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Seasonal Contribution of Isoprene-Derived Organosulfates to Total Water-Soluble Fine Particulate Organic Sulfur in the United States. YUZHI CHEN, Tracy Dombek, Jenny Hand, Zhenfa Zhang, Avram Gold, Andrew Ault, Keith Levine, Jason Surratt, University of North Carolina at Chapel Hill Organosulfates (OSs) are the most abundant class of organosulfur compounds (OrgS) in atmospheric fine particulate matter (PM2.5). Globally, isoprene-derived OSs (iOSs) are the most abundantly reported OSs. The methyltetrol sulfates (MTSs), formed from multiphase chemical reactions of isoprene-derived epoxydiols (IEPOX) with acidic sulfate aerosols, are the predominant iOSs. A recent study revealed that the heterogeneous hydroxyl radical (•OH) oxidation of fine particulate MTSs yields several highly oxygenated and functionalized OSs previously attributed to non-IEPOX pathways. By using hydrophilic interaction liquid chromatography interfaced to electrospray ionization high-resolution quadrupole time-of-flight mass spectrometry (HILIC/ESI-HRQTOFMS), iOSs were quantitatively characterized in PM2.5 collected from 20 ground sites within the Interagency Monitoring of Protected Visual Environments (IMPROVE) network during the 2016 summer and winter seasons. Total watersoluble sulfur (TWS-S) and sulfur in the form of inorganic sulfate (Sinorg) were determined by inductively coupled plasmaoptical emission spectroscopy (ICP-OES) and ion chromatography (IC), respectively. The difference between TWS-S and Sinorg was used as an upper bound estimate of water-soluble OrgS concentration. Significantly higher OrgS concentrations, coincident with elevated iOS concentrations, were observed only in summer. On average, iOSs (130 ± 60, up to 240 ng m-3) explained 29% (± 7%) of OrgS and 5% (± 2%) of organic matter (OM = 1.8*OC) in summertime PM2.5 collected from the eastern U.S. For the western U.S., iOSs (11 ± 6 ng m-3) account for 6% (± 5%) of OrgS and 0.7% (± 0.4%) of OM. This study provides critical insights into the abundance, prevalence, spatial variability of iOSs across the U.S.