

**Molecular distributions and isotopic compositions of organic aerosols over the western North Atlantic: Dicarboxylic acids, oxoacids,  $\alpha$ -dicarbonyls (glyoxal and methylglyoxal), lipid class compounds, sugars, and secondary organic aerosol tracers**

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Marine aerosols were collected over the western North Atlantic off the coast of Boston to Bermuda in August 2012 using a high-volume air sampler. Aerosol samples were analyzed for organic and elemental carbon (OC/EC), low molecular weight dicarboxylic acids and related compounds, lipid class compounds (n-alkanes, fatty acids and fatty alcohols), sugars and various secondary organic aerosol (SOA) tracers. Homologous series (C<sub>2</sub>-C<sub>12</sub>) of dicarboxylic acids (31-335 ng m<sup>-3</sup>) were detected with a predominance of oxalic acid. Diacids were found to be the most abundant compound class followed by monoterpene-SOA tracers > isoprene-SOA tracers > sugars > oxoacids > fatty alcohols > fatty acids >  $\alpha$ -dicarbonyls > aromatic acids > n-alkanes. The concentrations of these compounds were higher in the coastal site and decreased towards the open ocean. Interestingly, contributions of oxalic acid to total aerosol carbon increased from the coast (2.3%) to the open ocean (5.6%) near Bermuda.

Stable carbon isotope ratios of aerosol total carbon and individual diacids and oxoacids were determined using an isotope ratio mass spectrometer. The stable carbon isotopic composition of oxalic acid increased from the coast (-17.5‰) to the open ocean (-12.4‰), suggesting that photochemical aging of organic aerosols occurred during the atmospheric transport over the ocean. Stable carbon isotope ratios of bulk aerosol carbon also increased from the coast near Boston (-24.3‰) to the open ocean near Bermuda (-18.2‰), consistent with photochemical aging of organic aerosols.