Nuclear Instruments and Methods in Physics Research B 268 (2010) 1150–1154

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Plutonium as a tracer of soil and sediment movement in the Herbert River, Australia

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ARTICLE INFO

Article history:
Available online 8 October 2009

Keywords:
Plutonium
AMS
Isotope ratios
Sediment transport
Erosion

ABSTRACT

Plutonium fallout from atmospheric nuclear-weapons testing in the 1950s and 1960s constitutes an artificial tracer suitable for the study of recent soil erosion and sediment accumulation rates. Traditionally ^{137}Cs has been the fallout isotope of choice for such studies, but the plutonium isotopes confer a number of advantages, which can be realised using the ultra-sensitive detection technique of Accelerator Mass Spectrometry (AMS). As a first application of plutonium to a whole-of-basin study, Pu has been measured in both soil and sediment across the catchment of the Herbert River, which is one of the major rivers draining into Australia's Great Barrier Reef Lagoon. Its catchment includes undisturbed areas as well as regions of pasture and sugar cultivation. The Pu measurements allow the relative contributions of surface and gully erosion from the different land use areas to be determined, and permit the discharged material to be apportioned between the relevant sources.

1. Introduction

Fallout ^{137}Cs from nuclear-weapons testing has been widely used as an isotopic tracer for the study of soil and sediment movement (e.g. [1–6]). Much less use has been made of the plutonium (Pu) that was deposited world-wide at the same time, despite the nuclear-weapons tests of the 1950s and 1960s dispersing ~3 tonnes of ^{239}Pu, or six times as many atoms of Pu as ^{137}Cs, around the globe [7]. The development of the ultra-sensitive technique of Accelerator Mass Spectrometry (AMS) for plutonium has, however, allowed the potential advantages of Pu relative to ^{137}Cs to be realised.

Plutonium, like caesium, attaches strongly to soil particles upon reaching the earth's surface. In a previous study [8] we showed that ^{239}Pu and ^{137}Cs are both suitable tracers of soil erosion and sediment transport, but that ^{239}Pu has distinct advantages in terms of sensitivity, precision, sample size, and the number of samples that can be measured. In particular, ^{137}Cs requires ~100 g of the fine component (<10 μm) of the soil or sediment and γ-ray counting times of 2 days to achieve comparable sensitivity to that achieved with ^{239}Pu using only 4 g of soil and AMS counting times of a few minutes. Substantially longer counting times for plutonium are readily achievable, allowing higher sensitivity to be attained. Further, the sensitivity of ^{137}Cs is declining with time due to its half-life of 30 years, and indeed has declined to less than 35% of its original value in the ~50 years since the nuclear-weapons testing era, a problem exacerbated in the southern hemisphere where fallout was only one third of that in the northern hemisphere [7]. Plutonium, on the other hand, has barely declined at all. Measurements of plutonium by AMS offer a further advantage in that they are essentially background free, and consequently yield results with statistically superior precision over those obtained for ^{137}Cs using γ-ray spectroscopy [9]. AMS also offers considerably higher sensitivity than is possible with α-particle counting [9,10].

In the present study, we apply plutonium as a tracer to assess how land use in a river catchment that drains into the lagoon behind Australia's Great Barrier Reef is influencing the delivery of sediment to the lagoon. High sediment levels are known to adversely affect coral abundance [11], but whether the increased sediment loads in the river [12] translate into increased sediment delivery to the lagoon with consequent impacts on the health of the Great Barrier Reef ecosystem remains unclear [13]. Differences in the plutonium concentrations between surface soils and river sediments can be used to apportion erosional sources between surface or channel (gully or bank) erosion. Because AMS provides the ability to measure large numbers of samples in a relatively short period of time, these studies can be performed at a basin-wide scale. The data can then be used to validate and improve computer model predictions of erosion and soil loss. The data and the models provide crucial input for the development of management strategies and practices aimed at soil conservation, sustainable production and environmental protection.

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doi:10.1016/j.nimb.2009.10.121

2. Study area

The Herbert river catchment is located in tropical north-eastern Queensland, Australia (Fig. 1) and encompasses an area of approximately 10,000 km^2. Descriptions of the catchment can be found in [8,12], but of most interest to the present work is that it can be broken up into regions largely characterised by three distinct types of land use: pasture lands, sugar cultivation and relatively undisturbed forest.

