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To cite this version:

Alexandra Garzon-Garcia, J. Patrick Laceby, Jon Olley, Stuart Bunn. Differentiating the sources of fine sediment, organic matter and nitrogen in a subtropical Australian catchment. Science of the Total Environment, 2017, 575, pp.1384-1394. 10.1016/j.scitotenv.2016.09.219 . cea-02648527

HAL Id: cea-02648527
https://hal-cea.archives-ouvertes.fr/cea-02648527

Submitted on 24 Jun 2021

Differentiating the sources of fine sediment, organic matter and nitrogen in a subtropical Australian catchment

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Abstract

Understanding the sources of sediment, organic matter and nitrogen (N) transferred from terrestrial to aquatic environments is important for managing the deleterious off-site impacts of soil erosion. In particular, investigating the sources of organic matter associated with fine sediment may also provide insight into carbon (C) and N budgets. Accordingly, the main sources of fine sediment, organic matter (indicated by total organic carbon), and N are determined for three nested catchments (2.5 km^2, 75 km^2, and 3076 km^2) in subtropical Australia. Source samples included subsoil and surface soil, along with C_3 and C_4 vegetation. All samples were analysed for stable isotopes ($\delta^{13}C$, $\delta^{15}N$) and elemental composition (TOC, TN). A stable isotope mixing model (SIAR) was used to determine relative source contributions for different spatial scales (nested catchments), climatic conditions and flow stages. Subsoil was the main source of fine sediment for all catchments (82%, SD = 1.15) and the main N source at smaller scales (55-76%, SD = 4.6-10.5), with an exception for the wet year and at the larger catchment, where surface soil was the dominant N source (55-61%, SD = 3.6-9.9), though contributions were dependent on flow (59-680 m^3/s). C_3 litter was the main source of organic C export for the two larger catchments (53%, SD = 3.8) even though C_4 grasses dominate the vegetation cover in these catchments. The sources of fine sediment, organic matter and N differ in subtropical catchments impacted by erosion, with the majority of C derived from C_3 leaf litter and the majority of N derived from either subsoil or surface soil. Understanding these differences will assist management in reducing sediment, organic matter and N transfers in similar subtropical catchments while providing a quantitative foundation for testing C and N budgets.

Key words: carbon, nitrogen, erosion, vegetation litter, stable isotopes, fingerprinting

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1 Introduction

Soil erosion is a major source of sediment to aquatic systems (Cerdan et al., 2010; Milliman and Meade, 1983). Erosion processes may also influence the transfer of carbon, potentially playing a major role in carbon budgets (Cole et al., 2007; Lal, 2003; Scott et al., 2006). The transfer of nitrogen to aquatic systems is also affected by erosion processes (Quinton et al., 2010). In Australia, land use change following European settlement triggered significant gully and channel erosion (Bartley et al., 2006; Olley and Wasson, 2003; Prosser and Slade, 1994). Research on tropical and subtropical systems in eastern Australia has demonstrated that subsoil erosion (i.e., gully and channel erosion) is the dominant source of sediment (Caitcheon et al., 2012; Hughes et al., 2009; Olley et al., 2013a; Olley et al., 2013b). Although the significance of subsoil erosion sources for sediment in this region is well-documented, the sources of organic matter and nutrients have received less attention.

Carbon and nitrogen stable isotope ratios ($\delta^{13}C$, $\delta^{15}N$) and elemental composition have been widely used to determine the sources of organic matter supporting food webs in aquatic environments (Bunn et al., 2003; Finlay, 2001; Hein et al., 2003). They have also been used extensively to trace the contributions of catchment sources to sediment (Laceby et al., accepted; Mukundan et al., 2010; Papanicolaou et al., 2003) and to in-stream particulate organic matter (Cooper et al., 2015; Kendall et al., 2001; McCorkle et al., 2016). $\delta^{13}C$ discrimination of sources is derived primarily from photosynthetic pathways that result in distinct $\delta^{13}C$ fractionations. The majority of tree and temperate grass species follow the Calvin-Benson cycle (C_3) photosynthetic pathway ($\delta^{13}C$: mean -28‰) (Boutton, 1991; Fry, 2006; Schimel, 1993). Plants following the Hatch-Slack cycle (C_4) pathway consist mainly of grass and cropping species primarily found in warmer climates with limited water availability ($\delta^{13}C$: mean -13‰) (Coleman and Fry, 1991; Werth and Kuzyakov, 2010). The Crassulacean acid metabolism (CAM) plants have the potential to utilize both C_3 and C_4 pathways (Werth and Kuzyakov, 2010). $\delta^{13}C$ should discriminate between material derived from C_3 and C_4 plants in tropical and subtropical environments. Most of the nitrogen in the biosphere is atmospheric N_2 which has a near constant $\delta^{15}N$ of 0‰ (Peterson and Fry, 1987). The majority of nitrogen in the rest of the biosphere has $\delta^{15}N$ values between -10‰ to +10‰. In general, $\delta^{15}N$ fractionation is complex, with a multitude of nitrogen sources and internal transformations potentially altering nitrogen isotopic ratios (Evans, 2007; Finlay and Kendall, 2007; Shearer and Kohl, 1993).

