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Tracing of aerosol sources in an urban environment using carbonaceous, major ions, Sr isotope, and mineralogical compositional data

ANDREIA PAULA1, SONIA LOPES3, SARA RIBEIRO2, JOSE FRANCISCO SANTOS3, CARLA PATINHA2, EDUARDO ANSELMO3, ROSARIO SOARES4, REGINA DUARTE1,4, ARMANDO DUARTE1
1Department of Chemistry & CESAM, University of Aveiro, 3810-193 Aveiro, Portugal
2Department of Geosciences & GeoBioTec, University of Aveiro, 3810-193 Aveiro, Portugal
3Department of Chemistry & CICECO, University of Aveiro, 3810-193 Aveiro, Portugal

Understanding the chemical composition of atmospheric particulate matter (PM), particularly with a diameter less than 2.5 μm (PM_{2.5}), has become an emergent topic in atmospheric research due to its relation to climate change and serious health issues. In this study, a comprehensive assessment of the carbonaceous (organic carbon, water-soluble organic carbon, elemental carbon), major water-soluble inorganic ions (SO_4^{2-}, NO_3^-, NH_4^+, Cl^-, K^+, Na^+), and Sr isotopes of PM_{2.5} samples has been performed for a one-year period in an urban environment at the Western European Coast (Aveiro, Portugal). The concentration of each component was found to be highly dependent on the seasonal events, with the carbonaceous component being the major constituent of the PM_{2.5} in almost all seasons. The concentration of SO_4^{2-} is higher in samples collected during warm seasons than in those of colder seasons, which is indicative of secondary formation (mediated by sunlight) in the atmosphere [F. Zhang et al., 2014]. On the other hand, the concentration of the more volatile NO^+ ion is highest for samples collected in colder seasons, which can be explained by the combined effects of lower air temperature and an increase of biomass burning and vehicles emissions during these seasons [F. Zhang et al., 2014]. The equivalent molar ratio of cations to anions was also computed, indicating that the PM_{2.5} is more acidic during high air temperature periods probably due to the highest accumulation of acidic components (e.g., SO_4^{2-}) during this period [J. Zhang et al., 2014].

For the Sr/Sr isotope ratios, it was possible to distinguish the labile (acid-leached) from the natural (remaining residue) Sr components (Fig.1). The highest Sr/Sr ratios were observed for the residues embedded in the collection medium, being this indicative of a natural crustal dust source. The Sr/Sr isotope ratios verified for the leachates are of the same order of magnitude of the reference value for seawater, thus suggesting that the urban PM_{2.5} samples have also a predominant marine influence. These findings are also in agreement with the mineralogical composition data, which shows the presence of gypsum with a clear marine origin on almost all samples, besides feldspar with an anthropogenic origin, and aluminosilicates from soil dust.

Fig. 1: Sr/Sr isotope ratios in residues and leachates of PM_{2.5} samples collected in the city of Aveiro (Portugal) during one-year period and under different air mass trajectories.
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Long-term size-segregated particle (PM10, PM2.5, PM1) characterization study in the rural background of Germany – influence of mass inflow and season

GERALD SPINDLER1, ACHIM GRÖNER1, KONRAD MÜLLER1, BASTIAN STIEGER1, MARKUS WALLASCH2, YOSUHEI INUMA1, HARTMUT HERRMANN1
1Leibniz-Institute for Tropospheric Research (TROPOS), Permoserstraße 15, 04318 Leipzig, Germany
2Federal Environment Agency, Wörlitzer Platz 1, 66844 Dessau-Roßlau, Germany

Long-term studies allow quantifying the influence of decreasing emissions to the concentrations. The particle mass concentration is also influenced by particle formation from precursors and long-range transport. The rural-background site Melpitz (12°56'E, 51°32'N, 86 m asl) is located in Germany in the Saxony lowlands near Torgau in the glacial valley of the river Elbe (Spindler et al. 2013). The Melpitz station is integrated in ACTRIS and EMEP and representative for a large area in central Europe.

High-Volume (HV) quartz filter samples for particles < 10 µm aerodynamic-diameter (PM10) were collected daily (since November 1992). Particles PM2.5 and PM1 were collected daily respective every six days (since January 2003). The determination of the particle mass was done gravimetrically. Main water-soluble ions (NO3-, SO42-, Cl-, Na+, NH4+, K+, Mg2+, Ca2+) were analyzed by ion chromatography. The determination of OC and EC (available since 2003) performed by a two-step thermographic method (according to VDI 2465 part 2). PAH and n-alkanes were determined by CPP-GC-MS (Curie point 510°C) for PM10 samples in 2012.

Daily samples were evaluated at first as yearly means and quarterly means (per year), at second for spatial and seasonal discrimination days were sorted for air mass transport from a western sector (W, 210-320°, mostly maritime influenced) and a broad eastern sector (E, 35-140°, continental influenced) using backward trajectories (96 h) Days with a strong change in transport direction (often more local influenced by inversion) were excluded here. A seasonal discrimination was done for winter (Wi, November-April) and for summer (Su, May-October). The result are the four categories WNW, WIE, SuW and SuE in which 68% of the whole time (November 1992 until December 2014) can be sorted (53.3% for W, 15.1% for E).
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Andreaia Paula¹, Sónia Lopes¹, Sara Ribeiro², José Francisco Santos², Carla Patinha², Eduardo Anselmo², Rosário Teixeira³, Regina Duarte¹,³ Armando Duarte¹

¹ CESAM & Department of Chemistry, University of Aveiro, 3810-193 Aveiro, Portugal
² GeBioTec & Department of Geosciences, University of Aveiro, 3810-193 Aveiro, Portugal
³ CICECO & Department of Chemistry, University of Aveiro, 3810-193 Aveiro, Portugal

Introduction
Understanding the chemical composition of atmospheric particulate matter (PM), particularly with a diameter less than 2.5 μm (PM₂.₅), has become an emergent topic in atmospheric research due to its relation to climate change and serious health issues. This study aims at performing a comprehensive assessment of:
• carbonaceous species (organic carbon, water-soluble organic carbon, elemental carbon) and major water-soluble inorganic ions (SO₄²⁻, NO₃⁻, NH₄⁺, Cl⁻, K⁺, Na⁺) in PM₂.₅ samples collected over 1-year period in an urban environment at the Western European Coast (Aveiro, Portugal);
• ⁸⁷Sr/⁸⁶Sr isotope ratios and mineralogical composition of the urban PM₂.₅ samples.

Chemical Composition of PM₂.₅

- During the course of the measuring period, seasonal PM₂.₅ mass concentrations were extremely variable.
- The carbonaceous component is the major constituent of the PM₂.₅ samples.
- The average ambient concentrations of SO₄²⁻ and NO₃⁻ follow an opposite trend between warm and cold seasons, the reason for which is depicted in Fig. 2.

Conclusions
The highest ⁸⁷Sr/⁸⁶Sr ratios were found in the PM₂.₅ residues, being this indicative of a natural crustal dust source. The ⁸⁷Sr/⁸⁶Sr ratios of the PM₂.₅ leachates are of the same order of magnitude of the reference value for seawater, thus suggesting that the urban PM₂.₅ samples have also a predominant marine influence.

References

Acknowledgements
A similar profile of mineral phases was observed for all PM₂.₅ samples: 1) aluminosilicates are likely related to the resuspension of soil dust, 2) gypsum is typical of marine aerosols, and 3) feldspar has an anthropogenic origin.