3. Methods

A soil depth profile was collected in each of the three land use areas at the sampling sites indicated by open circles in Fig. 1. Sample preparation and measurement details can be found in [8]. Briefly, 4 pg (i.e. 10^{10} atoms) of ^{242}Pu spike was added to a 4 g subsample of the homogenised soil and the plutonium leached with hot nitric acid and purified using ion exchange columns. The ^{239}Pu concentration and ^{240}Pu/^{239}Pu ratio were determined by AMS using the 14UD pelletron accelerator at the ANU using the methodology described in [9]. Samples were also analysed for ^{137}Cs content at the CSIRO Land and Water laboratories in Canberra using HPGe detectors to count the 662 keV γ-rays that arise from ^{137}Cs decay [14]. This allowed determination of soil depth profiles for both elements.

In addition, samples of surface soil were collected from across the catchment. Samples from a region that represented a particular land type and use were combined. In effect the entire catchment was subdivided into 18 regions of either forest, pasture or sugar cultivation, with a representative (composite) sample from each region. The approximate sampling locations are represented by the black circles in Fig. 1. River sediment deposits were also collected from 21 regions along the river upstream of any tidal influence (squares in Fig. 1), and from nine selected tributaries (crosses in Fig. 1). In contrast to the depth profile samples where bulk material was used, only the $<10\ \mu\mathrm{m}$ grain-size component of the surface soil and sediment samples was analysed [8]. Selection of a specific grain-size provides a common basis for comparison between soil and sediment samples. Transport and deposition processes will modify the grain-size distribution of sediment relative to its parent soil, and plutonium and ^{137}Cs concentrations will depend on grain-size due to differences in the surface area available for binding, with both ^{239}Pu and ^{137}Cs expected to bind most readily to smaller soil particles [8,15].

4. Results and discussion

4.1. Depth profiles

Fig. 2 shows the ^{239}Pu and ^{137}Cs concentrations as a function of soil depth for the three depth profiles. The profiles are typical of

