1-1-2014

Very long hillslope transport timescales determined from uranium-series isotopes in river sediments from a large, tectonically stable catchment

P O. Suresh
Macquarie University, suresh.puthiyaveetil-othayoth@mq.edu.au

Anthony Dosseto
University of Wollongong, tonyd@uow.edu.au

Paul P. Hesse
Macquarie University

Heather K. Handley
Macquarie University

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Abstract

The uranium-series isotopic compositions of soils and sediments evolve in response to time and weathering conditions. Therefore, these isotopes can be used to constrain the timescales of river sediment transport. Catchment evolution depends on the sediment dynamic timescales, on which erosion imparts a major control. Erosion rates in tectonically stable catchments are expected to be lower than those in tectonically active catchments, implying longer sediment residence times in tectonically stable catchments. Mineralogical, elemental and isotopic data are presented for modern channel sediments, alluvial and colluvial deposits from the Murrumbidgee River, a large catchment in the passive margin highlands of south-eastern Australia and three of its tributaries from the headwaters to the alluvial plain. Low variability in Si-based Weathering Index indicates that there is little chemical weathering occurring in the Murrumbidgee River during sediment transport. However, quartz content increases and plagioclase content decreases downstream, indicating progressive mineralogical sorting and/or physical comminution with increasing transport distance. U-series isotopic ratios in the Murrumbidgee River trunk stream sediments show no systematic downstream variation. The weathering ages of sediments within the catchment were determined using a loss-gain model of U-series isotopes. Modern sediments from a headwater tributary, the Bredbo River at Frogs Hollow, have a weathering age of 76 ± 30 kyr but all other modern channel sediments from the length of the Murrumbidgee River and its main tributaries have weathering ages ~400 ± 180 kyr. The two headwater colluvial deposits have weathering ages of 57 ± 13 and 47 ± 11 kyr, respectively. All the alluvial deposits have weathering ages similar to those of modern sediments. No downstream trend in weathering age is observed. Together with the soil residence time of up to 30 kyr for ridge-top soils at Frogs Hollow in the upper catchment area of the Murrumbidgee River (Suresh et al., 2013), the current results indicate, for the first time, that sediments in the Murrumbidgee catchment are stored in hill slope for long time (~200 kyr) before carried by the river. The long residence times of sediments indicate a low erosion rate from the catchment. The sediment transport timescales estimated are up to two orders of magnitude higher than those reported for tectonically active catchments in Iceland (Vigier et al., 2006) and in the Himalayas (Granet et al., 2007), indicating the influence of tectonism on catchment erosion.

Disciplines

Medicine and Health Sciences | Social and Behavioral Sciences

Publication Details

Suresh, P. O., Dosseto, A., Hesse, P. P. & Handley, H. K. (2014). Very long hillslope transport timescales determined from uranium-series isotopes in river sediments from a large, tectonically stable catchment. Geochimica et Cosmochimica Acta, 142 442-457.

*Manuscript

Very Long Hillslope Transport Timescales Determined from Uranium-Series Isotopes in River Sediments from a Large, Tectonically Stable Catchment

P. O. Suresh[1*], A. Dosseto[2], P. P. Hesse[1] and H. K. Handley[3]

[1]Department of Environment and Geography, Macquarie University, Sydney, Australia

[2]Wollongong Isotope Geochronology Laboratory, School of Earth and Environmental Sciences, University of Wollongong, Australia

[3]GEMOC, Department of Earth and Planetary Sciences, Macquarie University, Sydney, Australia

*Corresponding author

Department of Environment and Geography, Macquarie University, Sydney NSW 2109, Australia. E-mail: suresh.puthiyaveetil- othayoth@mq.edu.au.

Current address

R & D Division, Gujarat State Fertilizers and Chemicals Limited, Fertilizer Nagar, Vadodara, Gujarat – 391750, India. Email: posuresh@gsfcltd.com. Phone: +91-2653092529. Fax: +91-2652240966

Abstract

The uranium-series isotopic compositions of soils and sediments evolve in response to time and weathering conditions. Therefore, these isotopes can be used to constrain the timescales of river sediment transport. Catchment evolution depends on the sediment dynamic timescales, on which erosion imparts a major control. Erosion rates in tectonically stable catchments are expected to be lower than those in tectonically active catchments, implying larger sediment residence times in tectonically stable catchments. Mineralogical, elemental and isotopic data are presented for modern channel sediments, alluvial and colluvial deposits from the Murrumbidgee River, a large catchment in the passive margin highlands of south-eastern Australia and three of its tributaries from the headwaters to the alluvial plain. Low variability in Si-based Weathering Index indicates that there is little chemical weathering occurring in the Murrumbidgee River during sediment transport. However, quartz content increases and plagioclase content decreases downstream, indicating progressive mineralogical sorting and/or physical comminution with increasing transport distance. U-series isotopic ratios in the Murrumbidgee River trunk stream sediments show no systematic downstream variation. The weathering ages of sediments within the catchment were determined using a loss-gain model of U-series isotopes. Modern sediments from a headwater tributary, the Bredbo River at Frogs Hollow, have a weathering age of 76 ± 30 kyr but all other modern channel sediments from the length of the Murrumbidgee River and its main tributaries have weathering ages ~400 ± 180 kyr. The two headwater colluvial deposits have weathering ages of 57 ± 13 and 47 ± 11 kyr, respectively. All the alluvial deposits have weathering ages similar to those of modern sediments. No downstream trend in weathering age is observed. Together with the soil residence time of up to 30 kyr for ridge-top soils at Frogs Hollow in the upper catchment area of the Murrumbidgee River (Suresh et al., 2013), the current results indicate, for the first time, that sediments in the Murrumbidgee catchment are stored in hill slope for long time (~ 200 kyr) before carried by the river. The long residence times of sediments indicate a low erosion rate from the catchment. The sediment transport timescales estimated are up to two orders of magnitude higher than those reported for tectonically active catchments in Iceland (Vigier et al., 2006) and in the Himalayas (Granet et al., 2007), indicating the influence of seismicity on catchment erosion.

Keywords

Murrumbidgee River, U-series isotopes, sediment residence time, alluvium, weathering age

1. Introduction

The evolution of uranium-series (U-series) activity ratios during weathering is affected by factors such as pH, the presence of organic matter, and time. U-series isotopes are expected to be in secular equilibrium (parent-daughter activity ratio = 1) in unweathered bedrock older than 1 Myr (Bourdon et al., 2003; Dosseto et al., 2008a). If the half-life of the parent isotope is longer than the daughter isotope, then in a period of ~5 half-lives of the daughter nuclide, the parent daughter activity ratio will reach secular equilibrium (Bourdon et al., 2003) (half-lives of ^{238}U, ^{234}U and ^{230}Th are 4.4683 x 10^9 years (Jaffey et al., 1971), 24.525 x 10^4 years and 75.69 x 10^3 years (Cheng et al., 2000), respectively). Fractionation between isotopes during geological processes such as chemical weathering induces radioactive disequilibrium. During chemical weathering, U is preferentially mobilized over Th (Chabaux et al., 2003). Oxidizing conditions prevail in most weathering environments and so U will be present as U^{6+}, which is soluble in waters as the uranyl ion, $U_{VI}O_2^{2+}$ and is stabilized by highly soluble and non-reactive carbonate complexes (Langmuir, 1978). Th will be present as Th^{4+}, which is insoluble. This causes elemental fractionation between U and Th, and affects the ($^{230}Th/^{234}U$) activity ratios of weathered material. In addition, the high energy involved in the radioactive decay of U-series isotopes can damage crystal lattices and enhance loss of the daughter nuclide by leaching from damaged recoil tracks, hence creating disequilibrium in the parent-daughter activity ratio (Kigoshi, 1971, Rosholt, 1983, Chabaux et al., 2003, Vigier et al., 2011). Also, if radioactive decay occurs near the surface of the grain a fraction of the daughter nuclide may be directly ejected from a mineral grain (Kigoshi, 1971; DePaolo et al., 2006). The degree of fractionation of U-series isotopes in soils and sediments can be used to determine the timescale of weathering and erosion processes, as the radioactive disequilibrium is time dependent.

Soil residence time (Table 1) and production rates have been determined in different climatic and geomorphic settings through modelling the evolution of uranium-series isotopes in soil (Mathieu et al., 1995; Dequincey et al., 2002; Dosseto et al., 2008b; Ma et al., 2010; Dosseto et al., 2012; Suresh et al., 2013). Using the same model, the weathering age (Table 1) of sediments transported by rivers in a variety of geographical locations and climatic settings have been determined by Vigier et al. (2001; 2005; 2006), Dosseto et al. (2006a; b, 2008b) and Granet et al. (2007; 2010). Variations are observed in calculated sediment weathering ages between catchments and are controlled by changes in climate, human activity, relief, tectonic activity and bedrock composition.

The dissolved and suspended loads of sediments in rivers largely show shorter transport timescales (a few kyr) (Dosseto et al., 2008b, Granet et al., 2010) relative to the coarser particle load, such as the bedload (in the order of ~100 kyr or more) (Dosseto et al., 2008b, Granet et al., 2010). Suspended sediments from tropical rivers flowing through basaltic terrain in the Deccan Traps have given residence times 55 – 84 kyr (Vigier et al., 2005), whereas those from rivers draining basaltic terrain in Iceland gave residence times 1 – 8 kyr (Vigier et al., 2006). The two order of magnitude difference in residence times of sediments from lowland Amazon Rivers (100 – 500 kyr) and upland Amazon Rivers (3 – 4 kyr) could be due to differences in catchment relief in the two regions (Dosseto et al., 2006a, b). Suspended sediments from the upper Ganga (Ganges) River and tributaries in the Himalayas gave residence times ~30 kyr, but those from the river on the Ganga plain showed much higher residence times (~350 kyr) (Granet et al., 2007). The longer residence times on the alluvial plains may be due to reworking of old sediments in the plain. Lower relief compared to the upper river basin in the plain could also imply slower transport. In summary, large variations in sediment residence times in rivers are observed showing the influence of different factors like climate, catchment geomorphology and glaciation, controlling sediment movements in the catchment areas. Studying the sediment transport timescales and the affecting factors in the Murrumbidgee catchment, a tectonically stable passive margin in the highland area of south-eastern Australia where periglacial conditions prevailed during the Last Glacial Maximum (LGM), will further our current understanding of soil and landscape evolution in large catchments. A consolidated study of sediments in alluvium, colluviums and modern channel is expected to provide new insights on their evolution throughout the catchment.

Soil processes in the upper Murrumbidgee catchment are affected by factors such as rainfall and topography (Suresh et al., 2013). Ridge-top soil residence times of approximately

30 kyr have been determined using U-Th isotopes in the upper catchment area of the Murrumbidgee River (Suresh et al., 2013). Heimsath et al. (2001) reported soil production rate of ~ 45 mm/kyr by measuring cosmogenic radionuclides of ^{26}Al and ^{10}Be concentrations in soils at this location. Using this rate, Yoo et al. (2007) estimated a lateral transport time of ~60 kyr for soils along a 50 m hillslope profile. Dosseto et al. (2010) estimated comminution ages (Table 1) of ≤ 50 μm size sediments from palaeochannels and the modern channel of the Murrumbidgee River. They reported comminution ages an order of magnitude lower for the post-Last Glacial Maximum (LGM) sediments than the Holocene and pre-LGM sediments. The authors attributed the lower sediment residence time (Table 1) of the post-LGM sediments to the erosion of materials from the upper catchment and the higher sediment residence time of the modern and pre-LGM sediments to the reworking of alluvial deposits.

This paper presents U-series isotope, mineralogical and major element data of modern sediments, alluvial and colluvial deposits from the Murrumbidgee River of south-eastern Australia. The data are used to estimate the weathering ages of sediments in the catchment. In combination with the soil residence times determined using U-series isotopes (Suresh et al., 2013) and long-term erosion rates determined using cosmogenic radionuclides (Fujioka et al., 2012), the data are used to constrain the timescales of sediment dynamics from initial weathering to final deposition in this large catchment. This will aid our understanding of the long-term evolution and sustainability of landscapes in large catchments. The proposed change in sediment source, pre- and post-LGM, in the Murrumbidgee catchment described by Dosseto et al. (2010) is also re-examined using the new results.

