Using ^{239}Pu as a tracer for fine sediment sources in the Daly River, Northern Australia

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Abstract. The Daly River drains a large (52500 km^2) and mainly undisturbed catchment in the Australian wet–dry tropics. Clearing and cropping since 2002 have raised concerns about possible increased sediment input into the river and motivated this study of its fine sediment sources. Using ^{239}Pu as a tracer it is shown that the fine sediments originate mainly from erosion by gullying and channel change. Although the results also indicate that the surface soil contribution to the river channel sediments from sheet erosion has increased to 5-22% for the Daly River and 7-28% for the Douglas River (a tributary of the Daly River) in 2009 *vs*. 3-6% for the Daly River and 4-9% for the Douglas River in 2005. This excess top soil likely originates from the cleared land adjacent to the Daly River since 2005. However, channel widening largely as a result of hydrologic change is still the dominant sediment source in this catchment.

1 Introduction

Any sustainable agricultural development of northern Australia will require implementation of successful soil management strategies. In this study, fine sediment sources are investigated in the Daly River catchment, northern Australia, Figure 1. This is one of the few regions identified for the development of dry land farming systems in the semi-arid tropics of Northern Australia, largely due to the presence of perennial river flow and the availability of large areas of potentially arable soils [1-7].

Agricultural development of the Daly River catchment began in the 1880's with land clearing for the establishment of the first pastoral leases, although prior to this, the vegetation would have been controlled through aboriginal mosaic burning practices [8]. At present ~50% of the catchment area still remains in use for grazing of native vegetation for beef cattle production [5, 9].

Large scale land clearing for intensive cattle production was carried out during 2000–2002 and over 200,000 ha was cleared, with an additional 110,400 ha earmarked to be cleared in the next decade [10]. In these recent developments, new commercial agricultural/pastoral lots (30,000 ha) were allocated in the Stray Creek area and additional land clearing occurred in the Douglas/Daly-Katherine corridor. Overall the total land clearings at present represent ~9% of the catchment area [11].

While it seems certain that land clearing which has occurred post-European settlement has increased the erosion rates and sediment delivery to the river network in this catchment, what is not certain is how much of the eroded sediment actually reaches the river network, and of this how much is stored within the network relative to that which is transported out to the ocean. These need to be quantified to allow implementation of appropriate catchment management strategies. This work provides new information on sediment sources within the Daly River catchment and will contribute to the development of such strategies.

2 Background

The atmospheric testing of nuclear weapons between 1945-1980 released many anthropogenic radioisotopes into the stratospheric global circulation system. These isotopes subsequently deposited on the Earth's surface where some of them strongly adsorbed on to soil particles. The movement of these particles across the landscape has become a powerful method for investigating soil redistribution by wind, water and tillage [12]. The most commonly used anthropogenic isotope to assess soil redistribution rates is caesium-137 (^{137}Cs). Many applications of ^{137}Cs to landscape studies exist (c.f.
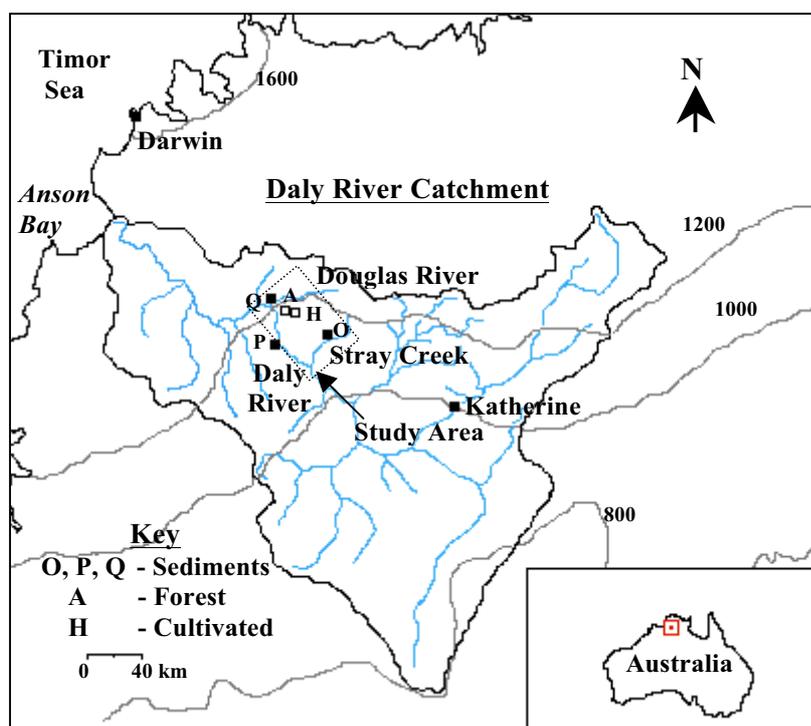
Fig. 1. The Daly River catchment. Rainfall isohytes are in mm/year.

