Origins of variability in the ^{230}Th/^{232}Th ratio in sediments

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Abstract Various geochemical tracers, including natural radioactivity, can be used to trace the source of sediment, both on hillslopes and in rivers. If the source of the sediment is to be correctly determined, we need to understand the nature of the material being traced and why the parameter being used to "fingerprint" the sediment varies. This paper examines the origins of variability in the ratio of ^{230}Th/^{232}Th in soils and sediments. It is shown that different particle-size and density fractions from soils formed from a uniform lithology have a constant ^{230}Th/^{232}Th ratio, the same ratio as that in the rock from which the soil was formed. Thus, fluvial transport of material derived from a uniform lithology will usually result in sediments with a uniform ^{230}Th/^{232}Th ratio, independent of particle size or grain density. The ^{230}Th/^{232}Th ratio can therefore be used to distinguish between sediments derived from different lithologies within a catchment, provided that the lithologies have distinctive ^{230}Th/^{232}Th ratios.

INTRODUCTION

Land degradation is a critical environmental issue, with significant implications for pastoral and forest production, water quality and storage, and agriculture. Tracing sediments and associated pollutants to their source offers a direct method of targeting conservation and reparation works. Sediment characteristics – such as clay mineralogy, sediment colour, mineral magnetic properties, lithogenic radionuclide ratios, and major element chemistry – have all been used to successfully identify sediment source areas (Woods, 1978; Grimshaw & Lewin, 1980; Walling & Kane, 1984; Caitcheon, 1993; Murray *et al.*, 1993; Olley *et al.*, 1993). However, no one tracer has proved to be universally applicable. If confidence is to be placed in the results of a sediment sourcing study then we need to understand how variability in the "signal" used to trace the sediment arises. This paper examines the causes of variability in the ^{230}Th/^{232}Th ratio in sediments.

Thorium-230 and ^{232}Th are naturally occurring lithogenic radionuclides in the ^{238}U and ^{232}Th decay chains respectively. Nuclides from these decay chains are distributed more or less uniformly through the regolith. Thorium-232 is the parent of the ^{232}Th decay series, whereas ^{230}Th is a daughter of the ^{238}U decay series. These nuclides are both long lived, with half-lives of 1.4×10^{10} years and 8.0×10^{4} years respectively. Thorium is highly particle-reactive (Moore, 1992), has low solubility, and is generally considered to be immobile in the surface environment.

Previously, observations of uniform $^{230}Th/^{232}Th$ ratios have been used to infer constancy in the types of accumulated detritus (Carpenter *et al.*, 1984; Short *et al.*, 1988). However, using variations in this ratio to locate the source of a material has only been done in uranium exploration (see Levinson *et al.*, 1982 for examples). This paper examines the mechanisms which cause variability in the $^{230}Th/^{232}Th$ ratio in modern sediments by examining this ratio in rocks and soils, and in soil particle-size and density fractions from a variety of rock types. These observations are then used to develop a model for the causes of variability in the $^{230}Th/^{232}Th$ ratio which can be applied to locating sources of sediment.

SAMPLING AND SAMPLE TREATMENT

Soil and rock samples were collected from five sites with different lithologies. Details of the sites and the number of samples analysed are summarized in Table 1, and the site locations are shown in Fig. 1. In each case the groups of soil samples were collected from above a known uniform lithology. The representative rock sample analysed from each site consisted of a number of fresh (unweathered) sub-samples which were combined to provide one averaged sample of the site lithology.

Fluvial transport results in sorting of soil material by particle size, and to a lesser extent by density (Krumbein & Sloss, 1963). In order to determine the effects of sorting on the $^{230}Th/^{232}Th$ ratio, sub-samples of soils from the five sites were separated by wet sieving into various size fractions (<63 μm, 63-125 μm, 125-250 μm, 250-500 μm, and >500 μm) and by heavy liquid separation (>2.95 g cm^{-3}) using sodium polytungstate (Callahan, 1987). Separation at this density produces the most marked contrast in mineralogy, so any effects of density sorting on the lithogenic radionuclides should be apparent in the fractions produced by this separation. The $^{230}Th/^{232}Th$ ratio was then determined for each particle size fraction and for the heavy mineral fraction (>2.95 g cm^{-3}).

Table 1 Sampling details for the eight sites.

Site	Lithology	Sampling area	No. of soil samples
Geebung Creek	granite	76.9 ha	18
McKeowns Valley	basalt	12 km^2	13
McKeowns Valley	mixed sediments	38 km^2	13
Black Mountain	sandstone	12 × 12 m	8
Whiteheads Creek	granite	2 soil profiles	8

ANALYTICAL METHODS

The concentrations of thorium isotopes ^{230}Th and ^{232}Th were determined by alpha-particle spectrometry using procedures described by Martin & Hancock (1992). The absolute concentrations of thorium isotopes in sediment and soil samples were

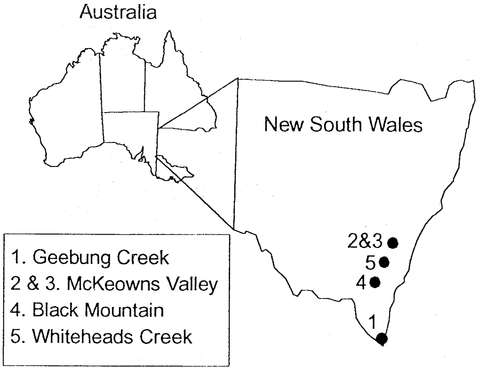

Fig. 1 Site location map.

determined either by using ^{229}Th tracer or from the ^{228}Th activity determined by gamma spectrometry (Murray *et al.*, 1987). The samples were normally counted for 48 h and recoveries were typically of the order of 70%.

