Field Investigations of Microbially Facilitated Calcite Precipitation for Immobilization of Strontium-90 and Other Trace Metals in the Subsurface

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Subsurface radionuclide and metal contaminants throughout the U.S. Department of Energy (DOE) complex pose one of DOE's greatest challenges for long-term stewardship. One promising stabilization mechanism for divalent ions, such as the short-lived radionuclide ^{90}Sr, is co-precipitation in calcite. We have previously found that that nutrient addition can stimulate microbial ureolytic activity, that this activity accelerates calcite precipitation and co-precipitation of Sr, and that higher calcite precipitation rates can result in increased Sr partitioning. We are conducting integrated field, laboratory, and computational research to evaluate the relationships between ureolysis and calcite precipitation rates and trace metal partitioning under environmentally relevant conditions, and investigating the coupling between flow/flux manipulations and precipitate distribution and metal uptake.

Our September 2010 experimental campaign at the Integrated Field Research Challenge (IFRC) site located at Rifle, CO was based on a continuous recirculation design; water extracted from well M-07 was amended with urea and molasses and re-injected into up-gradient well M-02. The recirculation experiment was followed by 14 months of geophysical and groundwater monitoring. Cross borehole electrical conductivity tomography indicated a very conductive, high porosity layer overlying a low conductivity unit within the recirculation cell. An increase in electrical conductivity, primarily in the top layer, was observed 40 days after the start of urea recirculation and was likely the result of urea hydrolysis occurring predominantly in the upper portion of the recirculation cell. This interpretation is supported by analysis of sediments from a core (BS-11-2) collected between the injection and extraction well 10 months following urea injection which showed high *ureC* gene copy numbers as well as much higher laboratory measured ^{14}C urea hydrolysis rates in the upper portion of the recirculation cell compared to the deeper portion of the cell.

Long term groundwater sampling of the injection and extraction wells showed an initial increase in urea concentration associated with injection activities followed by decreasing urea concentration and associated increases in ammonium and dissolved inorganic carbon (DIC) following the termination of injection. Preliminary estimates of urea loss yielded a first order rate constant for urea hydrolysis of 0.2 day^{-1}. This value is approximately 7 times higher than estimated for previous field experiments conducted in eastern Idaho. Additionally, DIC carbon isotope ratios were measured for the groundwater. Injected urea had a δ^{13}C of -40.62±0.36 ‰ compared to background groundwater DIC δ^{13}C of -16.62±0.27‰. Observed decreases in groundwater DIC δ^{13}C of up to -19.75‰ suggested that up to 41% of the post injection DIC resulted from urea hydrolysis. Aquifer solids incubated within the treatment zone retained high ureolytic activity 10 months after the cessation of active urea amendment. Sediments from background cores BS-01 collected in 2010, sediments recovered following 10 months incubation in experimental wells B-01, M-02, and M-04, and core BS-11-2 are being processed for trace metal speciation in different operationally defined sediment fractions as well as determination of the δ^{13}C for carbonate minerals.