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Effects of Water-soluble Organic Carbon on Aerosol pH

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Water soluble organic carbon (WSOC) is a ubiquitous and significant fraction of fine particulate matter. Despite advances in aerosol thermodynamic equilibrium models, there is limited understanding on the comprehensive impacts of WSOC on aerosol acidity (pH). We address this limitation by studying submicron aerosol that represent the two extremes in acidity levels found in the atmosphere: strongly acidic aerosol from Baltimore, MD, and weakly acidic conditions characteristic of Beijing, China. These cases are then used to construct mixed inorganic/organic single-phase aqueous particles, and thermodynamically analyzed by the E-AIM and ISORROPIA models in combination with activity coefficient model AIOMFAC to evaluate the effects of WSOC on the H⁺ ion activity coefficients (γ_{H^+}) and activity (pH). We find that addition of organic acids and non-acid organic species concurrently increases γ_{H^+} and aerosol liquid water. Under the highly acidic conditions typical of the eastern U.S. (inorganic-only pH ~1), these effects mostly offset each other, giving pH changes of < 0.5 pH units even at organic aerosol dry mass fractions in excess of 60%. Under conditions with weaker acidity typical of Beijing (inorganic-only pH ~4.5), the non-acidic WSOC compounds had similarly minor effects on aerosol pH, but organic acids imparted the largest changes in pH compared to the inorganic-only simulations. Organic acids affect pH in the order of their pK_a values (oxalic acid > malonic acid > glutaric acid). Although the inorganic-only pH was above the pK_a value of all three organic acids investigated, pH changes in excess of 1 pH unit were only observed at unrealistic organic acid levels (aerosol organic acid concentrations > 35 $\mu\text{g m}^{-3}$) in Beijing. The model simulations were run at 70%, 80%, and 90% relative humidity (RH) levels and the effect of WSOC was inversely related to RH. At 90% RH, WSOC altered aerosol pH by up to ~0.2 pH units, though the effect was up to ~0.6 pH units at 70% RH. The somewhat offsetting nature of these effects suggests that aerosol pH is sufficiently constrained by the inorganic constituents alone under conditions where liquid-liquid phase separation is not anticipated to occur.

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