2. Study Area

The Murrumbidgee River in south-eastern Australia is divided into three distinct geomorphic regions (Wallbrink et al., 1998). The upper catchment is a mountainous region with high relief (up to 2000 m above sea level) comprising an area of approximately 20,500 km^2 (Fig. 1). Burrinjuck Dam isolates the upper from the middle Murrumbidgee catchment (combined area 34,000 km^2). The middle catchment is characterized by rolling terrain with gullying (Wallbrink et al., 1998) and progressively decreasing tributary input to the river further downstream. The river enters the lower Murrumbidgee alluvial plain downstream of Narrandera to eventually merge with the Murray River. Average annual rainfall in the catchment area ranges from 1900 mm in the upper mountains to less than 350 mm on the plain (NSW Water, 2011).

Modern sediment samples (currently being transported by the river) were collected from riverbanks and bars at 7 locations throughout the length of the Murrumbidgee River, in the section between the confluence of the Bredbo River with the Murrumbidgee River in the mountainous, bedrock-confined upper catchment and Darlington Point in the alluvial plain (Fig. 1). These modern samples were either deposited or mobilized by recent flows. Modern channel sediments were collected above and below the confluences of the river with major tributaries, namely the Bredbo, Goodradigbee and the Tumut Rivers. A modern sediment sample from the upper Bredbo River was collected from Frogs Hollow, ~40 km upstream of Bredbo (Fig. 1). Alluvial deposits from four locations along the Murrumbidgee River were sampled at different depths, wherever possible (Table 2). These samples, from deep bank exposures, represent older and/or higher floodplain deposits of likely Holocene age. Two samples from the alluvial deposits at Wangrah Creek, a minor tributary in the upper catchment (studied by Prosser et al., 1994) area also were collected. Two colluvial deposit samples were collected from a gully near Bredbo. The colluvial sediments were deposited by runoff and sheetwash processes at the base of a ridge. A late Pleistocene dust deposit sample (deposition age = 21.6 ± 2 ka, determined by optically stimulated luminescence dating; in Fitzsimmons et al. 2013) was collected at a depth of 150 cm from McKenzie’s Waterhole Creek near Carcoar (Fig. 1; Hesse et al. 2003), to assess the contribution of aeolian material to the U-series isotope composition of river sediments.

3. Materials and Method

Modern sediments were collected from banks and bars of the river channel. Colluvial and alluvial deposits were sampled from natural bank exposures adjacent to the channel. The depositional ages of alluvial sediments are unknown, except for those from Wangrah Creek (MU8_low and up, 12,420 ± 150 yr BP; radiocarbon age; Prosser et al., 1994).

All of the samples were dried at 60°C overnight. Aliquots of each sample were placed in acid-cleaned polypropylene containers for X-ray diffraction (XRD), X-ray fluorescence (XRF) and U-series isotopic analysis.

Dried samples were powdered to ~10 µm size for XRD using an agate mortar and pestle. X-ray diffractograms were recorded using a PANalytical X'pert PRO MPD diffractometer with a 45 kV, 40 mA CuKα radiation X'celerator detector and Bragg Brentano geometry, conducting scans from 5 to 50° 2θ at 5° 2θ/min. Highscore Plus software version 2.2.4 with the ICDD PDF2 database by PANalytical and the basic Rietveld refinement option available in the software were used for mineral identification and quantification, respectively.

Sediment aliquots for major element analysis were powdered to less than 10 µm and prepared into 40 mm glass discs by fusion with lithium borate containing lanthanum oxide (Norrish and Hutton, 1969). Analyses were performed using a Philips PW2400 XRF instrument at Mark Wainwright Analytical Centre (University of New South Wales), following the procedure described by Norrish and Hutton (1969). Reproducibility of the results was determined by replicate sample analysis (3 samples) yielding an elemental oxide standard error below 1 % for all elements.

For U-series analysis, samples were ashed at 550 °C overnight. Approximately 2 g of ashed sample was leached with $Mg(NO_3)_2$ to remove ion exchangeable uranium and thorium from the grain surfaces and U-Th bound to the organic matter destroyed during ashing (Gleyzes et al., 2002). Approximately 100 mg of leached sample was weighed into 15 ml PFA vials and approximately 30 mg of a ^{236}U-^{229}Th tracer solution was added and weighed. The samples were then digested in a mixture of HCl, HNO_3, HF and $HClO_4$ in closed vials at 120 °C overnight. The sample solutions were then dried and then redissolved in 7M HNO_3. U and Th were separated and purified using an anion exchange resin (Biorad AG1X8) following a procedure described in Sims et al. (2008). Measurement of U and Th isotopes were performed on a Nu Instruments Nu-Plasma multi-collector ICPMS instrument following the procedure outlined in Turner et al. (2011). Reproducibility of the results was assessed by replicating the whole procedure for two samples. This yielded a reproducibility of 0.17 % for Th concentration, 0.8 % for U concentration, 1.2 % for (^{234}U/^{238}U) and 3 % for (^{230}Th/^{238}U). Accuracy was determined by measuring the U-series isotopic ratios and U-Th concentrations in TML-3, a standard rock sample (Table Mountain Latite, Williams et al., 1992; Sims et al., 2008). The measured concentrations and activity ratios are within 2σ error limits of published values (Table 2; Sims et al., 2008). The total procedural blank was 150 pg for U and 140 pg for Th, which are insignificant when compared to the U and Th amounts of sediment samples digested (~0.5 µg of U and ~2.5 µg of Th for 0.1g of sample).

4. Results

4.1. Mineralogy

All of the modern channel sediment samples contain > 60 wt. % quartz, except for the sample from the Bredbo River at Frogs Hollow (Table 3; Fig. 2). Albite, microcline and muscovite contents vary from 0 to 20 wt. %. Illite was detected only in some of the modern sediments (Table 3). All the alluvial deposit samples except MU8_up contain > 60 wt. % quartz (Table 3). Albite content varies from 0 to 20 wt. %, microcline from 0 to 10 wt. % and muscovite from 3 to 54 wt. %. Illite was detected only in the alluvial sample collected from Darlington Point (1.6 wt. %). Colluvial deposit samples contain 61 wt. % quartz and approximately 30 wt. % muscovite. Albite was detected (6 wt. %) in only one of the colluvial samples (Table 3). XRD analysis detected only quartz in the dust sample.

4.2. Particle size

All the modern sediment samples, except MU11 from the Bredbo River at Frogs Hollow, contain > 25 % mud (Table 3). Alluvial samples contain variable proportions of mud (< 63 µm) and sand (63 to 2,000 µm) (Table 3). The alluvial deposit sample MU3 from the deposits near the Bredbo – Murrumbidgee confluence is the coarsest, with only 2 % mud. The alluvial samples from a single deposit contain variable proportions of mud at different depth. For example, the sample MU8_up from 1.4 m depth at Wangrah Creek contains 74 % mud, whereas the sample MU8_low from 3.6 m depth contains 37 % mud. These variations reflect the mixed suspended (mud) and bed (sand) load nature of the Murrumbidgee River and reflect small variations of depositional environment within the river bed and proximal floodplain.

4.3. Major Elements

Major element data are given in Table S1 in the Appendix. All of the samples contain > 64 wt. % SiO_2, except sample MU8_up from the Wangrah Creek alluvial deposit, with 57 wt. % SiO_2. Greater than 10 wt. % Al_2O_3, ~ 2 to 9 wt. % Fe_2O_3 and > 2 wt. % K_2O were detected in all samples. An increase in SiO_2 and decrease in Al_2O_3 with decreasing depth is observed for alluvial deposits at Gundagai and colluvial samples from Bredbo gully. The dust sample contains 84 wt. % SiO_2, 6 wt. % Al_2O_3 and 4 wt. % Fe_2O_3 (Table S1).

4.4. U-series isotopes

U concentrations in the samples range from 1.7 to 6.3 ppm and Th concentrations vary between 9.6 and 21.8 ppm in alluvial deposits (Table 2). The highest U and Th concentrations were observed in the alluvial samples MU8_up from Wangrah Creek and MU19_low from Gundagai. In alluvial deposits from Gundagai, U and Th concentrations decrease with decreasing depth. The two colluvial samples from Bredbo have nearly identical U and Th concentrations. For the modern sediment samples, U and Th concentrations display a range similar to those of alluvial deposits.

All of the sediment samples have ($^{234}U/^{238}U$) activity ratios greater than 1, indicating the relative enrichment of ^{234}U. The ratios vary from 1.02 to 1.35 in the alluvial deposits and from 1.09 to 1.26 in the modern sediments (Table 2, Fig. 3). All of the sediment samples except MU3, MU11 and MU22 have ($^{230}Th/^{234}U$) ratios lower than 1, varying from 0.61 to 0.86. This also may correspond to enrichment of ^{234}U.

The dust sample contains 11.6 ppm Th and 1.7 ppm U (Table 1). It has a ($^{234}U/^{238}U$) ratio of 0.996 and ($^{230}Th/^{234}U$) ratio of 1.24 (Table 2).

5. Discussion

5.1. Mineral sorting and weathering

The presence of quartz, albite, microcline and muscovite in all sediments is consistent with the large area of granitic bedrock in the Murrumbidgee catchment. Quartz content increases and plagioclase content decreases downstream in the modern sediments (Fig. 2). This could be due either to chemical dissolution of plagioclase, preferential physical weathering of plagioclase over quartz, or due to mineral hydrodynamic sorting. The lack of evidence for chemical dissolution or physical breakdown (detailed below) of minerals points towards mineral sorting during river transport. The increasing extent of mineral dissolution with increasing stream length should correspond to a similar trend in weathering indices, which is not observed here. The Si-based weathering index (WIS = $SiO_2/(SiO_2+Al_2O_3+CaO+Na_2O)x100$) does not show a downstream increasing trend. The WIS is preferable to other indices of chemical weathering, such as the CIA or CIW (Harnois, 1988) because of the mobilization of Al during weathering in the soil profiles (Driscoll et al., 1985; White et al., 2008; Suresh et al. 2013; Suresh et al., submitted). The CIA and CIW indices consider Al to be immobile (Nesbitt and Young, 1982; Harnois, 1988). Suresh et al. (2013) reported mobility of Al in the soil of the Murrumbidgee catchment. The lack of a systematic downstream evolution of WIS values either in modern sediments or alluvial deposits of the Murrumbidgee River suggests that little chemical weathering occurs during sediment transport (Fig. 4). Physical breakdown of particles or progressive abrasion downstream should correspond to a decreasing trend in particle size distribution, which is not observed here (Table 3). Hydrodynamic sorting of minerals, which occurs depending on the settling velocity of minerals grains (related to their size, density and shape) and flow velocity, could have affected the distribution of the minerals in the sediments. Observations of mineral sorting have been reported in the Yamuna River in the Himalayas (Dalai et al., 2004). The absence of a downstream trend in WIS values may also imply rapid transport of sediments by the river.

Mobilization of elements from sediments has been commonly discussed in comparison to the composition of the average upper continental crust (UCC) (Taylor and McLennan, 1985; Dalai et al., 2004). All major element contents were averaged for the alluvial deposits, modern sediments and colluvium and then normalized to average UCC contents taken from McLennan (1995) and bedrock values taken from Chappell (1984) (Fig. 5). Normalised Na, Ca and Mg contents are all < 1, indicating loss during chemical weathering. Al, Si and K are comparatively immobile. Mn seems to be enriched in the modern and colluvial samples, which could possibly indicate anthropogenic input of Mn, but the huge error bars limits our ability to draw conclusions (Fig. 5).

