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1 **The challenges and opportunities of addressing particle size effects in sediment source**
2 **fingerprinting: a review**

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15 **Abstract:**

16 Tracing sediments back to their catchment sources using biogeochemical and physical fingerprints involves multiple
17 assumptions. One of the most fundamental assumptions is that these fingerprints are consistent during sediment
18 generation, transportation, and deposition processes. Accordingly, the biogeochemical fingerprints used to trace
19 sediment must remain constant, during detachment and redistribution, or they must vary in a predictable and
20 measurable way. One key challenge to this assumption is the sorting effect of particles by size during detachment,
21 mobilization, transportation and deposition processes. Owing to the notable effect of particle size on sediment
22 fingerprints, we believe it is important to review the main approaches used to address the effects of changes in
23 particle size composition on sediment fingerprints. The two main approaches to addressing particle size impacts on
24 fingerprint properties are: fractionation of source and sediment material to a narrow particle size range (e.g. isolation
25 of <10 µm or <63 µm fractions), and concentration corrections (e.g. normalising concentrations by parameters such
26 as specific surface area). These approaches are often used in combination. The utility of fractionation and corrections
27 to address particle size effects has received increasing attention and the relative merits of these procedures have
28 been subject to debate. Accordingly, alternative techniques to address particle size effects in sediment fingerprinting
29 studies are being adopted. For example, a tributary tracing technique or edge-of-field samplers may minimise particle
30 size effects on sediment source fingerprints. The interrelationships between particle size and biogeochemical tracer
31 properties suggest that particle size may also contribute to the formation of contrasts in sediment fingerprints
32 between sources. Indeed, there may be a significant opportunity to derive further sediment source information
33 through comprehensively investigating and unravelling the complexity of particle size–biogeochemical interactions.

34 **Key words:** Grain size; sediment fingerprinting; composite fingerprinting; sediment tracing; sediment provenance

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47 **1. Introduction**

48 Accelerated soil erosion impacts land and water quality worldwide. Although sediment is a natural
49 component of fluvial systems that provides fundamental structure to riverine landscapes and is essential
50 in many aquatic ecosystems (Vercruyssen et al., 2017; Wohl et al., 2015), the excess supply of fine sediment
51 from accelerated soil erosion often degrades riverine and coastal environments (McCulloch et al., 2003;
52 Owens et al., 2005; Walling and Collins, 2016) and contributes to the downstream transfer of particle-
53 bound contaminants (Gateuille et al., 2014; Yamashiki et al., 2014). Elevated suspended sediment loads
54 may also increase the cost of operating and maintaining water treatment and transportation infrastructure
55 (Clark, 1985). Knowledge of the relative contribution of different sources supplying sediment to riverine,
56 lacustrine and coastal systems is a crucial prerequisite to implementing efficient best practices necessary
57 to limit the off-site impacts of excessive sediment delivery (Belmont et al., 2011; Koiter et al., 2013b).

58 One increasingly adopted field-based approach to identifying sources supplying material to riverine,
59 lacustrine and coastal environments is sediment fingerprinting. Tracing sediments back to their primary
60 sources with fingerprinting techniques offers a direct method to identify the nature, location and relative
61 source contribution of sediment transported in waterways. Sediment properties such as mineral magnetic
62 parameters, fallout radionuclides, major and trace element geochemistry, and compound specific stable
63 isotopes (CSSI) have all been used to trace sediment sources (Blake et al., 2012; Caitcheon, 1993; Evrard
64 et al., 2011; Hancock and Revill, 2013; Murray et al., 1993b; Walling and Kane, 1984).

65 For properties to be effective tracers of sediment, they must differentiate between sediment sources
66 whilst behaving conservatively (Walling et al., 1993). Conservative behavior is characterized by constancy
67 in sediment properties, where the properties of sediment sources remain constant, or at the very least,
68 any variation in these properties should occur in a predictable and measurable way. In addition, properties
69 of the eroded sediment should remain constant through sediment detachment, transportation and
70 deposition processes, or again, vary in a predictable and measurable way (Belmont et al., 2014; Koiter et
71 al., 2013b; Motha et al., 2002; Olley et al., 2001).

72 Erosion and transport processes are selective regarding the particle size of the material affected.
73 Detachment processes that generate sediment for fluvial transport are particle size dependent. Clay
74 particles may resist detachment depending on the strength of their bond with the substrate whereas
75 coarse sand may resist detachment simply as a result of size and weight (Bradford et al., 1992; Poesen,

76 1992). Silt and fine sand are thus more subject to detachment and subsequent transport as they are lighter
77 and without bonds binding them to the substrate (Morgan, 2005; Poesen, 1992). Thereafter, the particle
78 size of suspended sediment directly influences settling velocities in aquatic systems (Gibbs et al., 1971)
79 resulting in the transport and deposition of suspended sediment being particle size selective (Viparelli et
80 al., 2013; Walling et al., 2000). Fluvial transport, including overland flow, produces changes in the
81 characteristics of the material being transported in comparison to the original source material. In general,
82 the average size of particles decreases, while the degree of sorting and the average roundness increases,
83 with distance travelled. These changes result from a combination of selective transportation, deposition,
84 and particle abrasion, with these processes acting over the entire landscape (Frings, 2008; Krumbein and
85 Sloss, 1951; Le et al., 2015; Moss and Walker, 1978).