[13]) including marine, lake and overbank sedimentation, catchment erosion and sediment source studies and wind erosion studies. However owing to the 30.07 year half life [14], ~70% of the ^{137}Cs from nuclear weapons testing has already decayed, and over the next few years it will drop closer to detection limits, especially in the southern hemisphere where fallout was much lower (~30%) than in the northern hemisphere. In addition the release of ^{137}Cs from local sources (e.g. Chernobyl accident) has complicated the application of ^{137}Cs in soil erosion studies in many locations around the world.

Recently, the anthropogenic plutonium ([239,240]Pu) radioisotopes ([239]Pu $t_{1/2}$=24100 years, [240]Pu $t_{1/2}$=6560 years, [14]) have been used as soil and sediment tracers [15-24]. Like [137]C, they fell out in rain and attached strongly to soil particles upon reaching Earth's surface, and like ^{137}Cs, they have been shown to be suitable tracers of soil erosion and sediment transport [16, 19, 21, 25]. On reaching Earth's surface the particle reactive Pu is held in the surface soil layers where it binds to hydrous oxide soil coatings [26]. Here it will persist for thousands of years because of its long half-life. Furthermore it provides a chronological maker as it was present in negligible amounts prior to anthropogenic production and release.

3 Sampling sites

The Daly River and its tributary the Douglas River flow through the north east of the study site. Stray Creek flows into the Daly River about 50 km upstream from the confluence of the Daly and Douglas Rivers. Flood sediment and sand deposits of these three waterways were collected from the channel beds and banks, Figure 1. The [239]Pu content in these fluvial samples has been used to indicate the amount of surface soil in the river sediment.

3.1 River-sediment sampling

At the Daly River, sediment samples were collected from sediment deposited on rocks lying at the water's edge in the river channel at site P. In the wet season these rocks would be submerged and covered by fine sediment, transported by the river. These samples are considered representative of the material transported downstream and eventually out of the catchment. The Douglas River has deposited sediments high on banks near its confluence with the Daly River. This material is deposited during floods and gradually builds up on the banks. Approximately 50 g of this material was collected from each of 10 locations, along ~100 m of the river bank at site Q, and then combined to make one representative sample. At Stray Creek the channel bed and channel banks were also sampled spatially at site O. As sediment deposition was limited here, it was necessary to collect sediments attached to leaves, twigs and low bushes.

3.2 Particle size analysis

To determine the fate of [239]Pu when binding to surface soils, two surface soil samples were separated into various particle size fractions. This was used to assess whether the [239]Pu was binding preferentially to different soil textures. Conventional sieving and settling techniques [27] were used to separate various size fractions of surface samples from a forest (site A) and a nearby cultivated soil (site H), after the samples were dispersed using an ultrasonic bath. Each fraction was prepared and the [239]Pu concentration measured as described in section 4.