5.2. Uranium and thorium concentration and activity ratios

U concentrations in the modern channel sediments do not show significant trends with stream length (Table 2). This may indicate that no significant leaching of U is occurring during transport. Th concentrations are much more variable than U. Positive correlations (correlation coefficient R = 0.86 for U and 0.6 for Th) exist between sediment mud content and U and Th concentration (Fig. 6). An increase in soil and sediment U and Th concentration with decreasing grain size has been reported by Baeza et al. (1995), Lee et al. (2004) and Suresh et al. (2013). In the modern channel sediment samples, U concentrations show a strong (R = 0.8) positive correlation with muscovite content (Fig. 7). Similar observations were reported in soil profiles from Frogs Hollow in the catchment area of the Murrumbidgee River (Suresh et al., 2013). Suresh et al. (2013) proposed that muscovite is the mineral phase dominating the U budget in the Frogs Hollow soil profiles. Our data suggest that this is also true in river sediments.

The lack of significant chemical weathering during river transport (Fig. 4) could suggest that there has been little fractionation of U-series isotopes during fluvial transport. A negative correlation exists between WIS and U and Th concentrations in alluvial or modern sediments (Fig. 8). These two observations together imply that no significant U or Th loss is occurring during transport due to chemical weathering. The decrease in U and Th concentration with increasing WIS may be the result of chemical weathering of sediments before reaching the river channel. Soils from Frogs Hollow in the upper Murrumbidgee catchment exhibit WIS values similar to those observed in the river sediments (Fig. 8).

($^{234}U/^{238}U$) activity ratios > 1 in all samples suggest an enrichment of ^{234}U over ^{238}U (Fig. 3). However, during chemical weathering, ^{234}U is preferentially removed from the solid phase, and therefore, ($^{234}U/^{238}U$) < 1 is expected in the residue of weathering (Chabaux et al., 2003; Dosseto et al, 2008a). The activity ratio ($^{234}U/^{238}U$) <1 is observed in the soil samples from Frogs Hollow in the upper catchment area of the Murrumbidgee River (Suresh et al., 2013). This again follows the suggestion that the removal of U and Th from the sediments occurs before entering in to the river stream. Sediment ($^{234}U/^{238}U$) activity ratios > 1 suggest input of ^{234}U from a fluid phase. During chemical weathering the ^{234}U leached from the solid phase will be concentrated in the fluid, driving the ($^{234}U/^{238}U$) ratio in the liquid > 1 (Dequincey et al., 2002; Chabaux et al., 2003; Robinson et al., 2004; Anderson et al., 2007; 2009; Vigier et al., 2011). The mineral phases formed from this fluid are characterised by the ($^{234}U/^{238}U$) ratio of the fluid phase, i.e. > 1 (Plater et al., 1992, Dequincey et al., 2002). The

^{234}U from the fluid phase may be retained by residual phases by several mechanisms discussed by Scott (1968), namely: 1. Incorporation into the lattices of clay minerals, 2. Adsorption to mineral surfaces, 3. Association to Al and Fe oxides and 4. Complexation by organic materials. ($^{234}U/^{238}U$) activity ratios > 1 in river sediments have been observed in Deccan rivers (Vigier et al., 2005), Amazonian rivers (Dosseto et al., 2006a) and Himalayan rivers (Granet et al., 2007).

The ($^{234}U/^{238}U$) ratios > 1 and ($^{230}Th/^{234}U$) ratios < 1 in sediments imply a net gain of U over loss to leaching and that the gained U has a higher ($^{234}U/^{238}U$) ratio than that in the leached U component (Dequincey et al., 2002). ($^{234}U/^{238}U$) or ($^{230}Th/^{234}U$) ratios in the modern sediments show no systematic downstream evolution. The ($^{230}Th/^{234}U$) ratios of all but two of the modern sediments are similar (averaged at 0.712 with 1σ standard deviation of 0.034). The only modern sediment with a ($^{230}Th/^{234}U$) > 1 is from the uppermost headwater location and thus is expected to have undergone the least in-channel transport. Colluvial samples also show ($^{230}Th/^{234}U$) ratios > 1. The ($^{234}U/^{238}U$) and ($^{230}Th/^{234}U$) > 1 in colluvial deposits could imply a net removal of U due to leaching over gain, and a more fractionated ($^{234}U/^{238}U$) ratio (> 1) in the gained component than in the leached (Dequincey et al., 2002). This could further imply that, for the sediments in the Murrumbidgee catchment, ($^{230}Th/^{234}U$) ratios are > 1 before sediments enter the river channel, where significant exchange of U between sediment and water would result in a net gain of ^{234}U, producing ($^{230}Th/^{234}U$) ratio < 1. For alluvial deposits, ($^{230}Th/^{234}U$) ratios decrease with increasing ($^{234}U/^{238}U$) (R = -0.9), indicating the evolution of these ratios with time (Fig. 3).

5.3. Weathering age of Sediments in the Murrumbidgee River catchment

The variation of U-series isotopic composition of sediments is a function of time and any loss or gain of isotopes. Vigier et al. (2001), Dosseto et al. (2006a, b) and Granet et al. (2007, 2010) used a model to quantify the time-variation of U-series isotopes in sediment and soil samples, considering the loss of isotopes by chemical weathering. Their model considered the present concentration of any radioactive isotope as a function of its production by decay from the parent (if present), and loss through its own radioactive decay and leaching. Dequincey et al. (2002) and Dosseto et al. (2008a; b) modified these models by incorporating any possible gain of isotope to the sediments by processes such as precipitation of secondary phases, dust deposition or physical illuviation in soil profiles. In this model, the time variation of the abundance of a given isotope N_j is:

$$\frac{dN_j}{dt} = \lambda_i \cdot N_i - \lambda_j \cdot N_j - w_j \cdot N_j + \Gamma_j \cdot N_{j,0} \quad (1)$$

where subscripts *i* and *j* refer to the parent and daughter isotopes, respectively, λ is the decay constant (yr^{-1}), $N_{j,0}$ is the initial isotope abundance in the unweathered bedrock, Γ and w are gain and loss coefficients, respectively (in yr^{-1}). The gain coefficient, Γ, represents the rate at which an isotope is incorporated to the sediments via dust deposition and/or secondary phase coating. Hence, the terms $\Gamma_j \cdot N_{j,0}$ in the combined form represents the rate at which the abundance of the isotope is increased due to gain. The loss or dissolution coefficient w determines the rate at which isotope loss occurs during chemical weathering. The coefficients w and Γ are assumed to be constant over the duration of weathering for each nuclide (Chamberlain et al., 2005; Ferrier and Kirchner, 2008). White and Brantley (2003) also suggested constant w values for a given mineral based on laboratory experiments of mineral weathering. The term t represents the time elapsed since the onset of weathering by which isotope fractionation occurred, and is termed the *sediment weathering age*, T_w (Table 1). Detailed discussion on the model is given in Dosseto et al. (2008b). This model can be solved for sediment samples from a given catchment area to determine the set of w and Γ values for each nuclide and the weathering age (T_w) for each sample by reproducing the observed ($^{230}Th/^{234}U$) and ($^{234}U/^{238}U$) activity ratios. For the set of 10 samples from the modern sediments, there are 20 input parameters (($^{230}Th/^{234}U$) and ($^{234}U/^{238}U$)) and 16 output parameters (10 weathering ages, three w values and three Γ values). The w and Γ for this set of samples for a given nuclide is assumed to be the same. For solving the model, boundary conditions are applied to w and Γ values of each nuclide to include values reported so far for these parameters either in nature or in laboratory experiments in the range for a set of samples. The reported values of w_{238}, and w_{234} vary between 10^{-6} and 10^{-4} yr^{-1} and w_{230} varies between 10^{-18} and 10^{-4} (Vigier et al., 2001, 2005, 2006, 2011; Dequincey et al., 2002; Chabaux et al., 2006; Dosseto et al., 2006a, b, c, 2008a, b, 2012; Ma et al., 2010; Suresh et al., 2013). For Γ values of ^{238}U and ^{234}U, the range used is 10^{-8} to 10^{-4}, and for ^{230}Th, the range used is 10^{-14} to 10^{-4} which includes published values for ^{238}U, ^{234}U and ^{230}Th isotopes (Dosseto et al., 2008a, b, 2012; Ma et al., 2010; Suresh et al., 2013).

For the Murrumbidgee sediment samples, the model was solved using codes written in Matlab for alluvial, colluvial and modern samples separately. The composition of the Devonian granitic bedrock from Cooma in the upper Murrumbidgee catchment, reported by Chappell (1984) is taken as the initial condition at the onset of bedrock weathering, representative of the catchment area. Since the bedrock is older than 1 Myr, ($^{230}Th/^{234}U$) and ($^{234}U/^{238}U$) ratios are considered to be at secular equilibrium (cf. Handley et al., 2013). The gain (Γ) and dissolution (w) coefficients can take different values for each nuclide. A constant w value implies a time-dependant evolution of chemical dissolution rate of an isotope, with a higher w value corresponding to the rate of reaction slowing down faster to reach a steady state, and vice-versa (White and Brantley, 2003).

While solving, the model equation (1) iteratively produces ($^{230}Th/^{234}U$) and ($^{234}U/^{238}U$) ratios and compares them with the measured ($^{230}Th/^{234}U$) and ($^{234}U/^{238}U$) to minimize the difference between the model-produced and measured ratios. Since the equation is highly non-linear, a set of solutions for each T_w, w and Γ value is generated. The mean value of each set will be taken as the final solution. The error value associated with each final solution is calculated using the 1σ standard deviation of the produced set of solutions. The leaching and gain coefficients for all nuclides and the T_w values of each sample are given in Table 4.

The weathering age of colluvial deposits from Bredbo Gully encompasses the vertical soil profile residence time, lateral transport time through the hillslope, and the storage time in the colluvial deposit. Two Bredbo Gully samples collected at 0.4 m and 1.7 m depth display weathering ages of 48 ± 11 and 57 ± 13 kyr, respectively (Table 4; Fig. 9). The modern sediment from the upper Bredbo River (Frogs Hollow) has a weathering age of 77 ± 31 kyr. There are no alluvial deposits observed upstream of this sampling site and sediments are thus expected to be delivered to the channel directly from the hillslope. All the other modern sediments and alluvial deposits have weathering ages that vary from 313 ± 142 to 451 ± 191 kyr. Note that ages of 316 ± 52 and 480 ± 78 kyr for modern sediments from the Murrumbidgee River were determined by Dosseto et al. (2010), using the comminution dating approach developed by DePaolo et al. (2006), which agree with our results.

Anthropogenic activities such as land clearing and agricultural practices have directly or indirectly led to increased soil erosion rates and catchment sediment yield, compared with pre-European settlement rates (Wasson et al., 1998; Olley et al., 2003). Conversely, the Burrinjuck Dam (constructed in 1920) is considered to be an excellent trap of river sediments (e.g. Wasson et al., 1987; Srikanthan and Wasson, 1993; Olley et al., 1997; 2003) and thousands of small farm dams have been constructed throughout the catchment. However, these significant changes to sediment load have not affected the overall source types or U-series characteristics of sediments in the river. Our results show that the sediments upstream and downstream of the Dam have the same weathering age, indicating that the trapping of sediments by the construction of the dam has not affected the nature of sediments downstream. Dosseto et al. (2010) also reported residence times of > 300 kyr for both pre-European (deposition age ~2.5 kyr) and post-European settlement sediments from the Murrumbidgee River downstream of the Burrinjuck dam.

