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Sediment residence times constrained by uranium-series isotopes: a critical appraisal of the comminution approach

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Keywords
sediment, times, residence, constrained, uranium, series, isotopes, critical, appraisal, comminution, approach

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Sediment residence times constrained by uranium-series isotopes: A critical appraisal of the comminution approach

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1. INTRODUCTION

The uranium-series (U-series) isotopes provide invaluable time chronometers for quantification of the rates of Earth’s surface processes over the last several 100 ka, such as silicate weathering and soil and sediment residence in the landscape (e.g., Osmond and Ivanovich, 1992; Vigier et al., 2001; Dequincey et al., 2002; Maher et al., 2004; Dosseto et al., 2008; Andersen et al., 2009 and see Chabaux et al., 2003; Vigier and Bourdon, 2011, for recent comprehensive reviews). Quantification of such rates is crucial to improve our understanding of the relationship between climate change, tectonics and landscape evolution throughout the Quaternary. Recent studies (e.g., DePaolo et al., 2006; Dosseto et al., 2010; Lee et al., 2010) of the fractionation between 234U and 238U in fine-grained sediment have
demonstrated the potential in constraining the comminution age of continentally-derived sediments, that is, the time elapsed since physical weathering of the source rock into fine-grained (<~50 µm) particles (DePaolo et al., 2006; Fig. 1). The comminution age includes the time that a particle has been mobilised in transport, held in temporary storage (e.g., soils and floodplains) and the time elapsed since final deposition to present day (Fig. 1). Therefore, if the deposition age of sediment can be constrained independently, for example via optically stimulated luminescence (OSL) dating, the residence time of sediment (e.g., a palaeochannel deposit) can be computed from:

\[ T_{res} = t_{com} - t_{dep} \]  

where \( T_{res} \) is the sediment residence time (the time between the formation of the silt-sized grain and final deposition, or transport time), \( t_{com} \) the comminution age of the sediment and \( t_{dep} \) is the deposition age of the sediment.

Where it is possible to compare calculated comminution ages of detrital sediments with independent age estimates, the comminution data yield reasonable timescales but maybe offset from independently constrained ages by several 100 ka (e.g., Lee et al., 2010). Clearly the comminution method is still in its infancy and has only been applied in a handful of studies. The technique also remains to be tested and examined for more complex fluvial environments in order to assess its real value as a dating technique. For example, in large catchment areas with potentially much longer transport times, more complex sediment transport pathways and a wide range in source rock lithology. Nevertheless, using the U-series comminution approach, residence times calculated for palaeochannel sediments of the Murumbidgee River in temperate, southeastern Australia (Dosseto et al., 2010) suggested a link between catchment erosion, climate and vegetation.

Here we present the first U-series study of palaeochannel sediments from Cooper Creek in the Lake Eyre Basin in central Australia. This study site affords a great opportunity to assess the comminution approach as the vast catchment area of the basin (~1.2 x 10^6 km^2) drains a wide variety of lithology (including fine-grained sedimentary rocks) and furthermore, the deposition age of the deposits has already been determined independently by Cohen et al. (2010). The objectives of the study are twofold: (1) to determine comminution ages and residence times of Cooper Creek sediments using common assumptions of input parameter values found in the present literature and, (2) estimate both individual parameter and combined comminution age uncertainties for a synthetic sample.

2. 234U–238U COMMINUTION AGE METHOD

2.1. Concept

The ‘comminution age’ dating model of DePaolo et al. (2006) hypothesises that measured disequilibria between U-series nuclides (234U and 238U) in fine-grained continental (detrital) sediments can be used to calculate the time elapsed since mechanical weathering of a grain to the threshold size (~50 µm). When 238U undergoes alpha decay to 234Th, the associated release of energy results in the physical displacement (recoil) of the 234Th nuclide from the original position of 238U. When this decay occurs in a mineral grain it can result in the physical ejection of 234Th from the solid (e.g., into a pore fluid) and lead to the subsequent depletion of 234U (the daughter of the ejected intermediate 234Th nuclide) relative to 238U (Kigoshi, 1971) (Fig. 2). In coarse-grained sediment (sand-sized particles and larger) 234U–238U disequilibria is only measurable at grain edges as the grain radius is much larger than the recoil length of ~30 nm (theoretical x-recoil length of feldspar; Ziegler et al., 1996). However, in fine-grained silt-sized or smaller detrital sediments this process leads to measurable 234U–238U disequilibria in bulk samples comprising fine-grained particles (Fig. 2). This disequilibrium can be used to calculate the comminution age (time since mechanical weathering) of the sediment (\( t_{com} \)) using the following equation (DePaolo et al., 2006):

\[ t_{com} = \frac{1}{\lambda_{234}} \ln \left( \frac{A_{\text{meas}} - (1 - f_s)}{A_0 - (1 - f_s)} \right) \]  

where \( \lambda_{234} \) is the 234U decay constant (2.82629 x 10^-6 yr^-1), using \( t_{1/2\text{C234U}} \) of 245,250 yr, from the compilation of Bourdon et al., 2003), \( A_{\text{meas}} \) is the measured (234U/238U) activity ratio (parenthesis around U-series isotope ratio denotes activity ratio) of the sediment, \( f_s \) is the recoil loss factor, defined as the fraction of 238U decays that result in the recoil loss of the intermediate nuclide 234Th, and \( A_0 \) is the initial (234U/238U) of the source rock.

2.2. Uncertainties in the comminution age calculation

2.2.1. (234U/238U) of the sediment source, \( A_0 \)

The (234U/238U) activity ratio of the source rock, \( A_0 \), has been assumed by previous authors (e.g., DePaolo et al.,
Lee et al., 2010; Dosseto et al., 2010) to be 1, i.e., in secular equilibrium. While this may be expected for unfractured, fresh, crystalline source rocks (e.g., Zeilinski et al., 1981; Rosholt, 1983), it is unclear whether sedimentary rock such as siltstone, formed of silt-sized ‘out-of-secular-equilibrium’ grains, will achieve secular equilibrium after 1 Ma (5 times the half-life of the daughter nuclide, $^{234}$U) (Fig. 2a). If the source rock is porous, permeable and/or fractured then it is possible that $^{234}$Th produced by post-depositional $\alpha$-recoil may continue to be lost, via removal by pore and migrating fluids. In this scenario, $A_0$ will never achieve secular equilibrium and therefore, possess $(^{234}$U/$^{238}$U) < 1 (Fig. 2a). The age of the source rock is also important to constrain as any source rock younger than 1 Ma could potentially be out of secular equilibrium. Furthermore, if the source rock is chemically weathered or if $^{234}$U has been preferentially leached from damaged $\alpha$-recoil sites (see Section 2.3.1), measurable fractionation of $^{234}$U from $^{238}$U may occur prior to physical comminution to the threshold grain size. In such a scenario, the residence time of a particle would represent the time between its isotopic alteration, in a profile, and the time of its final deposition. Whether the source rock is in $^{234}$U-$^{238}$U secular equilibrium needs to be tested for each study. $(^{234}$U/$^{238}$U) activity ratios upon the calculated comminution age of a synthetic sample.

2.2.2. Recoil loss factor, $f_a$

The largest source of uncertainty in the comminution age calculation is attributed to difficulties in the estimation of $f_a$, the fraction of $^{234}$Th lost via $\alpha$-recoil in a bulk sample. Even slight variations in estimated $f_a$ values can significantly affect inferences about weathering rates and comminution ages (e.g., DePaolo et al., 2006; Lee et al., 2010). Several approaches have been used to estimate $f_a$, and these are summarised in Maher et al. (2006) and Lee et al. (2010). Previous research (e.g., Maher et al., 2006; DePaolo et al., 2006; Lee et al., 2010) suggests that reasonable approximations of $f_a$ can be obtained by: (1) weighted geometric estimation of $f_a$ using sample grain size distributions and assumptions for surface roughness and grain aspect ratio, and (2) measurements of specific surface area (e.g., Brunauer–Emmett–Teller (BET) gas adsorption measurements) with an incorporated fractal correction to account for the significant difference between the size of the adsorbed gas molecule and the recoil length scale (Semkow, 1991; Bourdon et al., 2009).

In the geometric estimation, the amount of recoil is related to the size, sphericity and surface area (roughness) of the grain. A SEM image (Fig. 3) of the Cooper Creek palaeochannel sediment shows that mineral grains are rarely spherical or perfectly smooth and therefore, in reality, $f_a$ will be larger than would be assumed for the simple case of a spherical grain (Fig. 2b). Consequently, a weighted geometric model is preferred to yield reasonable estimations of $f_a$ (DePaolo et al., 2006).
method can significantly overestimate \( f_s \), as noted by DePaolo et al. (2006) and Maher et al. (2006). To overcome this scale issue, Bourdon et al. (2009) incorporated the theoretical fractal recoil model of Semkow (1991) that predicts \( f_s \) based on the BET surface measurements according to the following equation:

\[
\frac{d}{4} \left( \frac{2^{\beta - 1}}{4 - D} \right) \left( \frac{L}{L_s} \right)^{D_s} \cdot L \cdot S_{BET} \cdot \rho
\]

where \( D \) is the fractal dimension of the surface (which can be determined via BET data, see Bourdon et al., 2009 for details) and \( a \) is the diameter of the adsorbate gas molecule (N\(_2\) in most BET measurements). This means of estimating the recoil ejection factor has yielded ice core ages (based on the accumulation of \(^{230}\)Th recoil products from dust trapped within the ice) comparable to independently obtained ages (Aciego et al., 2011).

