

$(^{234}\text{U}/^{238}\text{U})$ disequilibrium in fresh waters: Tracing the sources of U from a monolithological catchment area

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The use of ($^{234}\text{U}/^{238}\text{U}$) radioactive disequilibrium in river waters as tracer of the water-rocks interactions requires a precise knowledge of both the sources and the processes controlling the supply of the U isotopes in river waters. The aim of this work is to investigate these points at the scale of a small monolithological catchment. For this purpose, the temporal variations of the U and Sr isotope ratios, as well as the U, Sr, Rb and Ba concentrations, were studied at the outlet of the experimental basin (0.8 km²) representing the springs of the Strengbach (Vosges, France). The waters were regularly collected between January 1996 and March 1998. Samples from a flood event of May 1994 were also studied. All these waters were filtered at 0.22 μm, acidified and stored into polypropylene bottles on site. The U, Sr, Ba and Rb contents were determined by ICP-MS, the U and Sr isotopic compositions on a VG Sector mass spectrometer.

The data reveal a significant decrease of the ($^{234}\text{U}/^{238}\text{U}$) activity ratio, from 1.02 to 0.96, when the flow increases, during the flood of May 1994, as well as during the yearly collection (Fig. 1). The trace-element concentrations of the waters collected during the year tend to decrease when the discharge of the stream increases: they decrease by 30% for the Sr (15 to 12 ppb) to about a factor two for U (0.2 to 0.1 ppb). These variations differ from those found for the flood event: when the discharge increases, the U content increases (from 0.2 to 0.4 ppb) and the Sr concentration strongly decreases (19 to 10 ppb). Such a discrepancy is also found for the dissolved organic carbon (DOC); its amounts increase during the flood event of May 1994 (A. Probst, pers. com.), whereas it remains fairly constant at 1.11–1.44 mg/L in the samples collected during the year.

The decrease of the ($^{234}\text{U}/^{238}\text{U}$) ratio when the flow increases, with values above and below the secular equilibrium, indicates that at least two main reservoirs have to be involved to explain the ($^{234}\text{U}/^{238}\text{U}$) ratio in the dissolved load of the Strengbach water at its spring. One with a slight

^{234}U enrichment is thought to represent a water which weathered the granitic bedrock in secular equilibrium. The low ($^{234}\text{U}/^{238}\text{U}$) disequilibrium of this end member agrees with mineralogical observations on similar granites, which have shown that some U bearing mineral phases have lost nearly all their U in a similar weathered granite (Pagel, 1982). Thus, the leaching of U from these minerals should induce no important $^{234}\text{U}/^{238}\text{U}$ fractionation. The second reservoir with a ($^{234}\text{U}/^{238}\text{U}$) ratio below equilibrium, suggests a mobilization of U from material already weathered. This simple mixing scheme model envisaged for the samples collected during the year in between the flood events, is favoured by the fairly good correlation between the ($^{234}\text{U}/^{238}\text{U}$) and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and between the ($^{234}\text{U}/^{238}\text{U}$) activity ratio and the reciprocal of the U contents (Fig. 2).

However, this mixing model cannot explain the geochemical variations observed during the flood event of May 1994. When the flood data are plotted in the two former diagrams, they define a trend significantly different to that of the other water samples. The interpretation of the whole data in terms of mixing implies the interference of a third component, with low $^{87}\text{Sr}/^{86}\text{Sr}$ and ($^{234}\text{U}/^{238}\text{U}$) ratios and a high U concentrations. This end member also relates to the increase of the DOC content. The soils developed in the Aubure catchment belong to the brown podzolic soils which are known to present high DOC contents in the upper horizons (O or A) and low DOC contents in the deeper horizons (C) or in the alterite (Thurman, 1985). Consequently, these different characteristics allow to explain the chemical composition of the waters during the flood event as recording a contribution of water flowing through the superficial horizons of the catchment. This water, with respect to its high concentration of organic matter, dissolves or complexes U. The importance of organic colloids in the transport of U in waters is now well recognized (Porcelli *et al.*, 1997; Dearlove *et al.*, 1991). The supply of U in the dissolved load of the

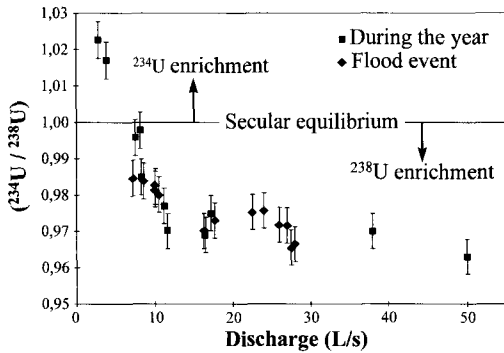


FIG. 1. Variations of ($^{234}\text{U}/^{238}\text{U}$) activity ratios against the discharge of stream.

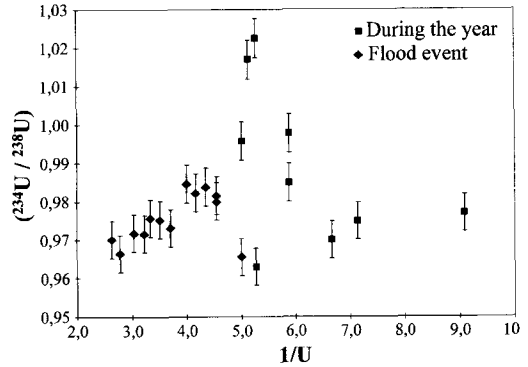


FIG. 2. ($^{234}\text{U}/^{238}\text{U}$) vs $1/U$.

Strengbach, at the outlet of the Aubure experimental basin, can thus be explained with three components: during the year, a bed rock component, predominant during very lean flows, and a 'weathered rock' component prevailing in higher flows. This 'weathered rock' end member is obviously different from the end member involved in the flood event of May 1994. With a ($^{234}\text{U}/^{238}\text{U}$) disequilibrium below unity and a low DOC content (about 1.5 mg/L), the 'weathered rock' end member is a water that can only come from deeper horizons of the soils and/or from alterite. In addition to these two components an U contribution from the superficial soil horizons is involved during the flood event. These results clearly show that an important part of the dissolved U transported by the Strengbach waters originates from U remobilized from material that was already weathered.

From an hydrological point of view, the whole data can be explained by assuming an aquifer located in the deeper horizons of the granitic alterite, the surface of which varying vertically with time. During lean flows, the aquifer is probably confined near the bedrock and the ($^{234}\text{U}/^{238}\text{U}$) ratio of the water tends

to be greater than unity. When the discharge increases, the surface of the aquifer raises, and could be extended into the weathering horizons and have a ($^{234}\text{U}/^{238}\text{U}$) ratio below unity. During the flood event, water from the superficial horizons of the soils becomes predominant.

As a whole, these results demonstrate that U in dissolved load of the rivers does not only represent the ^{234}U - ^{238}U fractionation induced during weathering processes of surface rocks, but it can also be influenced by U remobilization from material already weathered. The results also suggest that the ($^{234}\text{U}/^{238}\text{U}$) disequilibrium could become relevant to evaluate the contribution of different weathering horizons profiles to the chemical flux transported by river waters.

References

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