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Estimating residence times of fine sediment in river channels using fallout ^{210}Pb

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Abstract The residence time of fine-grained sediment (<10 μm diameter) within river channels is often assumed to be short, and has been rarely quantified. Here we investigate a new approach to estimate residence times of such material. Fine-grained sediments collected from along the Brisbane River, Queensland, Australia, have higher excess ^{210}Pb activities and ^{210}Pb/^{137}Cs ratios than sediment eroded from uncultivated, cultivated and channel bank sources in the catchment. We explore the possibility that these higher ^{210}Pb activities are related to residence of the sediment within the channel and propose a method for using this data to make a residence time calculation. The method gave a range of residence times of 0–21 years for the Brisbane sediments. Data from the adjacent Logan River system provide a range of 0–9 years, although both ranges were dependent on a number of assumptions regarding sediment thickness and ^{210}Pb fallout rates and amounts.

Key words residence time; fallout nuclides; land-use change; catchment management; Australia

INTRODUCTION

Many of the world's rivers have been adversely affected by human use of the surrounding landscape. Intensification of agriculture invariably leads to an increase in erosion and the greater potential for delivery of sediment and nutrients to rivers. Increasingly, efforts to redress some of the adverse effects have focused on turning off the source of the pollutants. In Australia, fine-grained sediment is recognized as a significant river contaminant (Donnelly *et al.*, 1995; Prosser *et al.*, 2001). European settlement 150 years ago triggered a massive increase in both hillslope and gully erosion (Eyles, 1977; Wasson *et al.*, 1998; Prosser *et al.*, 2001). In many areas the gully networks which formed shortly after European settlement continue to supply fine-grained sediment at rates far exceeding the pre-European condition (Olley & Wasson, in press). In many areas, catchment restoration efforts are focusing on revegetating these gully networks. There is no doubt that such efforts will eventually improve downstream water quality. How long before such improvements are seen remains unanswered, and it is difficult to quantify without realistic estimates of the residence time of the sediment in the channels. Here we explore a method for estimating such residence times using fallout radionuclide data.

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THEORY

All soil surfaces have been labeled by fallout ^{137}Cs and ^{210}Pb. Caesium has been deposited as a result of above ground nuclear weapons testing during the 1950s and 1960s and strongly sorbs onto fine-grained particles (Walling & He, 1997). Fallout ^{210}Pb, also known as excess or unsupported ^{210}Pb, is the product of atmospheric decay of naturally occurring ^{222}Rn. Upon deposition onto the soil surface it also binds strongly to fine-grained particles (Wallbrink *et al.*, 1999). Caesium has a half-life of 30 years, and ^{210}Pb$_{ex}$ 22.3 years. Deposition of ^{137}Cs ceased by 1970, whereas ^{210}Pb$_{ex}$ deposition is more or less constant. The ^{210}Pb$_{ex}$ to ^{137}Cs ratio in soils varies with depth; in the Murrumbidgee River catchment it ranged from about 9:1 at the surface to about 1:1 at a depth of 30 cm (Wallbrink *et al.*, 1998). The ratio between them in the bulk soil in a region is roughly constant from year to year allowing for decay and atmospheric inputs (Wallbrink & Murray, 1996).

Once a particle is transported into the fluvial system, it can accumulate additional fallout ^{210}Pb by direct deposition. As storage time in the river increases, the sediments would attain higher ^{210}Pb activities and ^{210}Pb/^{137}Cs activity ratios than those of the sediment eroded directly from the catchment. If the additional amount of ^{210}Pb added to the sediment during its storage time in the river can be estimated, then the time required to attain that amount can be calculated if the ^{210}Pb input rate is known.