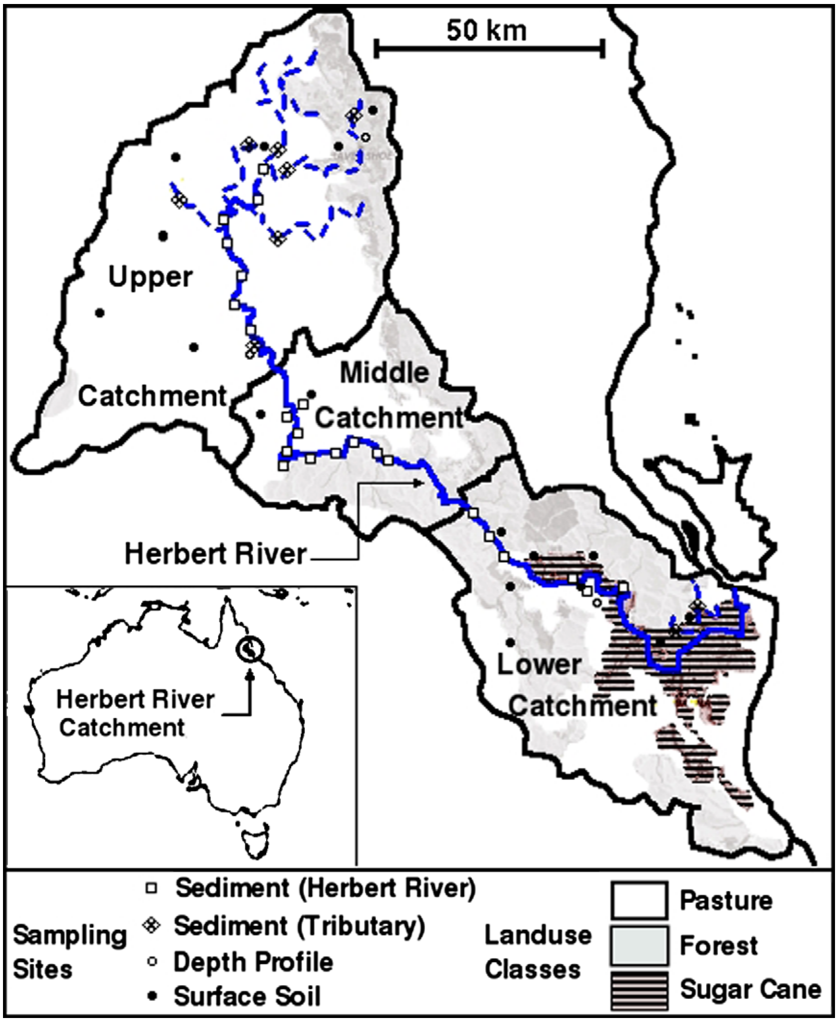

Fig. 1. Land use classes within the Herbert River catchment. The catchment has been divided into upper, middle and lower sub-catchments based largely on topology and land use. Squares and crosses indicate river sediment sample collection sites and circles indicate approximate soil sampling locations.

those seen elsewhere [16] and the two isotopes are strongly correlated in each case. At the two uncultivated sites (forest and pasture) the total inventory is found in the top 20 cm of the soil, and the shapes of the two profiles are similar and almost statistically indistinguishable. At the sugar cane site, the isotopes are found to greater depth and the profile is much flatter, reflecting mixing of the soil by cultivation. Since the measurements were made on whole soil samples, total inventories of the fallout isotopes can be determined at each site. These are listed in Table 1, along with the average annual rainfall at each site. In the pastured area the inventory is only half that at the other two sites, and strongly suggests a correlation with rainfall as has been observed elsewhere [4]. Data from more sites will, however, be required to substantiate this association.

4.2. Data from tributaries

Surface soil and sediment data were collected from a number of tributaries of the Herbert River and are shown in Table 2. Several of these tributaries were selected because they drain areas that are exclusively forest, pasture, or sugar cane. A clear contrast between the erosional processes under the three different land use regimes is evident.

In the forested catchment, the river sediment has a ^{239}Pu concentration that is ~80% of that in the surface soil, which indicates that most of the sediment has been derived from surface soil and that channel (gully or bank) erosion plays only a minor role in the forested environment. Sediment that is derived from gullies or river-bank collapse will generally be excavated from depths greater than 30 cm and hence can be considered to have a ^{239}Pu concentration indistinguishable from that of sub-soil.

In the pasture catchments, on the other hand, plutonium concentrations in the river sediment are only 6% and 12% of those in the surface soils, indicating that erosional loss in these areas is dominated by sub-soil i.e. gully development or river-bank collapse.

Table 1
^{239}Pu and ^{137}Cs isotope inventories determined from depth profiles at three widely-separated sites in the Herbert River catchment. An average soil density of 1.5 g/cm^3 was assumed. Rainfall values are taken from the ANUCLIM data set [17].

Land use area	Average annual rainfall (mm/yr)	^{239}Pu inventory (Bq/m^2)	^{137}Cs inventory (Bq/m^2)
Forest	1750	7.5 ± 0.4	246 ± 15
Pasture	780	3.6 ± 0.3	156 ± 12
Sugar cane	1800	6.9 ± 0.6	259 ± 32

Regions in which sugar cane cultivation is the predominant land use appear to be an intermediate case. Regular cultivation before planting may make the surface soil more susceptible to erosion in the sugar cane areas during periods of high rainfall relative to the uncultivated pastureland.

Four of the tributaries drain areas which are a mix of forest and pastureland. As might be anticipated from the discussion above, the fractions of surface erosion fall between the two end-members, although they vary by almost a factor of four despite similar proportions of pastureland in each case.

