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Fluoride Uptake by Calcium and Phosphate Amendments in Water in the Presence of Organic Matter

MR. MANIRATNAM PRAKASH AND ABHAS SINGH
Indian Institute of Technology Kanpur

Long-term consumption of water with high fluoride (F) concentrations (> 1.5 mg L^{-1}) may lead to dental and skeletal fluorosis. Although F may precipitate as the thermodynamically stable mineral fluorapatite ($Ca_5(PO_4)_3F_{(s)}$) in the presence of calcium (Ca) and phosphate (PO_4) in groundwater, the rate of precipitation is very slow (k = $10^{-28.2}$ µmoles. m^{-2} min^{-1}).[1] This rate can be enhanced by using fluorapatite as a seed (1 g L^{-1}).[2] However, in the presence of organic matter the extent and kinetics of F uptake in the form of fluorapatite is currently not known. This information is relevant to understand F uptake in contaminated soils. Hence, batch experiments were performed with F-spiked ultrapure water (resistivity ≥ 18.2 MΩ-cm) and synthetic groundwater containing major anions and cations consistent with the typical concentrations reported[3] in shallow groundwater of the middle Gangetic plain of India. To this water, synthetic (glucose ($C_6H_{12}O_6$)) and natural (humic acid) organic matter were added at aquifer-relevant concentrations of 1 mg L^{-1}, 10 mg L^{-1}, and 100 mg L^{-1}-carbon (C). The experiments were performed under the following conditions: (i) Seed + Ca + PO_4, (ii) Seed + Ca + PO_4 + organic carbon, (iii) Seed + No Ca + No PO_4 + organic carbon, and (iv) No Seed + Ca + PO_4 + organic carbon. Aqueous samples were collected at regular intervals for measurements of F, pH, Ca, PO_4, organic carbon, and different ions. Solid samples were also collected at the end of the experiments after 30 d for X-ray powder diffraction (XRD), scanning electron microscopy (SEM), and associated energy dispersive spectroscopy (EDS). For ultrapure water spiked with F, geochemical analysis of batch data shows that the presence of organic matter (glucose) in condition ii affects the kinetics of fluorapatite precipitation (Figs. 1A and 1B) by decreasing the pH of water. However, the F uptake kinetics remained unaffected in synthetic F-contaminated groundwater, even in the presence of organic matter, possibly because the system pH was well-buffered with bicarbonate (Fig 1C). These findings have implications in developing robust, suitable treatment approaches for F-contaminated shallow groundwater.

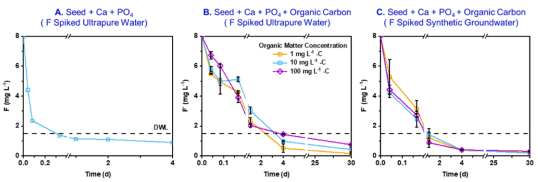

Fig. 1 Variation of fluoride (F) with time and organic matter under conditions supersaturated with respect to fluorapatite ($Ca_5(PO_4)_3F_{(s)}$) when fluorapatite was present as a seed and (A) no organic matter was added to ultrapure water; (B) variable organic matter (glucose) was added to ultrapure water; and (C) variable organic matter (glucose) was added to synthetic groundwater. Calcium and phosphate were initially added to each batch reactor to achieve supersaturation. All the experiments for conditions B and C were done in triplicate. For condition A, F was below (drinking water limit) DWL for ~8 h, whereas in conditions B and C, it took ~ 4 d and ~ 1 d, respectively.

References:
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