Both the weighted geometric and BET surface area methods described above will be used to estimate and compare values of \( f_s \), comminution age and sediment residence timescale for Cooper Creek palaeochannel sediments, employing the assumed input parameter values found in previously published comminution studies. A synthetic sample will also be used to constrain the comminution age uncertainty associated with the weighted geometric and surface area measurement (with fractal correction) methods.

### 2.3. Additional considerations

The comminution age of a sediment records the time since production of silt-sized material. The question of whether \(( ^{234}\text{U}/^{238}\text{U} )\) activity ratios are modified in the source rock prior to reduction of material to the fine grain size also requires consideration. Furthermore,after the threshold size is reached, dissolution of grain edges via chemical weathering or further physical weathering e.g., via attrition in the river, may remove the grain rims. Removal of the rims (which store the largest disequilibrium between \(^{234}\text{U} \) and \(^{238}\text{U} \)) could effectively reset the U-series clock, producing 
\(( ^{234}\text{U}/^{238}\text{U} )\) ratios closer to secular equilibrium than appropriate for that particle’s true comminution age. Future experiments are required to fully assess the importance of these processes on calculated comminution ages. For comminution age studies of palaeochannel sediments of the Lake Eyre Basin, and other areas, it is important to consider: (1) preferential loss of \(^{234}\text{U} \) through leaching, (2) addition of aeolian material, (3) influence of clay pellet aggregates, and (4) sample mineralogy and the effect of zircon.

#### 2.3.1. Preferential loss of \(^{234}\text{U} \) due to leaching

Preferential loss of \(^{234}\text{U} \) relative to \(^{238}\text{U} \) may occur via leaching of loosely bound recoiled \(^{234}\text{U} \) from damaged lattice sites (e.g., Fleischer, 1980; Andersen et al., 2009). This process could be of significance for both the initial source rock and the transported sediments themselves. Loss of \(^{234}\text{U} \) relative to \(^{238}\text{U} \) due to leaching, in addition to direct \( \alpha \)-recoil ejection of \(^{234}\text{Th} \) from the grain, complicates the assumptions underlying the comminution age method and
would generally lead to overestimates in the calculated ages. However, DePaolo et al. (2006) argue that it is possible to account for the observed range of $f_e$ values in North Atlantic. Pleistocene deep-sea sediments by $\alpha$-recoil alone without the need for preferential leaching of $^{234}$U. Maher et al. (2006) demonstrate that all of the measured $^{234}$U depletion in Pleistocene fluvial sediments from south central Washington can also be accounted for by the $\alpha$-recoil process. Porcelli and Swarzenski (2003) suggest that, in practice, loss of $^{234}$U via leaching along recoil tracks is only plausible when new surfaces are exposed or dry periods allow implantation of recoiled nuclides into adjacent phases. The quantification of the relative importance of leaching lost, versus recoil lost, $^{234}$U in each sample studied is difficult to constrain and is not directly addressed in this study.

### 2.3.2. Addition of aeolian material

The impact of dust on calculated U-series isotope residence timescales of soil and fluvial sediment has received little attention in previous studies, despite the fact that the typical grain size of aeolian material overlaps with that of interest in the comminution approach (<50 $\mu$m). The potential of aeolian material to modify bulk soil or fluvial U-series signatures will depend on its ($^{234}$U/$^{238}$U) ratio and its volume percentage contribution to the deposit. Locally sourced dust from nearby floodplains may have a similar ($^{234}$U/$^{238}$U) activity ratio to the fluvial sediments themselves and therefore, be indistinguishable from the fluvial signature. On the other hand, substantial input of distally derived dust, that has been circulating in the environment over a longer timescale than is representative of the fluvial deposit, may modify the residence time significantly.

The impact of aeolian material input on ($^{234}$U/$^{238}$U) ratios is important to consider for the present study area as a prevalence of dust deposits are recorded in the sedimentary record throughout the Quaternary in southeast Australia (e.g., Butler, 1956; Chen et al., 2002; Hesse and McIntosh, 2003; Cattle et al., 2009; Marx et al., 2011). The rate of dust accumulation in Australia during the late Pleistocene is suggested to have been much greater than during the Holocene (e.g., Hesse, 1994; Chen et al., 2002; McGowan et al., 2008).

In Australia, aeolian material is carried along two major pathways: to the east/southeast and northwest of the continent (Bowler, 1976; McIntosh, 1989; Hesse and McIntosh, 2003). Dust sourced from central Australia (e.g., Lake Eyre Basin (Fig. 4); Hesse and McIntosh, 2003) may be deposited either distally offshore in the Tasman Sea or in New Zealand (e.g., Hesse, 1994; Marx et al., 2009) or be deposited or more proximally on the Australian continent. The large catchment area of the Lake Eyre Basin and the westward/southwestward directed river discharge back towards Lake Eyre, means that flood events in Queensland and New South Wales will carry the top soil, including any recently deposited aeolian material (initially sourced from Lake Eyre, or further west of Lake Eyre), back to the interior. Multiple cycles of essentially eastward dust transport and westward fluvial drainage may therefore occur. The implication for U-series is that such long cycles of sediment transport and reworking in a fluvial-aeolian feedback system, may (1) continually mix recently weathered fluvial material with older reworked material or (2) mean that long residence times do not simply represent the fluvial sediment residence within a fluvial system but rather the combined fluvial and aeolian system as a whole. U-series measurements of Australian dust are urgently required to increase our understanding of the effects of dust additions to calculated residence times of fluvial sediment.

#### 2.3.3. Presence of clay pellets

The present day Cooper Creek floodplain and river valleys of the eastern part of the Lake Eyre Basin are characterised by clay-rich mud aggregates (Maroulis and Nanson, 1996). From the comminution perspective, the question raised is whether or not, if present, the silt- to sand-sized clay pellets disaggregate during wet-sieving and sample pre-treatment? Flume experiments show that the pellets are durable during transport and relatively resistant to disaggregation (Maroulis and Nanson, 1996). Therefore, if silt-sized clay pellets persist at the clay removal step, a percentage of secondary clay material (mineralogically speaking), is unlikely to be removed during centrifugation to remove the 0–2 $\mu$m fraction. This material may have unrepresentatively high ($^{234}$U/$^{238}$U) ratios relative to its amalgamated pellet grain size, and if present in abundance, will then perturb calculations of $f_e$ based on grain geometry or surface area. The use of sodium hexametaphosphate during sample pre-treatment is anticipated to deflocculate clays and help to alleviate this issue.

#### 2.3.4. Sample mineralogy

The $\alpha$-recoil length, $L$, varies for different minerals (see Section 6.2.1) and therefore, bulk sample $\alpha$-recoil length is dependant on sample mineralogy. Any uncertainty in sample mineralogy will consequently add extra uncertainty to the estimation of the recoil loss factor. This is probably the case for the present study in which sample mineralogy was not determined. Furthermore, the dominance of a mineral that is highly non-spherical/oblate (e.g., mica) may lead to additional uncertainty to the weighted geometric estimation of recoil loss factor, as this model assumes a linear relationship in grain aspect ratio between the largest and smallest grains.

The presence of zircon (or other high U concentration accessory minerals) is another factor that could potentially complicate the interpretations of palaeochannel sediment U-series disequilibria. The SEM image of GT2 in Fig. 3 clearly shows that zircon is present in the <53 $\mu$m sieved and leached fraction of Cooper Creek sediments, and over 25 grains were observed in the GT2 SEM sample stub. Zircon has a high affinity for the nuclides of interest, particularly compared to other silicate minerals such as quartz and feldspar (Blundy and Wood, 2003). Therefore, if digested along with the bulk rock sample, even minor amounts of zircon are expected to dominate U-series budget. The higher U–Th concentration and different recoil length, of zircon (22.7 nm, (theoretical) Ziegler et al., 1996, or 55 nm, (observed) Kigoshi, 1971), relative to that of feldspar (30 nm; (theoretical) Ziegler et al., 1996), may result in zircon dominance of bulk sample ($^{234}$U/$^{238}$U) ratios and control.
on estimated $f_a$ values. Whether the zircon is fluvially or aeolian derived may then be of great significance to the measured bulk sample $^{234}\text{U}-^{238}\text{U}$ disequilibria and any calculated comminution age of the sediment. We note that the sample digestion method used in this study (Section 5) is not expected to digest zircon. However, this issue needs to be considered further in future comminution studies. The impact of zircon could be tested by separation of the zircon fraction (silt-sized range) from a bulk sample, and subsequent processing of the zircon fraction as a separate
digested (in a pressure bomb) for U-series analysis. However, separation of fine-grained zircon from a sample of only a few grams may be challenging, and require micro-heavy liquid techniques.

