

MULTI-ANNUAL SOURCE APPORTIONMENT AND ABSORBING PROPERTIES OF ORGANIC AEROSOLS IN NORTHERN FRANCE

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Summary

To quantify accurately the adverse effects of the particulate matter (PM) fine fraction on climate, it is necessary to identify emission sources and associate them with aerosol radiative properties over long periods. Source apportionment is a useful tool to quantify PM contributions especially for organic aerosols (OA). Four years of real-time optical and chemical observations were combined to characterize OA sources and their absorbing properties at the ATOLL platform in Northern France. Positive Matrix Factorization (PMF) shows high contribution (74%) of oxygenated OA (OOA), followed by 14% of BBOA (due to biomass burning) and 12% of HOA (related to traffic) on average for the mass concentrations. Meanwhile, OA are responsible for 27% of light absorption at 370 nm, showing a significant contribution of Brown Carbon (BrC). A Multiple Linear Regression (MLR) analysis applied to determine source-specific Mass Absorption Efficiencies (MAE) of the OA factors highlights BBOA as the dominant source of BrC light absorption (64%).

Introduction

In Europe, more than 400,000 premature deaths have been attributed to PM exposure, making the study of its sources essential to improve air quality. Northern France is affected by relatively high PM concentrations exceeding the new daily PM_{2.5} concentration of 15 $\mu\text{g m}^{-3}$, recommended by WHO (EEA, 2021), due to heavy traffic, high urban density, significant agricultural activities overlapped with transnational pollution transport (UK, Benelux, Germany, etc.).

Methodology and Results

Four years (10/2016-12/2020) of near real-time Aerosol Chemical Speciation Monitor (ACSM) and Aethalometer (AE33) measurements were performed on the ATOLL (ATmospheric Observations in liLLe) platform located on the rooftop of a University of Lille building. Non-refractory submicron particles (NR-PM₁) were at 9.74 $\mu\text{g m}^{-3}$ on average and dominated by OA (45.4%). To investigate OA sources, a PMF analysis using the SoFi Pro software (Datalystica Ltd., Villigen, Switzerland) was performed and the rolling PMF algorithm applied in order to consider minor temporal changes in the source profiles.

We identified two primary OA – hydrocarbon-like OA (HOA, mainly related to traffic) and biomass burning OA (BBOA, mostly residential wood combustion) – and two secondary ones (OOA). HOA showed a constant contribution to OA throughout the year (seasonal averages: 10-14%), while BBOA varied from 8 to 15% with a peak in winter due to increased emissions. OOA factors – average contribution of 74% – were distinguished between their less and more oxidized fractions (LO-OOA and MO-OOA, respectively). We assessed the effect of OA factors on PM absorbing properties in the UV range by deriving their MAE. The BrC absorption coefficient was calculated at 470 nm then we applied a MLR model using PM mass concentrations of the OA factors to derive source-specific MAE. Overall, excellent agreements (r^2 0.72, slope 0.78 and RSME 1.66) between observations and calculations were obtained for unconstrained MAE values. The main BrC contributors were found to be the freshly emitted aerosols (BBOA and HOA, with 64% and 18% of total BrC absorption, respectively) followed by the two OOA factors (Fig. 1), concluding that, in Lille the predominant BrC source is biomass burning.

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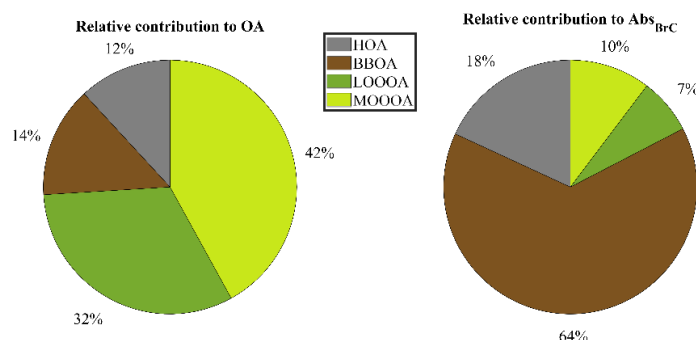


Fig. 1 Source apportionment of OA using rolling PMF algorithm (left) and relative contributions of OA factors to BrC absorption at 470 nm (right)