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# 1 **Differentiating the sources of fine sediment, organic matter and nitrogen in a** 2 **subtropical Australian catchment**

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## 13 **Abstract**

14 Understanding the sources of sediment, organic matter and nitrogen (N) transferred from terrestrial to  
15 aquatic environments is important for managing the deleterious off-site impacts of soil erosion. In  
16 particular, investigating the sources of organic matter associated with fine sediment may also provide  
17 insight into carbon (C) and N budgets. Accordingly, the main sources of fine sediment, organic matter  
18 (indicated by total organic carbon), and N are determined for three nested catchments (2.5 km<sup>2</sup>, 75  
19 km<sup>2</sup>, and 3076 km<sup>2</sup>) in subtropical Australia. Source samples included subsoil and surface soil, along  
20 with C<sub>3</sub> and C<sub>4</sub> vegetation. All samples were analysed for stable isotopes ( $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$ ) and elemental  
21 composition (TOC, TN). A stable isotope mixing model (SIAR) was used to determine relative source  
22 contributions for different spatial scales (nested catchments), climatic conditions and flow stages.  
23 Subsoil was the main source of fine sediment for all catchments (82%, SD = 1.15) and the main N  
24 source at smaller scales (55-76%, SD = 4.6-10.5), with an exception for the wet year and at the larger  
25 catchment, where surface soil was the dominant N source (55-61%, SD = 3.6-9.9), though  
26 contributions were dependent on flow (59-680 m<sup>3</sup>/s). C<sub>3</sub> litter was the main source of organic C export  
27 for the two larger catchments (53%, SD = 3.8) even though C<sub>4</sub> grasses dominate the vegetation cover  
28 in these catchments. The sources of fine sediment, organic matter and N differ in subtropical  
29 catchments impacted by erosion, with the majority of C derived from C<sub>3</sub> leaf litter and the majority of  
30 N derived from either subsoil or surface soil. Understanding these differences will assist management  
31 in reducing sediment, organic matter and N transfers in similar subtropical catchments while  
32 providing a quantitative foundation for testing C and N budgets.

33

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

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## 35 1 Introduction

36 Soil erosion is a major source of sediment to aquatic systems (Cerdan et al., 2010; Milliman and  
37 Meade, 1983). Erosion processes may also influence the transfer of carbon, potentially playing a  
38 major role in carbon budgets (Cole et al., 2007; Lal, 2003; Scott et al., 2006). The transfer of nitrogen  
39 to aquatic systems is also affected by erosion processes (Quinton et al., 2010). In Australia, land use  
40 change following European settlement triggered significant gully and channel erosion (Bartley et al.,  
41 2006; Olley and Wasson, 2003; Prosser and Slade, 1994). Research on tropical and subtropical  
42 systems in eastern Australia has demonstrated that subsoil erosion (i.e., gully and channel erosion) is  
43 the dominant source of sediment (Caitcheon et al., 2012; Hughes et al., 2009; Olley et al., 2013a;  
44 Olley et al., 2013b). Although the significance of subsoil erosion sources for sediment in this region is  
45 well-documented, the sources of organic matter and nutrients have received less attention.

46 Carbon and nitrogen stable isotope ratios ( $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$ ) and elemental composition have been  
47 widely used to determine the sources of organic matter supporting food webs in aquatic environments  
48 (Bunn et al., 2003; Finlay, 2001; Hein et al., 2003). They have also been used extensively to trace the  
49 contributions of catchment sources to sediment (Lacey et al., accepted; Mukundan et al., 2010;  
50 Papanicolaou et al., 2003) and to in-stream particulate organic matter (Cooper et al., 2015; Kendall et  
51 al., 2001; McCorkle et al., 2016).  $\delta^{13}\text{C}$  discrimination of sources is derived primarily from  
52 photosynthetic pathways that result in distinct  $\delta^{13}\text{C}$  fractionations. The majority of tree and temperate  
53 grass species follow the Calvin-Benson cycle ( $\text{C}_3$ ) photosynthetic pathway ( $\delta^{13}\text{C}$ : mean  $-28\text{‰}$ )  
54 (Boutton, 1991; Fry, 2006; Schimel, 1993). Plants following the Hatch-Slack cycle ( $\text{C}_4$ ) pathway  
55 consist mainly of grass and cropping species primarily found in warmer climates with limited water  
56 availability ( $\delta^{13}\text{C}$ : mean  $-13\text{‰}$ ) (Coleman and Fry, 1991; Werth and Kuzyakov, 2010). The  
57 Crassulacean acid metabolism (CAM) plants have the potential to utilize both  $\text{C}_3$  and  $\text{C}_4$  pathways  
58 (Werth and Kuzyakov, 2010).  $\delta^{13}\text{C}$  should discriminate between material derived from  $\text{C}_3$  and  $\text{C}_4$   
59 plants in tropical and subtropical environments. Most of the nitrogen in the biosphere is atmospheric  
60  $\text{N}_2$  which has a near constant  $\delta^{15}\text{N}$  of  $0\text{‰}$  (Peterson and Fry, 1987). The majority of nitrogen in the  
61 rest of the biosphere has  $\delta^{15}\text{N}$  values between  $-10\text{‰}$  to  $+10\text{‰}$ . In general,  $\delta^{15}\text{N}$  fractionation is  
62 complex, with a multitude of nitrogen sources and internal transformations potentially altering  
63 nitrogen isotopic ratios (Evans, 2007; Finlay and Kendall, 2007; Shearer and Kohl, 1993).