THE STUDY AREA

The Brisbane and Logan rivers are large mixed land-use catchments on the southeast coast of Queensland, Australia. They adjoin one another, have similar catchment areas of mixed lithology, and flow in an easterly direction to Moreton Bay. The effects of European land management practices in the Moreton Bay catchment are summarized in Caitcheon *et al.* (2001). Briefly, before European settlement the catchment was in a relatively stable condition, the impacts of European settlement led to significant increases in suspended sediment export. This resulted from extensive clearing, cultivation on the flood plains, over-grazing, and urban development. Only about 26% of the original vegetation remains over the entire catchment. Gully erosion is now extensive and is especially significant on granitic soils. Sediment yields increased rapidly in the nineteenth century, particularly in the first three decades following settlement. Caitcheon *et al.* (2001) also provide a detailed analysis of erosion in the catchment showing that the primary sediment sources are subsoil erosion (75%) from the gullies and channels and runoff (25%) from cultivated land.

MATERIALS AND METHODS

Catchment sampling

A total of 750 samples (groups of 10 from 75 areas) were collected from the Logan and Brisbane River catchments to characterize the ^{210}Pb and ^{137}Cs activities in recently eroded sediment (Fig. 1). Samples were collected where sediment was readily

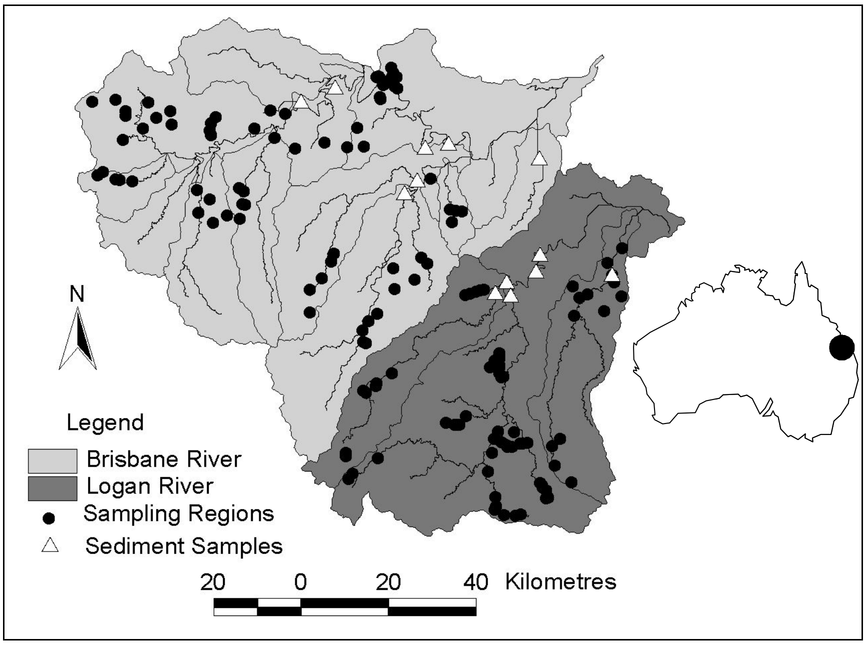

Fig. 1 Site location diagram, Brisbane and Logan rivers, southeast Queensland, Australia.

available for transport. Samples were divided into three erosion source classes: (a) uncultivated lands, (b) cultivated lands and (c) subsoil erosion from channels and gullies. These classes were then used as the basis for combining samples for analysis. Ten composite samples from each erosion class were analysed. The <10 μm fraction was separated from each of the combined samples by settling. All of the radionuclide analyses were undertaken on this <10 μm fraction. River sediments from the main channel of the Brisbane and Logan rivers were collected from the river bed during a period of low flow (Fig. 1). All samples were taken above the tidal limit. The <10 μm fraction of these sediments was removed by settling and used for subsequent radionuclide analyses.

Sample measurement by gamma spectrometry

The radionuclide concentrations were determined by routine high resolution gamma spectrometry, using the methods described by Murray *et al.* (1987). The detectors used in this study are "n"-type closed-ended co-axials. Detection limits are about ±0.3 Bq kg^{-1} for ^{137}Cs; ±3.0 Bq kg^{-1} for ^{210}Pb$_{ex}$ and ±0.1 Bq kg^{-1} for ^{226}Ra. Typical count times were one day.