4 Sample preparation for [239]Pu in soils and sediments

The sample preparation protocols for AMS were adopted from [25,28] with slight modifications. Typically, a 20 g aquilot of the sample was mixed with ~ 30 ml of MQ water

and a known amount (~ 4 pg) of ^{242}Pu tracer was added and the sample dried at 100°C for 12 hours. This was done to better facilitate the dispersal of the tracer in the sample. The sample was then ashed at 450°C for 8 hours to remove the organic components. Pu was leached from the sample by gently heating with ~ 40 ml of 8M nitric acid. The leachate was filtered and ~1g of NaNO$_2$ was added to convert the Pu to the 4+ oxidation state. The resulting solution was passed through preconditioned ion exchange columns containing 1.7g of BioRad AG 1-X8 100-200 mesh resin (Figure 3.9), followed by 25 ml of 8M HNO$_3$ and 70 ml of 12M HCl. The Pu was eluted from the resin with 25 ml of 0.1M NH$_4$I/12M HCl. The elutant was dried down and ~ 2ml of HNO$_3$ was added to remove the iodine.

After 1h of warming, ~ 2ml of HCl was added to remove ammonium nitrate and the solution was dried down. The residue was re-dissolved in 2ml of HNO$_3$ and 2mg of Fe was added as Fe(NO$_3$)$_3$. The resulting solution was evaporated to dryness and baked at 800°C for 8 hours. This step removes any residual chloride, converts the nitrate to oxide and yields plutonium dispersed in an iron-oxide matrix. The resulting iron-oxide was mixed with Ag powder in a 1:4 ratio by mass and pressed into aluminium sample holders.

4.1 AMS of ^{239}Pu

The Accelerator Mass Spectrometry (AMS) technique for detecting Pu ions with the 14UD tandem accelerator at the ANU is well established [29, 30]. Briefly, molecular PuO$^-$ ions extracted from the sample at the ion source were mass-selected by the injection magnet and directed into the accelerator operating at 4 MV. The molecular ions were then accelerated to the high voltage terminal and subsequently dissociated and stripped to Pu^{n+} ions by the gas stripper at a pressure of ~2 mTorr. The multiply-charged atomic ions were then further accelerated to the exit of the accelerator.

A energy analysing magnet selected Pu^{5+} ions of ~ 24MeV, which were then passed through a Wien (velocity) filter and counted individually by the propane-filled ionisation detector. The choice of the 5$^+$ charge state is a trade-off between charge state yield and the ability of the analysing magnet to bend the ions by the required 90°. The transmission efficiency (Pu^{5+}/PuO$^-$) through the accelerator is ~ 3 %. Thorium-232 was used to tune the electric and magnetic focusing elements of the beam transport system, as thorium is close in mass to the plutonium isotopes and is unlikely to lead to any cross-contamination background in the detector.

During a measurement sequence, switching between the different isotopes was effected by changing the injection magnet, terminal voltage and Wien filter settings. Typically, the ^{242,239}Pu isotopes were counted for periods of 1 and 2 min, respectively, and two cycles of the sequence followed by a concluding ^{242}Pu measurement were made for each sample. The mass of soil processed, the mass of ^{242}Pu spike (~4 pg), and the measured ^{239}Pu/^{242}Pu ratios were then used to deduce the ^{239}Pu concentrations.

5 Results and discussion

The effective distribution of particles and the respective ^{239}Pu concentrations across the size fractions of the samples from the forest (site A) and the cultivated (site H) sites are summarised in Table 1 and shown in Figure 2.

5.1 Particle size analysis

Site H is within ~500m of site A and although both soils show similar trends with particle size, there are significant differences in detail. In particular, the distribution in the cultivated soil (H) is lower in the smaller grain sizes. The lower ^{239}Pu levels in the fines of the cultivated site (H) is likely to be a result of the mixing of the upper soil layers (during ploughing) with ^{239}Pu deficient material from the lower levels and/or the loss of ^{239}Pu rich fines in erosion events.