64 Here, we use  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$ , total organic carbon (TOC), and total nitrogen (TN) to quantify the  
65 sources of organic matter and total nitrogen associated with fine particulate export ( $<63\ \mu\text{m}$ ) in a  
66 subtropical riverine system. First, we test whether  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$ , total organic carbon (TOC), and total  
67 nitrogen (TN) can be used to distinguish between organic matter derived from the four primary  
68 sources: subsoils, intact valley soils,  $\text{C}_3$  litter and  $\text{C}_4$  litter. These sources were identified as the most  
69 likely to contribute to fine particulate organic matter export during flow events in the area. Second,  
70 the relative contributions of these sources are determined at three different spatial scales in a series of

71 nested catchments using a modified sediment source fingerprinting approach (e.g. Collins and  
72 Walling, 2004; Davis and Fox, 2009; Koiter et al., 2013b). Third, variations in contributions from  
73 each of the sources between wet and dry years and base and event flow conditions are examined.

## 74 **2 Methods**

### 75 *2.1 Study Region*

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

81 Land use in the Logan River catchment is predominantly grazing (89%), native forest (7%) and  
82 cropping (2%) (Figure 1). The Logan River catchment geology consists of basalt (32%), arenite-  
83 mudrock (19%), arenite (17%), and alluvium (12%) (DME, 2008). Land use in the Knapp Creek  
84 catchment is also dominated by grazing (78%) with less than 22% of the native vegetation cover  
85 remaining. The geology of Knapp Creek primarily consists of arenite sandstone (81%) (DME, 2008).  
86 The majority of sediment produced in the Logan River catchment (Hancock and Revill, 2013) and the  
87 Knapp Creek subcatchment is derived from subsurface erosion sources (Lacey et al., 2015; Olley et  
88 al., 2009). There is extensive gully erosion throughout Knapp Creek, with at least 38 km of gullies.  
89 Most of this erosion is evident in the mid-catchment reaches where gullies are well-connected to the  
90 main channel (Olley et al., 2009). The Tilley Gully catchment, located in these mid-reaches (Figure  
91 1), is one of the top three sediment-yielding catchments in Knapp Creek, with an estimated sediment  
92 yield of more than 1000 T y<sup>-1</sup>. This is one sixth of the ~6000 T/y produced by gully erosion in the  
93 whole catchment (Olley et al., 2009). The catchment has been cleared for cattle grazing with less than  
94 20% of the original forest cover remaining. The Tilley Gully catchment is underlain by arenite  
95 sandstone (100%).

### 96 *2.2 Source sampling*

97 Source sampling includes the sampling of the primary sources of particulate material that may be  
98 potentially mobilized by rainfall events and transported downstream. Four potential sources were  
99 identified and sampled: subsoils (i.e. gully and channel banks), intact valley “non-gullied” surface  
100 soils, valley and gully grasses, and tree litter (fine sticks and leaves). These sources were selected  
101 after an extensive literature review (Hancock and Revill, 2013; Lacey et al., 2015; Olley et al.,  
102 2009), field investigations, and discussions with catchment managers and land-owners. Source  
103 sampling details are summarized in Table 1. Gully banks were sampled at five locations, separated by  
104 ~400 m, along Tilley Gully in April 2012 (Figure 1). Samples were taken from three horizontal strata  
105 differentiated by colour on bare gully banks. In total, 15 gully samples were collected. Channel banks  
106 were sampled in December 2013 at five locations along Knapp Creek (Figure 1). At each location,

107 two samples were composited for analysis, one near the base of the bank and the other near the top of  
108 the bank. Gully and channel bank samples were taken with an 18 cm<sup>2</sup> corer to a 10 cm depth.

109 Sampling locations for intact valley soils, valley and gully grasses, and tree litter were randomly  
110 selected from a 2.5 x 2.5 m grid covering an intact valley and an incising gully in the Tilley Gully  
111 catchment (Figure 1). Sample location was selected with the Sampling Design Tool for ArcGIS  
112 (10.0). The top 2 cm of intact valley soils were sampled with a trowel after vegetation was removed to  
113 ground level, between July and October 2011, in the dry season. In total, 24 samples were collected  
114 (Figure 1).

115 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

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

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

### 124 2.3 Sediment sampling

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

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

144 In the Logan River catchment, ten sediment samples were collected for analysis, also using a  
145 refrigerated autosampler. The sampling site was located 57 km downstream from Knapp Creek's  
146 junction with the Logan River (Figure 1). Samples were taken between January 27 and February 1,  
147 2013 and captured a high-flow event (366 mm of rainfall in 8 days, with a peak flow rate of 678 m<sup>3</sup>/s  
148 in the Logan River).

### 149 2.4 Sample Preparation

150 Samples collected to characterise the potential sources were processed using a method adapted  
151 from Gregorich *et al.* (2006) with sample processing details summarized in Table 1. The approach

152 was designed to mimic stream transport processes and to remove physically uncomplexed organic  
153 matter (not bound to mineral particles) from the soil and subsoil.

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

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

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

## 169 *2.5 Isotope and elemental analytical methods*

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

176  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values are reported in per mil (‰) relative to Pee Dee Belemnite (PDB) standard  
177 and relative to air  $\text{N}_2$ , respectively. The precision of  $\delta^{13}\text{C}$  was monitored with a sucrose standard over  
178 20 analysis runs reporting  $\delta^{13}\text{C} = -11.7$  ‰ (SD = 0.1, n = 84) and of  $\delta^{15}\text{N}$  with an IAEA-305a  
179 surrogate standard reporting  $\delta^{15}\text{N} = 0.2$  ‰ (SD = 0.3, n = 84). The precision of TOC and TN, reported  
180 in percent weight of dry sample (%) was monitored using an Acetanilide elemental standard over 20  
181 analysis runs reporting TOC = 3.4% (SD = 0.1, n = 84) and TN = 0.33% (SD = 0.005, n = 84). All  
182 data used in these analyses are provided in the supporting material (S1-S3).