RESULTS AND DISCUSSION

Parent rock and soil

The concentrations of ^{230}Th in the rock and soil samples from the sites are plotted against ^{232}Th concentrations in Fig. 2(a)-(e) and the rock and mean soil ^{230}Th/^{232}Th activity ratios for each of the sample groups are tabulated in Table 2. In Fig. 2(a), (b) and (c) the soil data are consistent with a single regression line passing through the origin, indicating these soils have uniform ^{230}Th/^{232}Th ratios. In Fig. 2(d) and (e) the data are clustered so no regression is possible. However, the ratios are still constant and a line has been fitted through the data and the origin. The above indicates that soils developed on a uniform rock type have uniform ^{230}Th/^{232}Th ratios. The ^{230}Th/^{232}Th ratios at each site for both soil and parent rock are clearly within analytical uncertainty of each other (Table 2), which also suggests that the lithology is the main factor controlling the ^{230}Th/^{232}Th ratios in soils.

The ^{230}Th/^{232}Th ratios in the soil samples range from 1.24 ± 0.03 for the samples from McKeowns Valley basalt to 0.532 ± 0.009 in samples from Whiteheads Creek granite. This suggests that whereas soils developed for a uniform rock type have a uniform ratio, soils developed on different rock types can have distinctive ^{230}Th/^{232}Th ratios.

THE EFFECTS OF FLUVIAL TRANSPORT

In order to compare the soil ^{230}Th/^{232}Th ratio to that in the sediments derived from them, it is necessary to examine the effects of sorting by particle size and density. The concentrations of ^{230}Th in various particle-size fractions are plotted against ^{232}Th concentrations in Fig. 3(a)-(e). In each case the data comes from particle-size fractions

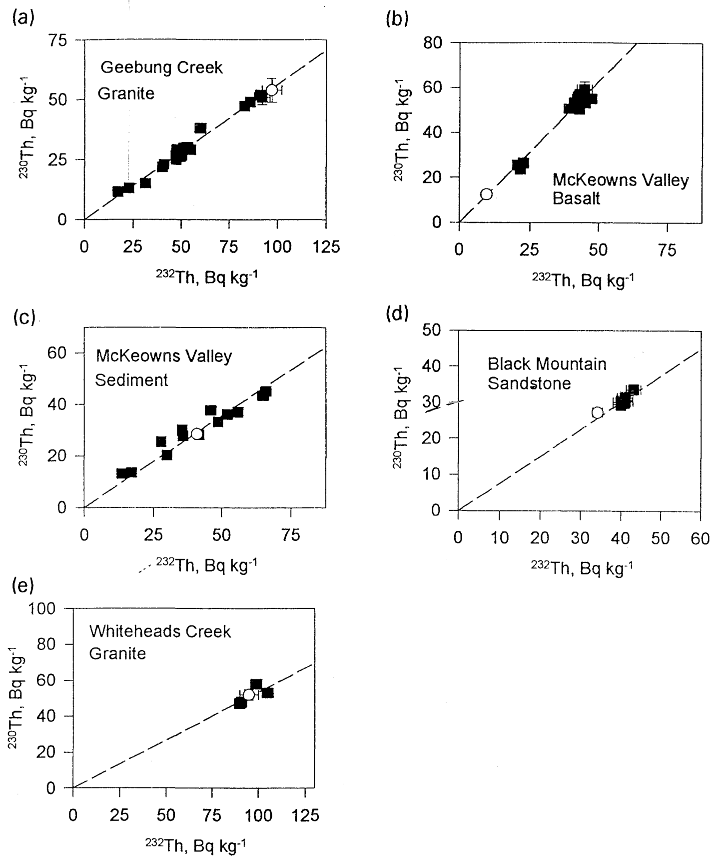

Fig. 2 Concentration of ^{230}Th against ^{232}Th in the rock (open circles) and soil samples (closed squares) from the five sites. Error bars represent uncertainties equivalent to one standard error on the mean (in most instances these are smaller than the symbol size). Regression lines (dashed line) are fitted through the soil data and the origin.

wet-sieved from the bulk soil samples. The mean bulk-soil ^{230}Th/^{232}Th ratio (from Table 2) is shown as a dashed line in each of the graphs. The data from the particle-size fractions are consistent with the bulk-soil ^{230}Th/^{232}Th ratios. The data tend to spread along the line in order of particle size, with the finer fractions having the highest

Table 2 The mean soil and parent rock ^{230}Th/^{232}Th ratios for each of the sample groups.