The leaching coefficients (w) for ^{238}U and ^{234}U estimated from the model for the sediments are consistent (within the large model errors; Table 4), showing that removal of these isotopes from the sediments takes place at comparable rates over the timescales of sediment evolution. The gain coefficients (Γ) for these isotopes in the sediment samples are also the same, within the large errors associated. Leaching coefficients estimated for modern sediments are similar to those determined for the suspended sediments in the Mackenzie River (Vigier et al., 2001) and for Amazon highland rivers (Dosseto et al., 2006b). Leaching coefficients of ^{238}U for the Ganga River and Narmada and Tapti rivers are an order of magnitude lower than those determined for the Murrumbidgee sediments. These differences probably indicate that leaching of U-series isotopes from sediments is controlled by the conditions of weathering in the rivers. Dosseto et al. (2008a) and Vigier et al. (2011) compiled ^{238}U leaching coefficients for sediments of different residence time and found a linear relationship. The leaching coefficient of ^{238}U observed here also conforms to the same relationship. White and Brantley (2003) reported that the dissolution rate of silicate minerals decreases significantly over kilo year timescales. This could possibly correspond to a decrease of leaching rate of ^{238}U. A general explanation is still to be reported for the observation of decreasing leaching coefficient with increasing weathering age. Keech et al. (2013) reported the leaching coefficients of U-series isotopes from soil samples in the soil chronosequence in Merced and proposed that the leaching process may not be uniform over the timescales of weathering. They argued that weathering rate can vary over time and hence assuming a first order leaching coefficient may not be appropriate in the case of sediments.

However, since the weathering rate represents the rate of mass loss and leaching coefficient represents the timescales of loss of an isotope, a first order assumption is still plausible. The coefficients reported here for the modern sediments, alluvial deposits and colluvial samples having different residence times are similar, and could possibly indicate steady state isotope leaching and gain coefficients over the time of weathering.

5.4. Timescales of sediment transfer and storage in the catchment

The stages of sediment evolution can be recognised as: (1) the soil profile residence time, (2) the lateral (colluvial) transport time, (3) transport time through the river and intermediate depositions and (4) time since the final deposition (Table 1). Suresh et al. (2013), using U-series isotopes, determined soil residence times of ~30 kyr on ridge tops in the upper catchment area of the Murrumbidgee River. Based on cosmogenic nuclide estimates of soil production rates (Heimsath et al), Yoo et al. (2007) modelled lateral residence time (Table 1) of soil at the same locality, using soil and saprolite geochemistry, to derive a lateral transport time of 60 kyr for a soil column of 1 m^2 base area to be transported 50 m downslope. The turnover times of sediments during river transport and alluvial deposits are discussed below.

The results presented here suggest that there is relatively rapid transfer of sediment through the channel system from the upper catchment to the lower catchment, including time spent in alluvial storage, with no detectable aging of sediments down the river (although weathering ages do carry very large uncertainties). Alluvial deposits occur along the upper Murrumbidgee (Fig. 10) and were sampled but showed weathering ages in the same range as the modern river sediments and with no downstream age trend, suggesting that alluvial storage in the upper catchment is of short duration. This can be tested by calculating the time required to fill the alluvial pockets in the upper catchment area. The total area of the sub-catchment contributing sediments to the Murrumbidgee River upstream of Burrinjuck Dam is 9673 km^2 (Verstraetan et al., 2007). Using the range of catchment denudation rates of 9 to 24 mm kyr^{-1} (Fujioka et al., 2012) and the area available for erosion, the volume of sediments exported by the river from the upper catchment area per year can be estimated to be 8.7×10^{-5} to 2.3×10^{-4} km^3yr^{-1}. The width of alluvial deposits in the upper catchment area has been estimated using Google Earth (Fig. 10). The total volume of sediment stored in these alluvial pockets can be calculated by taking the average width (500 m), thickness (considered to be 4 m, from the observed thicknesses of the two alluvial deposits sampled and the depth of the channel) and total length of the alluvial pockets (50 km, estimated from Google Earth), yielding a value of 0.1 km^3. Thus it can be inferred that the time required to fill the alluvial pockets is < 1 kyr (assuming 100% sediment trap efficiency), which implies that reworking of these alluvial sediments cannot account for the weathering ages over 300-500 kyr observed in modern river sediments. AMS radiocarbon measurements show that the upper limit of deposition age of floodplain deposits at Wangrah Creek is 12.4 ± 0.15 kyr (Prosser et al., 1994), with several substantial sediment flushing and filling episodes in the Holocene. They also reported the existence of remnant slope deposits older than 30 kyr in the Wangrah Creek catchment. This further implies that reworking of old alluvial deposits cannot account for residence timescales over 300-500 kyr for modern sediments.

Since the time spent by the sediments in the weathering profile, alluvial deposits and in transport by the river cannot account for the long residence time of sediments; it can be inferred that the ageing of sediment is occurring during hillslope transport. As mentioned above, Yoo et al. (2007) reported that colluvial transport time is 60 kyr through a 50 m downhill transect. The weathering ages of the two colluvial samples in this study are 48 ± 11 and 57± 13 kyr and the weathering age for sediment in the Bredbo River adjacent to the study locality of Yoo et al. (2007) at Frog's Hollow is 77 kyr, supporting this general timeframe.

To test whether hillslope transport could account for sediment weathering ages of hundreds of thousands of years, we determined the distribution of slope lengths in the Murrumbidgee catchment. Using 1s DEM data and ArcGIS the calculated median slope length in the upper Murrumbidgee catchment was estimated to be 265 ± 128 m. Assuming a linear relationship between slope length and sediment transport time and Yoo et al.'s (2007) rates, it can be estimated that the sediments spend ~ 220 ± 106 kyr residing on the hillslope prior to reaching the river channel. The assumption of a linear relationship between slope length and sediment transport time is justifiable as the universal soil loss equation model (USLE) considers that hillslope sediment delivery is linearly related to the slope length and steepness (Gallant, 2001; Lu et al., 2006; Verstraeten et al., 2007). On the basis of the calculated transport time, it can be concluded that the lateral residence time driven by transport through hillslope most likely accounts for the long residence time of modern sediments in the Murrumbidgee River, whereas the vertical soil residence time driven by soil production from the saprolite and fluvial transport time by the river account for a smaller proportion.

A shorter comminution age (42 ± 7 kyr) for a post-LGM palaeochannels of the lower Murrumbidgee River has been reported by Dosseto et al. (2010) in contrast to residence times over 300 kyr for modern sediments and over 100 kyr for pre-LGM palaeochannels sediments. They proposed that reworking of old (high residence time) alluvial deposits in the middle and lower valley was the dominant sediment source pre- and post-LGM, and younger (low residence time) hillslope soil or sediment from the upper catchment was the sediment source during the LGM. Faster downslope transport of sediments from the catchment area during the LGM is plausible, as the vegetation cover during LGM in the area was herb and grass-dominated (Singh and Geissler, 1985) and hence could have promoted erosion (Dosseto et al., 2010 and references therein). Nevertheless, the residence time of more than 300 kyr for the post LGM sediment deposits at Wangrah Creek suggests that such an effect must have been quite limited. Furthermore, extensive erosion of the hillslopes would remove very old soil material and therefore remove the source of very old sediment deposited in the upper catchment alluvium during the Holocene and transported today. Our results constrain the sources of young LGM sediment to fresh bedrock erosion (e.g. channel bed) or ridgetop soils, rather than colluvial soils or alluvium within the upper catchment.

5.5. Assessing the potential contribution of aeolian material

Holocene dust deposits from the Snowy Mountains in the upper catchment of the Murrumbidgee River have been analysed by Marx et al. (2011). Using the trace element data of the dust samples, they showed that the source of dust is the Murray-Darling basin and that all the dust samples have U/Th ratios > 1. The late Pleistocene dust sample analysed here has a U/Th ratio < 1, which may point towards a different source of dust prior to the Holocene. The sample preparation procedure followed by Marx et al. (2011) did not include removal of exchangeable phases, which may have an effect on U/Th ratios. The possibility of aeolian dust contribution to the Murrumbidgee sediments was tested following the binary mixing models suggested by Albarede (1995) for concentrations and ratios. No relationships were deducible from the model using U-series concentrations or activity ratios of the sediments and the dust, when considering the U and Th data of soil from Frogs Hollow (reported by

Suresh et al., 2013) or of the most downstream sediment sample or of the colluvial samples as the other end member for mixing. Correlations were absent when subsets of U – Th data (tributary-trunk stream, colluvial, alluvial or modern sediments) were considered in the binary mixing model. The non-uniform spread in the U and Th concentrations of the sediments with some of the values less than that of the dust (as shown in Fig. 8) also indicate that no significant mixing of sediments with dust is occurring, but more samples are required to thoroughly understand the potential dust contribution.

5.6. Broader Implications

A consolidated study of the evolution timescales of all the compartments of sediments in a single catchment (colluvial, alluvial and modern channel sediments) is reported for the first time here. The results have local as well as global implications. The residence timescales of the Murrumbidgee sediments discussed here modify the current understanding of influence of LGM on sediment transport by rivers in temperate Australia. Dosseto et al. (2010) concluded that during the LGM, fresh sediments were loaded to the Murrumbidgee River due to high hillslope connectivity. Their argument of reworking of alluvial deposits being the source of modern sediments of very long residence time after the LGM envisages large alluvial deposits in the upper catchment area, which are absent. The long residence times of alluvial and colluvial deposits in the upper catchment area reported here necessitate alternate explanations for the arguments of Dosseto et al. (2010). The young residence times of post-LGM sediments from the palaeochannels of the Murrumbidgee River could only be explained by proposing that they are either produced by bedrock incision or sourced from the ridgetops containing young soil. Prosser et al. (1994) suggested that the hillslopes were not well connected with the river channel during the LGM. This suggestion supports inferred long residence times of sediments, as this will correspond to aging of sediments in the hillslopes.

Long residence times of river sediments may correspond to slow erosion in the catchment (Dosseto et al., 2008a). Tectonic and climate regimes are known to affect denudation rates (von Blackenburg, 2006; Portenga and Bierman, 2011). The globally averaged erosion rate on catchments in tectonically active areas is over an order of magnitude higher than that for catchments in tectonically inactive areas (Portenga and Bierman, 2011).

Sediment residence times of 1-8 kyr have been reported for tectonically active catchments in Iceland (Vigier et al., 2006). Granet et al. (2007) reported ~ 1 kyr transfer time of sediments by the river Ganga draining the tectonically active upper Himalayan region. The long residence times (~320 kyr) of sediments in the Murrumbidgee River reported here may reflect the stable tectonic conditions of the catchment. Topography plays a major role in weathering and erosion. A global compilation of the slope and the relief data of drainage basins showed positive correlation with erosion rates (Portenga and Bierman, 2011). The relationship observed between the estimated slopelength and the large lateral residence time in the Murrumbidgee catchment supports their suggestion (provided the large lateral residence times of sediments correspond to a slow erosion rate).

6. Conclusions

1. The mineralogical, elemental and U-series isotopic characteristics of sediments carried by the Murrumbidgee River in the south-eastern Australia were determined. The mineralogy of the sediments is consistent with the granitic lithology of the catchment.
2. The Si-based Weathering Index does not vary systematically downstream, suggesting insignificant chemical weathering of sediments during transport in the Murrumbidgee River.
3. The concentrations and activity ratios of U-series isotopes of the river sediments do not show evidence of downstream evolution. Rapid exchange of U-series isotopes between water and sediments to reach a chemical equilibrium may be occurring. Alternatively, the lack of systematic downstream trends in geochemistry could imply rapid sediment transport by the river system.
4. Muscovite content in the sediments shows a positive correlation with U concentration. Muscovite content plays a major role in controlling U-series isotopes in sediments. A similar observation was reported for soils in the upper catchment of the Murrumbidgee River (Suresh et al., 2013).
5. Long lateral residence time of colluvial soil is inferred (~220 kyr). Soil residence time driven by hillslope transport and average slope length of the catchment indicates that of the total residence time in the catchment, sediments spend ~220 kyr in hillslope transport. This could further imply

slow erosion, consistent with the average erosion rate of 9 – 24 mm/kyr estimated using cosmogenic radionuclides (Fujioka et al., 2012).

