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Chemically-Resolved Aerosol Volatility Measurements from Two Megacity Field Studies

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The volatilities of different chemical species in ambient aerosols are important but remain poorly characterized. The coupling of a recently developed rapid temperature-stepping thermodenuder (TD, operated in the range 54–230oC) with a High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS) during field studies in two polluted megacities has enabled the first direct characterization of chemically-resolved urban particle volatility. Measurements in Riverside, CA and Mexico City are generally consistent and show ambient nitrate as having the highest volatility of any AMS aerosol species while sulfate showed the lowest volatility. Total organic aerosol (OA) showed volatility intermediate between nitrate and sulfate, with an evaporation rate of 0.6%•K-1 near ambient temperature, although OA dominates the residual species at the highest temperatures. Different types of OA were characterized with marker ions, diurnal cycles, and positive matrix factorization (PMF) and show significant differences in volatility. Reduced hydrocarbon-like OA (HOA, a surrogate for primary OA, POA), oxygenated OA (OOA, a surrogate for secondary OA, SOA), and biomass-burning OA (BBOA) separated with PMF were all determined to be semi-volatile. The most aged OOA-1 and its dominant ion, CO2+, consistently exhibited the lowest volatility, with HOA, BBOA, and associated ions for each among the highest. The similar or higher volatility of HOA/POA compared to OOA/SOA contradicts the current representations of OA volatility in most atmospheric models and has important implications for aerosol growth and lifetime. Our results strongly imply that all OA types should be considered semivolatile in models. The study in Riverside identified organosulfur species (e.g. CH3HSO3+ ion, likely from methanesulfonic acid), while both studies identified ions indicative of amines (e.g. C5H12N+) with very different volatility behaviors than inorganic-dominated ions. The oxygen-to-carbon ratio of OA in each ambient study was shown to increase both with TD temperature and from morning to afternoon, while the hydrogen-to-carbon ratio showed the opposite trend.