Here, we use $\delta^{13}C$, $\delta^{15}N$, total organic carbon (TOC), and total nitrogen (TN) to quantify the sources of organic matter and total nitrogen associated with fine particulate export (<63 µm) in a subtropical riverine system. First, we test whether $\delta^{13}C$, $\delta^{15}N$, total organic carbon (TOC), and total nitrogen (TN) can be used to distinguish between organic matter derived from the four primary sources: subsoils, intact valley soils, C_3 litter and C_4 litter. These sources were identified as the most likely to contribute to fine particulate organic matter export during flow events in the area. Second, the relative contributions of these sources are determined at three different spatial scales in a series of nested catchments using a modified sediment source fingerprinting approach (e.g. Collins and Walling, 2004; Davis and Fox, 2009; Koiter et al., 2013b). Third, variations in contributions from each of the sources between wet and dry years and base and event flow conditions are examined.

2 Methods

2.1 Study Region

The study was conducted in three nested catchments in Southeast Queensland, Australia: the Logan River catchment (3076 km^2), the Knapp Creek catchment (75 km^2) and the Tilley Gully catchment (2.5 km^2) (Figure 1). Total annual rainfall in the region is variable, ranging between 500 and 1400 mm y^{-1} (Kooralbyn station (BoM, 2013)), with the majority falling during the summer wet season. Mean annual monthly temperatures range between 12.6 and 26.5ºC.

Land use in the Logan River catchment is predominantly grazing (89%), native forest (7%) and cropping (2%) (Figure 1). The Logan River catchment geology consists of basalt (32%), arenite-mudrock (19%), arenite (17%), and alluvium (12%) (DME, 2008). Land use in the Knapp Creek catchment is also dominated by grazing (78%) with less than 22% of the native vegetation cover remaining. The geology of Knapp Creek primarily consists of arenite sandstone (81%) (DME, 2008). The majority of sediment produced in the Logan River catchment (Hancock and Revill, 2013) and the Knapp Creek subcatchment is derived from subsurface erosion sources (Laceby et al., 2015; Olley et al., 2009). There is extensive gully erosion throughout Knapp Creek, with at least 38 km of gullies. Most of this erosion is evident in the mid-catchment reaches where gullies are well-connected to the main channel (Olley et al., 2009). The Tilley Gully catchment, located in these mid-reaches (Figure 1), is one of the top three sediment-yielding catchments in Knapp Creek, with an estimated sediment yield of more than 1000 T y^{-1}. This is one sixth of the ~6000 T/y produced by gully erosion in the whole catchment (Olley et al., 2009). The catchment has been cleared for cattle grazing with less than 20% of the original forest cover remaining. The Tilley Gully catchment is underlain by arenite sandstone (100%).

2.2 Source sampling

Source sampling includes the sampling of the primary sources of particulate material that may be potentially mobilized by rainfall events and transported downstream. Four potential sources were identified and sampled: subsoils (i.e. gully and channel banks), intact valley “non-gullied” surface soils, valley and gully grasses, and tree litter (fine sticks and leaves). These sources were selected after an extensive literature review (Hancock and Revill, 2013; Laceby et al., 2015; Olley et al., 2009), field investigations, and discussions with catchment managers and land-owners. Source sampling details are summarized in Table 1. Gully banks were sampled at five locations, separated by ~400 m, along Tilley Gully in April 2012 (Figure 1). Samples were taken from three horizontal strata differentiated by colour on bare gully banks. In total, 15 gully samples were collected. Channel banks were sampled in December 2013 at five locations along Knapp Creek (Figure 1). At each location, two samples were composited for analysis, one near the base of the bank and the other near the top of the bank. Gully and channel bank samples were taken with an 18 cm^2 corer to a 10 cm depth.

Sampling locations for intact valley soils, valley and gully grasses, and tree litter were randomly selected from a 2.5 x 2.5 m grid covering an intact valley and an incising gully in the Tilley Gully catchment (Figure 1). Sample location was selected with the Sampling Design Tool for ArcGIS (10.0). The top 2 cm of intact valley soils were sampled with a trowel after vegetation was removed to ground level, between July and October 2011, in the dry season. In total, 24 samples were collected (Figure 1).

Table 1. Summary of source and exported fine sediment sampling and preparation prior to elemental and isotope analysis