4.3. Data from the river

A substantial body of sediment data has been collected from the main channel of the Herbert River, as indicated by the open squares in Fig. 1. These data have been supplemented by surface soil samples, which in the upper and lower catchments provide good coverage and give a good estimate of the average surface soil concentrations of ^{239}Pu and ^{137}Cs. Vehicular access to much of the middle catchment is, however, difficult, and possible only when it is dry. Consequently, surface soil samples (and three river sedi-

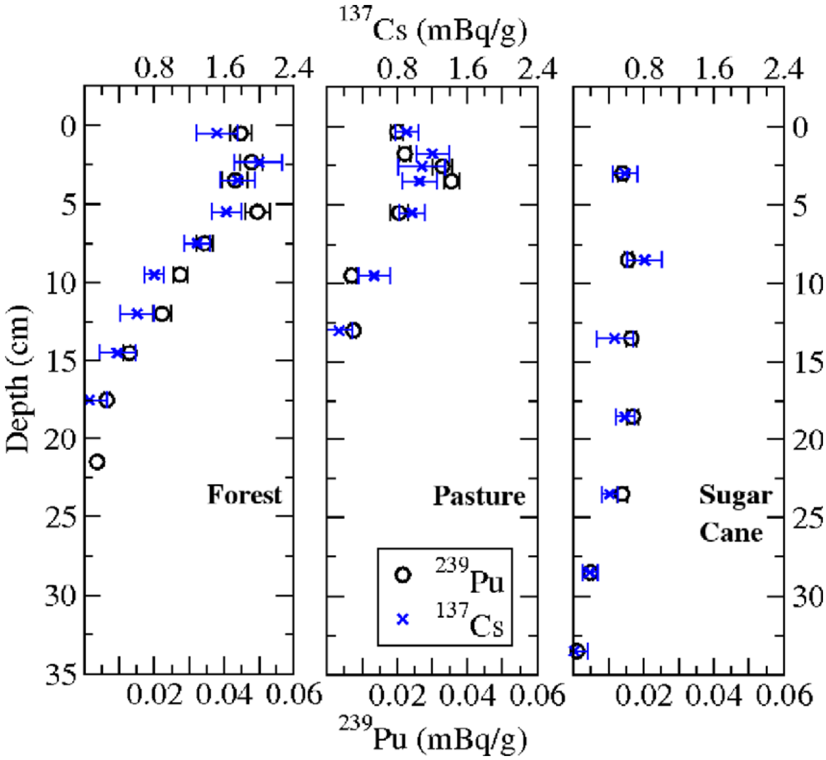

Fig. 2. ^{137}Cs and ^{239}Pu soil depth profiles from the forest (upper catchment), pasture (base of upper catchment) and sugar cane (lower catchment) areas in the Herbert River watershed.

Table 2

Average ^{239}Pu and ^{137}Cs concentrations in the <10 µm fraction of samples of surface soil and river sediment from nine tributary sub-catchments and the main channel of the Herbert River. Tributaries 1–9 progressively connect to the main river in the downstream direction.

Sub-catchment and land use	Sample type	^{239}Pu[a] (µBq/g)	^{137}Cs[a,b] (mBq/g)	^{137}Cs/^{239}Pu	Gully and river-bank erosion[c]
Forest					
Upper (tributary 1) (100% forest)	Soil	82 ± 6	–	–	17%
	Sediment	68 ± 5	–	–	
Pasture					
Upper (tributary 6) (100% pasture)	Soil	240 ± 13	–	–	88%
	Sediment	29 ± 3	–	–	
Upper (tributary 7) (100% pasture)	Soil	201 ± 11	–	–	94%
	Sediment	12 ± 2	–	–	
Sugar cane			–	–	
Lower (tributary 8) (100% sugar cane)	Soil	55 ± 3	–	–	69%
	Sediment	17 ± 1	–	–	
Lower (tributary 9) (100% sugar cane)	Soil	55 ± 3	–	–	42%
	Sediment	32 ± 3	–	–	
Mixed forest/pasture					
Upper (tributary 2) (52% forest)	Soil	118 ± 29	–	–	59%
	Sediment	48 ± 4	–	–	
Upper (tributary 3) (52% forest)	Soil	162 ± 9	–	–	54%
	Sediment	74 ± 5	–	–	
Upper (tributary 4) (48% forest)	Soil	122 ± 57	–	–	77%
	Sediment	28 ± 2	1.49 ± 0.20	53 ± 8	
Upper (tributary 5) (48% forest)	Soil	162 ± 9	–	–	90%
	Sediment	17 ± 2	–	–	
Herbert River main channel					
Upper (85% pasture)	Soil	164 ± 4 (82–240)	7.59 ± 0.26 (3.21–10.70)	46 ± 2	84%
	Sediment	27 ± 3 (20–38)	–	–	
Middle (52% forest)	Soil	161 ± 8 (49–240)	7.49 ± 0.47 (2.17–10.70)	47 ± 4	88%[d]
	Sediment	21 ± 2 (19–23)	1.58 ± 0.23 (0.74–2.81)	75 ± 11	
Lower (50% Sugar cane)	Soil	139 ± 9 (48–240)	6.54 ± 0.60 (0.74–9.9)	47 ± 5	80%[d]
	Sediment	30 ± 2 (21–38)	1.68 ± 0.11 (1.47–1.99)	56 ± 5	