## 183 *2.6 Statistical analysis and modelling*

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



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

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

199

$$\%E_{source_i} = \frac{E_{source_i} \times \%cont_i}{\sum_{i=1}^4 E_{source_i} \times \%cont_i} \times 100 \quad (1)$$

200

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

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

## 219 **3 Results**

### 220 *3.1 Source Discrimination*

221 The source discrimination potential of the measured properties was tested prior to modelling. In  
222 combination,  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$ , TOC and TN discriminate between all the different sources (Table 2).  $\delta^{13}\text{C}$   
223 discriminates between all sources, except intact valley soil and  $\text{C}_4$  litter.  $\delta^{15}\text{N}$  discriminates between  
224 litter and subsoil, and between litter and intact valley soil, with the two latter having higher  $\delta^{15}\text{N}$   
225 values than  $\text{C}_3$  or  $\text{C}_4$  litter. TOC discriminates between all sources, with  $\text{C}_4$  litter having the highest  
226 TOC content and subsoil the lowest. TN discriminates between subsoil and the other sources, but not  
227 between litter types or intact valley soil and  $\text{C}_3$  litter. Subsoil had a mean  $\delta^{13}\text{C}$  between  $\text{C}_3$  and  $\text{C}_4$   
228 vegetation litter. These four sediment properties in combination provide complete discrimination  
229 amongst all the sources and accordingly all sediment properties will be modelled in SIAR. Source  
230 samples are plotted with sediment samples in Figure 2.

### 231 *3.2 Tilley Gully*

232 Results for all Tilley Gully catchment sampling sites have been averaged as the variability between  
233 sampling sites was low (see standard deviation plotted in Figure 3). Results for individual sampling  
234 sites are provided in the supporting material (S4). Subsoil was the dominant source of exported  
235 sediment in Tilley Gully catchment during the wet season (2011-2012). The mean subsoil  
236 contribution to sediment was 97% (SD = 1) for all sampling sites. Intact valley soil,  $\text{C}_3$  litter and  $\text{C}_4$   
237 litter were minor sediment sources. Subsoil also contributed the most to organic matter export as  
238 indicated by TOC export (60%, SD = 6), and to TN (76%, SD = 5) in the Tilley Gully catchment  
239 (Figure 3).  $\text{C}_3$  litter contributed 21% (SD = 6) of TOC, followed by intact valley soil which  
240 contributed 11% (SD = 3). For TN, intact valley soil contributed 15% (SD = 4) while the other  
241 sources were insignificant (<8%). The Tilley Gully model results are provided as supporting material  
242 (S4).

### 243 *3.3 Knapp Creek*

244 There were significant differences in source contributions to exported sediment between the dry  
245 (2009-2010) and wet (2010-2011) years in Knapp Creek (Figure 4). Subsoil was the largest source of  
246 sediment for both climatic conditions with mean sediment contributions ranging from 72 to 94% (SD  
247 = 1 to 5) followed by intact valley soil ranging from 3 to 24% (SD = 1 to 6) (Figure 4). Intact valley  
248 soil contributions were significantly larger (21%) ( $p < 0.001$ ) for the samples collected in the wet year.  
249 All Knapp Creek model results are provided in the supporting material (S5).

250 While subsoil was the dominant source of sediment, it contributed less than 31% (SD = 5 to 9) of  
251 the organic matter (as indicated by TOC) during events. This reflects the low organic matter content  
252 of subsoil compared to other sources. There was a significantly lower contribution from subsoil to  
253 organic matter export (TOC) in the wet year ( $p < 0.001$ ) (Figure 4).  $\text{C}_3$  litter contributed on average 40

254 to 60% (SD = 9 to 10) of the organic matter export, with a significantly larger contribution occurring  
 255 in the dry year ( $p < 0.001$ ) (Figure 4). Intact valley soil and C<sub>4</sub> litter contributed on average 8 to 36%,  
 256 and 1 to 11%, respectively. Their contributions varied similarly, significantly increasing ( $p < 0.001$ )  
 257 for the wet year when the intact valley soil contributed a similar proportion than C<sub>3</sub> litter to organic  
 258 matter export (Figure 4).

259 The dominant source of TN varied between the wet and the dry year with subsoil being the main  
 260 source in the dry year (55%, SD = 11) and intact valley soil the most dominant source in the wet year  
 261 (61%, SD = 10) (Figure 4). C<sub>3</sub> litter contributed between 16 and 29% (SD = 7 to 10), and C<sub>4</sub> litter  
 262 contributions were negligible. In wet years, intact valley soil contributions were significantly larger ( $p$   
 263  $< 0.001$ ) than in the dry year (45% larger) (Figure 4).

264

265 Table 2.  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$ , TOC and TN of most probable sources during flow events (subsoil, intact valley  
 266 soil, C<sub>3</sub> litter and C<sub>4</sub> litter) and statistical analysis results for differences in  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$ , TOC and TN  
 267 between sources. T-tests (T) or Mann-Whitney U-test results are presented (statistical significant  
 268 differences determined at  $p < 0.05$ ).