6. The observation of long residence times (~400 kyr) of post-LGM deposits at Wangrah Creek in the Murrumbidgee catchment contrasts with the young residence times of post-LGM palaeochannel deposits reported by Dosseto et al. (2010). The proposal of Dosseto et al. (2010) that erosion of young soil from ridge top as the source of LGM sediments need to be revised. The sources of young sediments during LGM in the catchment could be bedrock incision or fresh bedrock weathering on the ridges.
7. Long weathering ages (> 320 kyr) are observed for the alluvial deposits and modern sediments in the catchment, except for the modern sediment sample collected from the Bredbo River at Frogs Hollow (77 kyr). The colluvial deposit samples also have an order of magnitude younger weathering ages (~50 kyr). Longer residence times of sediments in the catchment could correspond to the stable landscape, which has been relatively unaffected by tectonic activity or climatic changes.

Acknowledgements:

We are grateful to Norman Pearson, Peter Wieland and Russell Field for assistance during sample preparation and analysis. POS acknowledges an iMQRES scholarship supporting this research. AD acknowledges an ARC Future Fellowship FT0990447. HH acknowledges an ARC Linkage grant LP0990500 and an ARC Future Fellowship FT120100400. The analytical data were obtained using instrumentation funded by DEST Systematic Infrastructure Grants, ARC LIEF, NCRIS, industry partners and Macquarie University.

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Tables

Table 1. Definition of timescale terminology used.

Name	Definition	Determined by
Comminution age	Time since the production of fine grains (<50 µm) by bedrock weathering	^{238}U-^{234}U disequilibrium
Weathering age	Time since the onset of chemical weathering of bedrock to produce regolith.	^{238}U-^{234}U-^{230}Th disequilibrium
Soil residence time	Time since the conversion of saprolite into soil.	Weathering age of topsoil
Lateral residence time	Time spent by the soil on the hillslope.	1. ^{10}Be-derived soil production function and geochemical mass balance model (Heimsath et al., 2000). 2. Footslope colluvial weathering age
Sediment residence time	Time spent by the sediment grains from formation by bedrock weathering until final deposition.	Comminution age or weathering age minus deposition age (if applicable)

Table 2. Sampling localities and U-series data of leached sediment samples from the Murrumbidgee River and three of its tributaries.

Sample name	Place	Channel Length (km)¥	Sample Depth (m)	Th (ppm) £*	U (ppm) £*	(^{234}U/^{238}U)£	(^{230}Th/^{234}U)£
Alluvium							
MU8_low	Wangrah Creek	-40	3.6	16.62±0.03	3.82±0.03	1.21±0.01	0.67±0.02
MU8_up	Wangrah Creek	-40	1.4	21.70±0.04	6.33±0.05	1.35±0.02	0.62±0.02
MU3	Bredbo	0	4.1	10.69±0.02	1.77±0.01	1.02±0.01	1.07±0.03
MU18	Bundarbo	240	0.4	13.81±0.02	2.83±0.02	1.12±0.01	0.86±0.03
MU19_low	Gundagai	360	7.7	21.78±0.03	5.59±0.04	1.26±0.02	0.64±0.02
MU19_mid	Gundagai	360	3.6	18.46±0.03	3.12±0.02	1.08±0.01	0.82±0.02
MU19_up	Gundagai	360	0.25	12.49±0.02	2.36±0.02	1.09±0.01	0.84±0.03
MU22	Darlington Point	780	3	14.91±0.03	2.71±0.02	1.05±0.01	1.09±0.03
Colluvium							
MU6_low	Bredbo Gully	-1	1.7	16.61±0.03	2.61±0.02	1.27±0.02	1.11±0.03
MU6_up	Bredbo Gully	-1	0.4	16.95±0.03	2.63±0.02	1.06±0.01	1.09±0.03
Modern							
MU11	Frogs Hollow	-40	0	13.92±0.02	1.89±0.02	1.18±0.01	1.17±0.04
MU4	Bredbo	-1	0	15.13±0.03	2.92±0.02	1.12±0.01	0.73±0.02
MU5	Bredbo	-1	0	12.89±0.02	2.68±0.02	1.25±0.02	0.70±0.02
MU1	Bredbo	0	0	13.54±0.02	3.12±0.02	1.16±0.01	0.69±0.02
MU12	Taemas Bridge	160	0	9.65±0.02	2.16±0.02	1.19±0.01	0.74±0.02
MU15	Brindabella Valley	160	0	12.50±0.02	3.11±0.02	1.26±0.02	0.64±0.02
MU17	Bundarbo	240	0	17.79±0.03	4.42±0.04	1.19±0.01	0.75±0.02
MU20	Gundagai	360	0	12.52±0.02	4.23±0.03	1.22±0.01	0.49±0.02
MU16	Brungle	360	0	18.17±0.03	3.66±0.03	1.15±0.01	0.73±0.02
MU21	Darlington Point	780	0	10.73±0.02	2.19±0.02	1.09±.0.01	0.71±0.02
Dust							
MU24	Carcoar		1.5	11.61±0.02	1.70±0.01	1±0.01	1.24±0.03
Gravimetric standard							
TML-3 (n=2)				29.62±0.44	10.469±0.118	0.994±0.003	1.004±0.006

¥ Negative values indicate upstream Bredbo. * Concentrations are determined by isotope dilution. £ Reported with 2σ external errors

Table 3. Mineralogy, WIS and granulometric mud fraction data of the sediment samples from the Murrumbidgee River catchment.

Sample name	Quartz (%)	Albite (%)	Microcline (%)	Muscovite (%)	Illite (%)	WIS (%)[a]	Mud fraction (%)[b]
Alluvium							
MU8_low	71.2	6.4	0	22.4	0	83.4	37.3
MU8_up	45.9	0	0	54.1	0	72.3	74.4
MU3	72.7	19	0	8.2	0	86.5	2.2
MU18	78.4	14.3	4.1	3.2	0	84.4	37.3
MU19_low	62.8	12.9	3.9	20.5	0	80.4	78.3
MU19_mid	78.1	17.3	0	4.7	0	84.3	43.4
MU19_up	67.3	19.1	10.4	3.2	0	85.4	20.3
MU22	72.8	8	3.3	14.3	1.6	83.2	37.5
Colluvium							
MU6_low	61.1	0	0	39	0	78.0	32.5
MU6_up	61.9	6.6	0	31.6	0	83.0	30.4
Modern							
MU11	57.7	22.3	18.4	1.6	0	84.4	1.9
MU4	71.8	11.5	0	5.4	11.3	82.8	38.1
MU5	73.8	10.6	0	6.4	9.2	85.4	46.0
MU1	68.9	14.1	0	16.9	0	82.8	57.5
MU12	72.7	14	8.2	1.8	3.3	86.3	25.1
MU15	70.2	9.9	6.6	10.1	3.3	83.5	40.9
MU17	60.3	11.6	8.9	2	14	80.0	96.4
MU20	76.1	9.1	1.5	11.8	1.5	84.4	46.8
MU16	69.6	10.9	4.9	9.8	5.2	80.0	38.0
MU21	84.6	8.1	2.1	3.2	2	87.6	30.9
Dust							
MU24	100					92.4	

[a] From major element data see Table S1. [b] From particle size distribution measurement

Table 4. Modelled leaching and gain coefficients for U and Th isotopes and weathering ages.

	w_{238}	w_{234}	w_{230}	Γ_{238}	Γ_{234}	Γ_{230}
Alluvium	0.742±0.007	0.771±0.023	9.42±8.69	1.58±0.91	1.96±1.94	4.02±3.85
Colluvium	0.746±0.001	0.752±0.007	*3.6±0.04	*1.0±0.01	*0.7±0.4	*1.3±0.01
Modern	0.732±0.004	0.729±0.004	17.7±1.4	3.17±1.58	3.88±1.93	6.72±3.33

Sample name	**T_w (kyr)[$]**
Alluvium	
MU8_low	395±166
MU8_up	438±177
MU3	313±142
MU18	370±163
MU19_low	413±171
MU19_mid	334±146
MU19_up	355±155
MU22	339±154
Colluvium	
MU6_low	57±13
MU6_up	48±11
Modern	
MU11	77±31
MU4	369±168
MU5	397±180
MU1	402±181
MU12	403±187
MU15	418±189
MU17	419±189
MU20	451±191
MU16	381±173
MU21	371±169

All the coefficients are in 10^{-5} yr^{-1}
*Taken from Suresh et al. (2013), reported for the soil profile from Frogs Hollow.
The model errors reported are 2σ

Figure Captions

Fig. 1. The Murrumbidgee catchment area map (DEM data from Geoscience Australia). BR: Bredbo River. GR: Goodradigbee River, TR: Tumut River. MR: Murrumbidgee River. WC: Wangraha Creek. FH: Frogs Hollow. Locations of all the samples used in this study are shown. The numbers marking sample locations correspond to their MU numbers in Table 2. Suffix to the sample number reefers to the type of the sample (m: modern, a: alluvial deposit, c: colluvial deposit, d: dust deposit)

Fig. 2. Downstream variation of quartz and albite in the modern sediments from the Murrumbidgee River.

Fig. 3. U-series activity ratios of the sediment and dust samples. The error bars represent external analytical errors.

Fig. 4. Downstream variation of Si-based Weathering Index for modern, alluvial and colluvial sediments from the Murrumbidgee catchment.

Fig. 5. Average major element oxide concentrations of sediments normalized to those of the upper continental crust concentrations from McLennan (1995) and to those of the Cooma Graodiorite (Chappell 1984). A value < 1 indicates elemental mobilization, and a value > 1 indicates elemental gain.

Fig. 6. Variation of concentration of U and Th with mud content in the modern, colluvial and alluvial sediments. External analytical errors are smaller than the symbol size.

Fig. 7. Variation of concentration of U and Th with muscovite content in the modern sediment samples. External analytical errors are smaller than the symbol size.

Fig. 8. Variation of concentration of U and Th with WIS for the river modern, colluvial and alluvial sediments and dust samples. Soil data from Frogs Hollow in the upper catchment (Suresh et al., 2013) are also shown. External analytical errors are smaller than the symbol size.

Fig. 9. Weathering ages (T_w) of sediments from the Murrumbidgee River along the length of the stream. The error associated with T_w is the 1σ standard deviation

Fig. 10. Width of alluvial deposit pockets in the upper catchment area (upstream of Gundagai) of the Murrumbidgee River and two of its tributaries.

APPENDIX

Table S1. Major element data of all the samples from the Murrumbidgee catchment area.

