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Discussion

Soil formation rates determined from Uranium-series isotope disequilibria in soil profiles from the southeastern Australian highlands

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ABSTRACT

The sustainability of soil resources is determined by the balance between the rates of production and removal of soils. Samples from four weathering profiles at Frogs Hollow in the upper catchment area of the Murrumbidgee River (southeastern Australia) were analyzed for their uranium-series (U-series) isotopic composition to estimate soil production rates. Sequential leaching was conducted on sample aliquots to assess how U-series nuclides are distributed between primary and secondary minerals. Soil is increasingly weathered from bottom to top which is evident from the decrease in ($^{234}U/^{238}U$) ratios and increase in relative quartz content with decreasing soil depth. One soil profile shows little variation in mineralogy and U-series geochemistry with depth, explained by the occurrence of already extensively weathered saprolite, so that further weathering has minimal effect on mineralogy and geochemistry. Al_2O_3 is mobilized from these soils, and hence a silicon-based weathering index treating Al_2O_3 as mobile is introduced, which increases with decreasing soil depth, in all profiles. Leached and unleached aliquots show similar mineralogy with slight variation in relative concentrations, whereas the elemental and isotopic composition of uranium and thorium show notable differences between leached and unleached samples. Unleached samples show systematic variations in uranium-series isotopic compositions with depth compared to leached samples. This is most likely explained by the mobilization of U and Th from the samples during leaching. Soil residence times are calculated by modeling U-series activity ratios for each profile separately. Inferred timescales vary up to 30 kyr for unleached aliquots from profile F1 to up to 12 kyr for both leached and unleached aliquots from profile F2. Muscovite content shows a linear relationship with U-series derived soil residence times. This relationship provides an alternative method to estimate residence timescales for profiles with significant U-series data scatter. Using this alternative approach, inferred soil residence times up to 33 kyr for leached samples of profile F1 and up to 34 kyr for leached samples of profile F3 were determined. A linear relationship between soil residence times and WIS (Si-based Weathering Index) exists and is used to estimate soil residence times for profile F3 (up to 28 kyr) and F4 (up to 37 kyr). The linear relationship between soil depth and calculated residence time allows determination of soil production rates, which range from 10 to 24 mm/kyr and are comparable to the rates determined previously using cosmogenic isotopes at the same site (Heimsath et al., 2001b). This implies that at this site, on the highland plateau of southeastern Australia, soil thickness has reached steady-state, possibly as a result of stable tectonic conditions but despite variable climatic conditions over the timescale of soil development. Soil-mantled landscapes are the geomorphic expression of this balance between soil production and denudation, and our results show that in tectonically quiescent regions, this landscape can be achieved in less than 30 kyr.

1. Introduction

The availability of soil resources depends on the balance between soil production via weathering and loss by erosion. If the soil production rate is greater than or equal to the erosion rate, the soil resource is sustainable. In a compilation and review of soil production and erosion rates estimated for various climatic and geological settings, Montgomery (2007) reports an imbalance between agricultural soil loss and replenishment of soil resulting in a net soil loss; raising a concern for the availability of sustainable soil resources. To estimate soil production rates and therefore assess soil sustainability it is necessary to quantify the soil residence time, i.e. the time elapsed since conversion of saprolite or bedrock into soil.

Until recently, soil production rates were difficult to quantify, inferred from weathering rates by methods using stream water and

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sediment chemistry (Owens and Watson, 1979a, 1979b; Pavich, 1989; Wakatsuki and Rasyidin, 1992; Troeh et al., 2004). These studies relied on assumptions such as homogeneous bedrock composition or unvarying stream water composition representative of solute abundance over the timescale of soil development (Alexander, 1988; Owens and Watson, 1979a, 1979b). Over the past few years, two new techniques have emerged that have significantly improved our ability to quantify rates of soil production. Firstly, the measurement of cosmogenic nuclides in the material underlying the soil (saprolite or bedrock) can be used to determine soil denudation rates. Assuming a constant soil thickness over time, soil production rates can be determined as they are equal to denudation rates (Heimsath et al., 2000, 2001b). However, this technique cannot be used to ascertain imbalances between soil production and loss since both processes are assumed to operate in equilibrium. Secondly, uranium-series (U-series) isotopic compositions of soil/saprolite material in a depth profile can be modeled to quantify the residence time of weathered material within the profile (*soil or saprolite residence time*). This method is based on the principle that the U-series isotopes fractionate during chemical weathering and their abundance is time-sensitive due to their radioactive nature (e.g. Chabaux et al., 2003, 2013; Ma et al., 2010, 2013; Dosseto et al., 2008a, 2008b, 2012; Dequincey et al., 2002). For a system that has remained closed for more than 1 Myr, all daughter–parent activity ratios of the uranium decay chains (e.g. ($^{234}U/^{238}U$) or ($^{230}Th/^{234}U$); parentheses denote activity ratios throughout this article) will be equal to 1. This is termed *secular equilibrium*. The implication is that for a parent rock older than 1 Myr, its U-series isotopic composition is known; it will be in secular equilibrium irrespective of the lithology. During soil formation, U-series isotopes fractionate, creating *radioactive disequilibrium* between parent–daughter nuclide pairs. ^{234}U is preferentially mobilized compared to ^{238}U as a result of two processes: (i) the recoil loss of ^{234}Th from mineral grains due to the decay of ^{238}U within a few 10's of nm of the grain surface, and subsequent decay of ^{234}Th into ^{234}U in the surrounding medium (Kigoshi, 1971); (ii) preferential leaching of ^{234}U located in radiation-damaged crystal sites from which it can be easily mobilized. Thus, ($^{234}U/^{238}U$) activity ratios <1 should be observed in soils. Fractionation between U(^{234}U, ^{238}U) and ^{230}Th occurs as a result of the difference in their solubility. Oxidizing conditions prevail in most soils and so U will be present as U^{6+}, which is soluble in waters as the uranyl ion, $U_{VI}O_2^{2+}$, is stabilized by highly soluble and non-reactive carbonate complexes (Langmuir, 1978). Th will be present as Th^{4+}, which is insoluble. As a result, we expect ($^{230}Th/^{234}U$) ratios >1 in weathering profiles. Once radioactive disequilibrium is produced, a given parent–daughter system will return to secular equilibrium governed by natural radioactive decay over a timescale equivalent to approximately 5 half-lives of the daughter isotope. Thus, the ($^{230}Th/^{234}U$) activity ratio will attain secular equilibrium in ~400 kyr, whereas it will take a million years for ($^{234}U/^{238}U$). Addition of isotopes of U or Th due to processes such as illuviation, dust deposition, lateral transport or bioturbation will also contribute to the U-series isotope composition of soils and this needs to be taken into account when modeling U-series isotopic composition of weathering profiles (Chabaux et al., 2003; Dequincey et al., 2002).