3. REGIONAL CONTEXT OF THE STUDY AREA: THE LAKE EYRE BASIN

The Cooper Creek, together with the Diamantina and Georgina Rivers, form the major sediment transport pathways within the Lake Eyre Basin, one of the world’s largest (~1.2 × 10^6 km²), internally draining, semi-arid catchment areas (Fig. 4a and b). The Cooper Creek originates in Queensland at the confluence of the Thomson and Barco rivers and flows ~1500 km southwest towards Lake Eyre salt playa (deposcentre 15 m below mean sea level) in arid South Australia (Fig. 4b). The catchment drains approximately one-seventh of the Australian continent and is characterised by relatively flat relief, stream gradients of 0.1–0.2 m/km and a series of anabranching ephemeral channels known as the ‘Channel Country’. The catchment is geologically hosted by a series of Early Palaeozoic to Cainozoic embedded, gently warped basins and associated near-horizontal sedimentary sequences. A detailed geological background of the region can be found in Nanson et al. (2008).

Local sediment may be supplied to the study area via the erosion of the Innamincka Dome and its associated low-gradient fan, which spills out into the Strzelecki Desert west of Innamincka (Fig. 4). Cainozoic crustal warping produced the topographic high and the entire flow of the Cooper Creek presently passes through the Dome in a confined channel. The exposed surface of the Dome consists mainly of the Winton Formation (shales and sandstones in a kaolinitic matrix, with minor limestone with leaf impressions) and the Mt Howie Sandstone (kaolinitic pebbly-sandstone, pebbles of the Winton Formation) of Late Cretaceous age and the Tertiary Eyre Formation (fluviatile kaolinitic cross-bedded sandstone and minor shales) of Late Cretaceous age and the Mt Howie Sandstone (kaolinitic pebbly-sandstone, pebbles of the Winton Formation) of Late Cretaceous age and the Tertiary Eyre Formation (fluviatile kaolinitic cross-bedded sandstone and minor shales) and silcrete (silicified psammites and minor shales) of Late Cretaceous age and the Mt Howie Sandstone (kaolinitic pebbly-sandstone, pebbles of the Winton Formation) of Late Cretaceous age and the Tertiary Eyre Formation (fluviatile kaolinitic cross-bedded sandstone and minor shales) and silcrete (silicified psammites and minor shales) of Late Cretaceous age. A compilation of new and previously published alluvium thermoluminescence (TL) ages of the Lake Eyre Basin is presented by Nanson et al. (2008). The alluvial evidence suggests that more pronounced fluvial activity, relative to today, extended back to at least oxygen isotope stages (OIS) 8 and 7 (up to 280 ka) and peaked ~120–90 ka during OIS 5. A smaller peak is observed (~65–60 ka) in late OIS 4 and a smaller still (~45–40 ka) in mid OIS 3 (Nanson et al., 2008). An overall picture is envisaged of a significantly oscillatory wet-dry climate, superimposed upon a gradual trend of increasing aridity (which is noted Australia-wide) and a general decline in the magnitude of the wet episodes throughout the mid to late Quaternary (Nanson et al., 2008). The significant sandy fluvial deposits observed within Cooper Creek palaeochannels (e.g., Fig. 4c) suggests that larger more competent rivers than today delivered coarse sand to the Cooper Creek Fan over approximately the last 100 ka (Cohen et al., 2010). Due to high evaporation and low relief only significant monsoon rains produce enough water to reach Lake Eyre at present times, usually on a decadal timescale and which most recently occurred in 2010. The present day Cooper Creek floodplain is dominated by clay and sand-sized mud aggregates with sporadic sand dunes (Nanson et al., 1986; Maroulis and Nanson, 1996).

4. PALAEOCHANNEL SAMPLE DETAILS

The Cooper Creek palaeochannel samples were collected from six trench pits dug across the Gidgealpa palaeochannel (Fig. 4c), extending south from the Gidgealpa dune (GD, Fig. 4c). Field sampling procedures and the stratigraphy of the fluvial deposits are described in detail in Cohen et al. (2010). The Gidgealpa palaeochannel lies within the Strzelecki desert close to the ‘South branch’ of the Cooper Creek and is still inundated during significant floods. The adjacent alluvium was found, by Cohen et al. (2010), to be the source for transverse and linear dunes, demonstrating an important inter-relationship between dune and floodplain. OSL depositional ages of the fluvial sand units, underlying the muddy floodplain deposits, range from 119 ± 12 ka to 22 ± 5 ka (Fig. 4c and Table 1). To obtain...
sediment residence times for palaeochannel sediments calculated from comminution ages it is necessary to know the deposition age of the sediment. Therefore, this study focuses on the sandy units already dated by Cohen et al. (2010). Due to the lack of fine material in some of the dated units it was necessary to combine the dated samples with neighbouring sand-rich samples in order to obtain sufficient silt-sized material. This may add some additional uncertainty to the calculated residence times. The exact stratigraphic location of the samples used in this study is given in Table 1 and Fig. 4c.

5. ANALYTICAL TECHNIQUES

The <~50 μm fraction, typically 2–3 g of material per bulk sample (from ~200 to 300 g of predominantly sandy material), was obtained by wet-sieving (53 μm mesh size) using deionised water. Approximately 2 g of the retained fine material underwent a sequential extraction procedure to remove organic and exchangeable material, carbonate and Fe-, Mn-oxide secondary minerals following a methodology modified from Schultz et al. (1998). Stage 1: removal of exchangeable/adsorbed/organics. The sample was first heated at 98 °C for 30 min in 30 mL of NaOCl (pH 7.5). After cooling, the sample was centrifuged and the NaOCl supernatant discarded. This stage was repeated again and afterwards the sample was rinsed (rinse stage) by adding 10 mL of 18.2 MΩ water, centrifuging at 7000 rpm for 15 min and then discarding the supernatant. The rinse stage was carried out twice. Note that all supernatants (including 18.2 MΩ water) were removed carefully via pipette after each centrifuge step. Stage 2: removal of carbonates. Twenty millilitres of 1 M NaAc in HAc at pH 4 was added to the residue from Stage 1. The sample was continually agitated using a rotary mixer at 30 rpm for 2 h at room temperature. After centrifugation the supernatant was discarded. The rinse stage was then repeated twice. Stage 3: removal of amorphous and crystalline Fe-, Mn-oxides. 20 mL of 0.04 M NH₂OH.HCl was added to the residue of Stage 2. The sample was continually agitated using a rotary mixer at 30 rpm for 5 h. After centrifugation the supernatant was discarded. The rinse stage was then repeated twice. Stage 4: removal of clay-sized material. The clay-sized 0–2 μm fraction of the samples was removed by controlled centrifugation, following the United States Geological Survey centrifugation method (Open-File Report 01-041). Prior to centrifugation, approximately 50 mL of filtered 5% sodium hexametaphosphate solution was added to the sample to disperse the particles. The mixture was then sonicated with an ultrasonic probe for 20 s at 140 W and agitated overnight at 30 rpm using a rotary mixer. The centrifugation step was undertaken an additional three times beyond when the sample was considered clear of suspended matter to try to ensure complete removal of the clay-sized material. The potential fractionation of 234U/238U caused by the sequential leaching procedure in steps 1–3 was not determined in this study but this should be investigated in future studies to assess the reliability of the pre-treatment approach.

Uranium concentrations and 234U/238U isotopic ratios were determined on the post-leached residue at the Macquarie University U-series Research Laboratory. Approximately 0.1 g of sediment was spiked with a 236U–239Th tracer and digested in a mixture of concentrated acids (HClO₄–HF–HNO₃–HCl). Separation of U followed standard anionic resin chromatography detailed in Turner et al. (2011). Uranium concentrations, determined by isotope dilution, and U isotope ratios were measured on a Nu Instruments Multi-Collector ICP-MS at Macquarie University following the approach described by Turner et al. (2011). Internal accuracy (<0.3%) and precision (<0.1%) were assessed by regular analyses of the U010 and U005A solution standards. TML-3, a secular equilibrium rock standard, digested alongside the samples yielded (234U/238U) within error of secular equilibrium (Table 1). A replicate analysis of GT6 (that underwent sequential extraction steps 1–4, acid digestion and column processing in a separate sample batch) yielded (234U/238U) within 2σ (and analytical error) of the initial GT6 analysis (Table 1). Noting that the reagents in the sequential extraction procedure are discarded at the end of each step and the sample is then rinsed with Milli-Q water, the total procedural blank for the phase extraction, digestion and column separation procedure is <60 pg.

Scanning Electron Microscope (SEM) BSE images of gold-coated samples were acquired using a Zeiss EVO MA15 at the GAU (Geochemical Analytical Unit) at Macquarie University operating under 10 kV acceleration. The scanning electron microprobe images of GT2 in Fig. 3 show the clean nature of the processed samples after sieving to <53 μm, sequential leaching and clay removal.