Sample name	SiO_2 (wt. %)	TiO_2 (wt. %)	Al_2O_3 (wt. %)	Fe_2O_3 (wt. %)	Mn_3O_4 (wt. %)	MgO (wt. %)	CaO (wt. %)	Na_2O (wt. %)	K_2O (wt. %)	P_2O_5 (wt. %)	Cr_2O_3 (wt. %)	ZrO_2 (wt. %)	SrO (wt. %)	ZnO (wt. %)	NiO (wt. %)	BaO (wt. %)
Alluvium																
MU8_low	68.7	0.7	12.62	9.13	0.05	1.25	0.3	0.8	2.67	0.3	0.01	0.03	0.01	0.01	0.01	0.03
MU8_up	57.42	0.89	20.51	8.36	0.09	1.93	0.7	0.8	3.5	0.2	0.02	0.03	0.02	0.02	0.01	0.06
MU3	78.22	0.44	9.94	2.84	0.05	0.8	1	1.3	2.61	0.1	0.01	0.03	0.02	0.01	BLD	0.03
MU18	75.71	0.7	11.36	3.15	0.08	0.88	0.9	1.7	2.65	0.1	0.01	0.05	0.02	0.02	0.01	0.04
MU19_low	69.13	0.87	14.91	5.14	0.15	1.3	0.8	1.2	2.62	0.2	0.03	0.04	0.02	0.01	0.03	0.05
MU19_mid	76.4	0.67	11.66	3.21	0.07	0.87	0.9	1.7	2.81	0.1	0.01	0.06	0.02	0.01	0.01	0.04
MU19_up	77.99	0.46	10.75	2.42	0.05	0.69	0.8	1.8	3.16	0.1	0.01	0.04	0.01	0.01	BLD	0.04
MU22	72.52	0.75	13.28	4.15	0.02	0.84	0.5	0.9	2.33	0	0.01	0.05	0.07	0.01	BLD	0.04
Colluvium																
MU6_low	64.11	0.64	17.42	6.63	0.15	1.21	0.4	0.3	3.15	0.1	0.02	0.03	0.01	0.03	0.01	0.04
MU6_up	71.2	0.73	13.5	5.51	0.19	1.45	0.4	0.7	2.99	0.1	0.02	0.03	0.01	0.01	0.01	0.03
Modern																
MU11	76.58	0.18	10.41	1.73	0.04	0.39	1.3	2.4	3.5	0	BLD*	0.01	0.01	BLD	BLD	0.03
MU4	71.62	0.86	12.41	4.8	0.09	1.41	1.3	1.2	2.18	0.1	0.02	0.05	0.02	0.01	0.01	0.04
MU5	75.82	0.52	10.96	3.89	0.32	0.9	0.9	1.1	2.58	0.1	0.01	0.03	0.01	0.01	0.01	0.03
MU1	71.56	0.72	12.81	4.62	0.2	1.3	1	1.1	2.63	0.1	0.01	0.03	0.02	0.01	0.01	0.04
MU12	78.43	0.42	10.37	2.59	0.06	0.66	0.8	1.3	2.71	0.1	BLD	0.02	0.01	0.1	0.01	0.03
MU15	72.32	0.71	11.79	3.43	0.12	0.93	1.5	1	2.93	0.2	0.01	0.04	0.02	0.01	0.01	0.04
MU17	68.25	0.95	14.57	4.47	0.17	1.09	1.2	1.3	3.33	0.2	0.01	0.03	0.02	0.01	0.01	0.05
MU20	73.98	0.73	11.1	3.87	0.16	1.09	1.1	1.5	2.34	0.1	0.03	0.04	0.02	0.01	0.01	0.03
MU16	67.65	0.86	14.27	5.6	0.4	1.6	1.5	1.2	2.3	0.2	0.03	0.06	0.02	0.01	0.02	0.04
MU21	79.04	0.53	9.5	2.4	0.04	0.58	0.6	1.1	2.44	0.1	0.01	0.05	0.01	BLD	BLD	0.02
Dust																
MU24	83.61	1.09	6.37	3.86	0.08	0.2	0.1	0.3	0.9	0	0.01	0.08	0.01	BLD	BLD	0.01

*BLD stands for below the limit of detection

Figure 1

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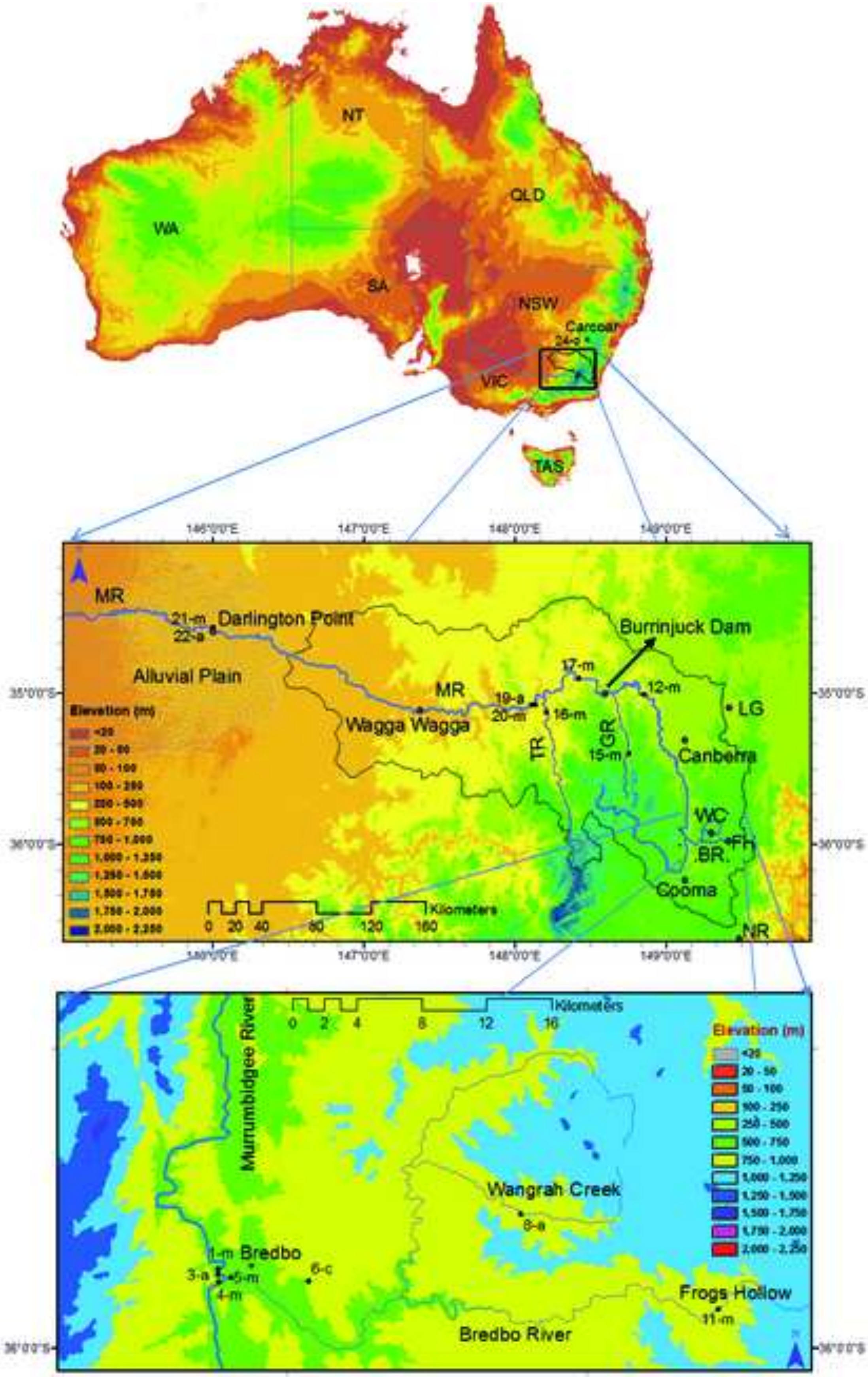

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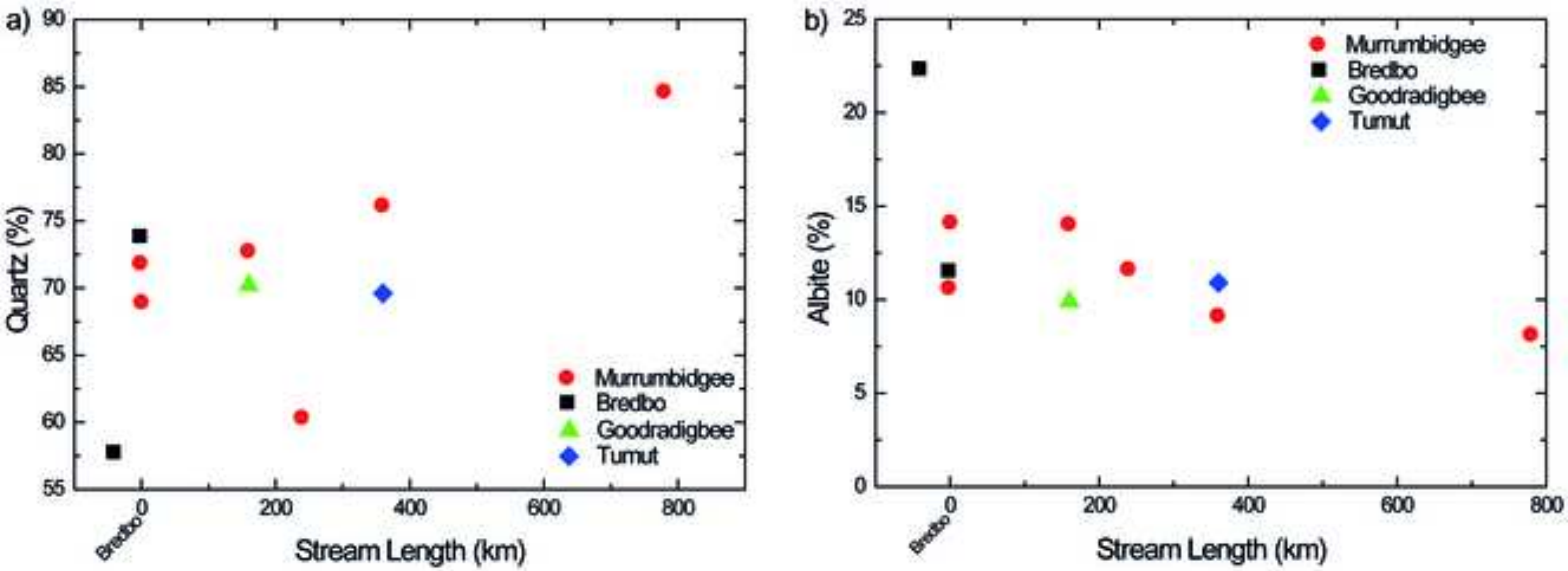

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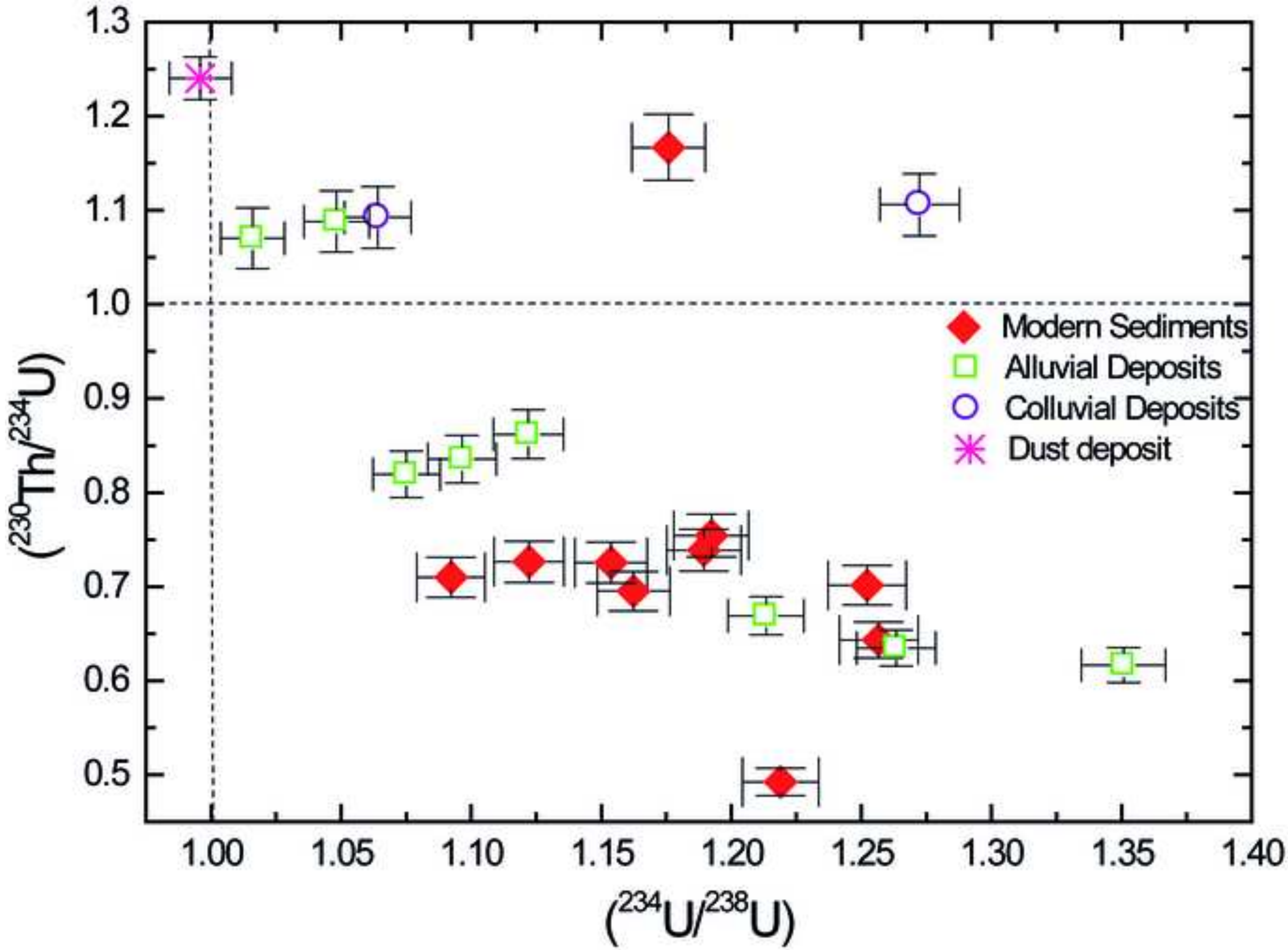