269

Source	$\delta^{13}\text{C}$ (‰)	SD	Subsoil	Intact valley soil	C <sub>3</sub> litter	C <sub>4</sub> litter
Subsoil	-18.9	1.6				
Intact valley soil	-14.3	0.9	***			
C <sub>3</sub> litter (tree)	-25.6	3.3	***	***		
C <sub>4</sub> litter (grass)	-13.5	1.1	***	-	***	
	$\delta^{15}\text{N}$ (‰)	SD	Subsoil	Intact valley soil	C <sub>3</sub> litter	C <sub>4</sub> litter
Subsoil	5.6	2.6				
Intact valley soil	3.4	0.8	- (t)			
C <sub>3</sub> litter (tree)	-1.5	1.0	*** (t)	*** (t)		
C <sub>4</sub> litter (grass)	-2.4	2.7	*** (t)	*** (t)	- (t)	
	TOC (%)	SD	Subsoil	Intact valley soil	C <sub>3</sub> litter	C <sub>4</sub> litter
Subsoil	0.6	0.2				
Intact valley soil	5.0	0.6	***			
C <sub>3</sub> litter (tree)	32.1	8.1	***	***		
C <sub>4</sub> litter (grass)	40.7	5.3	***	***	*	
	TN (%)	SD	Subsoil	Intact valley soil	C <sub>3</sub> litter	C <sub>4</sub> litter
Subsoil	0.05	0.02				
Intact valley soil	0.51	0.11	***			
C <sub>3</sub> litter (tree)	0.80	0.32	***	-		
C <sub>4</sub> litter (grass)	0.84	0.30	***	**	-	

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

270

### 271 3.4 Logan River

272 Subsoil contributed the majority of sediment sampled in the Logan River catchment for all flow  
273 stages, with the exception of the first sample (Figure 5a). Subsoil mean contributions to exported  
274 sediment increased with flow, with the largest contribution occurring just after peak flow and high  
275 contributions occurring during the receding limb of the hydrograph (mean > 60%, SD = 3 to 7)  
276 (Figure 5a). Intact valley soil was the second largest contributor with the largest contribution at the  
277 start of the high-flow event (mean 47%, SD = 11), and the lowest just after peak flow (4%, SD = 3)  
278 (Figure 5a). C<sub>3</sub> litter had a similar trend, with the largest contribution at the start of the high-flow  
279 event of 21% (SD = 6), and the lowest just after peak flow. C<sub>4</sub> litter contributions were minimal  
280 (<1%) (Figure 5a). The Logan River model results are included in the supporting material (S6).

281 C<sub>3</sub> litter was the main contributor of organic matter as indicated by TOC for all samples collected  
282 during the high-flow stages, with the largest contribution at the start of the event (70%, SD = 10), and  
283 a gradual decrease reaching the lowest contribution (45%, SD = 11), for the last sample taken (Figure  
284 5b). Intact valley soil was the second largest source of organic matter (Figure 5b) (10 - 43%, SD = 7  
285 to 10). C<sub>4</sub> litter contributed less than 5% (Figure 5b).

286 Intact valley soils were the main source of TN (mean = 55%, SD = 4) (Figure 5c), followed by C<sub>3</sub>  
287 litter (mean = 26%, SD = 3) with the exception of when subsoil contribution peaked (Figure 5c). C<sub>4</sub>  
288 litter was again a minor source (<5%). Subsoil contributions to fine particulate organic matter (TOC)  
289 and TN export increased with flow, reaching their highest value just after peak flow (25 and 46%, SD  
290 = 9 and 12) when they were the second largest and largest source, respectively (Figure 5b,c).

## 291 4 Discussion

292 Soil erosion exports large quantities of organic matter downstream (Lal, 2003; Ludwig and Probst,  
293 1996; Scott et al., 2006), redistributing landscape carbon and nitrogen pools laterally, vertically,  
294 and/or longitudinally from catchments to the marine environment (Gregorich et al., 1998; Ma et al.,  
295 2016; Ran et al., 2014). Recent research conducted on tropical and subtropical river systems in  
296 Australia has shown that subsoil erosion (gully and channel erosion) is the dominant source of  
297 sediment entering these waterways (Caitcheon et al., 2012; Hughes et al., 2009; Olley et al., 2013a).  
298 Although subsoils contribute the majority of sediment, the modelling of source and in-stream  
299 particulate material with  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$ , TOC and TN indicated that subsoils are not always the dominant  
300 source of fine particulate organic matter and nitrogen (TOC, TN).

301  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$ , TOC and TN were successfully used as complementary tracers of source contributions  
302 to fine particulate material export from intact “non-gullied” valley soils, subsoils, C<sub>3</sub> litter and C<sub>4</sub>  
303 litter. TOC discriminated between all sources. TN discriminated subsoils from other sources, with the  
304 former having a significant lower content, though not between litter types or intact valley soil and C<sub>3</sub>  
305 litter.  $\delta^{13}\text{C}$  did not discriminate between C<sub>4</sub> litter and intact valley soil, which derives its carbon input

306 mainly from this type of litter. Finally,  $\delta^{15}\text{N}$  discriminated between litter and soils with the latter  
307 having significantly higher  $\delta^{15}\text{N}$ .

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

321 Importantly, the main sources of sediment, organic matter and of TN are different. Subsoil is  
322 clearly the main sediment source (mean = 82%, SD = 1).  $\text{C}_3$  litter is the main organic matter source  
323 (mean = 42%, SD = 3). Subsoil is the main TN source (mean = 44%, SD = 3), with exceptions for the  
324 autosamplers in the wet year and larger catchment where intact valley soil was the main TN source  
325 (mean = 61%, SD = 10 and mean = 55%, SD = 4 respectively). This disproportionally large  
326 contribution of  $\text{C}_3$  litter to organic matter and intact valley soil to TN, relative to their contribution to  
327 sediment, occurs because of the larger TOC content in litter biomass and larger TN content in intact  
328 valley soil compared to subsoil.

#### 329 *4.1 Controls on source contribution: Spatial scale*

330 Subsoil was the main source of exported sediment varying from 49% during low flow conditions  
331 in the largest catchment to 97% in the smallest catchment. Differences in subsoil contributions to  
332 sediment export between catchments is likely related to the severity of erosion and to the magnitude  
333 of flows sampled.  $\text{C}_3$  litter was the main source of organic matter export (as indicated by TOC  
334 export), varying from 21% in the smallest catchment to 70% at low flows in the largest catchment.  
335 This contribution may be explained by the higher TOC% in litter biomass compared to subsoil: 49  
336 times more on average. Differences in source contributions to organic matter export between  
337 catchments were most likely due to tree cover density in the riparian area.