BET surface areas were measured by N₂ gas adsorption for sequentially leached and clay removed samples GT1 and GT6 (which were the only samples with sufficient fine-grained material for analysis). Measurements were carried out using a Micromeritics ASAP 2020 at Stanford University and using a Micromeritics Gemini VII 2390 at the Particle and Surface Sciences laboratory in Gosford, Australia to determine inter-laboratory variations in surface area measurements. The measurements produced by each laboratory are presented in the footnote to Table 2. The BET measurements made at each of the two laboratories were carried out on the same sample aliquot and therefore any variation in the result is likely due to measurement variation rather than sample heterogeneity.

Particle size distributions of the <53 μm bulk sediments (clay-included) were obtained using a Micromeritics Sedigraph III 5120 at Macquarie University. The clay-excluded particle size distributions were calculated by excluding the <2 μm measurements and re-normalising the data.

6. RESULTS AND DISCUSSION

6.1. (234U/238U) activity ratios

Sample pre-treatment was carried out to remove organic material and secondary minerals including carbonate from the <53 μm sieved fraction. Secondary minerals such as
Particle size distribution data for the clay-excluded samples are recalculated from the clay-included data, restricting the grain size to the 2–53 micron fraction. The recoil loss of its parent 234Th (Fig. 2). GT6 has a (234U/238U) activity ratio of the fluid they precipitate from and so most likely possess a (234U/238U) > 1 (Maher et al., 2006). Removal of that component is, therefore, expected to lower the (234U/238U) ratio of the bulk sample, and such is observed for samples GT2 and GT6. However, the (234U/238U) ratios of GT1, GT4 and GT5 increased after removal of the clay-sized fraction, suggesting that the clay material lost may have been primary and had low (234U/238U) due to high recoil efficiency in small, old, grains. Overall, removal of the clay fraction has shifted the clay-excluded data closer to secular equilibrium. (234U/238U) activity ratio broadly correlates with U concentration in both sets of differentially phase-extracted samples (R² = 0.74; Fig. 5b). The inclusion or exclusion of the clay-sized fraction is important to consider in future studies, as the estimated s-recoil loss factor of a bulk sediment sample will clearly be significantly controlled by the presence or absence of this size-fraction.

Table 2
Calculated recoil loss factors (fₙ values) for Cooper Creek palaeochannel sediments.

<table>
<thead>
<tr>
<th>fₙ</th>
<th>Sample</th>
<th>Minimum (1-Aₐₐₑₜₐₜ)</th>
<th>Geometric (λₛ 1–2)</th>
<th>Geometric (λₛ 1–17)</th>
<th>BET</th>
<th>BET</th>
<th>BET</th>
<th>BET</th>
<th>λₛ needed for min fₙ</th>
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<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>a</td>
<td>b</td>
<td>a</td>
<td>b</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.050</td>
<td>0.012</td>
<td>0.025</td>
<td>0.092</td>
<td>0.008</td>
<td>0.130</td>
<td>0.019</td>
<td>1–8</td>
</tr>
<tr>
<td>Clay-included</td>
<td>GT1</td>
<td>0.050</td>
<td>0.012</td>
<td>0.024</td>
<td>n.p.</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Clay-included</td>
<td>GT2</td>
<td>n.p.</td>
<td>0.012</td>
<td>0.030</td>
<td>1–12</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>Clay-included</td>
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<td>0.023</td>
<td>0.012</td>
<td>0.041</td>
<td>1–15</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Clay-included</td>
<td>GT4</td>
<td>0.035</td>
<td>0.011</td>
<td>0.035</td>
<td>1–22</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Clay-included</td>
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<td>0.043</td>
<td>0.011</td>
<td>0.029</td>
<td>n.p.</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Clay-included</td>
<td>GT6</td>
<td>n.p.</td>
<td>0.012</td>
<td>0.029</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Clay-excluded</td>
<td>GT1</td>
<td>0.022</td>
<td>0.011</td>
<td>0.041</td>
<td>0.029</td>
<td>0.003</td>
<td>0.063</td>
<td>0.014</td>
<td>1–8</td>
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<tr>
<td>Clay-excluded</td>
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<td>0.004</td>
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<tr>
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<td>0.012</td>
<td>0.041</td>
<td>1–7</td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Clay-excluded</td>
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<td>0.011</td>
<td>0.048</td>
<td>1–5</td>
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<td></td>
<td></td>
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<tr>
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<td>0.011</td>
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<td>1–9</td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Clay-excluded</td>
<td>GT6</td>
<td>n.p.</td>
<td>0.012</td>
<td>0.039</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

1-Aₐₐₑₜₐₜ is the minimum value for fₙ required to satisfy the comminution equation, as defined in Section 6.2. fₙ geometric is the weighted geometric estimation of fₙ calculated using Eq. (3) given in the text, measured particle size data, λₛ as shown, β from 1 to 10 and L = 30 nm. fₙ BET and fₙ BETfrac are calculated using Eqs. (4) and (5) given in the text. n.p., it is not possible to solve the equation for fₙ as the measured (234U/238U) activity ratio is greater than the corresponding A₀ (234U/238U) ratio.

Particle size distribution data for the clay-excluded samples are recalculated from the clay-included data, restricting the grain size to the 2–53 micron fraction. λₛ surface roughness; β, aspect ratio.

An assumed bulk density of 2670 kg/m³ was used in the surface area estimations of fₙ. λₛ needed for minimum fₙ is the range in surface roughness required to reach the minimum value of fₙ able to solve the comminution equation (assuming all other input parameter values remain the same).

Fe-, Mn-oxides, carbonates and clays are likely to incorporate U having the (234U/238U) ratio of the fluid they precipitate from and so most likely possess a (234U/238U) > 1. Therefore, secondary minerals are not representative of the primary detrital (234U/238U) activity ratio and any calculated comminution age would be erroneous. However, as the local sedimentary basement exposed on the Innaminka Dome is clay (kaolinite) rich, clay-sized, clay compositional material, that would usually considered as a secondary mineral, may be incorporated as primary source material in the palaeochannel deposits. U-series isotopic compositions were subsequently determined for both the clay-included (after sequential leaching Stage 3) and clay-excluded fractions (after phase extraction Stage 4) described in the analytical techniques.

U concentrations and (234U/238U) activity ratios of the Cooper Creek palaeochannel sediments are listed in Table 1 and presented on Fig. 5. The majority of clay-included and clay-excluded samples have (234U/238U) < 1, as expected for detrital fine-grained sediments that have lost 234U due to the recoil loss of its parent 234Th (Fig. 2). GT6 has (234U/238U) > 1 for both the clay-included and clay-excluded samples, suggesting either incomplete removal of secondary phases, which is supported by the higher U concentration of this sample, or significant implantation of 234Th (Table 1 and Fig. 5b). If the clay-sized component of the sample was formed secondary to the detrital grains, it is expected to have (234U/238U) > 1 (Maher et al., 2006).

6.2. Calculated comminution ages and residence times of Cooper Creek sediments based on ‘reasonable’ assumptions for input parameters

As described in Section 2, determination of a meaningful calculated comminution age, and therefore sediment residence timescale, principally depends on uncertainty in the estimation of the recoil loss factor (fₙ) and any uncertainty in the (234U/238U) of the starting material (A₀) (DePaolo...
et al., 2006). Subtle variations in estimated \( f_a \) values have been shown to greatly affect inferences about physical weathering rates (Maher et al., 2006). Following the comminution methodology and numerical bounds of Eq. (2), with a set \( ^{234}\text{U}/^{238}\text{U} \) (activity ratio of the source \( A_0 \)) and using the measured sample \( ^{234}\text{U}/^{238}\text{U} \) ratios \( A_{\text{mean}} \), a minimum value for \( f_a \) is required to satisfy the comminution equation, which is defined as: \( 1 - A_{\text{mean}} \). Note that, for measured samples with \( ^{234}\text{U}/^{238}\text{U} > 1 \) (GT2 and GT6 clay-included and GT6 clay-excluded samples), it is not possible to determine the minimum \( f_a \) theoretically, as the measured sample is required to have \( ^{234}\text{U}/^{238}\text{U} < A_0 \). The theoretical minimum \( f_a \) values for the Cooper Creek sediments are shown in Table 2 and Fig. 6a and b. In the following two sections, sample \( f_a \) values are calculated using input assumptions reflecting those in the present U isotope comminution literature.

6.2.1. Geometric calculation of \( f_a \) for Cooper Creek palaeochannel sediments

Table 2 shows the \( f_a \) values determined for Cooper Creek palaeochannel sediments using both the geometric weighted mean method (Eq. (3)) and surface area methods (Eqs. (4) and (5)) discussed above. For the geometric model, particle size distribution data is utilised but assumptions are required for the surface roughness factor \( \lambda_s \) and aspect ratio \( \beta \). The weighted geometric recoil loss factor was calculated for Cooper Creek palaeochannel sediments using two models, (a) assuming \( \lambda_s \) between 1 and 2, following DePaolo et al. (2006) and Dosseto et al. (2010) and (b) where \( \lambda_s \) varies between 1 and 17, the inferred range in surface roughness for the Kings River Fan sediments investigated by Lee et al. (2010). The surface roughness factor is taken to increase linearly with increasing grain diameter in both cases. The aspect ratio of grains, \( \beta \), is also likely to vary in natural sediment samples, as demonstrated by the largely spherical to elongated grains in the SEM image of GT2 (Fig. 3). Linearly varying \( \beta \) values ranging from 1 to 10 were used in the geometric calculations of \( \alpha \)-recoil loss factor, following DePaolo et al. (2006) and Dosseto et al. (2010), where a spherical grain represents \( \beta = 1 \) (largest grain) and an oblate spheroidal grain has \( \beta > 1 \) (smallest grain).