RESULTS

The ^{210}Pb$_{ex}$ and ^{137}Cs concentrations (Bq kg^{-1}) on the <10 μm fraction from samples collected from each of the three erosion source classes are shown in Fig. 2. As expected (Wallbrink *et al.*,1999) the concentrations of both nuclides are highest in uncultivated soils, and lowest in subsoil material from channels and gullies. Fine-grained sediments eroding from cultivated land have intermediate values. The mean $^{210}Pb_{ex}$ and ^{137}Cs ratios of the uncultivated, cultivated and channel bank sources are, 5.8 ± 0.4, 3.2 ± 0.1 and 2.6 ± 0.8 respectively, with a mean of 4.1 ± 1.4 (solid line in Fig. 2). This defines the average $^{210}Pb_{ex}$ to ^{137}Cs ratio of all the catchment sources (for the year 2001).

The fallout tracer concentrations on <10 μm sediments from the Brisbane and Logan rivers are also shown in Fig. 2. The mean ratio of the sediments is 15.7 ± 4.7 and 14.3 ± 1.3 respectively; these are inconsistent with those of the catchment erosion sources.

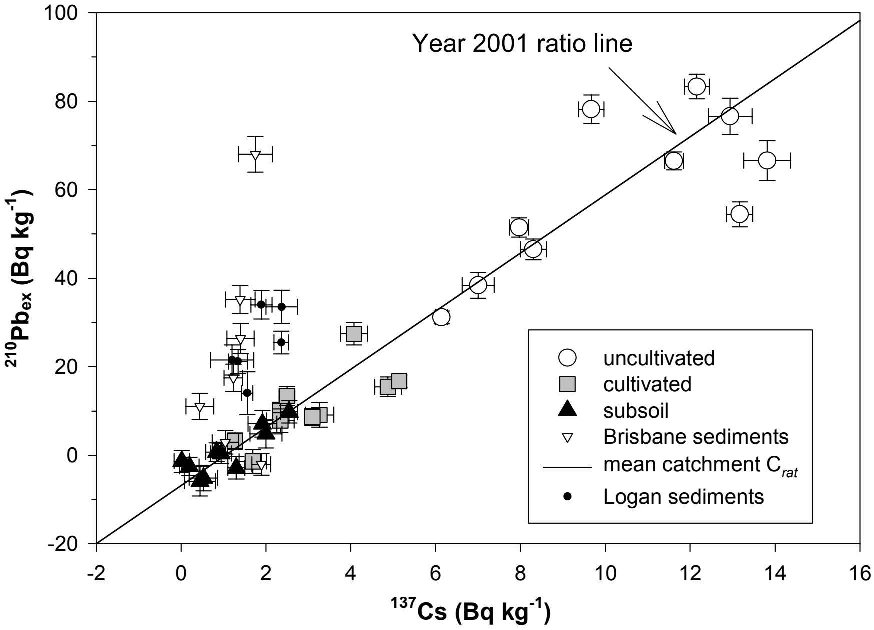

Fig. 2 Concentrations of ^{137}Cs and $^{210}Pb_{ex}$ on <10 μm deposited sediments and erosion sources of the Brisbane and Logan river systems.

DISCUSSION

The ratio of $^{210}Pb_{ex}$ to ^{137}Cs was much higher on the Brisbane River sediments (15.7 ± 4.7) than those of the catchment erosion source samples (ranges: 5.8 ± 0.4, 3.2 ± 0.1 and 2.6 ± 0.8). This results either from the preferential transport of particles with a higher $^{210}Pb_{ex}$ to ^{137}Cs ratio from the catchment into the river, or the direct addition of $^{210}Pb_{ex}$ to the sediment in the river channel. As no such higher ratios were measured, for the purpose of this paper we assume that the latter process dominates.

