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ABSTRACT TITLE: An Integrated Experimental and Modeling Study of U(VI) Bioreduction with Emulsified Vegetable Oil as Electron Donor from Laboratory to Field Scales

ABSTRACT: Bioreduction and immobilization of U(VI) in contaminated sites with simple substrates such as acetate and ethanol require daily to weekly injection to avoid reoxidation and remobilization. We hypothesized that slowly degraded and strongly retained substrates could lead to longer term U(VI) reducing conditions, therefore, decreasing the injection frequency from daily/weekly to monthly/quarterly. To test this hypothesis, we conducted microcosm and field tests with emulsified vegetable oil (EVO) as a slow release substrate, and developed a comprehensive model to quantify the microbiological and geochemical dynamics observed in these tests.

In these tests, EVO supported microbial reduction of nitrate, Fe(III), U(VI), and sulfate and CO₂ (methane production observed). In the field test, a single 2-h EVO injection resulted in decreased U concentration downstream (~50 m) by 80% for 100 days, and U discharge to a nearby creek by 50% for over 1 year. X-ray adsorption near edge spectroscopy (XANES) analysis of aquifer solids confirmed U(VI) reduction to U(IV). Pyrosequencing and quantitative PCR of 16S rRNA from monitoring well samples revealed a rapid decline in bacterial community richness and diversity, and indicated the selection of a narrow group of taxa rather than broad community stimulation after EVO injection. Based on the relative abundance of representative operational taxonomic units observed through DNA sequencing approaches, and known physiologies of closely allied species or genera, we developed a conceptual model in which Pelosinus catalyzes oil hydrolysis and ferments glycerol, Desulfotalea oxidizes long-chain fatty acids, and Comamonadaceae, Geobacter, and Desulfovibrio reduce nitrate, Fe(III), U(VI), and sulfate, and Methanobacteria, and methanomicrobia growth leads to methane production.

We implemented the conceptual model in a numerical biogeochemical model, which includes hydrolysis of EVO, production and oxidation of long-chain fatty acids (LCFA), glycerol, acetate, and hydrogen, reduction of nitrate, Fe(III), U(VI) and sulfate, and methanogenesis with growth and decay of multiple functional microbial groups. We parameterized the biogeochemical model by using growth rate parameter values from the literature and estimating EVO, LCFA and glycerol degradation rate coefficients based on microcosm test results.

Using a kinetic Langmuir isotherm to approximate EVO retention, we applied the biogeochemical model to simulate observed acetate, nitrate, Fe, U, and sulfate concentration dynamics, and the activities of multiple microbial functional groups during and after EVO injection. When the lab-determined parameters were applied in the field-scale simulation, the estimated rate coefficient for EVO hydrolysis was about 1 order of magnitude greater than that in the microcosms. Model results suggested that precipitation of LCFA with Ca in the groundwater created a secondary long-term electron donor source. The model predicted substantial accumulation of denitrifying and sulfate-reducing bacteria, and U(IV) precipitates. The accumulation was greatest near the injection wells and along the lateral boundaries of the treatment zone where electron donors mixed with electron acceptors in the groundwater. While electron acceptors such as sulfate have generally been considered to compete with U(VI) for electrons, this work highlighted their role in providing electron acceptors for microorganisms to degrade complex substrates thereby enhancing U(VI) reduction and immobilization.