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## Evolving mass spectra of the oxidized component of organic aerosol: results from aerosol mass spectrometer analyses of aged diesel emissions

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**Abstract.** The species and chemistry responsible for secondary organic aerosol (SOA) formation remain highly uncertain. Laboratory studies of the oxidation of individual, high-flux SOA precursors do not lead to particles with mass spectra (MS) matching those of ambient aged organic material. Additionally, the complexity of real organic particles challenges efforts to identify their chemical origins. We have previously hypothesized that SOA can form from the atmospheric oxidation of a large suite of precursors with varying vapor pressures. Here, we support this hypothesis by using an aerosol mass spectrometer to track the chemical evolution of diesel exhaust as it is photochemically oxidized in an environmental chamber. With explicit knowledge of the condensed-phase MS of the primary emissions from our engine, we are able to decompose each recorded MS into contributing primary and secondary spectra throughout the experiment. We find that the SOA becomes increasingly oxidized as a function of time, quickly approaching a final MS that closely resembles that of ambient aged organic particulate matter. This observation is consistent with our hypothesis of an evolving suite of SOA precursors. Low vapor pressure, semi-volatile organic emissions can form condensable products with even a single generation of oxidation, resulting in an early-arising, relatively less-oxidized SOA. Continued gas-phase oxidation can form highly oxidized SOA in surprisingly young air masses via reaction mechanisms that can add multiple oxygen atoms per generation and result in products with sustained or increased reactivity toward OH.

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