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Technical note

Assessment of a sequential phase extraction procedure for uranium-series isotope analysis of soils and sediments

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HIGHLIGHTS

- We tested a sequential extraction procedure for U-series isotope analysis on powdered rock standards (TML-3 and BCR-2) and a soil sample.
- The leaching procedure does not impart any artifactual isotope fractionation to the sample.
- Considerable U and Th are removed from the samples.
- Mineralogy of the sample is unaffected by the procedure.
- Redistribution of isotopes occurred during leaching.

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ABSTRACT

The study of uranium-series (U-series) isotopes in soil and sediment materials has been proposed to quantify rates and timescales of soil production and sediment transport. Previous works have studied bulk soil or sediment material, which is a complex assemblage of primary and secondary minerals and organic compounds. However, the approach relies on the fractionation between U-series isotopes in primary minerals since they were liberated from the parent rock via weathering. In addition, secondary minerals and organic compounds have their own isotopic compositions such that the composition of the bulk material may not reflect that of primary minerals. Hence, there is a need for a sample preparation procedure that allows the isolation of primary minerals in soil or fluvial sediment samples. In this study, a sequential extraction procedure to separate primary minerals from soils and sediments was assessed. The procedure was applied to standard rock sample powders (TML-3 and BCR-2) to test whether it introduced any artefactual radioactive disequilibrium. A new step was introduced to remove the clay-sized fraction ($< 2 \mu\text{m}$). Significant amounts (5–14%) of U and Th were removed from the rock standards during the procedure. No significant alteration in ($^{234}\text{U}/^{238}\text{U}$) and ($^{230}\text{Th}/^{238}\text{U}$) activity ratios of the rock standards occurred during the procedure. Aliquots of soil sample were subjected to the sequential extraction process to test how each step modifies the uranium-series activity ratios and mineralogy. Although no secondary minerals were detected in the unleached soil aliquots, the sequential leaching process removed up to 17% of U and Th and modified their activity ratios by up to 3%. The modification of the activity ratios poses a demand for careful means to avoid redistribution of isotopes back to the residual phase during phase extraction.

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1. Introduction

Chemical weathering plays a major role in the evolution of the Earth's surface. As such, it is important to constrain timescales of weathering processes during soil production and sediment transfer to better understand rates of landscape evolution. Over the

past decade, uranium-series (U-series) isotopes have been used to determine the production rates of soil from bedrock and the transport rates of sediments (e.g., Suresh et al., 2013; Vigier and Bourdon, 2011; Granet et al., 2010; Chabaux et al., 2008; Dosseto et al., 2012, 2008, 2006; DePaolo et al., 2006; Dequincey et al., 2002; Vigier et al., 2001; Rosholt, 1982). U-series isotopes are ideal tools to constrain such timescales as they fractionate during rock-water interaction and their decay rate is such that radioactive disequilibrium operates on timescales similar to that of weathering processes. For a system closed for more than one million years (e.g. bedrock older than lower-Pleistocene), the

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^{238}U – ^{234}U – ^{230}Th isotope system will be in secular equilibrium; i.e., the parent–daughter activity ratios will be 1. During weathering, ^{234}U may be preferentially leached from damaged crystal lattice sites created by the high energy alpha decay of ^{238}U (Rosholt, 1982). Additionally, if the decay of ^{238}U occurs near the surface of a soil or sediment grain, a fraction of the intermediate nuclide, ^{234}Th (parent of ^{234}U), may be ejected out of the grain due to the recoil energy of the decay, decreasing the ($^{234}\text{U}/^{238}\text{U}$) (herein, a ratio in parentheses denotes the activity ratio) (Kigoshi, 1971). In oxidising conditions, U will have an oxidation state of +6 and form the uranyl ion, $\text{U}_\text{VI}\text{O}_2^{2+}$, which is stabilized by highly soluble carbonate complexes at pH 5 to 8 (Langmuir, 1978). Th will be present as Th^{4+} , which is water-insoluble (at pH near 7). The difference in solubility, and hence mobility, causes elemental fractionation between U and Th, which contributes to the radioactive disequilibrium between ^{230}Th and ^{234}U . Since the activities of the isotopes are time dependent, it is possible to model their evolution in weathering profiles and sediments over time (Suresh et al., 2013; Dosseto et al., 2012, 2011, 2008, 2006; Chabaux et al., 2003; Dequincey et al., 2002; Vigier et al., 2001). Two approaches are used to estimate weathering timescales using U-series isotopes. The first utilises the fractionation of U and Th isotopes due to their differences in chemical mobility. The second approach uses the isotopic fractionation of ^{238}U and ^{234}U created by the recoil loss of ^{234}U from the surface of grains. The recoil length for the high energy alpha decay of ^{238}U is ~ 30 nm in most silicate minerals (Hashimoto et al., 1985; Kigoshi, 1971; Turkowsky, 1969) and therefore this fractionation is only measurable in grains of a few tens of micrometres or less. The measured ^{234}U – ^{238}U disequilibrium can be used to estimate the formation age (comminution age) of the grains following the comminution approach of DePaolo et al. (2006). This approach has been used to determine rates of soil formation and timescales of sediment transport and their relationships to past climate changes (Dosseto et al., 2010; Lee et al., 2010; Suresh et al., 2010; Lee 2009; DePaolo et al., 2006).