338 Although most of these catchments have been cleared of trees, tree litter contributes a larger  
339 proportion than grasses and intact valley soil to organic matter export, even though  $\text{C}_4$  grasses grow  
340 densely within headwater gullies.  $\text{C}_3$  inputs from riparian trees have a higher connectivity to the  
341 stream network than grassland litter, and riparian areas are likely to have higher net primary

342 production than grasslands in the upper catchments, which may explain this disproportionate  
343 contribution to organic matter export. It is also probable that cattle remove large quantities of grass  
344 biomass reducing the amount of C<sub>4</sub> litter present and available for export. Further, pasture has a  
345 higher contribution of below (roots) than above ground litter (Roscoe et al., 2001; Wedin et al., 1995)  
346 reducing the probability of pasture litter being exported.

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

#### 354 4.2 Controls on source contribution: climatic conditions

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

362 Total annual rainfall had an important role in determining subsoil, intact valley surface soil and  
363 vegetation litter contributions to exported sediments, organic matter and TN. In wet years, erosion  
364 rates are higher and gully growth would affect not only eroded bare banks, though potentially more  
365 protected and vegetated gully banks (Garzon-Garcia et al., 2015). Further, there would likely be more  
366 erosion occurring on intact valleys during wet years relative to dry years. This may explain why  
367 subsoil relative contributions to exported sediment, organic matter and TN were lower in wet than in  
368 dry years (22, 18 and 36% lower, respectively; SD = 5.2, 9.1, 10.5), and intact valley soil and C<sub>4</sub> litter  
369 contributions were higher in the wet year.

370 Similarly to subsoil, C<sub>3</sub> litter contributions to organic matter and TN export was larger in the dry  
371 year. Dry conditions may cause trees to shed a larger amount of leaves (Keith et al., 2012), slow litter  
372 decomposition (Hutchens and Wallace, 2002; Langhans et al., 2008) and reduce transport and export  
373 of tree litter due to a lower frequency of rain events (Webster et al., 1999). In wet years, there would  
374 be less tree litter available and most of it would be exported quickly and early in the wet season  
375 resulting in lower overall contributions. The increase in intact valley soil contributions during wet  
376 years may also have a role in explaining the decline in contributions from C<sub>3</sub> litter.

377 4.3 Controls on source contribution: flow stage

378 Flow stage influences the sources of organic matter (as indicated by TOC export) and TN during a  
379 high-flow event. The subsoil contribution was clearly dictated by the magnitude of the flow and thus  
380 became an important source of organic matter and the main source of TN immediately after peak  
381 flow. The C<sub>3</sub> litter contribution was larger at the beginning of the high-flow event and decreased  
382 gradually, but was the main organic matter source overall (55%, SD = 4). Large amounts of tree litter  
383 accumulate in dry headwater gullies during the dry season and possibly in between rain events  
384 (Garzon-Garcia et al., 2014; Webster et al., 1999). This would most likely occur in dry river bed  
385 channels, bordered by riparian trees. The presence of most of the C<sub>3</sub> vegetation litter close to  
386 waterways and its low density, which facilitates its transport compared to higher density subsoil, may  
387 explain the larger contribution of C<sub>3</sub> litter at the beginning of the sampled high-flow event (Figure 5).

388 The amount of previously deposited C<sub>3</sub> litter would also influence these source contributions,  
389 potentially being a major limiting factor. It is likely that at the start of the wet season, C<sub>3</sub> litter  
390 contributions during significant flow events would be larger as well, with longer time intervals  
391 between events. Previous research in the Logan River catchment concluded that exported sediment for  
392 lower magnitude events had higher TOC and TN concentrations, and as flow increased, the TOC and  
393 TN content of sediment decreased (Garzon-Garcia et al., 2015). It was hypothesized that TOC and TN  
394 source contributions vary with event magnitude and that vegetation litter could be an important  
395 source. Our findings support these hypotheses and those from other authors that have highlighted the  
396 importance of vegetation litter to export (Bellanger et al., 2004; Gomez et al., 2003; Juarez et al.,  
397 2011; Kao and Liu, 2000).

398 TN export from the sampled high-flow event had varying sources with a tendency for intact valley  
399 surface soil to have higher contributions, except for larger flows where subsoil became the main  
400 contributor to export. It is likely that the dominance of intact valley soil as a TN source is also related  
401 to subsurface flows of soluble N, considering dissolved solids would be present in isotopic analysis of  
402 autosampler samples which were freeze dried before analysis (Table 1). C<sub>3</sub> litter was also an  
403 important TN source, contributing more than subsoil sources for most of the flow stages. Detailed  
404 analysis of soluble and particulate TN loads and source contributions to each fraction is required in  
405 the future to determine the relevance of each source to N export.

406 When contributions to sediment export from intact valley soil and C<sub>3</sub> litter is higher than around  
407 10-20%, their contribution to TN export dominates, as occurred for most of the high-flow event  
408 (Figure 5) and for the wet year flow events in the Knapp Creek. When subsoil contributions to  
409 sediment export is higher than around 80%, subsoil becomes the main source of TN. Intact valley soil  
410 and C<sub>3</sub> litter contributions to TN export may be higher in certain catchment areas, where surface soil  
411 erosion is elevated and/or where there is larger presence of tree cover. There is likely a complex  
412 interrelationship between the distribution of the vegetative cover and the characteristics of the rainfall  
413 regime that governs organic matter and nutrient mobilisation and export.