Calculating sediment residence times

We model the river as a channel with sediment entering at the upper end (Fig. 3). This sediment deposits as a layer on the river bed, and is transported along the river as a result of a series of resuspension and deposition events. The thickness of the bottom sediment layer amenable to transport (S_{sed}) is assumed constant along the length of the

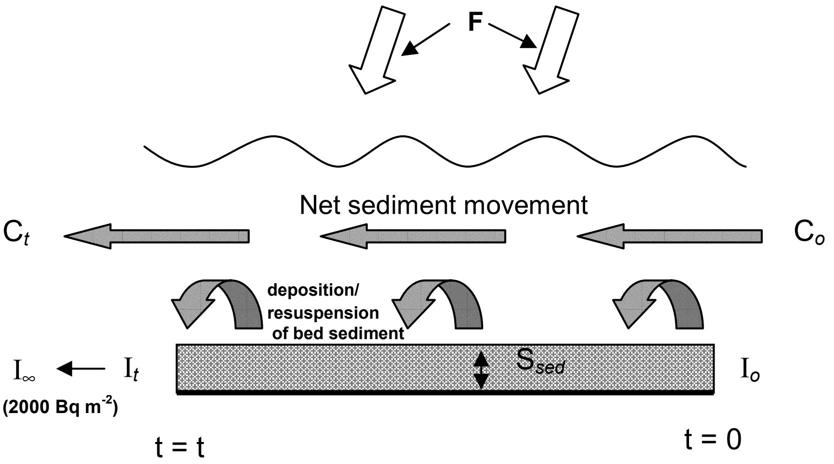

Fig. 3 Schematic of postulated model for deriving sediment residence times.

river, and vertically well-mixed. Sediment enters the river channel at time $t = 0$ with an initial excess ^{210}Pb activity, C_0, supplied by erosion from catchment sources.

Additional excess ^{210}Pb accumulates in the sediment as a result of fallout ^{210}Pb (F) depositing in the river channel. The fallout ^{210}Pb either deposits directly onto the sediment if the river bed is dry, or is scavenged and deposited onto the river sediments by the resuspension-deposition cycle. At time t, equal to the residence time of sediment in the river, sediment samples are collected and measured for their excess ^{210}Pb activity (C_t). This model is described by:

$$I_t = I_\infty(1 - e^{-\lambda t}) + I_0(e^{-\lambda t}) \tag{1}$$

where λ is the decay constant for ^{210}Pb (0.031 year^{-1}); I_0 is the initial inventory of excess ^{210}Pb within the mobile layer of river sediment derived from catchment erosion ($t = 0$, units Bq m^{-2}), and is given by $C_0 \times S_{sed}$; I_t is the inventory of excess ^{210}Pb within the mobile sediment layer at the time of sample collection ($t = t$), given by $C_t \times S_{sed}$; I_∞ is the inventory of excess ^{210}Pb which occurs when depositional flux and decay within the sediment profile reach equilibrium. Under steady-state conditions this occurs after 4–5 half-lives of ^{210}Pb (90–100 years).

In equation (1), the first term on the right-hand side describes ingrowth of excess ^{210}Pb accumulated by river sediment as a result of fallout. The second term on the right-hand side describes decay of excess ^{210}Pb associated with the sediment when it initially enters the river channel ($t = 0$).

