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Effect of citrate on the fate of uranyl phosphate in a soil column

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Due to the low solubility of uranyl phosphate minerals (UP), the deliberate precipitation of UP is a remediation strategy to limit the mobility of uranium contamination in the subsurface. A comprehensive understanding of the long-term stability of these precipitates, particularly in the presence of natural organic ligands, is necessary to determine whether the precipitation of UP is an effective remediation strategy or if these minerals could become secondary contamination sources of uranium in the subsurface. This study uses a combination of saturated soil column experiments, solid phase characterization, and batch dissolution experiments to explore the release of uranium from the UP chernikovite [$(H_3O)(UO_2)(PO_4)•3H_2O$] in soil with or without the presence of citrate, an organic ligand produced by plants and microbes that can impact the dissolution of chernikovite.

Column experiments demonstrate that citrate both mobilizes cations from the soil, such as aluminum, iron, magnesium, calcium, and sodium, and promotes the dissolution of chernikovite in a soil matrix. In a column receiving 1 mM citrate, the dissolved uranium concentrations in the effluent were over 10^4x greater than uranium concentrations in the effluent from a column without citrate. The concentration of uranium from the citrate column reached a maximum of 54 μM and then decreased even though most (86%) of the solid chernikovite remained within the source. X-ray diffraction analyses revealed differences in the length of the mineral interlayer between the initial chernikovite source and the sources recovered from the column experiments. These changes are most noticeable in the source recovered from the column without citrate and suggest that cations in the soil may be incorporated into the UP structure. In batch dissolution experiments, the sources recovered from the columns with citrate and without citrate produced 89x and 357x less dissolved uranium respectively than the initial chernikovite sources. These recovered sources had similar dissolved uranium concentrations as UP synthesized with potassium and calcium. These findings indicate that citrate is effective at dissolving UP in a soil matrix, but the chernikovite sources are changing to less soluble forms during the column experiments, likely due to interactions with cations, like sodium, potassium, and calcium.