Size-segregated long-term chemical analysis and source apportionment of the organic aerosol in the Northern hemisphere using offline aerosol mass spectrometry

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The development and field deployment of the Aerodyne aerosol mass spectrometer (AMS, Canagaratna et al., 2007) have greatly improved the air quality monitoring by providing real time measurements of the nonrefractory (NR) submicron aerosol (PM1) components. The application of factor analysis on the collected organic aerosol (OA) mass spectra enabled the efficient disentanglement of aerosol factors, which could be subsequently related to specific aerosol sources and processes. However, the cost and complex maintenance requirements of the AMS make its deployment at sufficient sites to determine regional characteristics impractical. Such limited amount of long-term source apportionment datasets represents a severe limitation in particular for model validation and for the development of appropriate mitigation strategies. Furthermore, the negligible transmission efficiency of the AMS inlet for supermicron particles prevents the characterization of their chemical nature and contributing sources.

To broaden the spatial and temporal coverage and size segregation capability of the AMS, we have adapted this instrument for the analysis of water extract of filter samples, which are routinely collected at many air quality sites. The method was applied to more than 2000 samples of particulate matter with aerodynamic diameters smaller than 1, 2.5, and 10 µm, collected at more than 20 urban and rural locations in the northern hemisphere (Figure 1), during summer and winter.

We show that at all sites OA is highly oxygenated, indicating that the secondary production of this fraction is very rapid, even during severe winter haze events such as those recurrent in China (Huang et al., 2014). By analyzing long-term data, we show that this secondary organic fraction exhibit a strong seasonality, and hence can be separated into a winter dominating fraction most-probably from anthropogenic precursors and a summer dominating fraction strongly affected by biogenic emissions. Such seasonality has not been previously observed as online AMS datasets are typically limited to weeks of measurements. Primary emissions, especially from residential heating (e.g. biomass and coal burning), may be a significant fraction of the total OA mass, mostly during winter but only at certain locations. By examining the spectral patterns of coarse particles, we could identify new factors related to primary biological emissions, which are dominated by carbohydrates and mostly associated with plant debris (Figure 2).



Figure 1: Location of the measurement sites. The number of sites per country is indicated in red.

Source apportionment data from the different locations and during different seasons will be presented and compared with additional measurements including radiocarbon data and molecular markers.



Figure 2: Size segregated sources of the organic aerosol in summer and winter at a rural site in Switzerland. These sources include hydrocarbon like OA (HOA), biomass burning OA (BBOA), water soluble primary biological OA (WSPBOA), winter oxygenated OA (W-OOA), summer oxygenated OA (S-OOA), and cellulose from biological emissions.

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