The \( \alpha \)-recoil length, \( L \), varies depending on grain mineralogy and is relatively poorly constrained at present. Current estimates lie between 20 and 40 nm in silicate minerals (see Maher et al., 2006 and references therein). The mineralogy of the Cooper Creek palaeochannel sediments has not been specifically determined and so an intermediate, theoretical value for feldspar (30 nm) (Ziegler et al., 1996) is used in this study following Maher et al. (2006), DePaolo et al. (2006) and Dosseto et al. (2010) and is similar to that used by Lee et al. (2010) \((L = 34 \text{ nm})\). As expected, the geometric calculations of \( f_a \) for both the clay-included and clay-excluded samples show that if a greater surface roughness is assumed (i.e., \( \lambda_s \) from 1 to 17 compared to 1–2), a larger amount of recoil loss is predicted (at least double) due to the increased surface area available (Table 2 and Fig. 6a and b). Using similar parameter range assumptions to previous authors (except for sample GT2 clay-excluded), the recoil factors produced using \( \lambda_s \) from 1 to 2 are lower than the minimum \( f_a \) required to solve the comminution equation. Weighted geometric model estimations of \( f_a \) produce values within the typical parameter range of \( f_a \) (0.01–0.1) given by Bourdon et al. (2009) but lie below the range of published \( f_a \) values for palaeochannel deposits from temperate Australia (grey stipple, \( y \)-axis Fig. 6) even for the clay-included samples.

6.2.2. Surface area measurement estimations of \( f_a \) for Cooper Creek palaeochannel sediments

Bulk sample surface area measurements by BET can also be used to calculate the \( \alpha \)-recoil loss factors. However, BET gas adsorption based estimates can greatly overestimate \( f_a \) due to the significant difference between the size of adsorbant gas molecule and the \( \alpha \)-recoil length (e.g., Maher et al., 2006). As discussed above, in principle it is possible to overcome this issue by considering a fractal recoil model (e.g., Semkow, 1991; Bourdon et al., 2009). Table 2 and Fig. 6b display the calculated \( \alpha \)-recoil loss fraction for two samples (clay-excluded GT1 and GT6) for which there was sufficient post-leaching material available for BET surface area analysis. BET measurements were carried out on the same sample aliquots at two different laboratories (see analytical techniques and Table 2 footnote for details and results) to investigate the effect of inter-laboratory surface area variations on calculated \( f_a \) values. In agreement with previous findings (e.g., Bourdon et al., 2009), the data demonstrate clearly that if the BET data are used without the fractal amendment then much higher \( f_a \) values are obtained (e.g., \( f_a = 0.092 \) and 0.008 for BET\(^a\) and BET\(_{\text{fract}}\), respectively; Table 2). The observed inter-laboratory variations in BET measurements (Table 2 footnote) produce relatively variable \( f_a \) values (compare BET\(_{\text{fract}}\) with BET\(_{\text{fract}}\)). In comparison to the weighted geometric method, the BET\(_{\text{fract}}\) \( f_a \) values are closest in magnitude to those of the geometric \((\lambda_s = 1–2) \) model. However, the BET\(_{\text{fract}}\) values of \( f_a \) are also lower than the minimum \( f_a \) required to solve the comminution age equation (Fig. 6b).

In summary, it is shown that \( f_a \) of a sample can vary significantly depending on whether or not the clay fraction is removed, which model of \( f_a \) estimation is used and the input values assumed. Sample GT1, which has both weighted geometric and fractal corrected surface area estimations of \( f_a \), shows that only the weighted geometric model (using \( L = 17 \) as the upper limit for surface roughness) lies above the minimum \( f_a \) value necessary to solve Eq. (2). This might suggest that, either the maximum assumed values of surface roughness for the Cooper sediments are too low, or alternatively, if we assume that the estimated recoil loss factor values are largely correct for each sample (e.g., for the BET\(_{\text{fract}}\) method), the inconsistencies between the \( f_a \) values and the theory of the comminution methodology (minimum \( f_a \)) may be explained by preferential loss of \( ^{234}\text{U} \) via leaching. In the latter case, preferential leaching of \( ^{234}\text{U} \) would lead to lower \( ^{234}\text{U}/^{238}\text{U} \) ratios and therefore, higher minimum \( f_a \) values. Regarding the former scenario, it is indeed plausible that the surface roughness is larger than that assumed here, based on previous studies (up to a maximum of 17). White et al. (1996) calculated the surface roughness of the
silicate fraction of Californian soils and show surface roughness factors of up to >600 maybe appropriate. Table 2 shows the minimum surface roughness required (5–48) using the weighted geometric estimation of $f_s$ to reach the theoretical minimum $f_s$ value (assuming that the other parameters remain fixed as before). For the Cooper Creek sediments, and the presently available data, it is not possible to determine the relative importance of each option but clearly the role of chemical erosion versus physical erosion on the final $^{234}$U/$^{238}$U of a given particle requires further consideration, and tighter constraints on surface roughness are needed for each sample. White et al. (1996) also show that the surface roughness for the primary silicate component of the soil increases with soil age, a factor not presently considered by the comminution methodology (i.e., changes in surface roughness with soil or deposit age).

6.2.3. Comminution age and sediment residence times of Cooper Creek palaeochannel sediments

Based on the ‘reasonable’ assumptions used for $f_s$ estimation detailed above, the residence times of Cooper Creek palaeochannel sediments for clay-excluded and clay-included samples have been calculated using Eq. (2) and are presented in Table 3 and Fig. 7. Few realistic comminution ages (and sediment residence times) were produced from the clay-included samples (Table 3). Recoil factors constrained by BET surface area measurements (assuming $A_0 = 1$) produce either negative residence times or do not give a solution to the comminution age equation. However, arithmetically valid residence times for the clay-excluded sediments (geometric model with $\lambda_0 = 1–17$) lie between 423 and 620 ka. For the clay-excluded samples, more solutions in age produced for the same sample depending on whether the clay-sized fraction is included or excluded (e.g., GT3 and GT4, Fig. 7), and also different relative patterns i.e., a decrease (clay-excluded) or increase (clay-included) in residence age for sediments deposited between ~100 and 35 ka, it is not possible to infer links between residence ages and geomorphologic processes in semi-arid Australia at this stage.

Within the context of the preceding discussion and due to the range of sample pre-treatment procedures, potential lab-dependent variations for surface area measurements (Table 2), the use of variable equipment to measure particle size distribution (e.g., sedigraph versus mastersizer) and variations in the assumptions involved to estimate sediment residence time, at present it is difficult to compare data sets produced by different research groups. Subsequently, in order to be able to directly compare the Cooper Creek palaeochannel sediment residence times with those for the Murrumbidgee River palaeochannel sediments in southeastern Australia (the only other data set produced using the U-series comminution approach in Australia), the samples and data should be processed in the same way and comminution ages calculated using the same assumptions. The Murrumbidgee samples were sequentially leached using the same methodology as that described here and were analysed with the clay fraction included. The recoil loss fraction was calculated using the weighted geometric model with the assumption that $\lambda_0 = 1–2$, $\beta = 1–10$. $A_0$ was assumed to be in secular equilibrium (i.e., equal to 1). However, the corresponding model for the Cooper Creek failed to produce comminution ages (Table 3) and therefore, it is not possible to make comparisons between residence times of temperate and semi-arid Australia.

6.3. Numerical estimation of comminution age uncertainties: Monte Carlo simulations

The comminution age error shown in Table 3 for the Cooper Creek sediments was calculated by propagating the error on the measured $^{234}$U/$^{238}$U ratio and including
a 16% uncertainty estimation for \( f_a \) (Table 3 footnote, after Dosseto et al., 2010). However, the true uncertainty on the comminution age, based on reasonable ranges of assumed input parameters, has yet to be fully constrained and is likely to be larger than that quoted for the Cooper Creek samples. In the following section, we present the results from a series of Monte Carlo simulations, carried out to estimate the uncertainty upon comminution age related to each individual input parameter in the calculation of \( f_a \) (weighted geometric and BET surface area measurement with fractal correction methods). The combined uncertainties for each \( f_a \) calculation method were then estimated, along with a total combined estimation of comminution age uncertainty based on the combined uncertainties of \( f_a \), \( A_0 \) and \( A_{\text{meas}} \).

6.3.1. Uncertainties associated with the weighted geometric determination of \( f_a \)

Due to the limitations of the Cooper Creek sediment samples in producing \( f_a \) values high enough to solve the comminution equation, uncertainty estimations were carried out using a synthetic sample for which a grain size distribution and reference comminution age (572 ka) were generated. The considered ranges of input parameters used in the Monte Carlo simulations are given in Table 4. Uncertainties were then estimated based on the probability distributions obtained from the Monte Carlo simulations. We adopt the 95% confidence limit of these distributions as a measure of total uncertainty. Full details of this procedure are given in Appendix A.