Sample name	Type of sample	Date of samples	Sample number	Bulking	Subsamples analyzed	Location	Tumble time	Sieve type and fraction size	Freeze dried
Intact valley soil (source)	Soil/sediment 0-2 cm	July-October 2011	24	N/A	7	Tilley Gully intact valley	1 hour	Wet sieved <63 um	N/A
Gully bank subsoil (source)	Soil/sediment 0-10 cm	April 2012	15	N/A	7	Tilley Gully banks	1 hour	Wet sieved <63 um	N/A
Channel bank subsoil (source)	Soil/sediment 0-10 cm	December 2013	5	Compositing 2 samples distributed vertically	5	Knapp Creek banks	1 hour	Wet sieved <63 um	N/A
Grass litter (source)	Vegetation litter	July-October 2011	37	N/A	7 from gullies 7 from intact valleys	Tilley Gully intact valley and gully	1 hour	Ground <0.5 mm	N/A
Tree litter (source)	Vegetation litter	July-October 2011	7	NA	7	Tilley Gully gully	1 hour	Ground <0.5 mm	N/A
Tilley Gully sediment (export)	Time-integrated sediment sample	December 2011-February 2012	10	Integrated wet season	10	Tilley Gully (3 sampling sites, 2 vertical positions, 2 replicates)	N/A	Dry sieved <63 um	N/A
Knapp Creek sediment (export)	Refrigerated autosampler	January 2010-October 2010	39	N/A	39	Knapp Creek (1 sampling site)	N/A	Settling columns <10um and wet sieved <63um post-settling	N/A
Logan River sediment (export)	Refrigerated autosampler	January 2013 – February 2013	10	N/A	10	Logan River (1 sampling site)	N/A	N/A	Freeze dried

Grasses (i.e., valley and gully) and tree litter were sampled between July and October 2011. At each sampling location (Figure 1), standing grass was cut at the ground surface level. Thereafter, leaf and woody litter were removed from a 0.20 x 0.20 m quadrant and packed separately into paper bags. In total, 37 grass samples were collected. Seven samples of tree litter were collected from the gully sites. No tree litter was present in the intact valley.

All soil and sediment samples were packed in plastic bags and transported on ice to the laboratory for analysis. Twelve subsoils (channel and gully banks), 7 intact valley soils, 7 gully grass, and 7 intact valley grass samples were analysed with preparation methods described below.

2.3 Sediment sampling

Sediment sampling includes the sampling of the particulate material that is being transported in suspension during high flow events. Time-integrated samplers (Phillips et al., 2000) were installed to collect representative fine particulate material from flow events that occurred during the 2011-2012 wet season in the Tilley Gully catchment. Sampling sites were located in two headwater gullies and at the outlet of this catchment (Figure 1). At each site, four time-integrated samplers were installed. They were installed at two different heights on both sides of the gullies. These sampling site locations were selected to understand the variability of source contribution at smaller scales (gully units and different gully depths) and to have replicate samples for each gully section depth. The samples were collected at the end of the wet season. In total, 10 samples were collected and analysed. At one site (G2) only the lowest samplers were inundated. Table 1 provides a summary of all sediment sampling information.

Fine sediment samples were collected from lower Knapp Creek during the 2009-2011 hydrologic years, using a refrigerated autosampler (Figure 1). The 2009-2010 hydrologic year had below average rainfall (658 mm) whereas the 2010-2011 hydrologic year had above average rainfall (1341 mm) (BoM, 2013). This refrigerated autosampler captured 5 and 4 flow events in each of these sampling years, respectively. The number of samples collected during an event was based on event duration and water level. The autosampler was triggered to start sampling when water levels rose 10 cm above the base flow level at each site and samples were subsequently taken at fixed time intervals. In total, 39 samples were collected and analysed from this autosampling station.

In the Logan River catchment, ten sediment samples were collected for analysis, also using a refrigerated autosampler. The sampling site was located 57 km downstream from Knapp Creek's junction with the Logan River (Figure 1). Samples were taken between January 27 and February 1, 2013 and captured a high-flow event (366 mm of rainfall in 8 days, with a peak flow rate of 678 m^3/s in the Logan River).

2.4 Sample Preparation

Samples collected to characterise the potential sources were processed using a method adapted from Gregorich *et al.* (2006) with sample processing details summarized in Table 1. The approach was designed to mimic stream transport processes and to remove physically uncomplexed organic matter (not bound to mineral particles) from the soil and subsoil.

Initially soil and subsoil samples were passed through an 8 mm sieve to remove large root biomass and litter, oven dried at 40°C and sieved (<2 mm) to remove large litter fragments and gravel. Then a subsample (20-30 g) was shaken in a tumbler for an hour with 100 mL of milli-Q water, suspended litter particles were removed with a vacuum pump and the remaining suspension was wet sieved (63 μm) to recover a water sample with suspended fines which was oven dried at 60°C for 48 h and hand-milled prior to analysis. Analysis was carried on the <63 μm particle size material.

Grass and tree litter samples were oven dried for at least 48 h at 60°C and ground (<0.5 mm). A 3 g subsample was shaken in a tumbler for an hour with 100 mL of milli-Q water, water was removed and grass and tree litter were redried at 60°C before C and N analysis.

Sediment samples from Tilley Gully were oven-dried at 60°C, dry-sieved to <63 μm and hand-milled prior to analysis. Knapp Creek automated event samples were fractionated (<10μm) with settling columns based on Stokes' Law. These samples were wet-sieved (63 μm) to remove large particulate organic matter post settling, oven dried at 60°C for 48 h, and ground with a stainless steel ball-mill grinder prior to analysis. The Logan River refrigerated event samples were frozen and freeze dried prior to analysis on the recovered material.

