr.it (D. Contini).

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hygroscopicity of inorganic species. This causes variations in the light scattering properties of aerosols with changes in the relative humidity (Deshmukh et al., 2012a). Among organic aerosols, water-soluble organic carbon (WSOC) has a relevant nucleating ability, mostly observed in cloud condensation nuclei (Cruz and Pandis, 2000; Decesari et al., 2005). The ability of water-soluble organic particles to form cloud condensation nuclei (CCN) has been explained by considering the effects of both the hygroscopic and the surface-active properties of WSOC, which, in turn, depend on the WSOC chemical composition (Shulman et al., 1996; Facchini et al., 1999).

The size distribution of fine particulate matter can provide information about the chemical and the physical processes affecting aerosol concentrations and compositions as they are transported in the atmosphere (Cabada et al., 2004). This may be particularly important in the Mediterranean basin, because it is a region subject to fluxes of anthropogenic and natural particles from northern Europe and from the African continent as a consequence of its geographic location. Furthermore, the Mediterranean area is characterised by complex meteorology that favours the ageing of pollutants, thus inducing high levels of aerosols and photo-oxidant gases (Rodriguez et al., 2002; Pateraki et al., 2013).

The contributions of the different natural and anthropogenic sources acting on a specific site are strongly size dependent. Recently, it has been shown that source apportionment of particle number populations can be successfully performed by statistically analysing the size distributions (Dall'Osto et al., 2012). Over the years, efforts have been made using different approaches to obtain knowledge about the composition of atmospheric aerosols as a function of size, using statistical receptor models on data collected with multi-stage impactors. However, the size-segregated contributions of the different sources are still not completely understood (Fang et al., 2006; Gietl and Klemm, 2009; Ny and Lee, 2011; Dordevic et al., 2012). The main limitations in the use of cascade impactors in source apportionment are related to the potential losses of particles in the impactors and to the generally limited number of species analysed. This is especially true in low-flow-rate impactors, so that analyses are often limited to water-soluble ions and carbonaceous species (Gietl and Klemm, 2009).

The objectives of this work were to characterise the size distributions of aerosols and of the water-soluble species (major ions and organic and inorganic carbon) in an urban background site in southeastern Italy. Furthermore, the data collected were used to perform size-segregated source apportionment of aerosols. The samples were collected with a 10-stage Micro-Orifice Uniform Deposit Impactor (MOUDI) and analysed to perform source apportionment using the Positive Matrix Factorization (PMF) receptor model to investigate the differences in the source attribution to the different size ranges and to the accumulation (stages S6–S10, size interval of 0.056–1 μm) and coarse (stages S1–S5, size interval of 1–18 μm) modes of aerosol. Losses of chemical species in the impactor sampling are evaluated, and their influence on the source apportionment results is assessed. Specific aspects related to the potentiality and limits of the size-segregated source apportionment based on water-soluble species are also discussed.

2. Methods and procedures

2.1. Experimental site and instruments

The measurement site was located in the experimental field of ISAC-CNR, inside the University Campus in Lecce (Apulia region, Fig. 1). The site is located in the Salentum peninsula approximately 4 km SW of the town of Lecce (SE Italy, coordinates 40°20′N, 18°06′E). The largest industrial settlements of the Apulia region are: the area of Taranto and the area of Brindisi, which are located between 30 km and 80 km to the W and NW of the site, respectively. The site can be considered an "urban background" that is not strongly influenced by traffic or industrial emissions (Belosi et al., 2006; Contini et al., 2010a). Aerosol samples were collected at 2.5 m above the ground using a Micro-Orifice Uniform Deposit Impactor (MOUDI II, MSP, Model 120R) operating at 30 l/min,

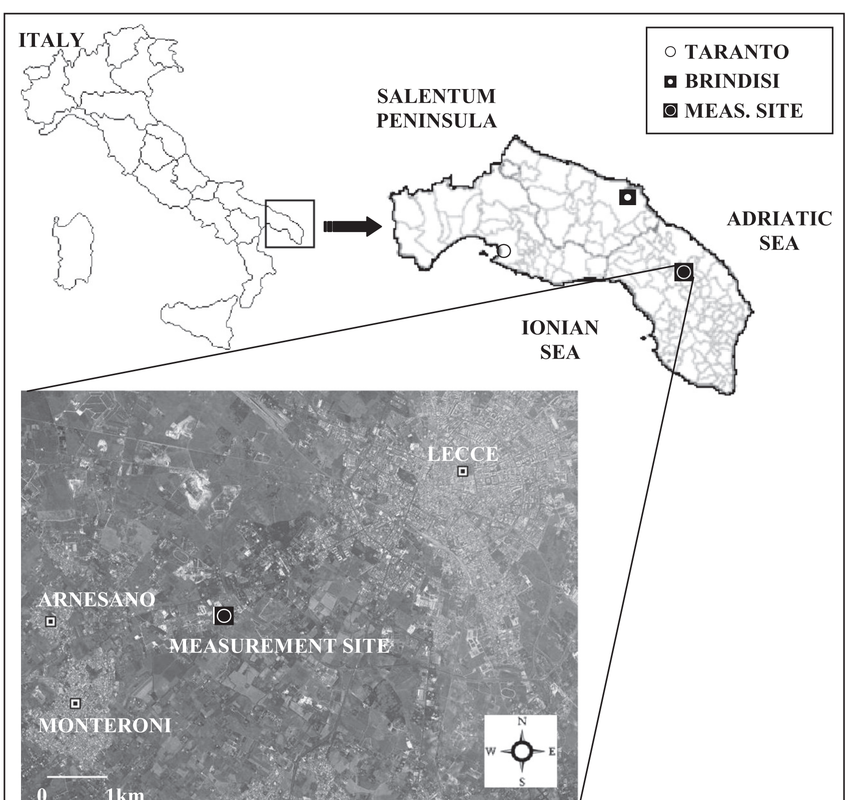

Fig. 1. Map of the measurement area, including the position of the urban areas of Lecce, Arnesano and Monteroni and the position of the main industrial sites of Taranto and Brindisi.