[a] Uncertainties include systematic and statistical errors and are $\sigma/\sqrt{(n-1)}$ for the sediment samples, where σ is the standard deviation of the set of n data points. Uncertainties for the soil samples are area-weighted standard deviations, as described in the text. Entries in parentheses give the range of scatter in the data.

[b] Cs analyses were not performed on the tributary samples as the smaller number of samples collected would have yielded statistically inferior results.

[c] Determined from Pu concentrations, Relative fraction of ^{239}Pu attributable to the sub-soil contribution to the river sediment; i.e. (^{239}Pu_soil/^{239}Pu_river sediment)-1.

[d] See text Section 4.4 for the derivation of these values.

ment samples) were collected only from the predominantly pastureland areas near the upper boundary of the middle catchment. Almost 50% of the middle catchment is comprised of pastureland, with the remainder covered in rainforest. We have assumed that the forest contributes relatively little sediment, and hence that the samples collected here are approximately representative of the middle catchment. Future sampling of river sediment and soil through the forested area will be required to confirm this assumption.

Average concentrations of ^{239}Pu and ^{137}Cs in soil and river sediment from the three major sub-catchments of the Herbert River are also presented in Table 2. Average soil concentrations were calculated by weighting each individual measurement with the area of the region that it represented. Average river sediment concentrations are simply weighted averages of the individual measurements from the main channel. Interpretation of the data from the upper catchment is straightforward since all of the sediment must originate in the catchment. The results clearly show that erosion is dominated by gullying and river-bank slumping, and given that the upper catchment is 85% pastureland, this is consistent with the results from the tributaries. Similar conclusions may be drawn from the limited data for the middle catchment. As noted above, these data are also from a predominantly pastureland region that is essentially a continuation of the upper catchment, and the results for the middle catchment most likely reflect this. Support for this conclusion can be found in the data from the mixed forest/pasture tributaries, which have similar levels of forest cover to that of the middle catchment. These tributaries also show erosion that is dominated by gullying and river-bank slumping, although to a lesser extent than that observed in the upper catchment. Interpretation of the data from the lower catchment is more complex because the sediment in the river is the sum of what is added locally and what is exported by the upper and middle catchments above. Nevertheless, average soil and sediment concentrations of plutonium differ little between the upper and lower catchments, and hence it is reasonable to conclude that the bulk of the sediment in the river in the lower catchment is sub-soil that has originated from gullies and river-bank collapse.

4.4. Comparison with models

Tables 3 and 4 compare the results from the present work with the predictions of the CSIRO SedNet model [18] for the Herbert River catchment. The SedNet model is specifically designed for Australian conditions and combines catchment topography, vegetation cover, rainfall and gully/river-bank stability to predict soil erosion and transport.

Table 3 shows the comparison for the nine tributaries studied in the present work. Listed is the relative fraction (in%) of ^{239}Pu attributable to the sub-soil contribution to the river sediment; i.e. (^{239}Pu_soil/^{239}Pu_river sediment)-1, this provides is a measure of the gullying and river-bank contribution. Clearly the SedNet model gives a reasonable account of the data, but with a tendency to underestimate the gullying contribution from the pastureland, and perhaps to overestimate it in the forested area.

