

6⁺-year source apportionment of fine carbonaceous aerosols at SIRTa (Paris region, France) using Aerosol Chemical Speciation Monitor (ACSM) and multi-wavelength Aethalometer (AE33)

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Keywords: submicron aerosol, long-term measurements, organic aerosol, black carbon, sources

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Introduction

Carbonaceous aerosols account for a major fraction of fine atmospheric particles and play a significant role on air quality and climate forcing. Although various primary sources and secondary transformation processes have already been identified and quantified worldwide, their characterization over long-term time periods remains poorly documented due to challenging highly-time resolved continuous observations. We report here the first source apportionment of carbonaceous aerosols for a 6⁺-year period from on-line in situ measurements. This unique dataset has been obtained from November 2011 to March 2018 at the SIRTa facility, a research monitoring platform which is representative of background air quality in the Paris region (France).

Methods

Submicron non-refractory aerosols were measured using an aerosol chemical speciation monitor (ACSM, Aerodyne Res. Inc.) with a 30-min time resolution, while equivalent mass concentration of BC was measured by a seven-wavelength aethalometer (AE33, Magee Sci.) with 1-min frequency. The relative source contributions of wood burning and traffic emissions to black carbon were estimated using the 'aethalometer model'. Positive matrix factorization, based on ME-2 engine, was applied to apportion the organic aerosol (OA) sources using the advanced Source Finder toolkit (SoFi).

Results

Two primary OA (POA) and two oxygenated OA (OOA) factors were identified and quantified over the entire studied period. POA factors were designated as hydrocarbon-like OA (HOA) and biomass burning OA (BBOA). The latter factor presented a significant seasonality with higher concentrations in winter with significant monthly contributions to OA (18-33%) due to enhanced residential wood burning emissions. HOA mainly originated from traffic emissions, but was also

influenced by biomass burning in cold months. OOA factors were distinguished between their less- and more-oxidized fractions (LO-OOA and MO-OOA, respectively). These factors presented distinct seasonal patterns associated with different atmospheric formation pathways. A pronounced increase of LO-OOA concentrations and contributions (50-66%) was observed during summer, which may be mainly explained by secondary OA (SOA) formation processes involving biogenic gas precursors. Conversely high concentrations and OA contributions (32-62%) of MO-OOA during winter and spring seasons were partly associated with anthropogenic emissions and/or long-range transport from northeastern Europe. The contribution of the different OA factors as a function of OA mass loading highlighted the dominant roles of POA during pollution episodes in fall and winter, and of SOA for highest springtime and summertime OA concentrations. Finally, long-term trend analyses using Mann-Kendall test indicated a decreasing feature (of about 200 ng m⁻³ yr⁻¹) for MO-OOA, very limited or insignificant decreasing trends for primary anthropogenic carbonaceous aerosols (BBOA and HOA, along with the fossil fuel and biomass burning black carbon components), and no trend for LO-OOA over the 6⁺-year investigated period.

This work is part of the COLOSSAL European COST Action CA16109. It has also been supported by the FP7 ACTRIS and H2020 ACTRIS-2 projects (grant agreements no. 262254 and 654109), the DIM-R2DS program, as well as by the French ministry of Environment through funding of the reference laboratory for air quality monitoring (LCSQA) activities. Y. Z. acknowledges the PhD Scholarship from the China Scholarship Council (CSC).