

Project 87016

Trace Metals in Groundwater & Vadose Zone Calcite: In Situ Containment & Stabilization of Strontium-90 & Other Divalent Metals & Radionuclides at Arid West DOE

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RESULTS TO DATE: Radionuclide and metal contaminants such as strontium-90 are present beneath U.S. Department of Energy (DOE) lands in both the groundwater (e.g., 100-N area at Hanford, WA) and vadose zone (e.g., Idaho Nuclear Technology and Engineering Center [INTEC] at the Idaho National Laboratory [INL]). In situ containment and stabilization of these contaminants is a cost-effective treatment strategy. However, implementing in situ containment and stabilization approaches requires definition of the mechanisms that control contaminant sequestration. We are investigating the in situ immobilization of radionuclides or contaminant metals (e.g., strontium-90) by their facilitated co-precipitation with calcium carbonate (primarily calcite) in groundwater and vadose zone systems. Our facilitated approach relies upon the hydrolysis of introduced urea to cause the acceleration of calcium carbonate precipitation (and trace metal co-precipitation) by a) increasing pH and alkalinity and b) liberating cations from the aquifer matrix by cation exchange reactions. Subsurface urea hydrolysis is catalyzed by the urease enzyme, which is produced in situ by native urea hydrolyzing microorganisms. Because the precipitation process tends to be irreversible and many western aquifers are saturated with respect to calcite, the co-precipitated metals and radionuclides will be effectively removed from the aqueous phase over the long term. We are currently conducting field based activities at both the INL Vadose Zone Research Park (VZRP), an uncontaminated surrogate site for the strontium-90 contaminated vadose zone at INTEC and at the strontium-90 contaminated aquifer of 100-N area of the Hanford site.

We have recently completed a set of five wells in that are completed in the surficial sediment of the VZRP accessing a perched water body at the shallowest basalt-sediment interface. This perched water body is analogous to the contaminated perched water bodies at INTEC. We are conducting urea injection experiment at VZRP with the goal of precipitating calcite in the perched water body as well as sequestering non-radiogenic strontium into any precipitated calcite. Prior to conducting the urea injections, pump tests are being conducted to characterize the hydraulic conductivity field in the vicinity of the wells. This testing will support the development of a fully coupled transport and biogeochemical model. A novel aspect of our modeling approach is our use of automated inverse techniques to parameterize the hydraulic conductivity of the flow field as-well-as constrains biotic and abiotic reaction rates. Our model has been used to simulate remediation scenarios and shows that although the initial groundwater is supersaturated with respect to calcite and that urea hydrolysis increase the degree of supersaturation, the system may exhibit a transient period during which calcite and co-precipitated strontium will redissolve at the conclusion of urea treatment. This result is a strongly influenced by both the advective flux of groundwater and the hydrolysis rate of urea and illustrates the importance of using fully coupled reactive transport models in the design and evaluation of remediation treatments for hazardous and radioactive waste.

Following the field experiments, pump tests will be repeated with the goal of identify zone of decrease hydraulic conductivity that may reflect calcite precipitation, the development of biofilms, or both. Additionally, if funding is available, post experiment collection of solid phase samples will be conducted to verify the amount and distribution of precipitated divalent metal in any produced calcites.

In addition to our research focused on the INL, intrinsic rates of urea hydrolysis (i.e., rates with no carbon source amendments to the system) have been measured for contaminated core and groundwater collected at the 100-N Hanford site. These results indicate that significantly more reactivity is associated with the sediments than with the groundwater and that the rates at the Hanford site are much higher than those observed for the Snake River Plain Aquifer beneath the INL.

