On the Interpretation of Oxygenated Organic Aerosols (and their Subtypes) Arising from Factor Analysis of Aerosol Mass Spectrometer Data

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Zhang et al. (EST 2005; ACP 2005) first performed factor analysis (FA) of Aerodyne Aerosol Mass Spectrometer (AMS) complete organic aerosol (OA) mass spectra. This study showed that an oxygenated organic aerosol (OOA) factor accounted for 2/3 of the OA mass at an urban site in Pittsburgh and strongly linked OOA to secondary organic aerosols (SOA). Many subsequent studies and the application of more powerful solution algorithms such as Positive Matrix Factorization (PMF) to the same FA problem have demonstrated the importance of OOA at most locations (e.g. Volkamer et al., GRL, 2006; Zhang et al., GRL and EST, 2007; Lanz et al., ACP, 2007 and EST, 2008; Ulbrich et al., ACPD, 2008; Nemitz et al., AST, 2008; Huffman et al., ACP 2009). Multiple studies have also identified several subtypes of OOA (e.g. OOA-1 and OOA-2). This type of analysis offers new insights because it provides some chemical resolution on the total OA mass with high time and size resolution, and bypasses the limitations of techniques that only analyze tracers and which may favor more reduced species. However the chemical resolution is limited and careful interpretation of the FA output is required, including the use of database spectra, time series of external tracers, tracer ratios, back-trajectory analyses, size-distribution analyses, etc. This presentation will address the interpretation of total OOA and its subfactors across a large range of locations in urban, suburban, rural, remote, and forested areas, and will compare with the results of other source apportionment techniques.

Based on data from multiple datasets we conclude that (1) anthropogenic SOA in and downwind of urban areas is an important source of OOA; (2) motor vehicles, meat cooking, and trash burning are unlikely to be sources of primary OOA; (3) SOA from biogenic and biomass burning precursors are also clear sources of OOA; (4) primary biomass burning OA (P-BBOA) typically shows significant differences with ambient OOA factors; (5) heterogeneous oxidation of urban POA may give rise to OOA although it can be at most a small contributor to ambient concentrations due to its limited mass emission in industrialized countries. Subtypes of OOA in multiple ambient datasets are shown to arise from (a) continued oxidative processing of emissions with increasing photochemical age; (b) partitioning of semivolatile SOA species under conditions of lower temperature and increased humidity; and (c) different source regions affecting a receptor site. We show through PMF analysis of thermal-denuder measurements that the more oxygenated OOA-1 component is is also less volatile. The interpretation and limitations of FA techniques, which currently need to assume a constant mass spectrum for each component, for cases in which the real spectra are evolving due to atmospheric aging will be discussed.