

Primary and secondary organic aerosol origin by combined gas-particle phase source apportionment

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Keywords: source apportionment, ME-2, organic aerosols, VOCs.

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Secondary organic aerosol (SOA), typically the dominant fraction of particulate organic mass (OA), remains poorly constrained. Its formation involves several unknown precursors, formation and evolution pathways and multiple natural and anthropogenic sources. In our work a combined gas/particle-phase source apportionment is applied to wintertime and summertime data collected in the megacity of Paris within the MEGAPOLI project to investigate SOA origin during both seasons (Crippa et al., submitted). Source apportionment was conducted by positive matrix factorization (PMF) analysis of combined mass spectra from an Aerodyne aerosol mass spectrometer (AMS) and a proton transfer reaction mass spectrometer (PTR-MS). PMF was implemented within a new source apportionment toolkit based on the multi-linear engine (ME-2) algorithm (Canonaco et al., in prep.).

The combined method improved apportionment of primary OA (POA) sources by the inclusion of gas-phase tracers. These tracers further allowed distinguishing between biogenic-influenced and continental/anthropogenic sources of SOA. We found that continental SOA was dominant during both seasons (24-50% of total OA), while contributions from photochemistry-driven SOA (9% of total OA) and marine emissions (13% of total OA) were also observed during summertime. A semi-volatile nighttime component was also identified (up to 18% of total OA during wintertime). The presence of benzene and PAN in the wintertime oxygenated continental factor suggested that this secondary fraction could be influenced by biomass smoke in aged air masses, consistent with ¹⁴C data.

Apportionment of the volatile organic compounds (VOCs, see Fig.1) indicated that

traffic was mostly the dominant source of the aromatic fraction. However, aged air masses (during winter) and nighttime air (during summer) were also significant sources. Cooking contributed an unexpectedly high fraction of the total aromatics, possibly due to the overlap of traffic and cooking emissions influenced by meteorology. The source apportionment of oxygenated VOCs revealed that secondary species dominated in both seasons, but especially summer. During wintertime acetic acid was also produced by wood burning emissions. Finally, biogenic emissions were produced by biomass burning during wintertime while “fresh” secondary species dominated in summer.

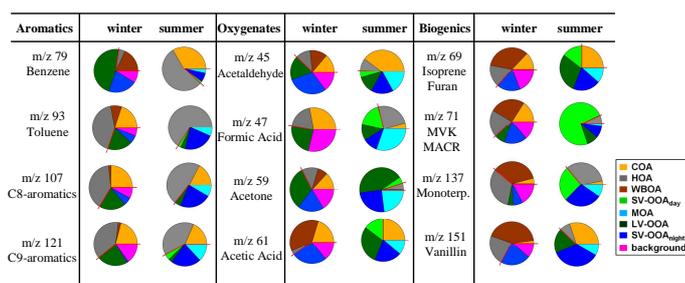


Figure 1. Source apportionment of aromatic, oxygenated and biogenic VOCs.

This research in the context of the MEGAPOLI project is financially supported by the European Community's Framework Program FP/2007-2011 under grant agreement n°212520, as well as the Swiss Federal Office for the Environment and Swiss National Science Foundation.

Canonaco, F., et al., in preparation.

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