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A Systematic Evaluation of Mineral-Based Reactive Amendments for In Situ Treatment of Fluoride-Contaminated Groundwater at a Former Aluminum Smelter Site

DIMITRI VLASSOPOULOS[1], DIMITRI VLASSOPOULOS[1], REBECCA GARDNER[1], RAJAT GHOSH[2] AND KRISTIN K GAINES[2]

[1]Anchor QEA LLC
[2]Alcoa
Presenting Author: dvlassopoulos@anchorqea.com

Mineral-based amendments were systematically evaluated for treatment of alkaline groundwater (pH 9-11) containing elevated levels of fluoride (80-1,800 mg/L) at a former aluminum smelter plant for purposes of identifying optimal reactive media for in situ remediation via permeable reactive barriers (PRBs) and soil mixing. Amendments tested included activated alumina (AA), various types of calcium phosphates (bone meal, bone char [BC], phosphate rock), carbonates (calcite, siderite), hydrotalcite, and magnesia, either alone or in combination. Batch and column tests were performed to rank media performance based on F removal efficiency, uptake capacity, binding strength (stability), and potential downgradient water quality impacts.

For all amendments, equilibrium F concentrations were approached within 2-4 days in batch tests except for MgO which was longer than 8 days. AA, BC, bone meal, and hydrotalcite performed best while phosphate rock, carbonates, and magnesium oxide were less reactive at the alkaline pH of the groundwater. F sorption isotherms showed a higher F uptake capacity for AA and hydrotalcite compared to BC and bone meal. Reaction of bone meal with groundwater increased dissolved phosphate, ammonium, and organic carbon concentrations, while BC only increased phosphate.

AA and BC are preferred for PRB applications due to secondary water quality concerns with bone meal and small particle size of hydrotalcite (~0.01 mm). Flow-through columns packed with AA or BC mixed with sand to simulating a PRB showed similar F uptake capacities (2.8-2.9 mg/g) despite different reaction mechanisms. F is sequestered in AA via strong surface complexes, while in BC, the primary mechanism was fluorapatite formation. Selective sequential extractions indicated a limited potential for F remobilization from the F-loaded amendments, with less F leached from AA than from BC at pH>10. A small fraction of phosphate (~0.05%) in soluble and weakly bound forms was released from the BC column but decreased over time. The soluble phosphate release was suppressed by including 3:1 BC/AA mix in a column.

These investigations form the basis for full-scale PRB design. Reactive transport simulations suggest that PRBs comprised of either AA/sand mix or BC/AA/sand mix could provide effective and long-term in situ treatment of F-containing groundwater.