

# Investigation of Uranium (VI) Uptake by Rhodochrosite and Its Effect on Uranium Mobility in Sediments

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Widespread contamination of India's groundwaters with uranium (U) has garnered increasing attention in recent years. Our previous investigations on oxic-anoxic Indian groundwaters revealed lower probabilities of U mobilization in the presence of elevated manganese (Mn), whose concentrations seemed to be controlled by rhodochrosite [ $\text{MnCO}_{3(s)}$ ] [1,2]. While several environmentally-relevant carbonate minerals [e.g.  $\text{CaCO}_{3(s)}$ ,  $\text{SrCO}_{3(s)}$ ,  $\text{BaCO}_{3(s)}$ ] have been previously reported to sequester U(VI), such capability of rhodochrosite is not known. In this contribution, the fate of dissolved U in under- and super-saturated conditions with respect to rhodochrosite was studied in completely mixed batch reactors at 25°C at 110 rpm. Pre- and post-experimental aqueous characterization of samples revealed  $\geq 50\%$  decrease in U in both conditions favoring precipitation and dissolution of  $\text{MnCO}_{3(s)}$  (Figure 1). Scanning electron microscopy and associated energy-dispersive X-ray spectroscopy and X-ray diffraction (XRD) on precipitated solids confirmed the presence of rhodochrosite. No U-bearing solid phases were detected, consistent with the undersaturated state indicated by saturation indices. Also, the possibility of redox conversion of U(VI) to U(IV) by Mn was thermodynamically found unfavourable. Detailed solid-phase characterizations using Raman spectroscopy, X-ray photoelectron spectroscopy and transmission electron microscopy showed U(VI) incorporation within  $\text{MnCO}_{3(s)}$  crystals. In order to evaluate the applicability of this mechanism in aquifer-relevant settings, batch leaching studies were conducted on contaminated sediments by systematically varying degrees of initial  $\text{MnCO}_{3(s)}$  supersaturation in two different influent matrices: ultrapure water (UW) and synthetic groundwater (SGW). Consistent with the results from controlled experiments, a negligible release of U was observed in both UW and SGW under highly supersaturated conditions of  $\text{MnCO}_{3(s)}$ . X-ray diffraction on leached sediments clearly indicated presence of  $\text{MnCO}_{3(s)}$ , which suggested that a similar mechanism that inhibited U mobilization from sediments was likely prevalent in aquifers in conditions where secondary precipitation of  $\text{MnCO}_{3(s)}$  was predominant.

[1] Mohapatra et. al. (2021), ACS Earth Space Chem., 5(10), 2700-2714.

[2] Sujathan and Singh (2023), Environ. Sci. Technol., 57(5), 1970-1986.

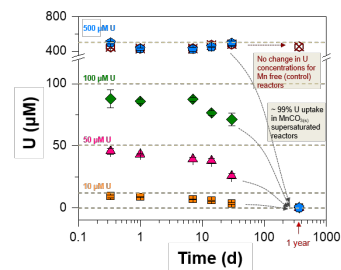


Figure 1. Variation of dissolved U in reactors spiked with Mn, dissolved inorganic carbonate (DIC) and varying concentrations of U. The initial Mn and DIC were 10 mM each except in control reactors where only 10 mM DIC was present. The markers represent mean value of three replicates and the error bars denote 1 $\sigma$  standard deviations of the means.