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Experimental investigation of uranium-series isotope mobility in a basaltic weathering profile

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The measurement of uranium (U)-series isotopes in regolith can be used to determine the formation rate of weathering profiles. This approach aims at following how the U-series isotope composition of primary minerals (i.e. those derived from the parent material) vary during the development of the weathering profile. Nevertheless, regolith samples are a complex mixture of primary minerals, secondary minerals that are the residue of primary mineral weathering, secondary minerals that precipitate from pore water, minerals derived from atmospheric deposition and organic matter. In this study, firstly we aim at isolating primary minerals and the secondary minerals derived from them, by evaluating a sequential extraction procedure designed to eliminate carbonates, Fe-Mn oxides and organic matter. Secondly, we investigate the behaviour of U-series isotopes during primary mineral dissolution by applying a mild HF/HCl etching solution to the residues of the sequential extraction. These experiments were performed on bedrock, saprolite and soil derived from a basaltic weathering profile in south-eastern Australia.

Results show that up to 50% of U is removed during sequential extraction, suggesting that (i) there is a large pool of labile U in the bedrock and (ii) secondary phases and organic matter account for a large fraction of the U budget in bulk saprolite and soil. Sequential extraction has little impact on the $(^{234}\text{U}/^{238}\text{U})$ activity ratio of bedrock and saprolite, whilst it shows a decrease in soil. This suggests that the pool of U removed from bedrock and saprolite has a $(^{234}\text{U}/^{238}\text{U})$ similar to that of primary minerals; but in the soil, the U removed (mostly from organic matter) is enriched in ^{234}U . This is expected as organic matter uptakes U from pore solutions, which are generally enriched in ^{234}U . During HF/HCl etching, the $(^{234}\text{U}/^{238}\text{U})$ of bedrock and saprolite is greater than 1. Sheng and Kuroda [1] previously proposed that $(^{234}\text{U}/^{238}\text{U}) > 1$ in rocks could be explained by the preferential dissolution of a soluble phase, depleted in ^{234}U relative to ^{238}U as a result of alpha recoil of ^{234}Th into a more resistant phase. In contrast, $(^{234}\text{U}/^{238}\text{U})$ ratios show little variation in the soil, suggesting further hydrolysis has little effect on the U isotope ratio of residual primary minerals in the soil.

These results illustrate the complexity of U-series isotope behaviour during chemical weathering. While sequential extraction is a recommended approach to isolate primary minerals and follow their isotopic evolution during regolith formation, it also uncovers that U-series isotope mobility is more diverse than previously postulated.

Reference

[1] Sheng, Z., Kuroda, P., 1986a. Isotopic fractionation of uranium: Extremely high enrichments of 234U in the acid-residues of a Colorado carnotite. Radiochim. Acta 39, 131-138.