An extractive electrospray ionization time-of-flight mass spectrometer (EESI-TOF) for online measurement of atmospheric aerosol particles

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Real-time, online measurements of atmospheric organic aerosol (OA) composition are an essential tool for determining the emissions sources and physicochemical processes governing aerosol effects on climate and health. However, the reliance of current techniques on thermal desorption, hard ionization, and/or separated collection/analysis stages introduces significant uncertainties into OA composition measurements, hindering progress towards these goals. To address this gap, we present a novel, field-deployable extractive electrospray ionization time-of-flight mass spectrometer (EESI-TOF), which provides online, near-molecular (i.e., molecular formula) OA measurements at atmospherically relevant concentrations without analyte fragmentation or decomposition. Aerosol particles are continuously sampled into the EESI-TOF, where they intersect a spray of charged droplets generated by a conventional electrospray probe. Soluble components are extracted and then ionized as the droplets are evaporated. The EESI-TOF achieves a linear response to mass, with detection limits on the order of 1 to 10 ng m(-3) in 5 s for typical atmospherically relevant compounds. In contrast to conventional electrospray systems, the EESI-TOF response is not significantly affected by a changing OA matrix for the systems investigated. A slight decrease in sensitivity in response to increasing absolute humidity is observed for some ions. Although the relative sensitivities to a variety of commercially available organic standards vary by more than a factor of 30, the bulk sensitivity to secondary organic aerosol generated from individual precursor gases varies by only a factor of 15. Further, the ratio of compound-by-compound sensitivities between the EESI-TOF and an iodide adduct FIGAERO-I-CIMS varies by only +/- 50%, suggesting that EESI-TOF mass spectra indeed reflect the actual distribution of detectable compounds in the particle phase. Successful deployments of the EESI-TOF for laboratory environmental chamber measurements, ground-based ambient sampling, and proof-of-concept measurements aboard a research aircraft highlight the versatility and potential of the EESI-TOF system.

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