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Hydrocarbon-like and oxygenated organic aerosols in Pittsburgh: insights into sources and processes of organic aerosols

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Abstract. A recently developed algorithm (Zhang et al., 2005) has been applied to deconvolve the mass spectra of organic aerosols acquired with the Aerosol Mass Spectrometer (AMS) in Pittsburgh during September 2002. The results are used here to characterize the mass concentrations, size distributions, and mass spectra of hydrocarbon-like and oxygenated organic aerosol (HOA and OOA, respectively). HOA accounts for 34% of the measured organic aerosol mass and OOA accounts for 66%. The mass concentrations of HOA demonstrate a prominent diurnal profile that peaks in the morning during the rush hour and decreases with the rise of the boundary layer. The diurnal profile of OOA is relatively flat and resembles those of SO_4^{2-} and NH_4^+ . The size distribution of HOA shows a distinct ultrafine mode that is commonly associated with fresh emissions while OOA is generally concentrated in the accumulation mode and appears to be mostly internally mixed with the inorganic ions, such as SO_4^{2-} and NH_4^+ . These observations suggest that HOA is likely primary aerosol from local, combustion-related emissions and that OOA is secondary organic aerosol (SOA) influenced by regional contributions. There is strong evidence of the direct correspondence of OOA to SOA during an intense new particle formation and growth event, when condensational growth of OOA was observed. The fact that the OOA mass spectrum from this event is very similar to that from the entire study suggests that the majority of OOA in Pittsburgh is likely SOA. O_3 appears to be a poor indicator for OOA concentration while SO_4^{2-} is a relatively good surrogate for this dataset. Since the diurnal averages of HOA track those of CO during day time, oxidation/aging of HOA appears to be very small on the time scale of several hours. Based on extracted mass spectra and the likely elemental compositions of major m/z 's, the organic mass to organic carbon ratios (OM:OC) of HOA and OOA are estimated at 1.2 and 2.2 $\mu\text{g}/\mu\text{gC}$, respectively, leading to an average OM:OC ratio of 1.8 for submicron OA in Pittsburgh during September. The C:O ratio of OOA is estimated at 1:0.8. The carbon contents in HOA and OOA estimated accordingly correlate well to primary and secondary organic carbon, respectively, estimated by the OC/EC tracer technique (assuming POC-to-EC ratio=1). In addition, the total carbon concentrations estimated from the AMS data agree well with

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those measured by the Sunset Laboratory Carbon analyzer ($r^2=0.87$; slope= 1.01 ± 0.11). Our results represent the first direct estimate of the OM:OC ratio from highly time-resolved chemical composition measurements.

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