

Size-Resolved Source Apportionment of PM₁₀ Organic Aerosol Measured with Aerosol Mass Spectrometry

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Atmospheric particles affect human health, climate and visibility. The extent and impact of these effects depend on both particle size and chemistry, which concurrently evolve in the atmosphere via direct emission or formation of new particles and subsequent condensation of semi-volatile species. Thus aerosol size and chemical composition are directly linked, and ideally should be measured simultaneously.

The Aerodyne aerosol mass spectrometer (AMS, DeCarlo et al., 2006) offers real-time, quantitative measurements of the size-resolved chemical composition of non-refractory submicron aerosol (organic aerosol (OA), Cl⁻, NO₃⁻, NH₄⁺, SO₄²⁻). The application of positive matrix factorization (PMF) techniques to AMS data have demonstrated that the mass spectra of bulk OA contain enough information to differentiate several factors, which can be related to different aerosol emission sources and/or formation processes. These factors typically include those related to primary emissions, such as hydrocarbon-like organic aerosol (HOA) from traffic, biomass smoke (BBOA), and cooking emission (COA). Secondary OA formed via the oxidation of volatile organic compounds can also be identified by this technique, termed oxygenated organic aerosol (OOA).

Most recently, the application of three dimensional PMF to online AMS data has been proposed to factorize the chemical composition data from particles in different size ranges (Ulbrich et al., 2012). However, such analysis is limited by the low signal-to-noise ratio of size-resolved AMS spectra (compared to those of bulk OA) and the negligible transmission efficiency of the conventional AMS for supermicron particles, although a newly developed aerodynamic lens system extends measurements into the PM_{2.5} range (Williams et al., in prep). Although supermicron particles constitute a considerable fraction of the total suspended particles, the chemical composition of their organic fraction remains largely unknown. It is thought that these particles include primary biological material (e.g. pollen, bacteria and spores) that can act as allergens and ice nuclei, affecting both public health and climate (Schneider et al., 2011).

In order to measure size-fractionated mass spectra, we explored the application of laboratory AMS measurements to aerosol filter samples. Such methodology based on offline sampling offers the separation of particles in discrete size ranges and allows measurement of particles through the PM₁₀ size range. The approach involves water extraction of the particulate matter from quartz filters by sonication and subsequent

nebulisation of the aerosol extract into the AMS. The OA extraction efficiency has been evaluated by comparison with AMS online measurements performed during the same period and is estimated at 80%. Method detection limits are estimated for organic aerosol, NO₃⁻, SO₄²⁻, and NH₄⁺ as 410 mgL⁻¹, 210 mgL⁻¹, 260mgL⁻¹, and 200 mg L⁻¹, respectively. Here, we present the application of the offline AMS approach to PM₁, PM_{2.5} and PM₁₀ filter samples collected at a rural site in Switzerland during summer and winter. Differences in aerosol fingerprints across particle sizes are examined and indicate a significantly different composition of coarse particles with higher contributions from fragments that are likely related to carbohydrates (e.g. m/z 29 and 60). Size-resolved PMF applied to these measurements will be discussed and compared with online AMS measurements to identify the contribution of primary biological particles.

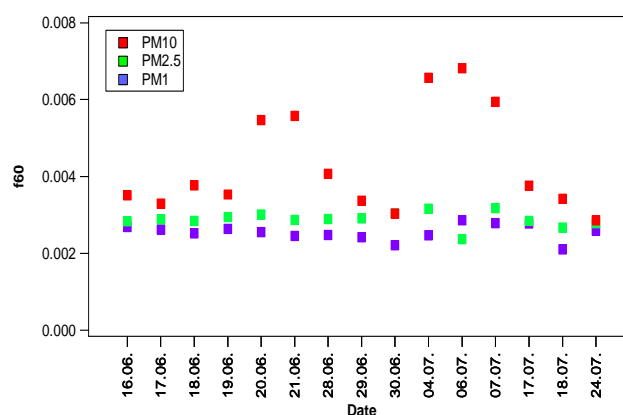


Figure 1. Time series of the f_{60} marker in 3 size fractions on 15 selected days with highest pollution during the EMEP campaign in Payerne 2012.

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DeCarlo, P.F. et al. (2006) *Anal. Chem.* **78**, 8281-8289.

Ulbrich et al. (2012) *Atmos. Meas. Tech.* **5**, 195-224.

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