The mass distributions at both sites appear to be normally distributed, with ~60% of the mass distribution being in the 50-250/275 μm median size range. The three finest (<50 μm) fractions contain ~80% of the ^{239}Pu activity in the forest soils and ~40% of the ^{239}Pu activity in the cultivated soil despite only ~19% of the total mass being found in these fractions. It should be noted that the cultivated soil lost 16.5% of its ^{239}Pu activity as a consequence of the loss of fines during the separation process. This activity loss, if added to the measured activity in the finer fractions, would result in a 'true' ^{239}Pu activity of the finer fraction in the cultivated soil of 56.5% of the total, which is closer to but still less than the proportion at the forested site. Above 50 μm, the ^{239}Pu concentrations were found to successively decrease as the grain size increased up to the >420 μm size fraction where the ^{239}Pu concentration increased again. This small increase is likely a consequence of the presence of aggregates of smaller grains in the >420 μm size fraction which were not broken up by the ultrasonic bath. The occurrence of the highest concentrations of fallout tracers in the finer soil size fractions has been observed previously. He and Walling, (1996) [31], for example have reported significant preferential adsorption of ^{137}Cs and ^{210}Pb by the finer (<63 μm) fractions in natural catchment soils. The effect is presumably due to the higher specific surface area per unit volume of finer fractions, and hence the availability of more ionic bonding sites per unit volume.

In addition the organic content and the ^{239}Pu concentration across the various size fractions exhibit a strong positive correlation (r^2>0.96). The >420 μm fractions (from both sites) were not included in the correlation data because of the likely presence of aggregates and macroscopic organic matter (twigs and leaves). Soil organic matter has been shown to preferentially adsorb Pu into its humic and fulvic acid phases (Livens and Baxter, 1988a and 1988b), even if they are present in small amounts. In this work the ^{239}Pu concentrations in the <10 μm fraction of the surface soils at the cultivated site H (WP077) and the forest site A (WP072), Table 1, are used as reference for the sheet eroded fine sediments from the uncultivated (forest) and cultivated areas respectively.

5.2 Fine sediment sources

Sediment budgets are used to characterise sediment inputs (sources), the storage of sediment, and sediment outputs (sinks) to the coast [32]. In the Daly River catchment, fallout tracers (^{137}Cs) have been previously used to identify the sources of fine sediment both in transport and deposited in the floodplains [33, 34]. Wasson *et al.* (2010) [33], identified the major sources of fine sediments in the Daly and Douglas Rivers using ^{137}Cs concentrations in sediments, channel material and surface soils collected in 2005. They estimated between 94-97% of the fine sediment in the Daly River and 91-96% of the fine sediment in the Douglas River was derived from gully/channel bank erosion. The lower end of these estimates were derived using the assumption that the ^{137}Cs-bearing component of the sediment was sourced from cultivated areas while the higher end was based on the assumption that the ^{137}Cs-bearing component of the sediment was sourced from uncultivated (forested) areas. In addition Wasson *et al.* (2010) [33], used OSL dating to identify the post 1975 layers in sediments in benches in the Daly River at Nancar (down stream of the study area in this work). From the ^{137}Cs content of these layers, they determined that 89-92% of the fine sediment in the Daly River between 1975-2005 was sourced from gully/ channel bank erosion. Thus they concluded that overall 89-97% of the fine sediments transported by the Daly River is of subsurface origin i.e., derived from gullying or channel bank erosion.

A similar study also using ^{137}Cs concentrations in surface soils, channel banks and channel sediments in the Daly River was conducted by Caitcheon *et al.* (2011)[34]. They determined the relative contributions of subsoil and surface sediment sources by fitting probability density functions to the ^{137}Cs concentration in the soil and subsoil samples. They estimated that 99% of the Daly River sediments are of sub-surface origin, further corroborating the higher end of the estimate by Wasson *et al.* (2010)[33].

