Novel Imaging Techniques, Integrated with Mineralogical, Geochemical, and Microbiological Characterizations to Determine the Biogeochemical Controls on Technetium Mobility in FRC Sediments

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Technetium-99 is a priority pollutant at numerous DOE sites, due to a combination of its long half-life (2.1×10^5 yr), high mobility as Tc(VII) (TcO_4^-; pertechnetate anion) in oxic waters, and bioavailability as a sulfate analog. Under anaerobic conditions, however, the radionuclide is far less mobile, forming insoluble Tc(IV) precipitates. As anaerobic microorganisms can reduce soluble Tc(VII) to insoluble Tc(IV), microbial metabolism may have the potential to treat sediments and waters contaminated with Tc. In previous studies we have focused on the fundamental mechanisms of Tc(VII) bioreduction and precipitation, and we have identified direct enzymatic (hydrogenase-mediated) mechanisms, and a range of potentially important indirect transformations catalyzed by biogenic Fe(II), U(IV), or sulfide. These baseline studies have generally used pure cultures of metal-reducing bacteria in order to develop conceptual models for the biogeochemical cycling of Tc. There is, however, comparatively little known about interactions of metal-reducing bacteria with environmentally relevant trace concentrations of Tc, against a more complex biogeochemical background provided by mixed microbial communities in the subsurface. This information must be available if in situ remediation of Tc(VII) contamination is to be successful at DOE sites.

The aim of this new project is to use a highly multidisciplinary approach to identify the biogeochemical factors that control the mobility of environmentally relevant concentrations of Tc(VII) in FRC sediments. We will use a combination of geochemical, mineralogical, microbiological, and spectroscopic techniques to determine the solubility and phase associations of Tc in batch sediment experiments ("progressive microcosms") where a sequence of terminal electron accepting processes is separated by time. Additional column experiments utilizing FRC sediments containing discrete biogeochemical zones will be challenged with low concentrations of ^{99m}Tc, and the mobility of the radionuclide imaged using a γ-camera. By comparing the pattern of Tc immobilization, with high-resolution studies of the mineralogy, geochemistry, and microbial ecology of the columns, we will further characterize the biogeochemical controls on Tc mobility in FRC sediments. Column experiments will also utilize similar approaches to determine the stability of immobilized reduced phases of Tc in the presence of oxidizing agents including nitrate. The effectiveness of strategies proposed to stimulate Tc(VII) reduction and precipitation in the subsurface will also be determined in both batch and column experiments. Finally the experimental results will be used to calibrate a modeling approach employing an established coupled speciation and transport code to provide parameters that could potentially be used to make predictions of the mobility of Tc in FRC sediments and other subsurface environments.