2.5 Isotope and elemental analytical methods

All samples were pelletized for TN and $\delta^{15}N$ analysis. For TOC and $\delta^{13}C$, samples were treated repeatedly with a 10% HCl solution to remove carbonates until there was no visual evidence of effervescence. Following the HCl treatment, samples were oven dried at 60°C for 48 h, pelletized, and weighed for analysis. Samples were combusted in a Sercon Europa elemental analyser with sample gas delivered to a Sercon Hydra isotope-ratio mass spectrometer at the Australian Rivers Institute, Griffith University, Nathan Campus, Brisbane.

$\delta^{13}C$ and $\delta^{15}N$ values are reported in per mil (‰) relative to Pee Dee Belemnite (PDB) standard and relative to air N_2, respectively. The precision of $\delta^{13}C$ was monitored with a sucrose standard over 20 analysis runs reporting $\delta^{13}C$ = -11.7 ‰ (SD = 0.1, n = 84) and of $\delta^{15}N$ with an IAEA-305a surrogate standard reporting $\delta^{15}N$ = 0.2 ‰ (SD = 0.3, n = 84). The precision of TOC and TN, reported in percent weight of dry sample (%) was monitored using an Acetanilide elemental standard over 20 analysis runs reporting TOC = 3.4% (SD = 0.1, n = 84) and TN = 0.33% (SD = 0.005, n = 84). All data used in these analyses are provided in the supporting material (S1-S3).

2.6 Statistical analysis and modelling

The potential of $\delta^{13}C$, $\delta^{15}N$, TOC and TN to discriminate between the sources was assessed using T-tests and Mann-Whitney U-tests. The Mann-Whitney U-test was used for non-parametric data and the ANOVA was used for comparisons of data with equal means and variance.

Isotopic and elemental data were modelled with SIAR V4 (Stable isotope analysis in R) (Parnell et al., 2010) to quantify the source of the recovered material collected during the flow events. SIAR uses Bayesian isotopic mixing models and model fitting with Markov chain Monte Carlo (MCMC) simulations of plausible values consistent with the data (n = 30000). Outputs include posterior distributions that represent a true probability density for the mixing contribution of the sources and an overall residual term (Parnell et al., 2010).

Source contributions to sediment (both inorganic and organic fractions) export for the nested catchments were first modelled in SIAR using $\delta^{13}C$, $\delta^{15}N$, TOC and TN with the concentration dependency of $\delta^{13}C$ and $\delta^{15}N$ corrected within the SIAR model. Second, to determine the relative contributions to organic matter (as indicated by TOC) and TN from the different sources, the contribution of each source to the exported TOC and TN was calculated with the SIAR model outputs as follows:

$$\%Esource_i = \frac{E\ source_i \times \%cont_i}{\sum_{i=1}^{i=4} Esource_i \times \%cont_i} \times 100 \qquad (1)$$

with $\%E\ source_i$ being the contribution of source i to TOC or TN with i varying from 1 to 4 to include all the 4 sources evaluated; $E\ source_i$, the mean TOC or TN content of source i obtained from elemental analysis of source samples and $\%cont_i$ the mean percent contribution of source i to sediment export as obtained from SIAR model outputs. The propagated standard deviation for each source TOC and TN contribution was calculated using SGUM v0.96 (Hall, 2010).

Data from the Tilley Gully catchment were grouped in a mixing model to obtain the distributions of organic matter (TOC) and TN source contributions for different sampling sites within this catchment. Mean and grouped standard deviations for the catchments are reported in the results and discussion. To understand the effects of climatic conditions, source contributions from a dry and a wet hydrologic year in the Knapp Creek catchment were compared. The mean and the standard deviation of these distributions are reported for individual source contributions in the results and discussion. Data from the high-flow event in the Logan River in 2013 are used to analyse the effect of flow stage on sediment, organic matter (TOC) and TN source contributions. For this catchment, each stage sample was modelled individually. The mean and the standard deviation of these distributions are reported for individual source contributions in the results and discussion. Differences in mixing model outputs for different climatic conditions were determined using ANOVA and Mann-Whitney U-tests. Statistical analyses were performed using R.3.0.1 and SigmaPlot 11.0 with statistical significance determined at the $\alpha = 0.05$ level.

3 Results

3.1 Source Discrimination

The source discrimination potential of the measured properties was tested prior to modelling. In combination, $\delta^{13}C$, $\delta^{15}N$, TOC and TN discriminate between all the different sources (Table 2). $\delta^{13}C$ discriminates between all sources, except intact valley soil and C_4 litter. $\delta^{15}N$ discriminates between litter and subsoil, and between litter and intact valley soil, with the two latter having higher $\delta^{15}N$ values than C_3 or C_4 litter. TOC discriminates between all sources, with C_4 litter having the highest TOC content and subsoil the lowest. TN discriminates between subsoil and the other sources, but not between litter types or intact valley soil and C_3 litter. Subsoil had a mean $\delta^{13}C$ between C_3 and C_4 vegetation litter. These four sediment properties in combination provide complete discrimination amongst all the sources and accordingly all sediment properties will be modelled in SIAR. Source samples are plotted with sediment samples in Figure 2.

