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Considerations for U-series dating of sediments: Insights from the Flinders Ranges, South Australia

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ABSTRACT

Uranium isotope ratios have been determined for the fine-grained detrital fraction of Pleistocene Wilkawillina valley-fill sediments, four local Proterozoic bedrock samples and fine-grained aeolian material from a sand dune deposit of the Flinders Ranges, South Australia. The aim was to quantify the comminution age, i.e. the time elapsed since physical weathering of the bedrock, and residence time of the valley-fill sediments and to place tighter constraints on input parameters for the comminution age calculation. Despite using two independent approaches for determination of the recoil lost fraction of ^{234}U from the sediment (weighted geometric and surface area estimates), samples fail to produce realistic comminution ages and hence, residence times. The issues involved in the ability to determine sediment comminution ages are discussed. The ($^{234}U/^{238}U$) activity ratio of the local bedrock is not in secular equilibrium, despite the bedrock being much older than 1 Ma, i.e. the timeframe for ^{234}U and ^{238}U to reach secular equilibrium in a closed system. Using the average Flinders Ranges bedrock ($^{234}U/^{238}U$) ratio instead of an assumed ($^{234}U/^{238}U$) activity ratio of unity for the source would significantly reduce calculated residence times. This result warrants concern for future studies using the comminution approach for which a secular equilibrium source ($^{234}U/^{238}U$) activity ratio is assumed. Significant input of aeolian material may modify the measured ($^{234}U/^{238}U$) activity ratios. Such input may be more tightly constrained in future studies using rare earth element and radiogenic isotopic data. Future comminution studies would benefit from further consideration of the importance of 1) leaching lost ^{234}U from source rock and bulk sediment samples, 2) wind deposition of fine-grained material and 3) the appropriateness and robustness of sample pre-treatment procedures.

1. Introduction

In order to quantify how fast a landscape responds to tectonic, climatic and human factors, accurate weathering rates and soil and/or sediment ages are required. The uranium-series (U-series) isotopes are a valuable tool for deriving the timescales of weathering and erosion processes (e.g., Plater et al., 1992; Scott et al., 1992; Vigier et al., 2001; Dequincey et al., 2002; Chabaux et al., 2003; Granet et al., 2007; Chabaux et al., 2008; Dosseto et al., 2008a, 2008b; Vigier and Bourdon, 2011). Recently, DePaolo et al. (2006) developed a method for dating the formation age of fine-grained sediments, in other words, the time elapsed to present day since physical weathering of source rock to a threshold grain size ($\leq \sim 50\ \mu m$), termed the '*comminution age*' (DePaolo et al., 2006; Dosseto et al., 2010; Lee et al., 2010; Handley et al., 2013). The comminution dating approach utilises ^{234}U–^{238}U disequilibrium in fine-grained sediments attributed to recoil loss of ^{234}U (see Section 2 for details). U-series recoil-loss dating has yielded reasonable timescale estimates when compared to independently constrained ages (e.g., Aciego et al., 2011), but can be offset by several 100 ka (Lee et al., 2010). Therefore, further testing and consideration of the methodology is required to improve the accuracy of ages produced and prove its value as a dating technique.

Here, we present a comminution study on Pleistocene alluvial deposits in the Flinders Ranges, South Australia. The uranium isotopes of local Proterozoic bedrock samples have also been determined to test whether the source material is in ($^{234}U/^{238}U$) secular equilibrium prior to the onset of physical weathering. We also constrain the potential ($^{234}U/^{238}U$) activity ratio of local aeolian material to examine the influence of external inputs on sediment residence time. We show that the bedrock samples have significant ^{234}U–^{238}U disequilibria. This has major implications for the comminution approach and the general assumption that the comminution chronometer does not start until the commencement of physical weathering of bedrock. We suggest that this is due to the preferential removal of ^{234}U from the source material via leaching from recoil-damaged sites.