414 *4.4 Implications and Further Research*

415 Our research indicates that C<sub>3</sub> litter is a significant source of organic matter exported from  
416 catchments affected by channel and gully erosion. Conversely, subsoil and intact valley soil were  
417 shown to be the most important source of exported TN, the former in severely gullied catchments and  
418 the latter when subsurface erosion contributed less than 80% to sediment export, which occurred in  
419 wetter years and at the peak of the hydrograph. These results highlight the importance of considering  
420 vegetation litter together with subsoils when tracing organic matter sources in catchments affected by  
421 gully erosion. These results also provide guidance for catchment management programs that aim to  
422 reduce fine sediment, carbon and nitrogen export in similar gullied catchments through identifying the  
423 importance of understanding the combination of erosional processes occurring in catchments at  
424 different scales, and the role of hydrology in driving these processes (i.e., dominance between  
425 headscarp retreat, gully incision and surface erosion).

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

436 **5 Conclusions**

437 This research has demonstrated that the sources of fine sediment, organic matter and N differ in  
438 subtropical catchments affected by gully erosion. While subsoil is clearly the main sediment source,  
439 C<sub>3</sub> litter dominates organic matter export, and N sources vary. Subsoil dominates N export at smaller  
440 scales, with an exception of the wet year and at the larger catchment, where intact valley soil was the  
441 main N source. The disproportionately large contribution of C<sub>3</sub> litter to organic matter export  
442 (measured as TOC export) and of intact valley soil to N export, relative to their contribution to  
443 sediment export, occurred because of the significantly larger TOC content in litter biomass and  
444 significantly larger TN content in intact valley soil relative to subsoil.

445 This novel application of sediment tracing and fingerprinting techniques to directly trace the  
446 different sources of carbon and nitrogen provides a unique approach to test catchment C and N  
447 budgets. Understanding differences in sediment, C and N sources in catchments degraded by erosion  
448 will assist management in reducing sediment, C and N transfers to the stream system in similar



449 subtropical catchments, while providing a quantitative foundation for restoring more natural C and N  
450 dynamics.

451

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460

## 461 **7 References**

462 Bartley R, Roth CH, Ludwig J, McJannet D, Liedloff A, Corfield J, et al. Runoff and erosion from  
463 Australia's tropical semi-arid rangelands: influence of ground cover for differing space and  
464 time scales. *Hydrological processes* 2006; 20: 3317-3333.

465 Bellanger B, Huon S, Velasquez F, Valles V, Girardin C, Mariotti A. Monitoring soil organic carbon  
466 erosion with  $d^{13}C$  and  $d^{15}N$  on experimental field plots in the Venezuelan Andes. *Catena*  
467 2004; 58: 125-150.

468 BoM. Historic annual rainfall totals for Kooralbyn station, Queensland (Archive). 2014, 2013.

469 Boutton TW. Stable Carbon Isotope Ratios of Natural Materials: II Atmospheric, Terrestrial, Marine  
470 and Freshwater Environments. In: Coleman DC, Fry B, editors. *Carbon Isotope Techniques*.  
471 Academic Press Inc., San Diego, 1991, pp. 173-186.

472 Bunn SE, Davies PM, Winning M. Sources of organic carbon supporting the food web of an arid zone  
473 floodplain river. *Freshwater Biology* 2003; 48: 619-635.

474 Caitcheon GG, Olley JM, Pantus F. The dominant erosion processes supplying fine sediment to three  
475 major rivers in tropical Australia, the Daly (NT), Mitchell (Qld) and Flinders (Qld) Rivers.  
476 *Geomorphology* 2012; 151: 188-195.

477 Cerdan O, Govers G, Le Bissonnais Y, Van Oost K, Poesen J, Saby N, et al. Rates and spatial  
478 variations of soil erosion in Europe: a study based on erosion plot data. *Geomorphology* 2010;  
479 122: 167-177.

480 Cole JJ, Prairie YT, Caraco NF, McDowell WH, Tranvik LJ, Striegl RG, et al. Plumbing the global  
481 carbon cycle: Integrating inland waters into the terrestrial carbon budget. *Ecosystems* 2007;  
482 10: 171-184.

483 Coleman DC, Fry B. Carbon Isotope Techniques. In: Paul EA, Melillo JM, editors. Isotopic  
484 Techniques in Plant, Soil and Aquatic Biology. Academic Press Inc., San Diego., 1991, pp.  
485 274.

486 Collins AL, Walling DE. Documenting catchment suspended sediment sources: problems, approaches  
487 and prospects. *Progress in Physical Geography* 2004; 28: 159-196.

488 Cooper RJ, Pedentchouk N, Hiscock KM, Disdle P, Krueger T, Rawlins BG. Apportioning sources of  
489 organic matter in streambed sediments: An integrated molecular and compound-specific  
490 stable isotope approach. *Science of the Total Environment* 2015; 520: 187-197.

491 Davis CM, Fox JF. Sediment fingerprinting: Review of the method and future improvements for  
492 allocating nonpoint source pollution. *Journal of Environmental Engineering* 2009; 135: 490-  
493 504.

494 DME. Queensland geological mapping (polygonised vector). State of Queensland Department of  
495 Mines and Energy, Brisbane, 2008, pp. Data Regional & 1:100000 sheet areas.

496 Evans RD. Soil nitrogen isotope composition. In: Michener R, Lajtha K, editors. *Stable Isotopes in  
497 Ecology and Environmental Science*. Blackwell Publishing, Malden, MA, 2007, pp. 83-98.

498 Finlay JC. Stable-carbon-isotope ratios of river biota: Implications for energy flow in lotic food webs.  
499 *Ecology* 2001; 82: 1052-1064.

500 Finlay JC, Kendall C. Stable isotope tracing of temporal and spatial variability in organic matter  
501 sources to freshwater ecosystems. In: Michener R, Lajtha K, editors. *Stable Isotopes in  
502 Ecology and Environmental Science*. Blackwell, Malden, MA, 2007, pp. 283-333.