In the first set of simulations, the input parameters of surface roughness \( (\lambda_s) \), aspect ratio \( (\beta) \) and \( \alpha \)-recoil length \( (L) \) were assigned small individual uncertainties of 1% (i.e., \( \pm 0.5\% \) of the values used to generate the reference age), keeping \( A_0 \) and \( A_{\text{meas}} \) fixed at 1 and 0.975, respectively (Appendix A). The results show that if the input parameters \( L \), \( \lambda_s \) and \( \beta \) are known within 1% of their true values, the uncertainty on the calculated comminution reference age of 572 ka is \( \pm 14 \) ka (at the 95% confidence limit; Fig. 8). It is highly likely that the uncertainty on each assumed input parameter is presently greater than 1%. Therefore, the uncertainty on the reference comminution age has also been calculated for each individual parameter based on what we consider to be a feasible uncertainty (or range) for each parameter. All other parameters in the simulation were fixed at the initial values (Appendix A).

Surface roughness \( (\lambda_s) \) is assumed to vary linearly with grain diameter. However, there is significant uncertainty on the appropriate limits of surface roughness (Sections 6.2.1 and 6.2.2). While a value of 1 (smooth surface) is commonly assigned to the smallest grains, values for the largest grains vary between 2 and 17 in the U-series comminution literature (Dosseto et al., 2010 and Lee et al., 2010, respectively). Here we explored a range of 7–30 for the largest grains; this range is considered to represent a reasonable uncertainty on this parameter considering silicate surface roughness values of up to 600 may be possible (White et al., 1996; Section 6.2.2). Monte Carlo simulations ran with this choice of \( \lambda_s \) produced a large range in \( \lambda_{\text{com}} \) between 169 and 801 ka, resulting in a significant uncertainty of \( \pm 316 \) ka. It was not possible to use a range of 1–2 as the lower limit of \( \lambda_s \) in our model (cf. DePaolo et al., 2006; Dosseto et al., 2010) as this produced \( f_a \) values below the minimum \( f_a \) limits of the equation and therefore, could not produce solutions based on the chosen combination of other input parameters values and \((^{238}\text{U}/^{235}\text{U})\) ratios used.

![Fig. 6. Geometric and surface area estimations of recoil loss factors \( f_a \) for (a) clay-included (0–53 μm fraction), and (b) clay-excluded (2–53 μm fraction) Cooper Creek palaeochannel sediments following methodologies described in the text and notation used in Table 2. 1–2 and 1–17 refer to the range in surface roughness used in the weighted geometric estimation of \( f_a \), \( 1-\text{Ameas} \) represents the minimum value of \( f_a \) required for the comminution age equation to be solved (see Section 6.2). Bars on the y-axis exemplify published ranges in recoil loss factor for: Murrumbidgee River palaeochannel sediments, southeast Australia (0–53 l), Hanford, Washington, USA (Maher et al., 2006; DePaolo et al., 2006) (dark grey solid bar).](image-url)
As for surface roughness, the aspect ratio ($\beta$) is typically modeled as a linear function of grain diameter, with the largest value (ellipsoidal grains) assigned to the smallest grains and a value of 1 (spherical grains) assigned to the largest grains in the sample. Based on previous studies that assume a value of 10 for the smallest grains (e.g., Dosseto et al., 2010), we allowed $\beta$ of the smallest grains to vary between 9 and 12 in our Monte Carlo analysis. Results from these simulations produced $t_{com}$ ages of 393–733 ka at the 95% confidence level and therefore, an uncertainty of ±170 ka in the comminution age.

As noted in Section 6.2.1, the $\alpha$-recoil lengths for individual minerals are relatively poorly constrained at present (Maher et al., 2006). Assumed values lie in the range of 30 nm (Maher et al., 2006; DePaolo et al., 2006; Dosseto et al., 2010) to 34 nm (Lee et al., 2010). Therefore an uncertainty of $30 \pm 2$ nm was used in the Monte Carlo simulations. This produced a range of $t_{com}$ between 492 and 678 ka and an uncertainty of ±93 ka (Table 4).

Using the combined uncertainties of $\lambda_s$, $\beta$, and $L$ in a Monte Carlo simulation, a significant uncertainty of ±283 ka (at a confidence level of 95%) is obtained for the comminution age. The corresponding $f_a$ values produced for the total combined uncertainty range from 0.025 to 0.083 (Table 4).

### 6.3.2. Uncertainties associated with the BET and fractal dimension determination of $f_a$

Similarly, in order to explore the uncertainty associated with the BET surface area with fractal correction $f_a$ method, it is necessary to first generate a model comminution age. The input assumptions used in the determination of a synthetic age are detailed in Appendix A (Step 2). Using $f_a$ values from Table 2, calculated Cooper Creek palaeochannel sediment comminution ages and corresponding residence timescales, $T_{res}$ (ka) using $f_a$ values from Table 2.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$t_{com}$</th>
<th>Error</th>
<th>Geometric</th>
<th>$T_{res}$</th>
<th>Error</th>
<th>Geometric</th>
</tr>
</thead>
<tbody>
<tr>
<td>Clay-included</td>
<td>GT1</td>
<td>Fail</td>
<td>Fail</td>
<td>19.1</td>
<td>3.09</td>
<td>88</td>
</tr>
<tr>
<td></td>
<td>GT2</td>
<td>n.p.</td>
<td>n.p.</td>
<td>151</td>
<td>24.4</td>
<td></td>
</tr>
<tr>
<td></td>
<td>GT3</td>
<td>Fail</td>
<td>279</td>
<td>45.1</td>
<td>161</td>
<td>26.0</td>
</tr>
<tr>
<td></td>
<td>GT4</td>
<td>Fail</td>
<td>235</td>
<td>5.05</td>
<td>342</td>
<td>55.4</td>
</tr>
<tr>
<td></td>
<td>GT5</td>
<td>Fail</td>
<td>157</td>
<td>25.4</td>
<td>122</td>
<td>19.7</td>
</tr>
<tr>
<td>Clay-excluded</td>
<td>GT1</td>
<td>Fail</td>
<td>138</td>
<td>40.9</td>
<td>151</td>
<td>24.4</td>
</tr>
<tr>
<td></td>
<td>GT2</td>
<td>22.3</td>
<td>31.3</td>
<td>19.1</td>
<td>88</td>
<td>14.2</td>
</tr>
<tr>
<td></td>
<td>GT3</td>
<td>Fail</td>
<td>253</td>
<td>59.0</td>
<td>Fail</td>
<td></td>
</tr>
<tr>
<td></td>
<td>GT4</td>
<td>Fail</td>
<td>157</td>
<td>25.4</td>
<td>Fail</td>
<td></td>
</tr>
<tr>
<td></td>
<td>GT5</td>
<td>Fail</td>
<td>364</td>
<td>25.4</td>
<td>Fail</td>
<td></td>
</tr>
</tbody>
</table>

$T_{res}$ (sediment residence time) is calculated using the $f_a$ values given in Table 2 and Eqs. (1) and (2) in the text.

$n.p.$ It is not possible to derive $T_{res}$ as the measured ($^{234}$U/$^{238}$U) activity ratio is greater than the corresponding $A_0$ ($^{234}$U/$^{238}$U) ratio. Fail, model failed to solve the comminution age equation due to the associated $f_a$ value being less than 1-$A_{\text{meas}}$ (see Section 6.2).

All $T_{res}$ calculated using BET surface area estimations of $f_a$ gave either negative sediment residence timescales or were not solvable (therefore, they are not shown).

Errors are the analytical uncertainties at the 2σ level calculated by propagation of the error on ($^{234}$U/$^{238}$U) and the recoil fraction (16%) following Dosseto et al. (2010).
the input parameter values given, a reference comminution age of 404 ka was obtained. This age is not identical to that generated by the weighted geometric method due to the different input parameters involved (e.g., bulk density, fractal dimension and the diameter of the adsorbant gas molecule in the BET surface area (fractal) method as opposed to surface roughness and aspect ratio as input parameters in the weighted geometric method). The combination and range of input parameter values required therefore resulted in slightly different initial ages being generated.

Using the same approach as above, the assumed input parameters of the method (Eq. (5)): total surface area \( (\text{BET}_{\text{fractal}}) \), fractal dimension \( (D) \), density \( (\varrho) \) and \( \Delta -\text{recoil} \) length scale \( (L) \) were varied by 1% in a Monte Carlo simulation, i.e., \( \pm 0.5\% \) of the initial values used to generate the reference age (Appendix A). This resulted in a small range in comminution age of 401–407 ka and therefore, a small uncertainty of \( \pm 3 \) ka. The uncertainty of each individual parameter was then assessed in turn using the parameter ranges specified in Table 4.