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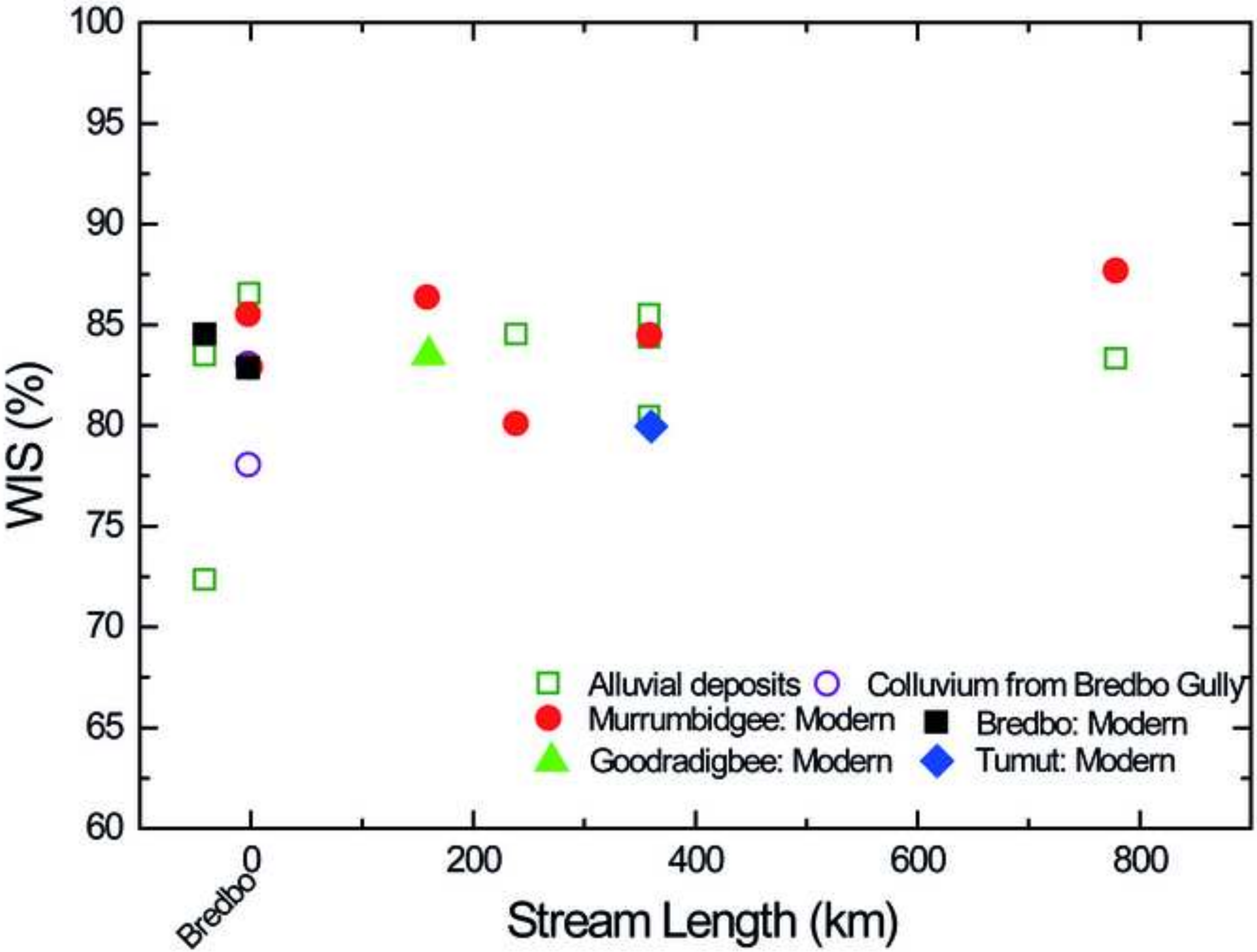

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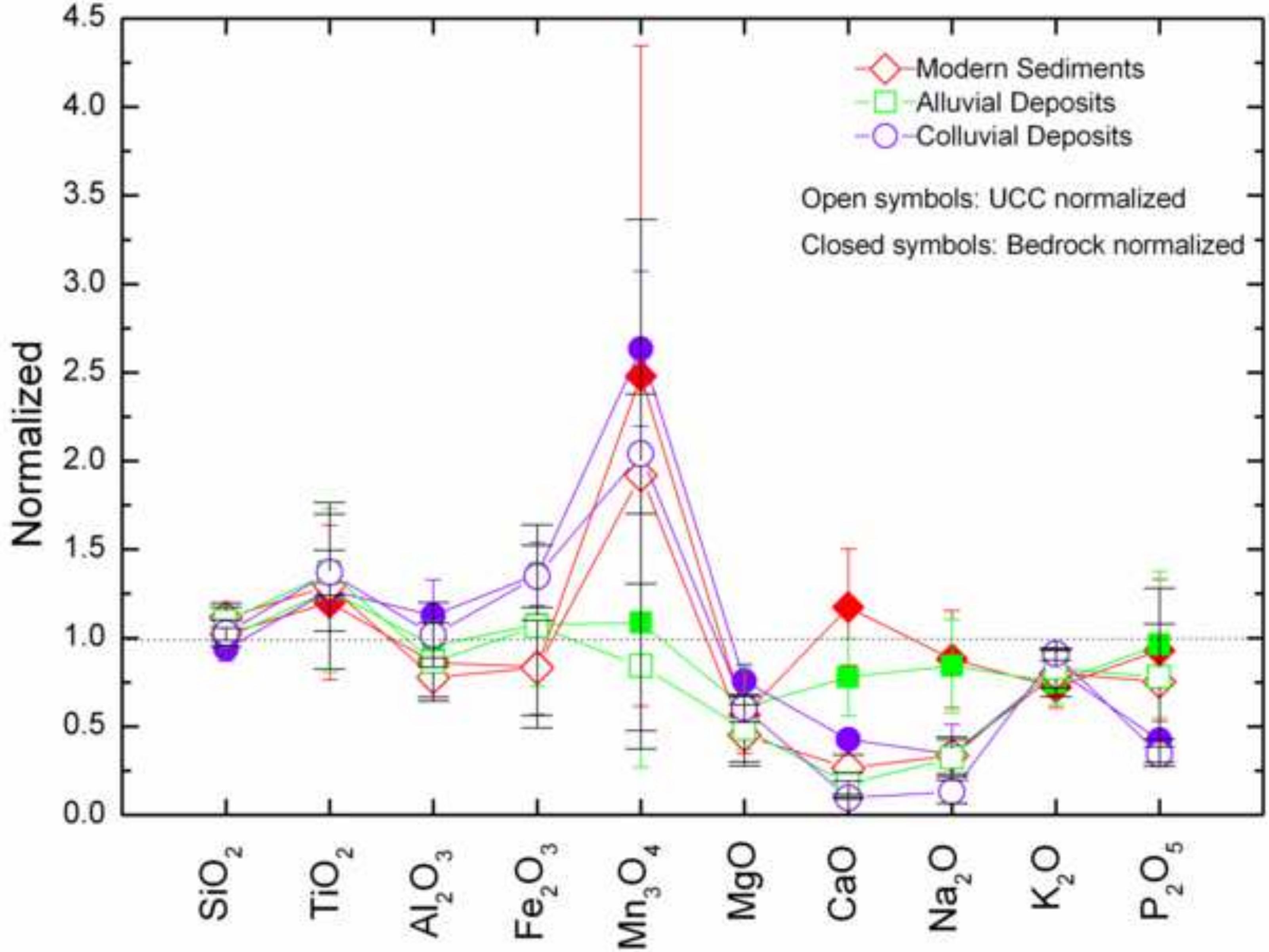

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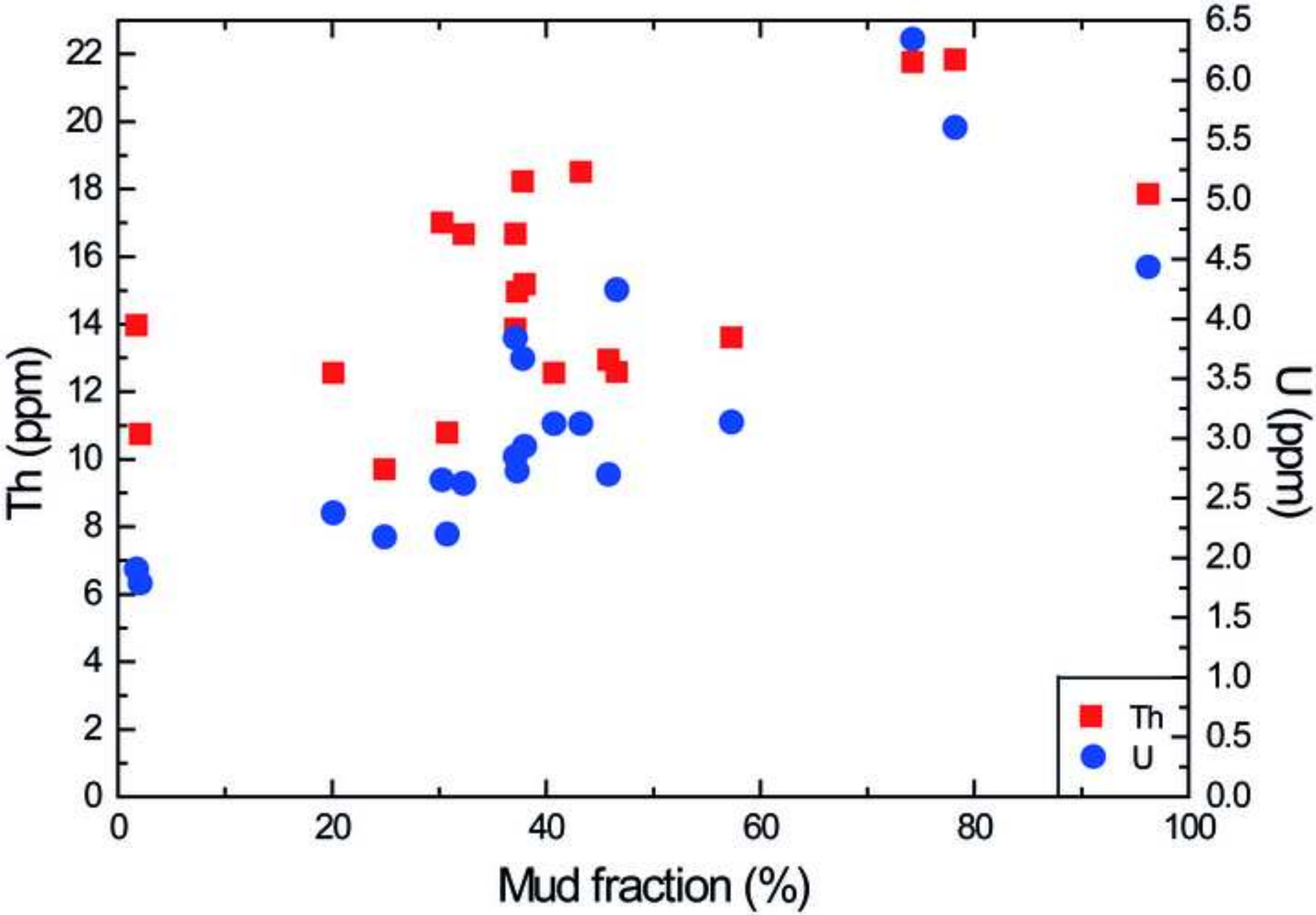