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2. Comminution age theory

The energy associated with the alpha decay of ^{238}U (half-life, $t_{1/2}=$ 4.5 Ga) to ^{234}Th ($t_{1/2}=24$ days) results in the recoil (physical displacement) of the daughter ^{234}Th nuclide from the initial parent location within a mineral. This recoil distance varies depending on mineralogy but is estimated to range between 20 and 50 nm in common silicate minerals (see Maher et al., 2006 and references therein). ^{234}Th then decays to ^{234}U ($t_{1/2}=245$ ka) via the intermediate ^{234}Pa nuclide ($t_{1/2}=7$ h). If recoil takes place within recoil–length distance of the grain edge then ^{234}Th may be physically ejected from the grain. In large grains (sand-size and larger) the recoil loss of ^{234}Th (hence ^{234}U) is insignificant due to the large volume to surface ratio of the grains. However in fine-grained material (~50 μm or less) the recoil loss of ^{234}Th creates a measurable disequilibrium between the parent, ^{238}U, and 'great-granddaughter', ^{234}U, nuclides, i.e. (^{234}U/^{238}U) ratios of <1 (where the parenthesis denotes an activity ratio). In comminution age theory, the magnitude of ^{238}U–^{234}U disequilibrium in fine-grained sediment is therefore related to the timescale of radioactive decay and proportion of recoil loss of ^{234}Th. A detailed description of the U-series comminution theory and methodology can be found in DePaolo et al. (2006) and Lee et al. (2010). The calculated comminution age of sediment is defined as the period of time elapsed since weathering of bedrock into fine-grained material to present day (Fig. 1). This includes the time a grain has spent in temporary storage e.g., in soils and floodplains and in transport prior to final deposition. To quantify the length of time that the sediment has resided in the catchment since mechanical weathering, prior to final deposition (T_{res}) (Fig. 1), the U-series comminution equation of DePaolo et al. (2006) can be utilised:

$$t_{\mathrm{com}} = -\frac{1}{\lambda_{234}} \ln\left[\frac{A_{meas}-(1-f_\alpha)}{A_0-(1-f_\alpha)}\right]$$

where λ_{234} is the ^{234}U decay constant (2.82629×10^{-6} a^{-1}; using $t_{1/2(^{234}\mathrm{U})}$ of 245,250 a, Bourdon et al., 2003), A_{meas} is the measured (^{234}U/^{238}U) activity ratio of the sediment, f_α is the recoil loss factor, defined as the fraction of ^{238}U decays that result in the recoil loss of the intermediate nuclide ^{234}Th from the grain, and A_0 is the initial (^{234}U/^{238}U) of the source rock. The sediment residence, or transport time (T_{res}), can then be calculated if the deposition age (t_{dep}) of the sediment is known, by simply subtracting the deposition age from the comminution age (Fig. 1).

Previous authors (DePaolo et al., 2006; Lee et al., 2010; Handley et al., 2013) have shown that comminution ages are highly dependant on the value used for the recoil lost fraction (f_α). Estimates of f_α can be produced using a number of different methods (see Maher et al., 2006 and Lee et al., 2010 for summaries of the different approaches). The most commonly employed methods are at present based upon either a weighted geometric estimation (DePaolo et al., 2006) using sample grain size distributions and assumptions for surface roughness and grain aspect ratio, or measurements of specific surface area (e.g., Brunauer–Emmett–Teller (BET) gas adsorption measurements) with an incorporated fractal correction (Semkow, 1991; Bourdon et al., 2009; Aciego et al., 2011) to account for the significant difference between the size of the adsorbed gas molecule (commonly N_2: 0.354 nm) and the recoil length scale (~20–50 nm; Hashimoto et al., 1985; Ziegler et al., 1996) (see Appendix A for f_α calculation equations and input parameter details). As pointed out by previous authors of the few comminution studies undertaken so far (e.g., DePaolo et al., 2006; Lee et al., 2010; Handley et al., 2013) the technique holds significant potential, but much more work is required before it can be considered as an accurate dating tool.