In this study [239]Pu in sediments and channel material from the three major streams which occur in the study area, namely the Daly and Douglas Rivers and Stray Creek, were measured to see if [239]Pu could be used as a sediment source tracer in a similar fashion to [137]Cs. The locations of the sediment sampling sites are shown in Figure 1. Stray Creek sediments had the lowest [239]Pu concentration at 7.7 ± 0.5 μBq/g. Further downstream on the Daly River the fine sediment [239]Pu concentration increases to 18.5 ±1.6 μBq/g. The [239]Pu concentrations in sediments near the Douglas side of the confluence of the Douglas and Daly rivers was 23.6 ± 1.6 μBq/g, which is slightly larger but similar to the [239]Pu concentrations in Daly River sediments. The [239]Pu concentrations indicate that the areas under cultivation and land clearing activities in the Douglas Daly region have increased the surface soil contribution to the river networks with slightly more surface sediment entering the Douglas River compared to the Daly River. In contrast, in Stray Creek catchment, which has limited cultivation and is dominated by grazing, the [239]Pu concentration is the lowest.

Table 1. [239]Pu activities and in various soil size fractions in surface soil samples from forest (A) and cultivated (H) areas of the Daly River catchment.

Particle Size range (μm)	Organic (mg/g)	[239]Pu (μBq/g)	Recovered [239]Pu (μBq/g)	Recovered[1] [239]Pu (%)	Mass[1] (%)
1. A, 3-8 (cm)					
<10	137.1	361.4 ± 20.3	17.3 ± 1.0	34.3	4.8
10-25	112.2	305.7 ± 20.5	9.2 ± 0.6	18.2	3.0
25-50	60.1	113.1 ± 7.2	12.7 ± 0.8	25.1	11.2
50-100	16.9	26.9 ± 1.7	5.8 ± 0.4	11.6	21.7
100-250	7.0	9.9 ± 1.3	3.8 ± 0.5	7.5	38.5
250-420	7.1	6.1 ± 0.8	1.0 ± 0.1	1.9	15.8
+420	225.9	36.3 ± 4.7	0.7 ± 0.1	1.4	2.0
Total			**50.5 ± 1.5**	**100**	**97.0**
A. 3-8 (cm)			**50.5 ± 2.7**		
2. H, 0-10 (cm)					
<10	73.6	84.3 ± 7.2	2.8 ± 0.2	10.1	3.3
10-25	69.9	81.2 ± 8.2	3.6 ± 0.4	12.9	4.4
25-50	42.9	38.5 ± 3.1	4.3 ± 0.3	15.3	11.1
50-100	22.1	21.6 ± 1.6	5.8 ± 0.4	20.6	26.7
100-275	26.8	10.5 ± 1.3	3.4 ± 0.4	12.1	32.2
420-275	20.8	15.4 ± 2.8	0.9 ± 0.2	3.1	5.6
+420	56.6	32.3 ± 4.2	2.5 ± 0.3	9.0	7.8
Total			**23.2 ± 0.9**	**83.1**	**91.1**
H. 0-10 (cm)			**27.9 ± 1.5**		

Note: [1]The percentages do not add to 100 due to losses in the separation process. The recovered [239]Pu is per g of soil fraction whereas the [239]Pu concentration is per g of soil.

Table 2. Sediment source contribution estimates from ^{239}Pu concentrations in the river sediments of the Douglas Daly region.

Site	Sub-catchment	^{239}Pu (µBq/g) Sediment	^{239}Pu (µBq/g) Soil[4]	Gully River/ Bank erosion (%)			
				^{239}Pu (2009)[1]	^{137}Cs (2005)[2]	^{137}Cs (1975-2005)[2]	^{137}Cs (2011)[3]
P	Daly River	18.5 ± 1.6	361 ± 20 (A) 84.3 ± 7.2 (H)	78-95	94-97	89-92	99
Q	Douglas River	23.6 ± 1.6	361 ± 20 (A) 84.3 ± 7.2 (H)	72-93	91-96	n.d.	n.d.
O	Stray Creek	7.7 ± 0.5	361 ± 20 (A) 84.3 ± 7.2 (H)	91-98	n.d.	n.d.	n.d.