Rearranging equation (1) gives

$$\frac{I_t - I_\infty}{I_0 - I_\infty} = e^{-\lambda t} \tag{2}$$

and thus the sediment residence is determined from

$$t = -\frac{1}{\lambda}\ln\left(\frac{I_t - I_\infty}{I_0 - I_\infty}\right) \tag{3}$$

I_∞ is determined from F/λ. The value of F for Brisbane is given by Turekian *et al.* (1977) as 65 Bq m^{-2}. Thus I_∞ is estimated to be 2000 Bq m^{-2} year^{-1}. We calculate C_0

from the product of the ^{137}Cs activity of the river sediment (1.3 ± 0.2 Bq kg^{-1}) and the ^{210}Pb$_{ex}$/^{137}Cs ratio of sediment being delivered to the river. The ^{210}Pb$_{ex}$/^{137}Cs ratio was determined to be 2.7 ± 0.7, by weighting the proportionate contributions from the major input sources determined by Caitcheon *et al,* (2001). Thus using Brisbane River data, C_0 is found to be 3.5 ± 1.1 Bq kg^{-1} (i.e.1.3 * 2.7).

Application of equation (3) also requires an estimate of S_{sed}, the value of which has a significant effect on the calculated residence time (Fig. 4). Although not explicitly measured, visual examination at the time of sampling indicates that mobile sediment in the Brisbane and Logan rivers is probably limited to a layer less than 20 mm thick. Consequently discrete thicknesses between 1 and 20 mm have been used to construct Fig. 4. As an example of the effect of S_{sed} on t, the most downstream value of ^{210}Pb$_{ex}$ activity in the Brisbane River sediment (C_t = 26.3 ± 3.4 Bq kg^{-1}) yields a value of t = 6 years for S_{sed} = 20 mm and t = 0 years for S_{sed} = 1 mm.

All the C_t concentrations on sediment measured in the Brisbane and Logan river systems, as well as their locations, are given in Fig. 5. Using the relationship of Fig. 4, they provide a range of residence times of 0–21 years for the Brisbane and between 0 and 9 years for the Logan, depending on S_{sed}. Their mean residence times (derived from the average of their C_t values, and using S_{sed} = 20 mm) is 5.1 and 5.7 years respectively. The data also show that residence time does not increase monotonically downstream. This is not unexpected as the river is not a simple closed channel and C_t values will be affected by inputs from tributaries and internal processes such as bank erosion. Thus, it is likely that river reaches will need to be treated as a series of discrete segments in which the residence of the sediment depends on local inputs and hydraulic conditions.

A simple manipulation of the known river sediment loads and channel dimensions can be used to check our method. For example, approximately 400 000 t year^{-1} (or 200 000 m^3 year^{-1}) of fine sediment is delivered to the mouth of the Brisbane River annually (Caitcheon *et al.*, 2001). A conservative estimate of total channel length in

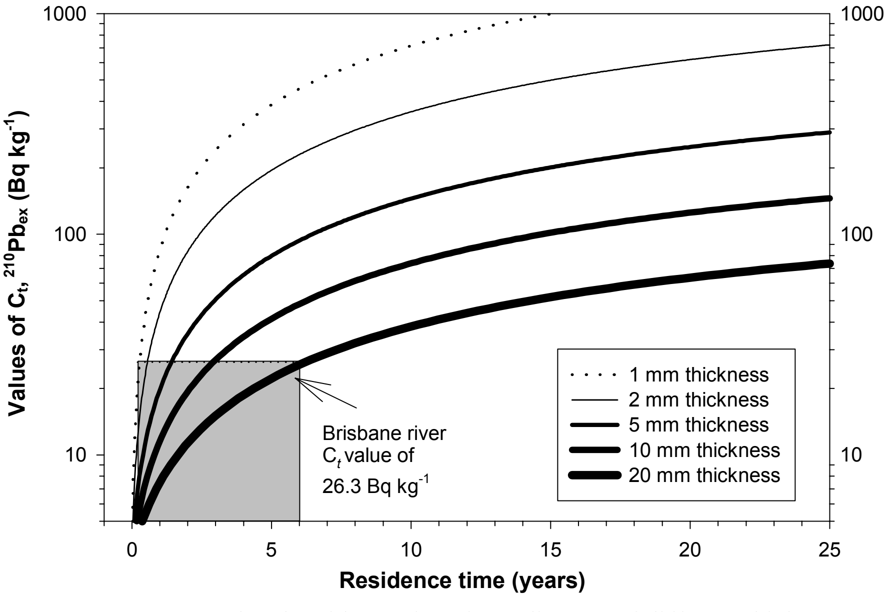

Fig. 4 Postulated residence time for sediments of different thickness to acquire the C_t concentration measured on deposited Brisbane River sediments.