3.2 Tilley Gully

Results for all Tilley Gully catchment sampling sites have been averaged as the variability between sampling sites was low (see standard deviation plotted in Figure 3). Results for individual sampling sites are provided in the supporting material (S4). Subsoil was the dominant source of exported sediment in Tilley Gully catchment during the wet season (2011-2012). The mean subsoil contribution to sediment was 97% (SD = 1) for all sampling sites. Intact valley soil, C_3 litter and C_4 litter were minor sediment sources. Subsoil also contributed the most to organic matter export as indicated by TOC export (60%, SD = 6), and to TN (76%, SD = 5) in the Tilley Gully catchment (Figure 3). C_3 litter contributed 21% (SD = 6) of TOC, followed by intact valley soil which contributed 11% (SD = 3). For TN, intact valley soil contributed 15% (SD = 4) while the other sources were insignificant (<8%). The Tilley Gully model results are provided as supporting material (S4).

3.3 Knapp Creek

There were significant differences in source contributions to exported sediment between the dry (2009-2010) and wet (2010-2011) years in Knapp Creek (Figure 4). Subsoil was the largest source of sediment for both climatic conditions with mean sediment contributions ranging from 72 to 94% (SD = 1 to 5) followed by intact valley soil ranging from 3 to 24% (SD = 1 to 6) (Figure 4). Intact valley soil contributions were significantly larger (21%) ($p < 0.001$) for the samples collected in the wet year. All Knapp Creek model results are provided in the supporting material (S5).

While subsoil was the dominant source of sediment, it contributed less than 31% (SD = 5 to 9) of the organic matter (as indicated by TOC) during events. This reflects the low organic matter content of subsoil compared to other sources. There was a significantly lower contribution from subsoil to organic matter export (TOC) in the wet year ($p < 0.001$) (Figure 4). C_3 litter contributed on average 40 to 60% (SD = 9 to 10) of the organic matter export, with a significantly larger contribution occurring in the dry year ($p < 0.001$) (Figure 4). Intact valley soil and C_4 litter contributed on average 8 to 36%, and 1 to 11%, respectively. Their contributions varied similarly, significantly increasing ($p < 0.001$) for the wet year when the intact valley soil contributed a similar proportion than C_3 litter to organic matter export (Figure 4).

The dominant source of TN varied between the wet and the dry year with subsoil being the main source in the dry year (55%, SD = 11) and intact valley soil the most dominant source in the wet year (61%, SD = 10) (Figure 4). C_3 litter contributed between 16 and 29% (SD = 7 to 10), and C_4 litter contributions were negligible. In wet years, intact valley soil contributions were significantly larger ($p < 0.001$) than in the dry year (45% larger) (Figure 4).

Table 2. $\delta^{13}C$, $\delta^{15}N$, TOC and TN of most probable sources during flow events (subsoil, intact valley soil, C_3 litter and C_4 litter) and statistical analysis results for differences in $\delta^{13}C$, $\delta^{15}N$, TOC and TN between sources. T-tests (T) or Mann-Whitney U-test results are presented (statistical significant differences determined at $p < 0.05$).

Source	**$\delta^{13}C$ (‰)**	**SD**	**Subsoil**	**Intact valley soil**	**C_3 litter**	**C_4 litter**
Subsoil	-18.9	1.6				
Intact valley soil	-14.3	0.9	***			
C_3 litter (tree)	-25.6	3.3	***	***		
C_4 litter (grass)	-13.5	1.1	***	-	***	
	$\delta^{15}N$ (‰)	**SD**	**Subsoil**	**Intact valley soil**	**C_3 litter**	**C_4 litter**
Subsoil	5.6	2.6				
Intact valley soil	3.4	0.8	- (t)			
C_3 litter (tree)	-1.5	1.0	*** (t)	*** (t)		
C_4 litter (grass)	-2.4	2.7	*** (t)	*** (t)	- (t)	
	TOC (%)	**SD**	**Subsoil**	**Intact valley soil**	**C_3 litter**	**C_4 litter**
Subsoil	0.6	0.2				
Intact valley soil	5.0	0.6	***			
C_3 litter (tree)	32.1	8.1	***	***		
C_4 litter (grass)	40.7	5.3	***	***	*	
	TN (%)	**SD**	**Subsoil**	**Intact valley soil**	**C_3 litter**	**C_4 litter**
Subsoil	0.05	0.02				
Intact valley soil	0.51	0.11	***			
C_3 litter (tree)	0.80	0.32	***	-		
C_4 litter (grass)	0.84	0.30	***	**	-	

(t) T-test, (-) Not significant, (*) Significant at $p < 0.05$,
(**) Significant at $p < 0.01$, (***) Significant at $p < 0.001$

3.4 Logan River

Subsoil contributed the majority of sediment sampled in the Logan River catchment for all flow stages, with the exception of the first sample (Figure 5a). Subsoil mean contributions to exported sediment increased with flow, with the largest contribution occurring just after peak flow and high contributions occurring during the receding limb of the hydrograph (mean > 60%, SD = 3 to 7) (Figure 5a). Intact valley soil was the second largest contributor with the largest contribution at the start of the high-flow event (mean 47%, SD = 11), and the lowest just after peak flow (4%, SD = 3) (Figure 5a). C_3 litter had a similar trend, with the largest contribution at the start of the high-flow event of 21% (SD = 6), and the lowest just after peak flow. C_4 litter contributions were minimal (<1%) (Figure 5a). The Logan River model results are included in the supporting material (S6).