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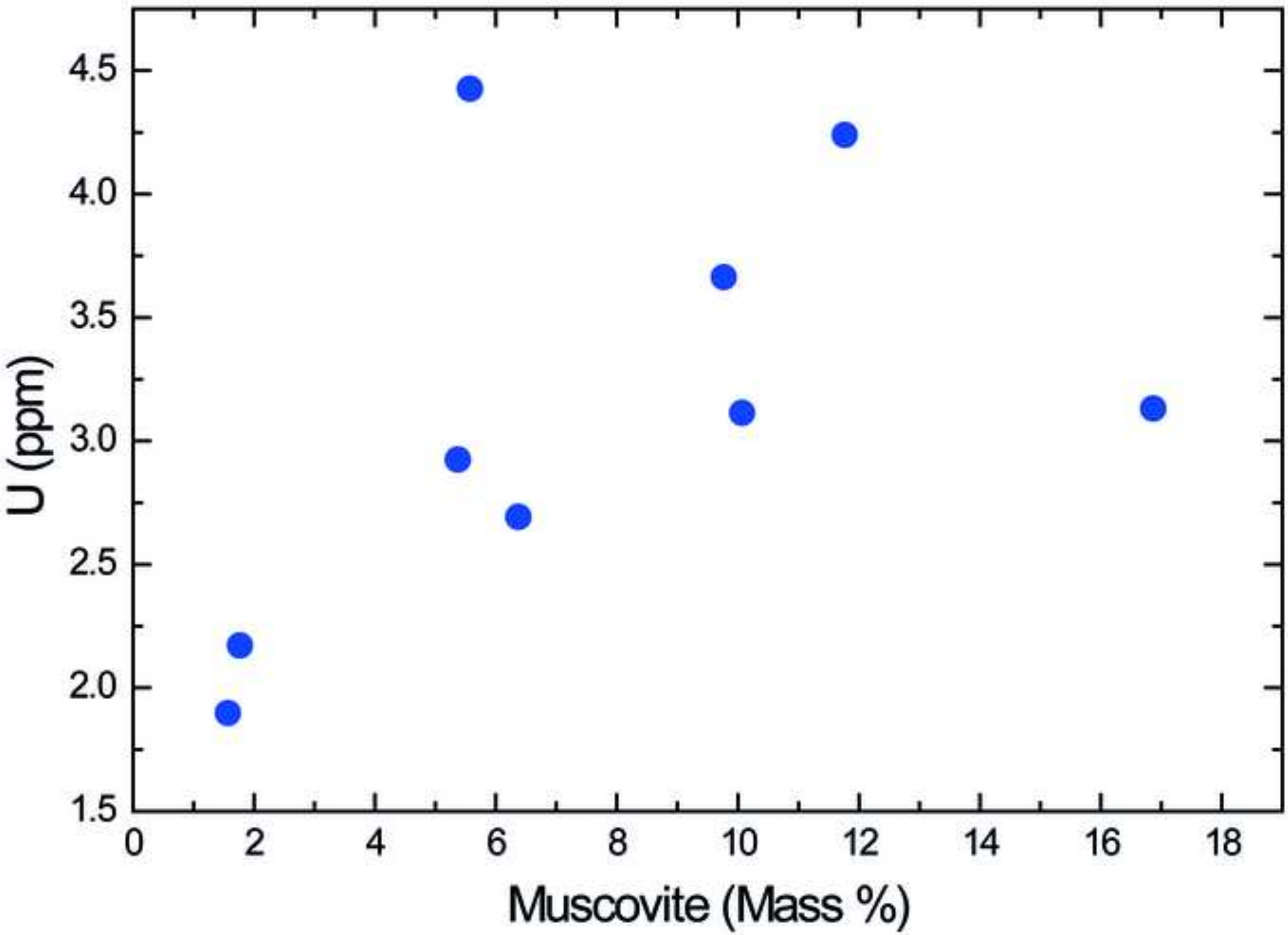

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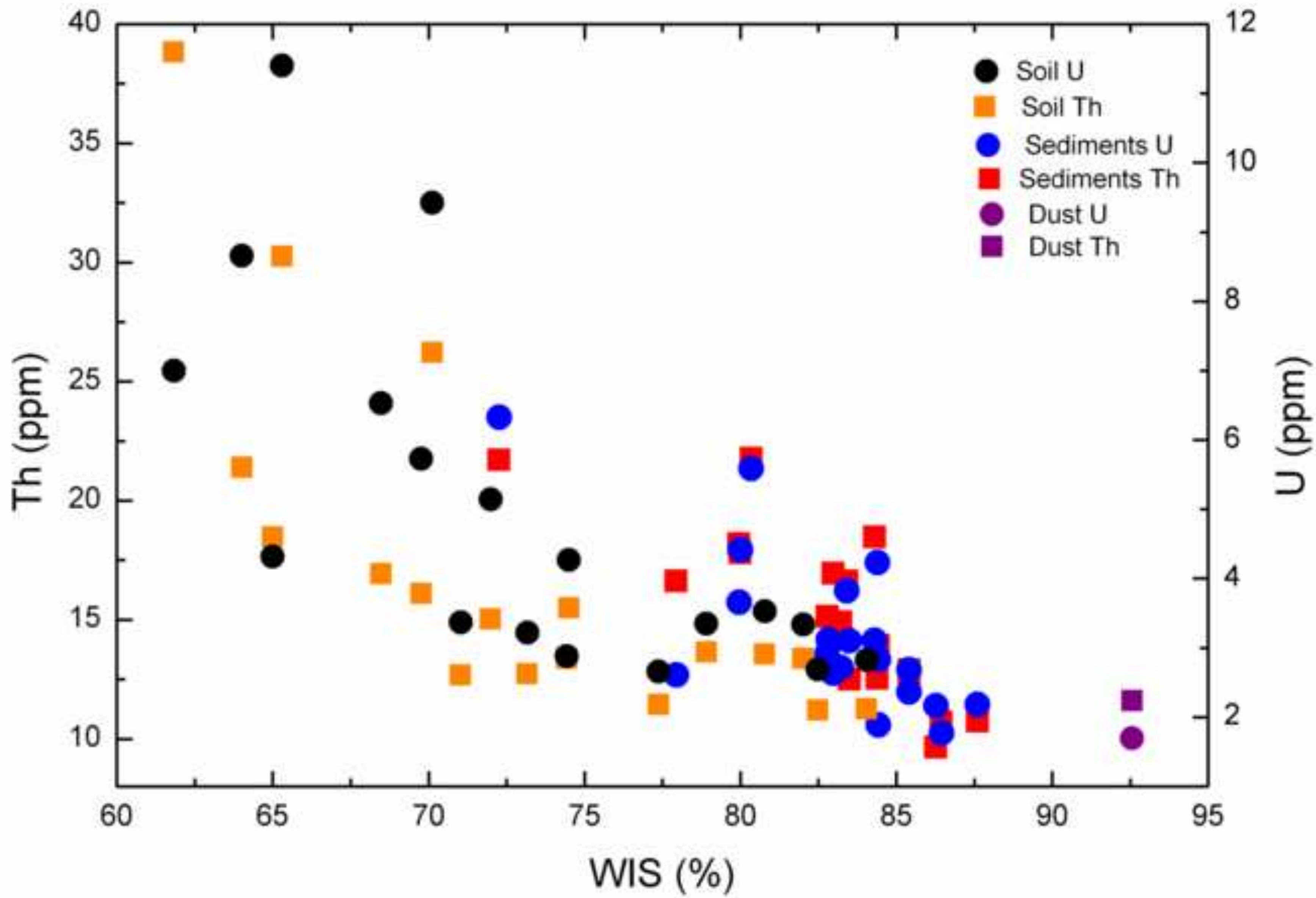

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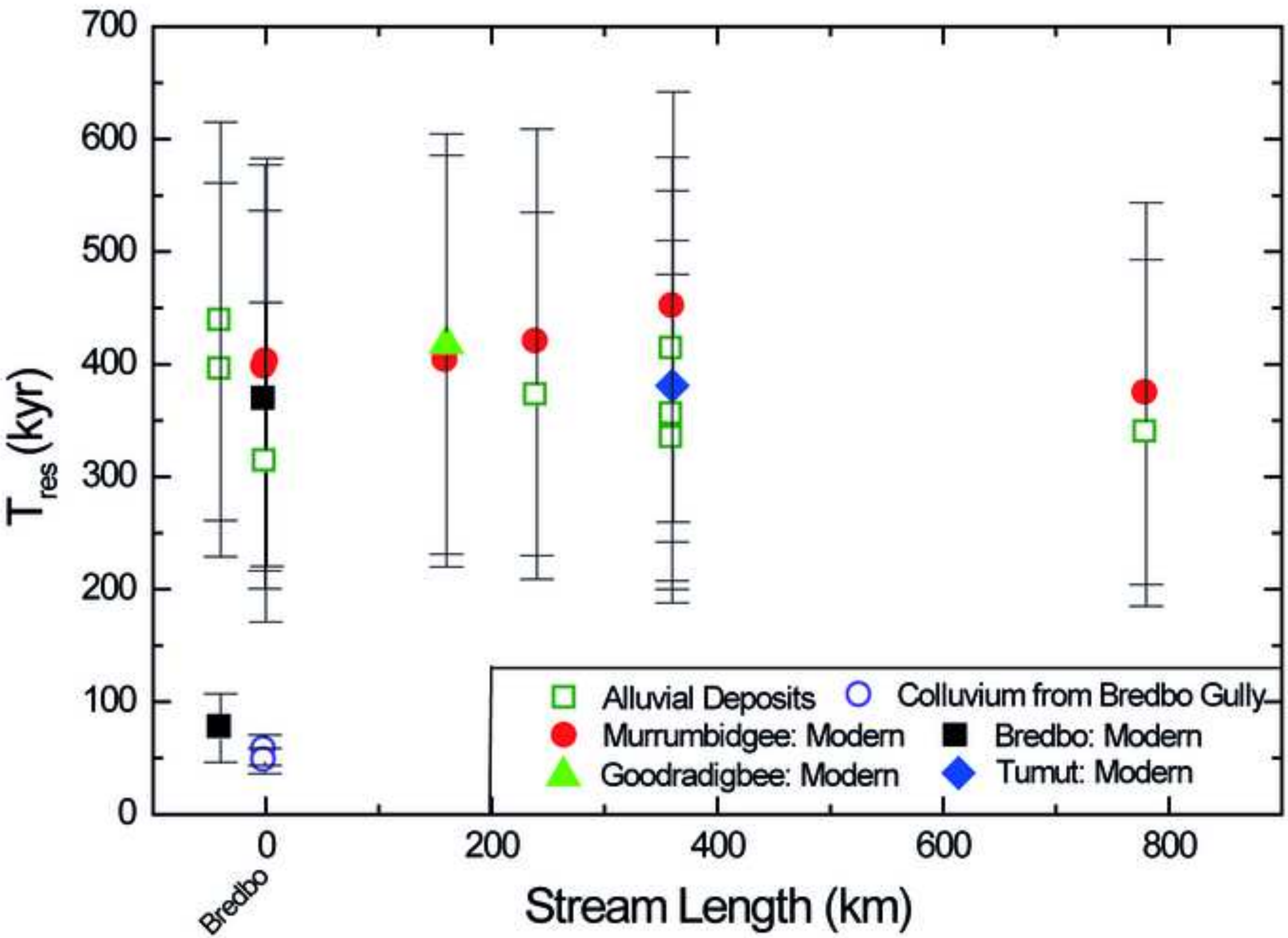

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