The tendency of the model to underestimate the contribution from gullying and river-bank collapse is more evident at the larger scale, as seen in Table 4. Note that in deriving the gully/river-bank contributions from the data for the combined ‘upper + middle’ catchment for example, it was assumed that the appropriate

Table 3
Relative proportions of surface and sub-soil (i.e. collapsed gully/river-bank) material in sediments from tributaries and the main channel of the Herbert River, based on the results of the ^{239}Pu data (reproduced from Table 2), and the SedNet model estimates.

Catchment	Land class	Gully/River-bank contribution (%)		
		^{239}Pu	SedNet	SedNet/^{239}Pu
Tributary 1	Forest	17 ± 8	26	1.5
Tributary 2	Forest/pasture	59 ± 11	53	0.9
Tributary 3	Forest/pasture	54 ± 4	45	0.8
Tributary 4	Pasture/forest	77 ± 11	45	0.6
Tributary 5	Pasture/forest	90 ± 1	80	0.9
Tributary 6	Pasture	88 ± 2	66	0.8
Tributary 7	Pasture	94 ± 1	78	0.8
Tributary 8	Sugar cane	69 ± 3	55	0.8
Tributary 9	Sugar cane	42 ± 6	55	1.3

Table 4
Relative proportions of surface and sub-soil (i.e. collapsed gully/river-bank) material in Herbert River sediment based on the results of the present work and the equivalent SedNet model estimates.

Catchment	Gully/River-bank contribution (%)			
	^{239}Pu[a]	^{137}Cs[b]	SedNet	SedNet/^{239}Pu
Upper	84 ± 2	–	62	0.7
Upper + middle	88 ± 2	80 ± 3	55	0.6
Upper + middle + lower	80 ± 2	75 ± 3	47	0.6

[a] See text Section 4.4 for the derivation of these values.
[b] Samples from the upper catchment were not analysed for ^{137}Cs content.

^{239}Pu concentration in the sediment was that in the middle catchment, since it contains contributions from both the upper and middle catchments. Sediment samples from the upper catchment have not yet been analysed for ^{137}Cs content. The relevant ^{239}Pu concentrations in soil were, however, the area-weighted average over both catchments, with the caveat discussed above that the forested section of the middle catchment was not included. Similar considerations apply to the combined 'upper + middle + lower' catchment. The major benefit to be derived from a basin-wide survey of plutonium concentrations such as reported here is that it allows the model to be 'tuned' to reflect more accurately the observed concentrations. It can then be used with more confidence to predict the impacts of changes in grazing or cultivation strategies on sediment loads in the river, and ultimately on the delivery of sediment to the Great Barrier Reef Lagoon.

5. Conclusion

The ultra-sensitive detection technique of AMS makes it viable to use ^{239}Pu to study soil and sediment movement in large scale catchments such as the Herbert River. Smaller sample sizes, shorter counting times, the absence of background and the superior statistical precision for AMS ^{239}Pu measurements, compared to those for ^{137}Cs, should make ^{239}Pu the tracer of preference for such studies, particularly in the Southern Hemisphere where fallout levels were lower than in the North. The data presented here for the Herbert River catchment indicate that it is possible to apportion the sources of sediment material between surface and sub-soil gully/river bank erosion in the different land use areas, and confirm that gully/river-bank erosion is the dominant source of sediment in pastured and cultivated areas. This finding is similar to other recent research in the Great Barrier Reef catchments which have determined that channel erosion dominates end of catchment sediment yields, particularly when grazing is the primary land use [19,20]. In contrast the data indicate that surface erosion is the dominant contributor in undisturbed forested areas.

The throughput of the AMS system allows large numbers of samples to be measured with good precision. The data provide valuable input for the management of the river catchments and are important for quantifying and predicting the effects of future changes in land use. Models such as SedNet have a crucial role to play in this regard. Isotopic tracers provide a valuable tool for testing and developing such models. The data presented here highlight areas where SedNet could be in need of improvement: most notably the ^{239}Pu data clearly indicate that the model appears to underestimate the gully/river-bank (sub-soil) contribution to the total sediment load.

Acknowledgements

This work was supported by Discovery Grant DP0557122 from the Australian Research Council.

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