C_3 litter was the main contributor of organic matter as indicated by TOC for all samples collected during the high-flow stages, with the largest contribution at the start of the event (70%, SD = 10), and a gradual decrease reaching the lowest contribution (45%, SD = 11), for the last sample taken (Figure 5b). Intact valley soil was the second largest source of organic matter (Figure 5b) (10 - 43%, SD = 7 to 10). C_4 litter contributed less than 5% (Figure 5b).

Intact valley soils were the main source of TN (mean = 55%, SD = 4) (Figure 5c), followed by C_3 litter (mean = 26%, SD = 3) with the exception of when subsoil contribution peaked (Figure 5c). C_4 litter was again a minor source (<5%). Subsoil contributions to fine particulate organic matter (TOC) and TN export increased with flow, reaching their highest value just after peak flow (25 and 46%, SD = 9 and 12) when they were the second largest and largest source, respectively (Figure 5b,c).

4 Discussion

Soil erosion exports large quantities of organic matter downstream (Lal, 2003; Ludwig and Probst, 1996; Scott et al., 2006), redistributing landscape carbon and nitrogen pools laterally, vertically, and/or longitudinally from catchments to the marine environment (Gregorich et al., 1998; Ma et al., 2016; Ran et al., 2014). Recent research conducted on tropical and subtropical river systems in Australia has shown that subsoil erosion (gully and channel erosion) is the dominant source of sediment entering these waterways (Caitcheon et al., 2012; Hughes et al., 2009; Olley et al., 2013a). Although subsoils contribute the majority of sediment, the modelling of source and in-stream particulate material with $\delta^{13}C$, $\delta^{15}N$, TOC and TN indicated that subsoils are not always the dominant source of fine particulate organic matter and nitrogen (TOC, TN).

$\delta^{13}C$, $\delta^{15}N$, TOC and TN were successfully used as complementary tracers of source contributions to fine particulate material export from intact “non-gullied” valley soils, subsoils, C_3 litter and C_4 litter. TOC discriminated between all sources. TN discriminated subsoils from other sources, with the former having a significant lower content, though not between litter types or intact valley soil and C_3 litter. $\delta^{13}C$ did not discriminate between C_4 litter and intact valley soil, which derives its carbon input mainly from this type of litter. Finally, $\delta^{15}N$ discriminated between litter and soils with the latter having significantly higher $\delta^{15}N$.

Issues associated with the conservativeness of biochemical properties (including $\delta^{13}C$, $\delta^{15}N$, TOC and TN) used for sediment fingerprinting have been raised in recent research (Koiter et al., 2013a; Laceby et al., 2015). Changes in biochemical properties occurring when fine particulate matter is eroded from source soils and directly transported downstream would be mainly caused by some biological processing (e.g., mineralization) taking place at first contact with water. To take into account the possible alterations of the biochemical properties of fine particulate sources due to rapid biological processing, source samples were processed for elemental and isotopic analysis in a manner designed to mimic stream transport. This was done to remove physically uncomplexed organic matter (e.g litter, plant remains) from the mineral fraction (Gregorich et al., 2006) in order to be able to use it as a “pure” end member, and to simulate fast biological processing that takes place on short time scales (1-2 days) that may change source isotopic and elemental composition. As we are fingerprinting sediment transported during high flow events, it is likely that no significant additional changes on biochemical properties would occur other than those mimicked in the lab.

Importantly, the main sources of sediment, organic matter and of TN are different. Subsoil is clearly the main sediment source (mean = 82%, SD = 1). C_3 litter is the main organic matter source (mean = 42%, SD = 3). Subsoil is the main TN source (mean = 44%, SD = 3), with exceptions for the autosamplers in the wet year and larger catchment where intact valley soil was the main TN source (mean = 61%, SD = 10 and mean = 55%, SD = 4 respectively). This disproportionally large contribution of C_3 litter to organic matter and intact valley soil to TN, relative to their contribution to sediment, occurs because of the larger TOC content in litter biomass and larger TN content in intact valley soil compared to subsoil.

4.1 Controls on source contribution: Spatial scale

Subsoil was the main source of exported sediment varying from 49% during low flow conditions in the largest catchment to 97% in the smallest catchment. Differences in subsoil contributions to sediment export between catchments is likely related to the severity of erosion and to the magnitude of flows sampled. C_3 litter was the main source of organic matter export (as indicated by TOC export), varying from 21% in the smallest catchment to 70% at low flows in the largest catchment. This contribution may be explained by the higher TOC% in litter biomass compared to subsoil: 49 times more on average. Differences in source contributions to organic matter export between catchments were most likely due to tree cover density in the riparian area.