503 Fry B. *Stable Isotope Ecology*: Springer. New York., 2006.

504 Garzon-Garcia A, Olley J, Bunn S. Controls on carbon and nitrogen export in an eroding catchment of  
505 south-eastern Queensland, Australia. *Hydrological Processes* 2015; 29: 739-751.

506 Garzon-Garcia A, Olley J, Bunn S, Moody P. Gully erosion reduces carbon and nitrogen storage and  
507 mineralization fluxes in a headwater catchment of southeastern Queensland, Australia.  
508 *Hydrological Processes* 2014; 28: 4669-4681.

509 Gomez B, Trustrum NA, Hicks DM, Rogers KM, Page MJ, Tate KR. Production, storage, and output  
510 of particulate organic carbon: Waipaoa River basin, New Zealand. *Water Resources Research*  
511 2003; 39: 1161-1168.

512 Gregorich EG, Beare MH, McKim UF, Skjemstad JO. Chemical and biological characteristics of  
513 physically uncomplexed organic matter. *Soil Science Society of America Journal* 2006; 70:  
514 975-985.

515 Gregorich EG, Greer KJ, Anderson DW, Liang BC. Carbon distribution and losses: erosion and  
516 deposition effects. *Soil & Tillage Research* 1998; 47: 291-302.

517 Hall BD. SGUM. Measurement Standards Laboratory of New Zealand, Lower Hutt, New Zealand,  
518 2010.

519 Hancock GJ, Revill AT. Erosion source discrimination in a rural Australian catchment using  
520 compound-specific isotope analysis (CSIA). *Hydrological Processes* 2013; 27: 923-932.

521 Hein T, Baranyi C, Herndl GJ, Wanek W, Schiemer F. Allochthonous and autochthonous particulate  
522 organic matter in floodplains of the River Danube: the importance of hydrological  
523 connectivity. *Freshwater Biology* 2003; 48: 220-232.

524 Hughes AO, Olley JM, Croke JC, McKergow LA. Sediment source changes over the last 250 years in  
525 a dry-tropical catchment, central Queensland, Australia. *Geomorphology* 2009; 104: 262-275.

526 Hutchens JJJ, Wallace JB. Ecosystem linkages between Southern Appalachian headwater streams and  
527 their banks: leaf litter breakdown and invertebrate assemblages. *Ecosystems* 2002; 5: 80-91.

528 Juarez S, Rumpel C, MChunu C, Chaplot V. Carbon mineralization and lignin content of eroded  
529 sediments from a grazed watershed of South-Africa. *Geoderma* 2011; 167-168: 247-253.

530 Kao SJ, Liu KK. Stable carbon and nitrogen isotope systematics in a human disturbed watershed  
531 (Lanyang-Hsi) in Taiwan and the estimation of biogenic particulate organic carbon and  
532 nitrogen fluxes. *Global Biogeochemical Cycles* 2000; 14: 189-198.

533 Keith H, Van Gorsel E, Jacobsen KL, Cleugh HA. Dynamics of carbon exchange in a *Eucalyptus*  
534 forest in response to interacting disturbance factors. *Agricultural and Forest Meteorology*  
535 2012; 153: 67-81.

536 Kendall C, Silva SR, Kelly VJ. Carbon and nitrogen isotopic compositions of particulate organic  
537 matter in four large river systems across the United States. *Hydrological Processes* 2001; 15:  
538 1301-1346.

539 Koiter A, Lobb D, Owens P, Petticrew E, Tiessen K, Li S. Investigating the role of connectivity and  
540 scale in assessing the sources of sediment in an agricultural watershed in the Canadian  
541 prairies using sediment source fingerprinting. *Journal of Soils and Sediments* 2013a; 13:  
542 1676-1691.

543 Koiter A, Owens P, Petticrew E, Lobb D. The behavioural characteristics of sediment properties and  
544 their implications for sediment fingerprinting as an approach for identifying sediment sources  
545 in river basins. *Earth-Science Reviews* 2013b; 125: 24-42.

546 Laceby JP, Huon S, Onda Y, Vaury V, Evrard O. Do forests represent a long-term source of  
547 contaminated particulate matter in the Fukushima Prefecture? *Journal of Environmental*  
548 *Management* accepted.

549 Laceby JP, Olley J, Pietsch TJ, Sheldon F, Bunn SE. Identifying subsoil sediment sources with carbon  
550 and nitrogen stable isotope ratios. *Hydrological Processes* 2015; 29: 1956-1971.

551 Lal R. Soil erosion and the global carbon budget. *Environment International* 2003; 29: 437-450.

552 Langhans SD, Tiegs SD, Gessner MO, Tockner K. Leaf-decomposition heterogeneity across a  
553 riverine floodplain mosaic. *Aquatic Sciences* 2008; 70: 337-346.

554 Ludwig W, Probst J. Predicting the oceanic input of organic carbon by continental erosion. *Global*  
555 *Biogeochemical Cycles* 1996; 10: 23-41.

556 Ma WM, Li ZW, Ding KY, Huang B, Nie XD, Lu YM, et al. Soil erosion, organic carbon and  
557 nitrogen dynamics in planted forests: A case study in a hilly catchment of Hunan Province,  
558 China. *Soil & Tillage Research* 2016; 155: 69-77.

559 McCorkle EP, Berhe AA, Hunsaker CT, Johnson DW, McFarlane KJ, Fogel ML, et al. Tracing the  
560 source of soil organic matter eroded from temperate forest catchments using carbon and  
561 nitrogen isotopes. *Chemical Geology* 2016.

562 Milliman JD, Meade RH. World-wide delivery of river sediment to the oceans. *The Journal of*  
563 *Geology* 1983: 1-21.