the catchment is about 1000 km, average width is about 100 m. If we assume a mean residence of 5 years (see above) then on average, an amount equivalent to the annual sediment load should be stored on ~200 km of river channel length per year. By dividing the annual sediment mass by this area of channel we derive a value of 10 mm sediment depth. This is within the range of our field observations and within a factor of 2 of the S_{sed} value used in deriving the residence time estimate. Given the uncertainties in both approaches, we believe they are in reasonable agreement.

In applying our method we have assumed that no desorption of ^{137}Cs from the sediments has occurred during transport, the catchment sampling has characterized the full range of erodible ^{137}Cs and ^{210}Pb soil activities, the ^{210}Pb$_{ex}$ is thoroughly mixed within the sediment layer and that a constant deposition of ^{210}Pb$_{ex}$ has occurred over the sediment storage time. All these assumptions are reasonable for river segments that are in approximate steady state between input and output of fine-grained sediment. Given the relatively short transit times expected for fine-grained sediment, steady state is a reasonable assumption for river reaches on the scale of one to tens of kilometres.

The uncertainties and time frame of the method are also limited by the precision of the analytical method used to measure ^{210}Pb, and the half-life of ^{210}Pb. Using the data presented in this paper, this practically limits the precision of the method to a range of 2–80 years. Given that the approach concerns the mobile fine-grained fraction of river sediment, we believe this time frame is consistent with the sediment residence times of most river systems with regular high flow events. The exception may be dryland rivers with very infrequent transport (flood) events.

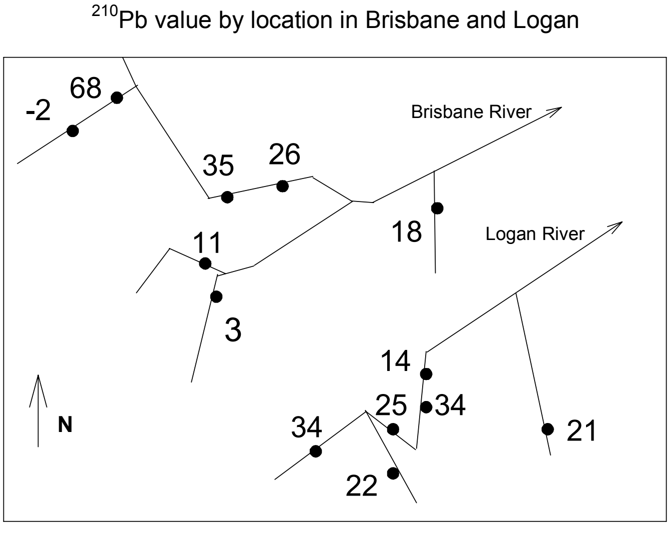

Fig. 5 Location of ^{210}Pb$_{ex}$ (C_t) values in the Brisbane and Logan river systems, Queensland, Australia.

CONCLUSIONS

The residence time of fine-grained (<10 μm) sediment within channels is often assumed to be short. In this paper we explored a new approach to quantify such residence times. The method produced a range of residence times for deposited Brisbane (and Logan) river sediments of 0–21 and 0–9 years respectively; given various assumptions regarding sediment thickness, and $^{210}Pb_{ex}$ fallout rates and amounts. The most likely catchment erosion source of the sediment was from subsoil erosion of channels and gullies. These estimated residence times have implications for catchment management. In particular the timeframes over which the benefit (or impact) of changes in catchment land use may be observed downstream.

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