Although most of these catchments have been cleared of trees, tree litter contributes a larger proportion than grasses and intact valley soil to organic matter export, even though C_4 grasses grow densely within headwater gullies. C_3 inputs from riparian trees have a higher connectivity to the stream network than grassland litter, and riparian areas are likely to have higher net primary production than grasslands in the upper catchments, which may explain this disproportionate contribution to organic matter export. It is also probable that cattle remove large quantities of grass biomass reducing the amount of C_4 litter present and available for export. Further, pasture has a higher contribution of below (roots) than above ground litter (Roscoe et al., 2001; Wedin et al., 1995) reducing the probability of pasture litter being exported.

Subsoil was the main source of TN for the smallest and intermediate catchments and just after peak flow in the largest catchment. Intact valley surface soil was also a relevant source of TN, varying from 15% in the smallest catchment to 68% towards the end of the high-flow event sampled in the largest catchment. This is explained by intact valley soil having a 10 times higher TN content than subsoil. Accordingly, differences in source contributions to TN export between catchments were most likely derived from differences in subsoil erosion rates relative to surface soil erosion rates (see Figures 3 to 5).

4.2 Controls on source contribution: climatic conditions

The largest difference between results occurred for subsoil and intact valley surface soil contributions to sediment, organic matter (as indicated by TOC) and TN export in the wet year (Figure 4), with a lower subsoil and larger intact valley soil contribution. It is possible that intact valley soil contributes a large fraction of the dissolved solids during wet years. Dissolved solids are likely to be present during isotopic analysis of samples that have been previously freeze-dried and sampled with the refrigerated autosampler stations in comparison to material sampled with the time integrated samplers (See Table 1).

Total annual rainfall had an important role in determining subsoil, intact valley surface soil and vegetation litter contributions to exported sediments, organic matter and TN. In wet years, erosion rates are higher and gully growth would affect not only eroded bare banks, though potentially more protected and vegetated gully banks (Garzon-Garcia et al., 2015). Further, there would likely be more erosion occurring on intact valleys during wet years relative to dry years. This may explain why subsoil relative contributions to exported sediment, organic matter and TN were lower in wet than in dry years (22, 18 and 36% lower, respectively; SD = 5.2, 9.1, 10.5), and intact valley soil and C_4 litter contributions were higher in the wet year.

Similarly to subsoil, C_3 litter contributions to organic matter and TN export was larger in the dry year. Dry conditions may cause trees to shed a larger amount of leaves (Keith et al., 2012), slow litter decomposition (Hutchens and Wallace, 2002; Langhans et al., 2008) and reduce transport and export of tree litter due to a lower frequency of rain events (Webster et al., 1999). In wet years, there would be less tree litter available and most of it would be exported quickly and early in the wet season resulting in lower overall contributions. The increase in intact valley soil contributions during wet years may also have a role in explaining the decline in contributions from C_3 litter.

4.3 Controls on source contribution: flow stage

Flow stage influences the sources of organic matter (as indicated by TOC export) and TN during a high-flow event. The subsoil contribution was clearly dictated by the magnitude of the flow and thus became an important source of organic matter and the main source of TN immediately after peak flow. The C_3 litter contribution was larger at the beginning of the high-flow event and decreased gradually, but was the main organic matter source overall (55%, SD = 4). Large amounts of tree litter accumulate in dry headwater gullies during the dry season and possibly in between rain events (Garzon-Garcia et al., 2014; Webster et al., 1999). This would most likely occur in dry river bed channels, bordered by riparian trees. The presence of most of the C_3 vegetation litter close to waterways and its low density, which facilitates its transport compared to higher density subsoil, may explain the larger contribution of C_3 litter at the beginning of the sampled high-flow event (Figure 5).

The amount of previously deposited C_3 litter would also influence these source contributions, potentially being a major limiting factor. It is likely that at the start of the wet season, C_3 litter contributions during significant flow events would be larger as well, with longer time intervals between events. Previous research in the Logan River catchment concluded that exported sediment for lower magnitude events had higher TOC and TN concentrations, and as flow increased, the TOC and TN content of sediment decreased (Garzon-Garcia et al., 2015). It was hypothesized that TOC and TN source contributions vary with event magnitude and that vegetation litter could be an important source. Our findings support these hypotheses and those from other authors that have highlighted the importance of vegetation litter to export (Bellanger et al., 2004; Gomez et al., 2003; Juarez et al., 2011; Kao and Liu, 2000).

TN export from the sampled high-flow event had varying sources with a tendency for intact valley surface soil to have higher contributions, except for larger flows where subsoil became the main contributor to export. It is likely that the dominance of intact valley soil as a TN source is also related to subsurface flows of soluble N, considering dissolved solids would be present in isotopic analysis of autosampler samples which were freeze dried before analysis (Table 1). C_3 litter was also an important TN source, contributing more than subsoil sources for most of the flow stages. Detailed analysis of soluble and particulate TN loads and source contributions to each fraction is required in the future to determine the relevance of each source to N export.

