Column-Scale Evaluation of *In Situ* Uranium Immobilization in Sediments Amended with Phosphate

Abstract

Phosphate amendments can be added to U(VI)-contaminated subsurface environments to promote *in situ* remediation. The transport of U(VI) in phosphate-treated systems can be controlled by non-equilibrium processes involving advective and diffusive transport and adsorption-desorption or dissolution-precipitation reactions. The primary objective of this study was to evaluate the impacts of phosphate addition on the transport of U(VI) through contaminated sediments. Sediments (< 2mm size fraction) from a site in Colorado were used in column experiments with flow rates that correspond to a typical groundwater velocity of 1 m/day. In the absence of phosphate, the sediments took up 1.8±0.2 μg U/g of sediments. If phosphate was not present when uranium-free influents were introduced, then all of the U(VI) was desorbed from the sediments within 100 pore volumes. In contrast, more than 75 % of the adsorbed uranium was retained when phosphate was present in the uranium-free influent. When an influent containing both uranium and phosphate was introduced, significant continued uptake of uranium was observed. The sediments took up 36 μg U/g of sediments during 334 pore volumes of operation.

Sequential extractions of sediments taken from the columns that had seen only uranium or phosphate revealed that uranium was uniformly distributed along the length of the columns and was primarily in ion-exchangeable and weak acid-soluble forms, which suggested that adsorption was the dominant mode of immobilization. For the sediments that were loaded with both uranium and phosphate together, the uranium was still primarily associated in ion-exchangeable and weak acid-soluble forms but with higher contribution of uranium in weak acid soluble forms and with these contributions increasing as the column operation progressed. The response of dissolved uranium concentrations to stopped-flow events indicated that intraparticle diffusion is a rate-limiting step for uranium adsorption-desorption and was affecting the uranium transport in the sediments. The results obtained provide insights into uranium-phosphate reactions and they can be used to design effective remediation strategies for phosphate based *in situ* remediation.