Site	Rock type	Soil	Rock	>2.95 g cm^{-3}
Geebung Creek	granite	0.571± 0.010	0.56 ± 0.06*	0.565 ± 0.009
McKeowns Valley	basalt	1.25± 0.02	1.30 ± 0.09	1.27 ± 0.05
	mixed sediments	0.75 ± 0.03	0.70 ± 0.03	0.75 ± 0.06
Black Mountain	sandstone	0.744 ± 0.003	0.80 ± 0.04	0.77 ± 0.05
Whiteheads Creek	granite	0.532 ± 0.009	0.55 ± 0.04	0.56 ± 0.03

* Calculated from XRF determination of U and Th in ppm and converted to Bq kg^{-1}.

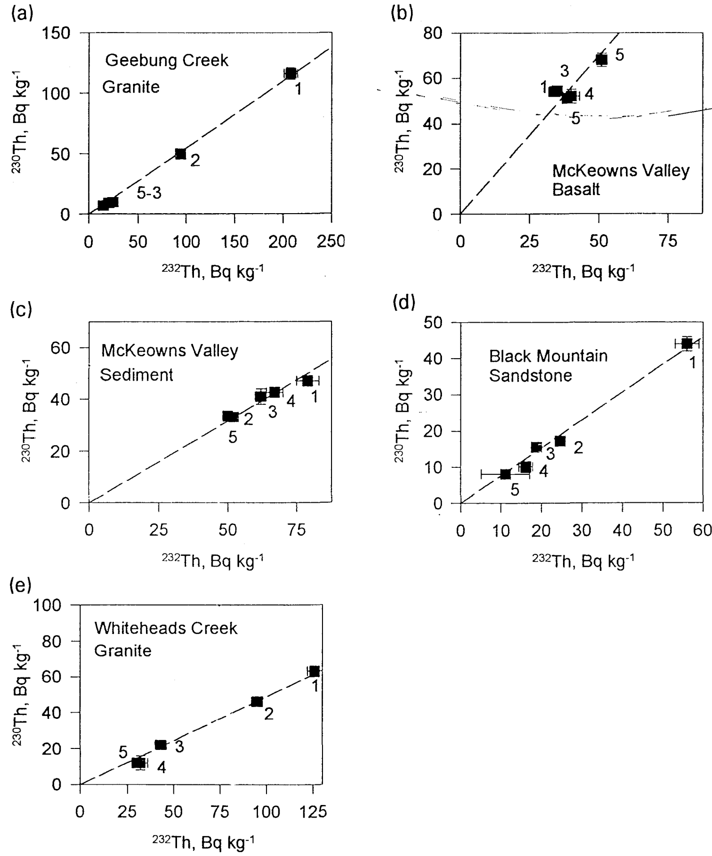

Fig. 3 Concentration of ^{230}Th against ^{232}Th in the various particle size fractions from each of the five sites. The numbers in each figure indicate the various size fractions in order of increasing particle size. Error bars as for Fig. 2. Lines shown represent the mean soil ^{230}Th/^{232}Th ratios from Table 2.

concentrations. Scott (1968) and Megumi *et al.* (1982) observed similar increases in various lithogenic radionuclide concentrations as particle size decreased. These data indicate that whereas the absolute concentrations of the thorium isotopes may vary as a function of particle size, the ^{230}Th/^{232}Th ratio remains constant and within analytical uncertainty of the ratio in bulk soil.

In order to investigate the effects of sorting by density, sub-samples of the bulk soils used for the particle-size study were separated at a density of 2.95 g cm^{-3}. In this way the primary and secondary heavy minerals were separated from quartz, feldspar, and clays. The heavy mineral fraction (>2.95 g cm^{-3}) was then analysed and the ^{230}Th/^{232}Th ratios are given in Table 2. In each case the ^{230}Th/^{232}Th ratio of the heavy mineral fraction is within analytical uncertainty of that in bulk soil. The lighter fraction (<2.95 g cm^{-3}) must therefore also have the same ratio as the bulk soil. Therefore, sorting of soil by density produces fractions that have the same ^{230}Th/^{232}Th ratio as the bulk soil.

CONCLUSIONS

From the above experiments it has been shown that sorting soil material either by particle size or density will produce fractions with the same $^{230}Th/^{232}Th$ ratio as the bulk soil, and this ratio in the bulk soil is the same as that in the parent rock. We therefore conclude that the variability of the $^{230}Th/^{232}Th$ ratio in sediments is probably dominated by the $^{230}Th/^{232}Th$ ratio in the parent rock from which the sediments are ultimately derived, and that this ratio is unlikely to be affected by fluvial sorting processes. This finding, combined with the site-to-site variability in the ratio (1.24 ± 0.03 to 0.532 ± 0.009), strongly suggests that the $^{230}Th/^{232}Th$ ratio can be used to distinguish sediments derived from different rock types if the latter have distinctive $^{230}Th/^{232}Th$ ratios.

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