Abstract

The University of Manchester

Saifeldin Mohammed Babiker Siddeeg

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Geochemistry of natural radionuclides in uranium-enriched river catchments

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Radionuclides from natural U-series in sediments from two river catchments in the UK have been studied. The aim was to gain insight into the behaviour of ^{238}U, ^{234}U, ^{230}Th and ^{226}Ra in real natural systems enriched in uranium. A radiochemical method for radium separation followed by alpha spectrometric measurement has been developed. The method allowed use of ^{225}Ra, in equilibrium with the parent ^{229}Th, as a yield determinant, and has been applied in ^{226}Ra concentrations measurements in the selected areas of study.

U-series progeny, ^{238}U, ^{234}U, ^{230}Th and ^{226}Ra, in totally dissolved sediments from the valley of the River Noe and the fraction leached by aqua regia, have been measured. Total sediment contents ranged from 9 ± 2 to 184 ± 8 $Bq.kg^{-1}$ for uranium, 9 ± 3 to 200 ± 13 $Bq.kg^{-1}$ for thorium and 18 ± 1 to 179 ± 8 $Bq.kg^{-1}$ for radium. The activity concentrations in the leached fractions, compared with the total, were 46% for uranium, 54% for thorium and 56% for radium, on average. The radionuclides showed extensive disequilibrium and this suggested a complex leaching/accumulation of uranium as well as an impact of organic matter and secondary minerals.

Uranium and radium have been geochemically characterised in sediments from near the South Terras abandoned uranium mine, Cornwall. Background activity concentration levels of uranium in sediments ranged from 13 ± 3 to 290 ± 14 $Bq.kg^{-1}$, with radium from 42 ± 4 to 424 ± 23 $Bq.kg^{-1}$. Elevated concentrations of uranium and radium were measured in two samples, S3 with 1820 ± 36 $Bq.kg^{-1}$ for uranium and 940 ± 53 $Bq.kg^{-1}$ for radium; and S7 with 4350 ± 53 $Bq.kg^{-1}$ for uranium and 1765 ± 48 $Bq.kg^{-1}$ for radium. Sequential chemical extraction for the two samples revealed that both uranium and radium were associated with organic and carbonate fractions, with 25 % of the uranium in the resistant phase of S7. $^{234}U/^{238}U$ activity ratios of the sequential extraction fractions showed different trends in the sediments, and this was linked to the impact of organic matter and/or exchange between water and sediment. Uranium-bearing minerals in association with potassium, calcium, iron, manganese and arsenic have been identified in these sediments.