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Individual Aerosol Droplet pH Measurement via a Ratio-metric Raman Method Using Aerosol Optical Tweezers: Evaluation of Thermodynamic and Activity Models

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Measuring pH in individual aerosol droplet is essential for understanding and estimating physicochemical processes within aerosol microenvironments. Recently, aerosol optical tweezers coupling with Raman spectroscopy have been applied to measure the pH of single trapped microdroplets by utilizing conjugate acid-base equilibrium to infer pH shifts. However, such measurements are easily affected by many factors such as variations in detecting volumes and laser intensities, making it hard to directly determine these acid and base concentrations through their respective peak areas. To overcome these problems and accurately measure the concentrations of SO_4^{2-} and HSO_4^- within individual $NaHSO_4$ microdroplets, in this study a ratio-metric spectroscopic method is developed based on the peak area ratio of $\nu(SO_4^{2-})/\nu(OH)$ and $\nu(HSO_4^-)/\nu(OH)$. Combined with the ion balance and ion activity coefficients, droplet pH is determined unambiguously. These experiment results were further used to evaluate the performance of activity models and thermodynamic models associated with aerosol pH, ion concentration and activity coefficient predictions. Pitzer, Simonson, and Clegg (PSC) model provides the best predictions of ion activity coefficients Extended Aerosol Inorganics Model vision IV (E-AIM IV) works well over a wide $NaHSO_4$ concentration range (0.4-8.8 mol/kg), while ACCENT Pitzer model predictions have extremely good agreement with the experiment results in low $NaHSO_4$ concentration condition (≤2.0 mol/kg). By contrast, ISORROPIA II shows relatively poor performance as compared with E-AIM IV.