[1]Gully /river bank erosion (%) = $(1-^{239}\text{Pu}_{sediment}/^{239}\text{Pu}_{soil}) \times 100$. [2]Wasson *et al*. (2010). [3]Caitcheon *et al*. (2012). [4]The ^{239}Pu activity in the <10µm (fine) fraction of the surface soils (0-10 and 0-3cm) from a cultivated (H) and a forest (A) site were used as reference for the sheet eroded fine sediments from the Douglas Daly area.
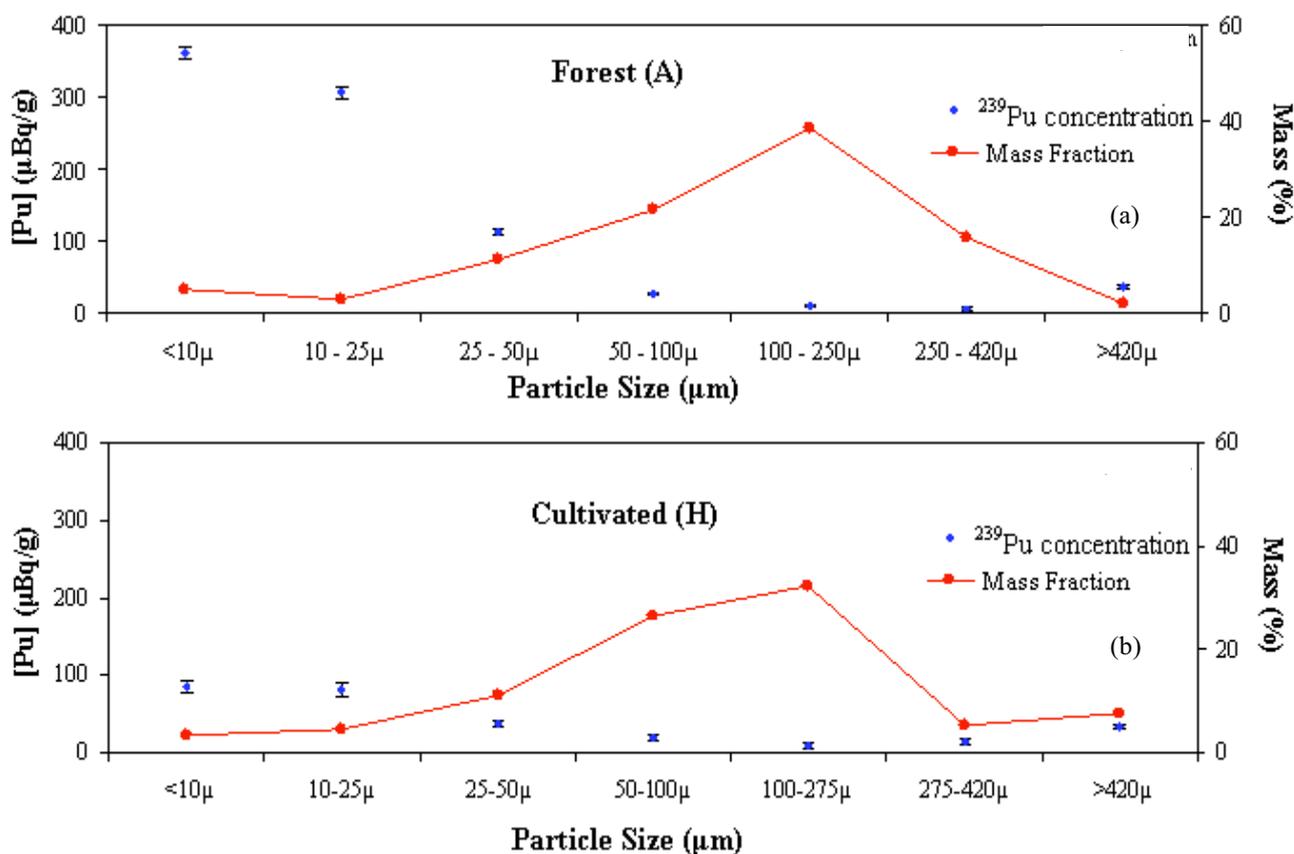