When contributions to sediment export from intact valley soil and C_3 litter is higher than around 10-20%, their contribution to TN export dominates, as occurred for most of the high-flow event (Figure 5) and for the wet year flow events in the Knapp Creek. When subsoil contributions to sediment export is higher than around 80%, subsoil becomes the main source of TN. Intact valley soil and C_3 litter contributions to TN export may be higher in certain catchment areas, where surface soil erosion is elevated and/or where there is larger presence of tree cover. There is likely a complex interrelationship between the distribution of the vegetative cover and the characteristics of the rainfall regime that governs organic matter and nutrient mobilisation and export.

4.4 Implications and Further Research

Our research indicates that C_3 litter is a significant source of organic matter exported from catchments affected by channel and gully erosion. Conversely, subsoil and intact valley soil were shown to be the most important source of exported TN, the former in severely gullied catchments and the latter when subsurface erosion contributed less than 80% to sediment export, which occurred in wetter years and at the peak of the hydrograph. These results highlight the importance of considering vegetation litter together with subsoils when tracing organic matter sources in catchments affected by gully erosion. These results also provide guidance for catchment management programs that aim to reduce fine sediment, carbon and nitrogen export in similar gullied catchments through identifying the importance of understanding the combination of erosional processes occurring in catchments at different scales, and the role of hydrology in driving these processes (i.e., dominance between headscarp retreat, gully incision and surface erosion).

A larger amount and a wider distribution of source samples is necessary to further validate these results along with examining the role of autochthonous sources like algae and macrophyte biomass. An examination of soluble sources to C and N is also warranted as they may give insight into the role of intact valley soil as an important N source. Including soluble sources together with complexed and uncomplexed (not bound to mineral particles) organic matter would provide more insight into the carbon and nitrogen budgets in stream systems. In particular, the supply of undecomposed organic matter sources like vegetation litter has been proposed to be a limiting factor in the restoration of soils and gullies in degraded catchments (Garzon-Garcia et al., 2014; Post and Kwon, 2000). Differences in organic matter and N bioavailability in the aquatic environment, related to potential sources, would complement this research and guide soil and catchment management prioritisation.

5 Conclusions

This research has demonstrated that the sources of fine sediment, organic matter and N differ in subtropical catchments affected by gully erosion. While subsoil is clearly the main sediment source, C_3 litter dominates organic matter export, and N sources vary. Subsoil dominates N export at smaller scales, with an exception of the wet year and at the larger catchment, where intact valley soil was the main N source. The disproportionally large contribution of C_3 litter to organic matter export (measured as TOC export) and of intact valley soil to N export, relative to their contribution to sediment export, occurred because of the significantly larger TOC content in litter biomass and significantly larger TN content in intact valley soil relative to subsoil.

This novel application of sediment tracing and fingerprinting techniques to directly trace the different sources of carbon and nitrogen provides a unique approach to test catchment C and N budgets. Understanding differences in sediment, C and N sources in catchments degraded by erosion will assist management in reducing sediment, C and N transfers to the stream system in similar subtropical catchments, while providing a quantitative foundation for restoring more natural C and N dynamics.

6 Acknowledgements

We express our thanks to Rene Diocares and Rad Bak at the Stable Isotope Laboratory - Australian Rivers Institute, Griffith University, for their assistance with the elemental and isotope analysis of samples. We acknowledge SEQHWP, the SEQ Catchments Load Monitoring Program at DSITI and the Chemistry Centre, and particularly Rob de Hayr and Belinda Thomson for helping us get access to high-flow event samples from the Yarrahappini station. We especially thank Tanya Ellison for her tireless assistance with fieldwork and express our gratitude and appreciation to Mark and Nia Tilley, Tilley Gully catchment landowners, for allowing us access to their land.

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Figure Captions

Figure 1. Location of the study region, catchment land use, and source sampling sites (in Tilley Gully catchment and Knapp Creek catchment) and fine sediment sampling sites (in Tilley Gully, Knapp Creek and Logan River catchments) with land use data provided by the Queensland Government, Australia.

Figure 2. Mean TOC and $\delta^{13}C$ along with TN and $\delta^{15}N$ for fine sediment and potential sources for Tilley Gully catchment (a), Knapp Creek catchment (including climatic conditions: dry and wet year) (b) and the Logan River catchment at Yarrahappini, subtropical Australia (c). All source values and their statistical difference test results are in Table 2. Error bars depict standard deviations.

Figure 3. Mean percent contributions from subsoil, intact valley soil, C_3 and C_4 litter to particulate organic matter (TOC) and TN export in Tilley Gully catchment for the integrated wet season 2011-2012. Error bars show propagated standard deviations.

Figure 4. Mean percent contributions to exported fine sediment (a), particulate organic matter (TOC) (b) and particulate TN (c) from subsoil, intact valley soil, C_3 and C_4 litter during high-flow events in a dry year (2009-2010) and in a wet year (2010-2011) in the Knapp Creek catchment. Error bars show standard deviations (a) and propagated standard deviations (b, c).

Figure 5. Mean percent contributions to exported fine sediment (a), and particulate organic matter (TOC) (b) and TN (c) from subsoil, intact valley soil, C_3 and C_4 litter for different sampling times corresponding to different flow stages, during a high-flow event sampled in January-Februrary 2013 at Yarrahappini in the Logan River. Error bars show propagated standard deviations.