564 Mukundan R, Radcliffe DE, Ritchie JC, Risse LM, Mckinley RA. Sediment fingerprinting to  
565 determine the source of suspended sediment in a southern piedmont stream. *Journal of*  
566 *Environmental Quality* 2010; 39: 1328-1337.

567 Olley J, Ward D, Pietsch T, McMahon J, Laceby P, Saxton N, et al. Rehabilitation priorities Knapp  
568 Creek. Phase 2a Report. Final Report. Healthy Country Project, 2009.

569 Olley JM, Brooks A, Spencer JS, Pietsch T, Borombovits DK. Subsoil erosion dominates the supply  
570 of fine sediment to rivers draining into Princess Charlotte Bay, Australia. *Journal of*  
571 *Environmental Radioactivity* 2013a; 124: 121-129.

572 Olley JM, Burton J, Smolders K, Pantus F, Pietsch T. The application of fallout radionuclides to  
573 determine the dominant erosion process in water supply catchments of subtropical South-East  
574 Queensland, Australia. *Hydrological Processes* 2013b; 27: 885-895.

575 Olley JM, Wasson RJ. Changes in the flux of sediment in the Upper Murrumbidgee catchment, SE  
576 Australia, since European settlement. *Hydrological Processes* 2003; 17: 3307-3320.

577 Papanicolaou AN, Fox JF, Marshall J. Soil Fingerprinting in the Palouse Basin, USA Using Stable  
578 Carbon and Nitrogen Isotopes. *International Journal of Sediment Research* 2003; 18: 278-284.

579 Parnell AC, Inger R, Bearhop S, Jackson AL. Source partitioning using stable isotopes: Coping with  
580 too much variation. *PlosOne* 2010; 5.

581 Peterson BJ, Fry B. Stable Isotopes in Ecosystem Studies. *Annual Review of Ecology and*  
582 *Systematics* 1987; 18: 293-320.

583 Phillips JM, Russell MA, Walling DE. Time-integrated sampling of fluvial suspended sediment: a  
584 simple methodology for small catchments. *Hydrological Processes* 2000; 14: 2589-2602.

585 Post WM, Kwon C. Soil carbon sequestration and land-use change: processes and potential. *Global*  
586 *Change Biology* 2000; 6: 317-327.

587 Prosser IP, Slade CJ. Gully formation and the role of valley-floor vegetation, southeastern Australia.  
588 *Geology* 1994; 22: 1127-1130.

589 Quinton JN, Govers G, Van Oost K, Bardgett RD. The impact of agricultural soil erosion on  
590 biogeochemical cycling. *Nature Geoscience* 2010; 3: 311-314.

591 Ran L, Lu XX, Xin Z. Erosion-induced massive organic carbon burial and carbon emission in the  
592 Yellow River basin, China. *Biogeosciences* 2014; 11: 945-959.

593 Roscoe R, Buurman P, Velthorst EJ, Vasconcellos CA. Soil organic matter dynamics in density and  
594 particle size fractions as revealed by the  $^{13}\text{C}/^{12}\text{C}$  isotopic ratio in a Cerrado's oxisol.  
595 *Geoderma* 2001; 104: 185-202.

596 Schimel DS. Theory and Application of Tracers. Vol 3: Academic Press, Inc., San Diego., 1993.

597 Scott DT, Baisden WT, Davies-Colley R, Gomez B, Hicks DM, Page MJ, et al. Localized erosion  
598 affects national carbon budget. *Geophysical Research Letters* 2006; 33.

599 Shearer G, Kohl DH. Natural Abundance of  $^{15}\text{N}$ : Fractional Contribution of Two Sources to a  
600 Common Sink and Use of Isotope Discrimination. In: Knowles R, Blackburn H, editors.  
601 Nitrogen Isotope Techniques. 2. Academic Press, Inc., San Diego, 1993.

602 Webster JR, Benfield EF, Ehrman TP, Schaeffer MA, Tank JL, Hutchens JJ, et al. What happens to  
603 allochthonous material that falls into streams? A synthesis of new and published information  
604 from Coweeta. *Freshwater Biology* 1999; 41: 687-705.

605 Wedin DA, Tieszen LL, Dewey B, Pastor J. Carbon isotope dynamics during grass decomposition and  
606 soil organic matter formation. *Ecology* 1995; 76: 1383-1392.

607 Werth M, Kuzyakov Y.  $^{13}\text{C}$  fractionation at the root-microorganisms-soil interface: A review and  
608 outlook for partitioning studies. *Soil Biology and Biochemistry* 2010; 42: 1372-1384.

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611 **Figure Captions**

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

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

621 Figure 3. Mean percent contributions from subsoil, intact valley soil,  $\text{C}_3$  and  $\text{C}_4$  litter to particulate  
622 organic matter (TOC) and TN export in Tilley Gully catchment for the integrated wet season 2011-  
623 2012. Error bars show propagated standard deviations.

624 Figure 4. Mean percent contributions to exported fine sediment (a), particulate organic matter  
625 (TOC) (b) and particulate TN (c) from subsoil, intact valley soil,  $\text{C}_3$  and  $\text{C}_4$  litter during high-flow  
626 events in a dry year (2009-2010) and in a wet year (2010-2011) in the Knapp Creek catchment. Error  
627 bars show standard deviations (a) and propagated standard deviations (b, c).

628 Figure 5. Mean percent contributions to exported fine sediment (a), and particulate organic matter  
629 (TOC) (b) and TN (c) from subsoil, intact valley soil,  $\text{C}_3$  and  $\text{C}_4$  litter for different sampling times  
630 corresponding to different flow stages, during a high-flow event sampled in January-February 2013  
631 at Yarrahappini in the Logan River. Error bars show propagated standard deviations.