We performed detailed molecular dynamics simulation studies for activity coefficients of a variety 1:1 aqueous electrolytes. Previous simulation studies focused on calculating chemical potentials at the standard state using a variety of techniques including thermodynamic integration [1,2,3] and the osmotic ensemble Monte Carlo method [4,5,6]. We seek to extend these studies by performing accurate calculations of the activity coefficients including rigorously calculating the Debye-Huckel limiting behavior. The chemical potentials from which we calculate the activity coefficients are estimated by calculating the free energy change via the Bennett's acceptance ratio method of adding a single ion pair to the system of interest. The ion pair is inserted fractionally and the free energy change is calculated for each interval. We compare the activity coefficients obtained for a variety of water and ion models to experimental results across a range of salt concentrations and temperatures. We also calculate osmotic coefficients and test for thermodynamic consistency via the Gibbs-Duhem equation.

[1] Sanz E, Vega C. Solubility of KF and NaCl in water by molecular simulation. *J. Chem. Phys.* 2007; 126(1): 014507.

[2] Aragones JL, Sanz E, Vega C. Solubility of NaCl in water by molecular simulation revisited. *J. Chem. Phys.* 2012;136(24): 244508.

[3] Ferrario M, Giovanni C, Eckhard S, Thierry C, Pierre T. Solubility of KF in water by molecular dynamics using the Kirkwood integration method. *J. Chem. Phys.* 2002;117(10): 4947-4953.

[4] Lísal M, Smith WR, Kolafa J. Molecular simulations of aqueous electrolyte solubility: 1. The expanded-ensemble osmotic molecular dynamics method for the solution phase. *J. Phys. Chem. B* 2005;109(26):12956-12965.

[5] Moučka F, Martin L, Škvor J, Jirsák J, Nezbeda I, Smith WR. Molecular simulation of aqueous electrolyte solubility. 2. Osmotic ensemble Monte Carlo methodology for free energy and solubility calculations and application to NaCl. *J. Phys. Chem. B* 2011;115(24): 7849-7861.

[6] Moučka F, Martin L, Smith WR. Molecular simulation of aqueous electrolyte solubility. 3. Alkali-halide salts and their mixtures in water and in hydrochloric acid. *J. Phys. Chem. B* 2012;116(18): 5468-5478.