3. Study area

The Flinders Ranges of South Australia (Fig. 2a) are a series of north–south striking ridges of folded, uplifted and dissected, largely sedimentary, Proterozoic and Cambrian rocks (Preiss, 1987). The relatively softer siltstones and shales have been eroded to form valleys and lower elevation rounded hills, while the more weathering-resistant quartzites and sandstones form prominent ridges and peaks as well as a small number of largely enclosed draining basins such as Wilpena Pound and Wilson's Pound (Fig. 2a). The ranges are flanked by several low elevation, internally draining playa lakes such as Lake Torrens, Lake Frome and Lake Callabona (e.g., Fig. 2a) that act as sediment sinks for eroded, fluvial material from the Flinders Ranges. The ranges are one of the most tectonically active regions of Australia and a significant proportion of the present day relief above the piedmont surface (up to 600–1000 m) is attributed to late Miocene to Recent tectonic uplift (e.g., Sandiford, 2003; Quigley et al., 2006). ^{10}Be cosmogenic isotope studies at sites in the Flinders Ranges suggest that the present relief between valley floors and range summits may have been generated in as little as approximately 4 Ma (Quigley, et al., 2007a). The present day climate is arid to semi-arid with mean annual precipitation between <200 mm (east and far north) and >400 mm (range ridges) and annual evaporation exceeding 2000 mm.

Significant (up to 18 m thick) late Pleistocene silt- and clay-rich valley fill deposits, now heavily incised, are documented in both the western and eastern draining catchments of the Flinders Ranges (e.g., Williams et al., 2001; Williams and Nitschke, 2005; Haberlah et al., 2010a, 2010b). Such fine-grained fluvial deposits are not accumulating today. The deposits were initially considered to be lake sediments (Cock et al., 1999) but are more recently described as slope wash deposits, dominated by aeolian sourced-material accumulated in either a 'fluvial wetland' or resulting from flood events (Williams et al., 2001; Haberlah et al., 2010a, 2010b; Haberlah and McTainsh, 2011). Some individual beds, only a few centimetres thick, can be traced for over a 100 m (Williams et al., 2001). The aeolian material is thought to be sourced predominantly from Lake Torrens to the west of the Flinders Ranges (Fig. 2a; Williams and Nitschke, 2005). This lake remained dry to ephemeral during the Quaternary and has accumulated more than 300 m of sediment since the Eocene (Williams and Nitschke, 2005). Based on the prominence of a dated travertine structure, which lies above the present playa surface, the floor of Lake Torrens is inferred to have been lowered by ~2.5 m by wind erosion during the last glacial (Schmid, 1990). The elevated Flinders Ranges act as a dust trap for wind-blown sediment travelling eastwards across the continent (Bowler, 1976; Hesse and McTainsh, 2003). Discontinuous shallow mantles of red-brown very fine sandy or silty clay, that outcrop on the summits and ridges of the Flinders Ranges (irrespective

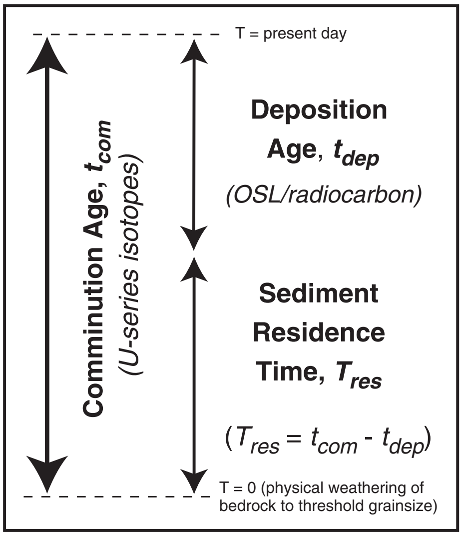

Fig. 1. Schematic diagram showing the relationship between comminution age, deposition age and sediment residence time. Modified from Dosseto et al. (2010). OSL, optically stimulated luminescence.