Title: **The role of citric acid in the transport of uranium(VI) through saturated porous media: The application of surface chemical models to transport simulations of bench-scale experiments**

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Abstract: Predicting the transport of contaminants through soil and sediments requires a model that incorporates chemical processes into the advection-dispersion equations. In most aquatic systems, species of dissolved organic matter (DOM) constitute an important pool of ligands available for complexing trace metals. In the presence of organic ligands, trace metals may form ternary (e.g., particle/metal/ligand) surface complexes. In addition, DOM may sorb to particle surfaces and (1) compete against metal ions for surface sites or (2) modify the sorptive characteristics of the original particle surface.

In this study, controlled laboratory batch and column experiments, in conjunction with a predictive multicomponent transport model, have been performed to study the effects of citric acid on the transport of uranium (VI) through saturated porous media under variable chemical conditions. Citric acid was used as a “model” organic ligand and surrogate for DOM. The sorbent used for the study was a quartz sand with an average particle size of 225 µm, whose surface properties were determined by a mixture of Si-, Fe-, and Al-surface groups (pH_{pzc} = 6.3).

Experimental results show that presence of citric acid increases the mobility of U(VI) due to the formation of U(VI)-citrate complexes and competition of citric acid against U(VI) for the surface sites. The formation of U(VI)-citric acid complexes is regulated by the dissolution of iron from iron oxide coatings in the sand as a consequence of the formation of iron/citric acid complexes. The asymmetrical breakthrough curves indicate a non-linear adsorption isotherm and/or kinetically controlled adsorption/desorption surface reactions. Multicomponent transport simulations with kinetically controlled surface reactions successfully simulate this asymmetrical behavior and the “forward” simulation (i.e., “prediction”) of other experimental conditions. The multicomponent transport model improved our ability to accurately simulate the effects of U(VI)/citrate complexes on U(VI) transport relative to the use of distribution coefficients (K_d) derived from batch data. Our transport simulations also suggest that for an accurate simulation of U(VI) transport in the presence of citric acid, the following processes and/or interactions need to be explicitly considered: (1) iron oxyhydroxide dissolution; (2) U(VI)/citrate and Fe^{3+}/citrate interactions; and (3) a mixed metal U(VI)-Fe^{3+}-citrate complex.