

Plutonium speciation and formation of nanoparticles with natural organic matter in contaminated environment

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Production Association 'Mayak' is the facility that is responsible for spent nuclear fuel reprocessing, isotope production and radioactive waste management. For decades it was also responsible for the separation of plutonium for military purposes. As a result of activities in 50's and 60's of XX century, large surface and subsurface contamination by different radionuclides occurred. In this study plutonium speciation in bottom sediments and water from natural open reservoirs is reported by various methods including TEM, SEM-EDX, sequential extraction and AsFFF coupled with organic carbon analysis and ICP-MS.

According to plutonium inventory in different reservoirs, its major fraction (>90 %) is in the bottom sediments from the reservoirs. According to the sequential extraction the partitioning of plutonium is governed by the interaction with high molecular weight natural organic matter (NOM) in the bottom sediments.

Despite most of Pu was found in the bottom sediments, small fraction of this element was also found in water. Its speciation was analysed by various methods including Asymmetric Field Flow Fractionation coupled with total carbon analysis and ICP-MS. It was established that Pu was bound to stable nanocolloids with the hydrodynamic diameter of 1-2.5 nm. This corresponds to the fraction of low-molecular weight NOM, presumably fulvic acids. The separate sampling was done to separate enough amount of this NOM for STXM and C-13 NMR examination that confirmed that this fraction correspond to aquatic fulvic acids. The AsFFF separation also indicate the presence of the NOM with the hydrodynamic diameter of 8-15 nm, but no plutonium was detected bound to this fraction.

Steady-state U-series systematics recorded in two deep weathering profiles, India

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Weathering exerts a profound influence on the elemental and isotopic inventories of nutrients in the ocean. Whilst many laboratory experiments have provided a rationale for preferential release or retention of elements or isotopes, empirical studies of deep weathering or lateritic profiles, and suspended sediment have returned more complex answers. This situation is also true for ²³⁴U, whose activity in the ocean is elevated yet few weathering studies have succeeded in documenting the complementary ²³⁴U-depleted residuum.

We present a new comprehensive major, trace element and U-series isotope study of two deeply weathered profiles developed across superimposed (c. 65 Ma) basaltic lava flows in the NE Deccan region of north-central India (c. 22°N, 79°E). In the upper profile, the chemical index of alteration (CIA) correlates strongly with loss on ignition ($r^2 = 0.972$). The lowest CIA of 40 is found in the centre of unweathered corestones and increases both upwards to the thin soil and downwards to the contact with the lower flow. High-precision trace element data show fine-scale but significant and correlated variations in element ratios such as La/Gd, Eu/Eu* and Y/Ho. By contrast, high-field strength elements (HFSE) are only mildly affected by weathering, and Th/U has remained relatively constant (4.04 ± 0.71) across the profile. (²³⁴U/²³⁸U) ranges from 0.79 in the most weathered samples, to 1.01 in the unweathered basalt, showing only weak correlation with CIA and LOI. Preferential ²³⁴U loss is strongly correlated to ²³⁰Th loss ($r^2 = 0.841$) along the ²³⁴U/²³⁰Th equiline suggesting young steady state behaviour.

The lower profile is more homogeneously weathered, with CIA ranging from 0.71 to 0.79, whilst Y/Ho is consistently subchondritic (22.5 ± 0.34) similar to that of the most strongly weathered samples in the upper profile. HFSE are also fractionated: both Zr/Hf and Nb/Ta increase downwards through the profile, whilst Th/U is generally higher than in the upper profile, ranging from 4.8 to 7.4, indicating a general loss of U. U-series confirm strong disequilibria with (²³⁴U/²³⁸U) ranging from 0.53 in the more weathered samples to 0.97, and is remarkably strongly correlated with depth ($r^2 = 0.981$). Again, ²³⁴U loss is strongly correlated ($r^2 = 0.969$) to ²³⁰Th loss along the ²³⁴U/²³⁰Th equiline.

In both flows, preferential weathering occurs towards the lower flow contacts. Young, steady-state loss of daughter nuclides confirms experimental studies regarding preferential solubility and, unlike data for suspended sediment, provides an explanation for the ²³⁴U excess in river and seawater.