The approaches discussed above are based theoretically on the evolution of U-series isotopes in primary mineral grains since the onset of bedrock weathering, assuming that these isotopes are in secular equilibrium prior to weathering, i.e., that the bedrock is older than 1 Ma. However, several previous studies (e.g., Suresh et al., 2013; Dosseto et al., 2012, 2008, 2006; Dequincey et al., 2002) have analysed bulk soils and/or sediments, which are complex mixtures of primary minerals, secondary phases and organic materials. Separation of primary minerals from the bulk material for U-series analysis is expected to significantly improve the accuracy of the results obtained from the approaches described above. Efforts have been made to separate primary minerals for the comminution age approach using U isotopes by Dosseto et al. (2010), Lee et al. (2010) and Suresh et al. (2010). However, the effect of the leaching procedure and separation of

clay-sized fraction on the U-series isotopes of the primary minerals still needs to be tested.

In order to test the reliability and examine the effects on nuclide activity ratios and elemental concentrations of phase extraction procedures proposed here to isolate the primary mineral grains of soil/sediments, we have carried out sequential leaching of soil sample aliquots using a procedure adopted and slightly modified from Schultz et al. (1998) to remove exchangeable and adsorbed fractions, organic materials, carbonates, and amorphous and crystalline Fe–Mn oxides (Table 1). We have added an additional step to physically remove the clay-sized particles by centrifugation (discussed in detail below). The residues from each step of the sequence were analysed for mineralogy, U and Th elemental and isotopic composition and particle size distribution to assess the effects of the removal of the different phases. In addition, powdered standard rock samples were subjected to the same sequential leaching and clay-size fraction removal procedure to assess whether the procedure induces any isotopic fractionation.

2. Materials and method

In order to determine whether the sequential leaching and clay separation procedure itself produces radioactive disequilibrium, we analysed USGS geochemical reference materials TML-3 and BCR-2. TML-3 (Table Mountain Latite) is a Pliocene lava from Sonora, California, which has U-series isotopes in secular equilibrium (Sims et al., 2008; Turner et al., 2001; Williams et al., 1992). BCR-2 (Basalt Columbia River) also has U-series isotopes in secular equilibrium (Prytulak et al., 2008; Hoffman et al., 2007). Both standards were subjected to the sequential leaching and clay removal procedure. A soil sample from a previously studied weathering profile, developed over granitic bedrock at Frogs Hollow, south-eastern Australia (Suresh et al., 2013, 2010) was used to investigate how the mineralogical, elemental and isotopic composition of soil material evolves during sequential leaching and the selective removal of different phases. The Frogs Hollow soil sample is ideal for such a study as secondary phases were not detectable by X-ray diffraction (XRD), which therefore, permits determination of how the leaching and clay-separation procedure affects U-series isotopes in primary minerals in soil.

All reagent solutions were prepared using Millipore Milli-Q water with 18.2 M Ω cm resistivity. Acid-washed polypropylene bottles were used for the storage of solutions. To remove exchangeable, adsorbed and organic phases from the soil, a solution of sodium hypochlorite in hydrochloric acid was prepared, using Merck Ultrapur sodium hypochlorite (6–14% strength) and Teflon distilled hydrochloric acid (Table 1). Sodium acetate solution, prepared with Merck Suprapur[®] sodium acetate and

Table 1
The leaching and clay separation procedure.

Leached fraction	Reagents	Process
(1) Exchangeable/adsorbed/organics	Sodium hypochlorite at pH 7.5 (15 ml/g of sample)	Add reagent to the sample. Heat in the oven at 98 °C for 30 min. Centrifuge at 7000 rpm for 15 min then discard the supernatant. Repeat these steps, add 10 ml ultra-pure water and centrifuge to rinse.
(2) Carbonates	1 M sodium acetate, adjusted to pH 4 with acetic acid (10 ml/g of sample)	Add reagent to the sample. Agitate at room temperature for 2 h using a rotary mixer. Centrifuge at 7000 rpm for 15 min and discard supernatant. Repeat these steps. Afterwards add 10 ml ultra-pure water, centrifuge at 7000 rpm for 15 min then discard the supernatant.
(3) Amorphous and crystalline Fe–Mn oxides	0.04 M hydroxylamine hydrochloride (10 ml/g of sample)	Add reagent to the sample. Agitate at room temperature for 5 h, centrifuge at 7000 rpm for 15 min, discard supernatant, add 10 ml Milli-Q water, centrifuge at 7000 rpm for 15 min then discard the supernatant.
(4) Clay	5% sodium hexa meta phosphate solution, filtered at 0.45 μm , 50 ml	Add reagent to the sample. Ultrasonicate at 190 W for 20 s. Agitate overnight. Centrifuge at 1500 rpm for 34 s and break-stop the centrifuge in 13 s to avoid resettling of clay. Discard supernatant, and repeat centrifugation step with Milli-Q water until the supernatant is clear.

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