Additive Surface Complexation Modeling of Uranium(VI) Adsorption onto Quartz-Sand Dominated Sediments

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Nuclear materials processing and U mining and milling resulted in many acidic U(VI) groundwater plumes during the Cold War. The F-Area of the Savannah River Site (SRS) in South Carolina (USA) is an example where groundwater has been contaminated by a large, persistent U(VI)-containing acidic plume during 1955-1988. Although many U(VI) contaminated aquifers (e.g., the F-Area) are composed predominantly of quartz-sand sediments, understanding of geochemical mechanisms governing U(VI) adsorption to quartz-sand dominated sediemnts under acidic conditions is still limited. We conducted U(VI) adsorption experiments using quartz-sand dominated plume sediments and reference minerals quartz, goethite and kaolinite. We developed a new humic acid adsorption method to determine the relative surface area abundances of individual minerals of the sediments. This method plays a key role in successful application of the mineral component-additivity (CA) based surface complexation modeling (SCM) for understanding of geochemical mechanisms governing U(VI) adsorption. Our experimental results indicate that quartz has stronger U(VI) adsorption ability per unit surface area than goethite and kaolinite at pH ≤ 4.0. Our modeling results indicate that the binary mineral (goethite/kaolinite) CA-SCM under-predicts U(VI) adsorption to the quartz-sand dominated sediments at pH ≤ 4.0. The new ternary mineral (quartz/goethite/kaolinite) CA-SCM with the addition of quartz component provides excellent predictions, suggesting quartz-sand adsorbs U(VI) more at pH near 3.0, kaolinite (exchange sites) contributes more at pH near 4.0, while goethite dominates at pH > 4.5. Potential influences of dissolved Al, Si and Fe on U(VI) adsorption are also discussed. This work is part of the Sustainable Systems Scientific Focus Area (SFA) program at LBNL, a project supported by the US Department of Energy for understanding and predicting long-term groundwater plume mobility and natural attenuation.