## Size-segregated source apportionment of aerosol using water soluble components

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The aerosol size distributions and the contents of chemical species in the different size-ranges play an important role in determining visibility, air quality and human health effects. The aim of this work is to perform a Source Apportionment (SA) study to evaluate the source contributions to each size range using only water soluble major components and discussing the potentiality and the limits of the approach used. Aerosol samples were collected in an urban background site (Lecce, SE of Italy), using a 10-stages Micro-Orifice Uniform Deposit Impactor (MOUDI II, MSP, Model 120R) operating at 30 l/m. The impactor was equipped with aluminium substrates (47 mm, MSP Corporation) and the 10 stages had nominal cut-off sizes ( in µm): 10, 5.6, 3.2, 1.8, 1.0, 0.56, 0.32, 0.18, 0.10 and 0.056. Teflon filters (47 mm, Pall Corporation) were used as back-up filters as well as sampling substrates for simultaneous measurement with a standard low-volume PM10 sampler (Tecora) used for evaluation of losses in the impactor. The average loss resulted in 19% ( $\pm$  2%) with the major losses associated to nitrate  $(35\% \pm 10\%)$ . Between February and October 2011, 21 samples were collected: 18 samples were sampled for 48 h and 3 sample for 72 h. Collected samples were stored in Petri dishes at 4 °C until analysed. Gravimetric analysis was performed using a microbalance (Sartorius CP2/F, sensitivity 1µg). Soluble ionic species were determined via High Performance Ion Chromatography (HPIC, Dionex DX-500 System), for  $SO_4^{2-}$ ,  $NO_3^{-}$ ,  $NH_4^{+}$ ,  $Cl^{-}$ ,  $Na^+$ ,  $K^+$ ,  $Mg^{2+}$  and  $Ca^{2+}$ , and with a Shimadzu TOC analyzer (model 5050), for Water Soluble Organic Carbon (WSOC) and Water Soluble Inorganic Carbon (WSIC) fractions. Chemical analysis show that measured ions accounted, on average, for 39.6% of PM10, WSOC accounts for 13.1% while WSIC for 1.8% of PM10.

The average size distributions of collected aerosol and of water soluble species were fitted to lognormal distributions. Aerosol presented a bimodal distribution that include an accumulation mode (Mass Median Diameter =  $0.35 \pm 0.02 \mu$ m), accounting in average terms for  $51 \pm 4$  % of PM10, and a coarse mode (MMD =  $4.5 \pm 0.4 \mu$ m), accounting in average terms for  $49 \pm 4$ % of PM10. A bimodal distribution was observed for SO<sub>4</sub><sup>2-</sup>, NH4<sup>+</sup>, WSOC and K<sup>+</sup> (more abundant in the accumulation mode) and for Cl<sup>-</sup>, Ca<sup>2+</sup>, WSIC and NO<sub>3</sub><sup>-</sup> (more abundant in the coarse mode). Na<sup>+</sup> and Mg<sup>2+</sup> presented a mono-modal distribution in the coarse range. Size-segregated composition of collected aerosol was used for SA using the EPA PMF3.0 software. A different procedure was used with respect to similar works

reported in literature (Chan et al., 2000; Fang et al., 2006), using two input datasets, representing the accumulation mode and the coarse mode. The concentrations of a specific species in the different size ranges were considered as independent samples of the same species in the PMF analysis rather than independent species of the same sample. Five sources were individuated in the accumulation mode (marine, nitrate and crustal. sulphate, combustion/traffic emission) and four sources in the coarse mode (marine, crustal, sulphate and nitrate). The size-segregated contributions were reported in Figure 1. There are some differences in the sources observed in the two modes: the accumulation mode nitrate, of secondary origin, is different from the nitrate observed in the coarse mode, that is mainly sodium nitrate and nitrate salts associated to crustal dust. Results showed that it has been possible to evaluate the source contributions in the different sizeranges as well as the contribution to the accumulation and coarse mode separately. In general, SA results were in agreement with indirect estimates of marine and ammonium sulphate contributions. However, there are limits to the application of the PMF model limited to the water soluble species. Specifically, in the accumulation mode it was not possible to distinguish between traffic source and the contribution of other combustion sources (i.e., biomass burning). In the coarse mode it was not possible to separate the sodium nitrate factor from a contribution of nitrate of crustal/resuspension origin.



Figure 1. Size-segregated source apportionment (the indicated diameters are the geometric mean diameters).

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