Primary Organic Aerosol Source Identification and Aging Observed by Hourly Measurements of Organic Molecular Markers in Urban Shanghai, China

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Molecular markers in ambient organic aerosol (OA) are valuable in providing source information and insights to formation process of OA. Their traditional quantification is based on offline analysis of filter samples, hugely hindering the utility of the tracer data due to the coarse time resolution and labor-intensive nature. In this study, hourly organic molecular markers in fine particulate matter were measured using a recently commercialized Thermal desorption Aerosol Gas chromatography-mass spectrometry (TAG) at an urban location in Shanghai, China during a three-week campaign from 9 November-3 December 2018. Anhydro sugars, fatty acids, aromatic acids, and polycyclic aromatic hydrocarbons (PAHs) were examined in detail. Their diurnal variations showed characteristic features representing the corresponding emission source activities. For example, stearic acid showed a clear peak around 7 pm, in accordance with the enhanced cooking activities during mealtime. Diagnostic ratios of related maker species of different reactivity provided unique information in uncovering the source information and tracking evolution of OA in the atmosphere. For example, Levo/Manno and Levo/K⁺ ratio analysis identified crop residue burning as the major form of biomass burning. Ratios of unsaturated and saturated fatty acids gave unambiguous indication of atmospheric degradation of unsaturated fatty acids after emissions. Furthermore, oleic acid to stearic acid ratio was highly correlated with O/C ratios, suggesting the possible utility of oleic acid as a model compound to examine the heterogeneous reaction of cooking-related OA. PAH ratio-ratio plots helped tag varying influences of major combustion sources associated with air masses of different origins, with coal combustion and biomass burning dominant under the influence of long range transport air mass and vehicle emissions dominant under local/median range air mass influence. This study demonstrated the utility of the hourly organic markers in capturing the dynamic change of the source emissions and atmospheric ageing, providing observational evidence to support their use in rapid source
apportionment.