The U-series approach has been used by Dosseto et al. (2008a) to study soil production rates on the escarpment of the eastern side of the Great Dividing Range in southeastern Australia. The reported soil production rates ranging from 12 to 77 mm/kyr are comparable to the range of denudation rates given by Heimsath et al. (2000) (10 to 50 mm/kyr). In this study, we apply the U-series approach to soil profiles at Frogs Hollow in the highlands (Fig. 1a), west of the escarpment site studied by Dosseto et al. (2008a). Compared to the escarpment area, the highland region is characterized by lower relief, erosion rates, mean annual temperature and rainfall. Pleistocene periglacial effects may also have been more important in the highlands, compared to the escarpment (Galloway, 1965; Barrows et al., 2001). At the Frogs Hollow study site, Heimsath et al. (2001b) determined soil production rates using cosmogenic nuclides ^{26}Al and ^{10}Be. With an assumption of constant soil thickness over time, the determined soil production rates vary from 10 mm/kyr for 70 cm-thick soil profiles to 50 mm/kyr for 20 cm-thick profiles, with soil production rate inversely related to soil thickness and slope curvature. Heimsath et al. (2001b) tested the steady-state soil thickness assumption by measuring a cosmogenic nuclide profile in a protruding tor. However, although this experiment constrains how soil is being denuded (and Heimsath et al. (2001b) show that at this site, soil was stripped during a rapid event more than 150 kyr ago), it does not allow determination of how soil thickness has evolved through time. Here, we use U-series isotopes measured in soil and saprolite material as an alternative approach to infer soil production rates, and compare the results to those of Heimsath et al. (2001b). The advantage of the U-series isotope technique is that steady-state soil thickness is not a required assumption, although other assumptions are needed and these are presented below. Comparing the soil production rates estimated from soil residence times determined using U-series activity ratios of soil profiles from Frogs Hollow with those determined using cosmogenic radionuclides (Heimsath et al., 2001b) is expected to put insight into the sustainability of soil-mantled landscape at the studied site.

Yoo et al. (2007) developed a model to study timescales of soil transport on hill slopes, combining chemical weathering and physical erosion. One of the important requirements for the model is the determination of soil residence time due to soil production from bedrock. To apply their model to estimate hill slope soil transport time at Frogs Hollow, Yoo et al. (2007) used the soil production rates determined by Heimsath et al. (2001b), which are inferred from soil denudation rate. The U-series isotope method discussed here gives a more direct method for determining soil residence time.

2. Field description

Frogs Hollow is a small area (0.04 km^2) in the upper catchment of the Murrumbidgee River (Fig. 1). The river flows from the Great Dividing Range of southeastern Australia towards the west, where it joins the Murray River and flows into the Southern Ocean. The study locality at Frogs Hollow is at an elevation of 930 m, 80 km south–southeast of Canberra, 75 km from the coast, and ~12 km west of the eastern escarpment (Fig. 1a). A detailed site description is given by Heimsath et al. (2001b). The underlying bedrock at this site is Devonian granite (Richardson, 1976). The average summer temperature is 19 °C and the average winter temperature is 5 °C. Annual average rainfall is 500 mm (Bureau of Meteorology, NSW, 2010). Patches of dry schlerophyl woodland are present above a grass understory. Samples were collected from four pits (F1–F4) dug at the most convex points on ridge tops, and were taken from both sides of a colluvial hollow to avoid any contribution from laterally transported soil (Fig. 1b). Profiles F1 and F2 are on the western ridge, where F2 is located 80 m south of F1 and 6 m lower in elevation. Profiles F3 and F4, on the eastern ridge of the hollow, are 100 m apart and F4 is at an elevation 3 m lower than that of F3. The two ridges are ~75 m apart.

Samples were collected at 8 cm intervals by inserting a cylindrical steel pipe horizontally into the pit wall. The four soil profiles were chosen on the basis that they replicate the same geomorphic and pedological conditions: high convexity on top of the ridges (Fig. 1). The expectation was that they would replicate profile properties and residence times. However, several differences in