Inter-laboratory variations in BET surface area measurements for the same Cooper Creek sample are highlighted in Table 2 (footnote); therefore, an uncertainty of \( \pm 2 \) m\(^2\)/g was assigned to the initial surface area of 8 m\(^2\)/g. This yielded a maximum uncertainty of \( \pm 118 \) ka associated with the reference model age of 404 ka. If a more conservative uncertainty of \( \pm 0.7–1 \) is considered (e.g., the uncertainty assigned to \( \Delta -\text{recoil} \) measured (\( {\text{U}}_{234}/\text{U}_{238} \)) ratio (0.975). For the weighted geometric method, the simulation produced an uncertainty of \( \pm 80 \) ka (not shown).

The uncertainty explored for \( D \), the fractal dimension, was determined using a conservative estimate based on the inter-laboratory variation observed in this parameter for the Cooper Creek samples (Table 2, footnote). Using a range in \( D \) from 2.45 to 2.55 (\( \pm 0.05 \) of the initial value), the simulations produced a \( t_{\text{com}} \) ranging from 310 to 540 ka, equivalent to an associated uncertainty of \( \pm 115 \) ka.

Varying the average bulk density, \( \varrho \), from 2700 kg/m\(^3\) \((\text{Bourdon et al., 2009; Dosseto et al., 2010)}\) to 2670 kg/m\(^3\) (a value typically assumed for upper crust materials in Bouguer gravity corrections) produced little uncertainty on the modelled comminution age (\( \pm 4 \) ka, Table 4). Therefore, while a representative sample density should be used, reasonable variation in the assumed value has little impact upon the comminution age uncertainty relative to the other input assumptions.

The length scale of \( \Delta -\text{recoil} \), \( L \), was assigned the same uncertainty to that used in the weighted geometric model (28–32 nm, see above). This produced an uncertainty of \( \pm 24 \) ka in the computed \( t_{\text{com}} \), significantly smaller than that obtained for the geometric model (\( \pm 93 \) ka; Table 4). This is not surprising, however, given the larger uncertainties associated with the individual parameters in the weighted geometric model.

6.3.3. Uncertainties associated with variations in the source rock \( \text{U} \) isotope ratio

The majority of the bedrock exposed within the Lake Eyre Basin is significantly older 1 Ma and therefore, it should be safe to assume that, prior to the onset of physical weathering, the \((\text{U}_{234}/\text{U}_{238}) \) ratio of the source rock \( (A_{0}) \) is in secular equilibrium. However, as noted in Section 2.2.1, fine-grained sediments within sedimentary rocks may be held in a steady state of \( \text{U}_{234}/\text{U}_{238} \) disequilibrium (Fig. 2a) or the source rock may have experienced preferential loss of \( \text{U}_{234} \) prior to physical erosion (e.g., by leaching of \( \text{U}_{234} \) from recoil damaged sites), in which case, the \((\text{U}_{234}/\text{U}_{238}) \) ratio of the source rock may be less than unity. If, for example, the \((\text{U}_{234}/\text{U}_{238}) \) ratio of the source is unknown, but lies between 0.98 and secular equilibrium (1.00), this produces an uncertainty of \( \pm 178 \) ka (Table 4) using the weighted geometric model. In the situation that source activity ratios of greater than 1 are also possible (e.g., due to the implantation of \( \text{U}_{234} \text{Th} \) without subsequent leaching of \( \text{U}_{234} \) from the damaged site), for example, up to 1.02, this translates to greater relative uncertainty of \( \pm 265 \) ka (not shown). For the surface area method if \( A_{0} \) is unconstrained and lies between 0.98 and 1, this produces an expected similar uncertainty of \( \pm 136 \) ka (Table 4) in comminution age.

6.3.4. Total combined uncertainties in comminution age

To calculate a total combined uncertainty, which includes not only the uncertainty associated with \( A_{0} \) estimation, but also the uncertainty associated with the source \( (A_{0}) \) and measured sample \( (A_{\text{meas}}) \) \((\text{U}_{234}/\text{U}_{238}) \) activity ratios, Monte Carlo simulations were performed for both the weighted geometric and BET surface measurement (with fractal correction) approaches using the ranges of input variables detailed in Table 4. In both simulations, \( A_{0} \) was taken to range from 0.98 to 1 (as above) and the uncertainty assigned to \( A_{\text{meas}} \) was \( \pm 3\% \) (0.972–0.978) on the synthetic measured \((\text{U}_{234}/\text{U}_{238}) \) ratio (0.975). For the weighted geometric model, the simulation produced a combined uncertainty of \( \pm 278 \) ka and the surface area measurement method produced an uncertainty of \( \pm 224 \) ka. These uncertainties of approximately \( \pm 50\% \) of the true ages are clearly non-trivial and while it may be argued that some of the parameters have smaller uncertainties than those used here (e.g., BET surface area measurements), the majority of input assumptions remain relatively poorly constrained at present (e.g., sample dependent \( L \) value) and in some cases our ranges may be perceived as conservative.

6.4. Approach for the comparison of comminution ages from different studies

With a wide variety of pre-treatment procedures and recoil loss factor estimation methods (including discrepancies between studies in assumed input parameter values) already published in the relatively limited U-series comminution dating literature, it is desirable to standardise procedures for future studies to be able to directly compare estimated comminution ages from different studies or research groups.

First of all, a consensus is required on the best pre-treatment procedure for the removal of unwanted secondary phases and on whether the clay-sized fraction should or should not be removed as part of the pre-treatment procedure. Our study clearly shows the significant differences in
comminution age produced depending on whether the clay-sized grain fraction is removed or not. To determine the most suitable sequential extraction method a detailed study is required to compare presently published sequential phase extraction methods, their efficiency of unwanted phase removal and to assess any modification to U concentration and, in particular, (234U/238U) activity ratio for a range of sediment types (glacial, fluvial and aeolian origin) which may have different secondary phase coatings. The sediment size-fraction(s) for which U-series studies are carried out on may have different secondary phase coatings. The sediment types (glacial, fluvial and aeolian origin) which is required to compare presently published sequential phase extraction methods, their efficiency of unwanted phase removal and to assess any modification to U concentration and, in particular, (234U/238U) activity ratio for a range of sediment types (glacial, fluvial and aeolian origin) which may have different secondary phase coatings. The sediment size-fraction(s) for which U-series studies are carried out on may have different secondary phase coatings.

The next step required is to use the same estimation method for the recoil loss factor. In the comparison of the weighted geometric and BET surface area (fractal) methods in this study (1% uncertainty model, Table 4), the BET surface area (fractal) method carries slightly less uncertainty and therefore, we recommend this option as the preferred method of recoil loss factor determination. However, it would be useful to extend the Monte Carlo approach taken here to include an assessment of the uncertainty in alternative recoil loss factor calculation methods (see Maher et al., 2006; Lee et al., 2010 for summaries of potential $f_s$ estimation models). The same assumptions (between studies) for any unknown input parameter values and their ranges are also required. Our suggested assumptions for unknown input parameter values are given in Table 5. We do not imply that the input values given and the surface area (fractal) model for $f_s$ estimation provide the least uncertain approach, but rather that it will facilitate comparison of data from separate studies, which is not the case at present.

**7. SUMMARY AND CONCLUSIONS**

The comminution approach for the calculation of sediment residence timescales was investigated using U isotope measurements of the fine-grained fraction of sandy palaeochannel sediments from semi-arid Australia. The comminution age is shown to be highly dependent on the estimated fraction of $\alpha$-recoil, which is in turn dependent on the chosen method of $f_s$ estimation and the inherent parameter assumptions (e.g., surface roughness factor and grain aspect ratio). In several cases, the selected parameter values (based upon similar assumptions used in the literature) failed to produce high enough recoil loss factors for Cooper Creek sediments to allow valid solutions to the comminution equation.