Fig. 2. ^{239}Pu activities in various soil size fractions in surface soils samples and distribution by weight of the bulk soil sample amongst the various particle size ranges from (a) forest (A) and (b) cultivated (H) areas of the Daly River catchment.

The percentage contributions from surface and channel sources to the sediment load in the three stream systems is determined below. The estimated fine sediment contribution to the river network of the ^{239}Pu deficient sub-surface layers from gully and river bank erosion are shown in Table 2. As noted earlier (*section 5.1*) the ^{239}Pu concentrations (84.3 ± 7.2 and 361.4 ± 20.3 µBq/g) in the <10 µm fraction of the surface soils (0-10 and 3-8 cm) at the cultivated site H (WP077) and the forest site A(WP072), Table 1, are used as reference for

the sheet eroded fine sediments from the uncultivated (forest) and cultivated areas respectively. The sediment source estimates were derived using the assumption that the ^{239}Pu bearing component of the sediment was sourced either from cultivated areas or from uncultivated (forested) areas. The actual contribution will be intermediate as it includes both sources. Stray Creek fine sediments have the highest source contribution from gully and channel bank erosion at 91-98 %, followed by the Daly River at 78-95% and Douglas River at 72-93%.

Overall the ^{239}Pu derived contributions of fine sediment from gully and channel banks in the Daly River of 78-95% compares well with the 89-92% long term average of Wasson *et al.* (2010).

Furthermore, the results of this study indicate surface soil input from sheet erosion of 5-22% for the Daly River and 7-28% for the Douglas River in 2009 *vs*. 3-6% for the Daly River and 4-9% for the Douglas River in 2005 estimated by [33]. This suggests (i) surface soil contributions along the Douglas River channel are slightly higher relative to the Daly River channel and (ii) surface soil contributions from the cleared land adjacent to the Daly River have increased since 2005, likely because of changes in land use i.e. increase in land clearing and cultivation activities since the earlier study. Moreover there was a huge sheet and rill erosion event in the study area in January 2008 and a few months later another [35]. This occurred as a result of heavy rainfall and likely enhanced surface soil input in the river which suggests that the timing of sampling relative to rainfall events is also important.

6 Conclusions

^{239}Pu has been used to identify sediment source contributions arising from gully/channel bank erosion. The gully/channel bank sediment contributions were estimated from comparison of ^{239}Pu concentrations in fine channel sediment with ^{239}Pu concentrations in fine fractions of surface soils from cultivated and forest sites. The gully/channel bank sediment contributions range from 78-95%, 72-93% and 91-98% for the Daly River, Douglas River and Stray Creek networks respectively. The lower end of these estimates is derived assuming that all of the ^{239}Pu-bearing sediments were sourced from cultivated fields while the higher end estimate assumes that all of the ^{239}Pu-bearing sediment was from forest areas (the actual situation would likely be intermediate). Moreover these estimates are comparable to, albeit with slightly lower limits than, estimates derived from earlier (sampling in 2005) ^{137}Cs studies. This indicates that there may be a slight increase in delivery of eroded surface soils to the river network as a consequence of sheet and rill erosion events and land clearing in the intervening period between the sampling (2005-2009) i.e. the clearing of forests and reduction of the riparian vegetation cover resulting in more sediments reaching the channels. However the dominant source of sediment in these rivers is still overwhelmingly from channel bank erosion.

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