Remediation mechanisms of uranium mill-tailing site at Ningyo-toge, Japan, under the circumneutral condition

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Ningyo-toge is the only uranium mine that was operated in Japan. Various toxic elements such as U, As, and Ra are still present in the mine wastewaters, of which the concentration except for Ra in the wastewater decrease below the reguratory limit while transported through channels to a pond. At this unique site with the mine wastewaters at near neutral pH, the mechanisms of decreasing their concentrations in the wastewaters are not fully understood. In order to understand the fundamental processes of natural remediation at this site, we have investigated the wastewaters and solids from upstream to the pond at the downstream.

Wastewater and sediment samples were collected from Ningyo-toge remediation site, Okayama, Japan and analyzed using a variety of analytical techniques including ICP-AES, -MS, XRD, SEM, synchrotron-based XAFS, DLS and TEM. Wastewater containing 0.18 mg/L of dissolved oxygen (DO) was contacted with oxygenated waters and the DO increased to 1.65 mg/L. Simultaneously dissolved ferrous iron was oxidized to form ferrihydrite nanoparticles as small as <~10 nm, which are associated with minor As and U. Large aggregates of ferrihydrite were sedimented and soluble Mn^{2+} was subsequently adsorbed to ferrihydrite and partially oxidized as revealed by XANES, although no discrete Mn phase formed (Mn : 60.6 umol/L). In contrast, the ferrhydrite nanoparticles as suspended colloids were transported to the pond in downstream, where the wastestream is competely oxidized (DO = 3.09 mg/L). The ferrihydirte colloids further formed aggregates as large as 0.98 um in diameter and precipitated. Subsequently, Mn adsorbed to and accumulated on the surface of ferrihydrite aggregate by oxidation to Mn^{4+} was promoted at the surface with high DO in solution, eventually forming fibrious birnessite nanocrystals (Mn : 9.57 umol/L). It is plausible that Ra concentration decreases by adsorbing to birnessite. Thus, at this specific site, Fe hydroxides nanoparticles play a key role on the instant sequestration of U and As, and subsequent formation of Mn dioxides that can remove Ra from the wastewater.