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## The changing nature of particulate organic carbon and the relation to aerosol liquid water

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Particulate organic carbon (OC) mass concentrations demonstrate decreasing trends in many regions across the contiguous US (CONUS). We investigate decadal trends in specific total organic carbon (TOC) volatility fractions OC1, OC2, OC3, and OC4 as defined and reported at 121 locations in the Interagency Monitoring of PROtected Visual Environments (IMPROVE) monitoring network from 2005-2016 for 23 chemical climatology regions across the CONUS. Volatility fraction OC2 drives ubiquitous decadal decreases in TOC, and OC3 mass concentrations increase. The largest changes in OC2 and OC3 occur in the eastern US. In four focus regions (Northeast, Appalachia, West Texas, and Northwest), OC fraction mass concentrations are converted to organic mass (OM) using region-specific OM:OC ratios. GEOS-Chem simulations reproduce and correlate strongly ( $R^2 > 0.7$ ) with OM fraction decadal trends. Decreases in aerosol liquid water (ALW) concentrations are tightly linked to observed change in individual TOC thermal fractions, and aerosol products derived from aqueous-phase isoprene oxidation predicted by GEOS-Chem. These results lend insight to changing chemical regimes with implications for particle phase state, viscosity, and oxidation state.