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Effect of Magnesium on Fluoride Removal from Groundwater Using Calcium Phosphate-Based Amendments

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Fluoride (F) is a major geogenic groundwater contaminant. While various methods have been developed for treating F, calcium and phosphate-based amendments have garnered recent interest. These methods involve using either sources of calcium and phosphate or solids such as brushite ($CaHPO_4{\cdot}2H2O$), hydroxyapatite ($Ca_5(PO_4)_3OH$), tricalcium phosphate ($Ca_3(PO_4)_2$), and octacalcium phosphate ($Ca_8H_2(PO_4)_6{\cdot}5H_2O$) to immobilize fluoride. Exposing F-containing systems to these solids results in the formation of fluorapatite ($Ca_5(PO_4)_3F$), which is thermodynamically the most stable F-containing mineral. Even though calcium phosphate-based F immobilization is efficient in simpler water matrices, its extent and kinetics is limited by the co-ions present in groundwater. To systematically understand the effects of major groundwater ions on fluoride uptake in systems containing (a) calcite and phosphate, and (b) brushite, batch experiments were performed in a synthetic groundwater matrix with varying levels (0-10 mM) of Na^+, Ca^{2+}, K^+, Mg^{2+}, Cl^-, SO_4^{2-}, and dissolved inorganic carbon (DIC). The matrix was prepared by considering the average analyte concentrations of groundwater collected from a contaminated site. Additional batch experiments were performed to understand Mg^{2+} interaction with fluorapatite, which was chosen as the starting solid in F-free synthetic groundwater matrix. Time-dependent aqueous samples were collected from the reactors and analyzed for major cations, major anions, and DIC, respectively, by ICP-MS, IC, and TOC analyzers. Solids collected at the end of the experiment were characterized by XRD, FTIR, and Raman spectroscopy. Results indicated that variable levels of Na^+, K^+, and Cl^- had minimal effect on F removal by fluorapatite formation. However, increasing Ca^{2+} in these systems aided F removal. While increasing SO_4^{2-} and DIC concentrations mildly hindered the F removal processes, Mg^{2+} had a significant inhibitory effect (Fig.1). Combined geochemical analysis and solid phase characterization suggested that when Mg was present above a critical concentration, fluorapatite precipitation was inhibited and an unknown calcium phosphate phase likely formed in both the systems. Also, a Mg-bearing apatite phase coprecipitated in the above processes rather than the adsorption of Mg onto fluorapatite, which was noticed at below critical concentrations. Results from this study will likely help devise appropriate treatment methods for different groundwater matrices.

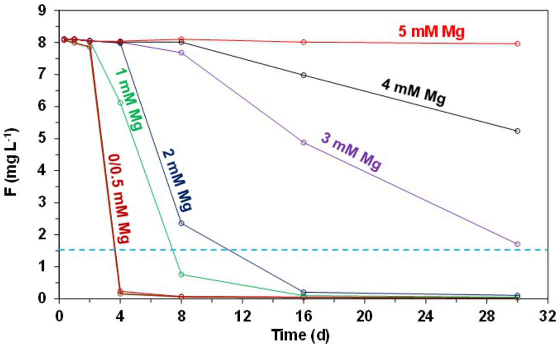

Figure 1. Time dependent variation in fluoride concentration in calcite + phosphate system in varied initial magnesium levels. The reactors were setup with calcite, phosphate, and fluoride in synthetic groundwater matrix.