---

### Table 4

The input parameter values and uncertainty estimations of Monte Carlo simulations.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Range considered</th>
<th>Range in $t_{com}$ (ka)</th>
<th>Uncertainty on $t_{com}$ (±ka)</th>
<th>Associated $f_s$ range</th>
<th>Solutions (out of 25,000)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Weighted geometric method</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Surface roughness, $\lambda_s$</td>
<td>1–7</td>
<td>1–30</td>
<td>169 801 316</td>
<td>0.026 0.066</td>
<td>All</td>
</tr>
<tr>
<td>Aspect ratio, $\beta$</td>
<td>1–9</td>
<td>1–12</td>
<td>393 733 170</td>
<td>0.028 0.037</td>
<td>All</td>
</tr>
<tr>
<td>Length of $\alpha$-recoil, $L$ (nm)</td>
<td>28</td>
<td>32</td>
<td>492 678 93</td>
<td>0.029 0.033</td>
<td>All</td>
</tr>
<tr>
<td>Source rock $^{234}$U/$^{238}$U, $A_0$</td>
<td>0.98</td>
<td>1</td>
<td>209 564 178</td>
<td>0.031 0.031</td>
<td>All</td>
</tr>
<tr>
<td>Combined $f_s$ uncertainty</td>
<td>126</td>
<td>692 283</td>
<td></td>
<td>0.025 0.083</td>
<td>24,843</td>
</tr>
<tr>
<td>Combined $t_{com}$ uncertainty</td>
<td>17</td>
<td>572 278</td>
<td></td>
<td>0.023 0.083</td>
<td>24,754</td>
</tr>
<tr>
<td><strong>Surface area (fract.) method</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fractal dimension, $D$</td>
<td>2.45</td>
<td>2.55</td>
<td>310 540 115</td>
<td>0.031 0.043</td>
<td>All</td>
</tr>
<tr>
<td>Total surface area, BET tot</td>
<td>8</td>
<td>12</td>
<td>253 488 118</td>
<td>0.033 0.049</td>
<td>All</td>
</tr>
<tr>
<td>Density, $\rho$ (kg/m$^3$)</td>
<td>2670</td>
<td>2700</td>
<td>396 404 4</td>
<td>0.037 0.037</td>
<td>All</td>
</tr>
<tr>
<td>Length of $\alpha$-recoil, $L$ (nm)</td>
<td>28</td>
<td>32</td>
<td>381 429 24</td>
<td>0.034 0.040</td>
<td>All</td>
</tr>
<tr>
<td>Source rock $^{234}$U/$^{238}$U, $A_0$</td>
<td>0.98</td>
<td>1</td>
<td>126 397 136</td>
<td>0.037 0.037</td>
<td>All</td>
</tr>
<tr>
<td>Combined $t_{com}$ uncertainty</td>
<td>401</td>
<td>407 3</td>
<td></td>
<td>0.037 0.037</td>
<td>All</td>
</tr>
<tr>
<td>Combined $t_{com}$ uncertainty</td>
<td>197</td>
<td>554 179</td>
<td></td>
<td>0.028 0.059</td>
<td>All</td>
</tr>
<tr>
<td>Combined $t_{com}$ uncertainty</td>
<td>25</td>
<td>472 224</td>
<td></td>
<td>0.028 0.059</td>
<td>All</td>
</tr>
</tbody>
</table>

Parameter ranges shown are those used in the estimation of the individual parameter uncertainties in comminution age (see Section 6.3 and Appendix A for more details).

Quoted $t_{com}$ ranges show the $t_{com}$ distribution produced by the simulation at 95% confidence limit. The uncertainty in $t_{com}$ equates to half of the calculated range in $t_{com}$.

For the surface area models, the diameter of the adsorbant nitrogen gas molecule (a) for BET analysis (0.35 nm) and the $^{234}$U decay constant ($\lambda_{diss} = 2.82629 \times 10^{-6}$, Bourdon et al., 2003) are considered constant.

1% uncertainty models were carried out by varying the input parameters shown in parenthesis by ±0.5% of the initial value used in the calculation of the reference model ages (572 ka and 404 ka for the weighted geometric and surface area estimations of $f_s$, respectively).

Models for the estimation of combined $f_s$ uncertainties in $t_{com}$ use the individual parameter ranges given in the table.

Models for the estimation of combined $f_s$ uncertainties use the individual parameter ranges given in the table, including that given for $A_0$ and an assumed uncertainty of ±3% in $A_{meas}$ (0.972–0.978). See Appendix A and Section 6.3 for further details.

Where the number of accepted simulations are less than the total run (25,000) some of the random models produced $f_s$ values below the minimum $f_s$ required to arithmetically solve the comminution equation (see the text for further details).
±220–280 ka. Much smaller uncertainties for the weighted geometric and surface area measurement (with fractal correction) methods are attained (±14 and ±3 ka, respectively) if the input parameters incorporated in the recoil loss factor models are known within 1% and there is no associated uncertainty in the source rock and measured sample (234U/238U) ratios. However, attaining such low uncertainties in the characterisation of the input parameters will be challenging. The BET surface area (fractal) estimation carries slightly less uncertainty in the determination of recoil loss factor (based upon a 1% uncertainty input parameter model), compared to the weighted geometric estimation of recoil loss factor.

Standardisation of pre-treatment procedures and \( f_s \) estimation method is required before the comminution ages and sediment residence times yielded in separate studies can be compared. A suggestion for generic assumed input parameter values for such a comparison are given here. The potential for preferential leaching of 234U (relative to 238U) during the pre-treatment leaching procedure should also be examined in future studies to assess the reliability of the leaching approach.

Clearly, the comminution technique affords great potential to determine the formation age of sediments providing that the present uncertainty of input parameters can be reduced, which should be a major goal for researchers in this field. Future directions include the measurement of U isotopes in catchment-representative source rocks, particularly where fine-grained sedimentary rocks (e.g., mudstones and siltstones) or fractured rocks dominate. This is required to confirm the assumed secular equilibrium value of this input parameter and place tighter constraint on the (234U/238U) variability of the source. Additional consideration of complicating factors such as whether the clay-sized fraction should be included or removed during sample pre-treatment, the effects of pre-treatment leaching on sample (234U/238U) activity ratios, the addition of dust and control of zircon are also required before the absolute comminution ages can be considered meaningful.

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Monte Carlo simulations were carried out using a synthetic sample to investigate the uncertainty in comminution age attached to each individual input parameter, as well as to determine a combined uncertainty estimation for each recoil loss estimation model, incorporating uncertainties in the source rock and measured sample (234U/238U) ratios. Using realistic ranges of input values, the total combined uncertainty in comminution age can amount to

Fig. 8. Example output from a Monte Carlo simulation (see Section 6.3 and Appendix A for further details on the model details). (a) Histogram of the comminution ages (valid comminution ages allowed by the assigned ranges in input parameters) obtained from 25,000 random models. (b) Cumulative frequency of comminution age in (a). The age range within the 95% confidence limit can be taken as a first-order measure of the uncertainty in the computed comminution age. The solution shown is for an uncertainty of ±0.5% in the initial input parameters used in the calculation of the reference model age (572 ka) for: surface roughness \((L_s)\) aspect ratio \((\beta)\) and s-recoil length \((L_s)\), using the weighted geometric model (with fixed source and sample 234U/238U ratios). The results show that if the input parameters \(L, L_s, \beta\) are known within 1% of their true values, the uncertainty on the calculated comminution reference age of 572 ka is ±14 ka, or ±2.4% (at the 95% confidence limit).
Table 5  
Suggested input parameter values to permit comparison between U-series comminution age studies.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Suggested value</th>
<th>Comment</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Weighted geometric method (Eq. (3))</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Surface roughness, ( \lambda_a )</td>
<td>1–10</td>
<td>Assuming a linear increase from the smallest to the largest grain</td>
</tr>
<tr>
<td>Aspect ratio, ( \beta )</td>
<td>10–1</td>
<td>Assuming a linear decrease from the smallest to the largest grain</td>
</tr>
<tr>
<td>Length of ( \alpha )-recoil, ( L (\text{nm}) )</td>
<td>30</td>
<td>Theoretical value for feldspar (Ziegler et al., 1996)</td>
</tr>
<tr>
<td>( ^{234} \text{U} ) decay constant, ( \lambda_{234} )</td>
<td>2.82629 ( \times 10^{-6} \text{yr}^{-1} )</td>
<td>After Bourdon et al. (2003)</td>
</tr>
<tr>
<td>Source rock ( { ^{234} \text{U} }/ { ^{238} \text{U} } ), ( A_0 )</td>
<td>1</td>
<td>Assuming no source rock disequilibrium</td>
</tr>
<tr>
<td>Measured ( { ^{234} \text{U} }/ { ^{238} \text{U} } ), ( A_{\text{meas}} )</td>
<td>As measured</td>
<td></td>
</tr>
<tr>
<td><strong>Surface area (frac.) method (Eq. (5))</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fractal dimension, ( D )</td>
<td>As determined</td>
<td>Following Bourdon et al. (2009)</td>
</tr>
<tr>
<td>Total surface area, ( \text{BET}_{\text{tot}} )</td>
<td>As measured</td>
<td></td>
</tr>
<tr>
<td>Density, ( \rho (\text{kg/m}^3) )</td>
<td>2670</td>
<td>Upper crust value used for Bouguer gravity corrections</td>
</tr>
<tr>
<td>( \text{N}_2 ) adsorbate gas molecule diameter (nm)</td>
<td>0.35</td>
<td>Theoretical value for feldspar (Ziegler et al., 1996)</td>
</tr>
<tr>
<td>Length of ( \alpha )-recoil, ( L (\text{nm}) )</td>
<td>30</td>
<td></td>
</tr>
<tr>
<td>( \delta_{234} )</td>
<td>2.82629 ( \times 10^{-6} \text{yr}^{-1} )</td>
<td>Theoretical value for feldspar (Ziegler et al., 1996)</td>
</tr>
<tr>
<td>Source rock ( { ^{234} \text{U} }/ { ^{238} \text{U} } ), ( A_0 )</td>
<td>1</td>
<td>After Bourdon et al. (2003)</td>
</tr>
<tr>
<td>Measured ( { ^{234} \text{U} }/ { ^{238} \text{U} } ), ( A_{\text{meas}} )</td>
<td>As measured</td>
<td>Assuming no source rock disequilibrium</td>
</tr>
</tbody>
</table>

**APPENDIX A. SUPPLEMENTARY DATA**

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.gca.2012.10.047.

**REFERENCES**