86 Not only are sediment transport processes particle-size selective, the properties used to trace sediments
87 may have different affinities to various particle size fractions. For example, fallout radionuclides are
88 preferentially bound to clay minerals owing to the higher number of potential sorption sites (Fan et al.,
89 2014; Lomenick and Tamura, 1965; Tamura, 1964). Magnetic minerals occur in soil and sediment as
90 aggregated concretions, discrete fine grains, and particle coatings on very fine grains (Oldfield, 1991;
91 Smith, 1999). Different geochemical elements are contained within the mineral matrix or adsorbed
92 (Stumm and Morgan, 2012). Organic matter may coat grains or be bound within the mineral matrix (Keil
93 and Mayer, 2014; Mayer, 1999). Ultimately, the properties used to trace sediment may have different
94 affinities (e.g. preferential adsorption/absorption) for various particles size fractions that in combination
95 with the selective transport of fine-grained material may affect sediment source fingerprinting results.

96 While there are multiple literature reviews published on sediment source fingerprinting (Collins and
97 Walling, 2002; D'Haen et al., 2012; Davis and Fox, 2009; Guzmán et al., 2013; Haddadchi et al., 2013; Koiter
98 et al., 2013b; Owens et al., 2016; Smith et al., 2013), and a recent emphasis on modelling approaches
99 (Cooper et al., 2014; Haddadchi et al., 2014; Laceby and Olley, 2015; Palazón et al., 2015b; Zhang and Liu,
100 2016), few studies have investigated the effects of particle size on sediment source signatures (Olley and
101 Murray, 1994; Russell et al., 2001; Smith and Blake, 2014). Here, we review the effects and challenges
102 (section 2), approaches (sections 3 and 4) and opportunities (section 5) of particle size selectivity within
103 the sediment source fingerprinting technique. We mainly focus on riverine environments, specifically
104 particle size selectivity as sediment moves from hillslopes into and through river channels, but the
105 concepts and examples presented are also relevant for similar applications in other aquatic systems such
106 as lakes, reservoirs, estuaries and the coastal zone.

107 **2. Sediment Property Predictability**

108 Sediment generation, transportation and deposition processes are known to be particle size selective,
109 where fine particles generally have a greater probability of being detached and transported further than
110 coarse particles (McLaren and Bowles, 1985; Walling and Moorehead, 1989). This particle size selectivity
111 often results in potential differences in biogeochemical tracer property concentrations in detached
112 material relative to their sources (He and Walling, 1996; Horowitz and Elrick, 1987). Accordingly, this
113 section will review the impacts of particle size selectivity on a variety of biogeochemical properties used
114 in sediment source fingerprinting research. We focus on some of the main properties – such as fallout
115 radionuclides, carbon and nitrogen parameters, elemental geochemistry and mineral magnetic properties
116 – but recognise that many of the findings are also likely to be relevant to other biogeochemical properties
117 used as fingerprints such as colour parameters and DNA.

118 **2.1 Fallout Radionuclides (^{137}Cs , $^{210}\text{Pb}_{\text{ex}}$, ^7Be)**

119 Caesium-137 (^{137}Cs , $T_{1/2}= 30$ y) and excess lead-210 ($^{210}\text{Pb}_{\text{ex}}$, $T_{1/2}= 22$ y) have been widely used to determine
120 the relative contributions of sediment from different erosion processes to waterways (Ben Slimane et al.,
121 2016; IAEA, 2014; Matisoff et al., 2002; Owens et al., 2012; Smith et al., 2011; Wallbrink et al., 1998;
122 Walling and Woodward, 1992) (Table 1). As both ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ are concentrated near the soil surface,
123 as they are atmospheric fallout products, sediments eroded from rill or sheet erosion often have high ^{137}Cs
124 and $^{210}\text{Pb}_{\text{ex}}$ activity concentrations (Walling, 2005), whereas sediments eroded from subsoil channel bank
125 or gully erosion processes have low ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ activity concentrations (Belmont et al., 2014; Olley et
126 al., 2013; Wallbrink et al., 1999). Comparing ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ in suspended sediments and sediments
127 generated by these different erosion processes generally allows for the relative sediment contributions
128 from these different erosion processes to be ascertained (Ben Slimane et al., 2013; Wallbrink and Murray,
129 1993; Wallbrink et al., 1999; Walling, 2003).

130

Table 1: Examples of research utilizing only fallout radionuclides to trace sediment sources (SSA refers to specific surface area).

Reference	Country	Area (km ²)	Parameters	Particle Size (µm)	Corrections or other approaches to address particle size dynamics	Pre-treatment before analysis	Raw Data Available
Belmont et al., 2011	United States	2880	¹⁰ Be, ²¹⁰ Pb _{ex} , ¹³⁷ Cs	<64, <125 ^a	SSA	--	Yes
Belmont et al., 2014	United States	880	¹⁰ Be, ²¹⁰ Pb _{ex} , ¹³⁷ Cs	<64, <125 ^a	--	--	Yes
Blake et al., 2009	Australia	<1	⁷ Be, ²¹⁰ Pb _{ex} , ¹³⁷ Cs	8 fractions ^b	Enrichment factors	Dispersed ^c	Partial ^d
Bonniwell et al., 1999	United States	389	⁷ Be, ²¹⁰ Pb _{ex} , ¹³⁷ Cs	--	Ratios	--	Yes
Evrard et al., 2010	Mexico	3, 9, 12	⁷ Be, ²¹⁰ Pb _{ex} , ¹³⁷ Cs	--	Sc Comparison	--	Partial ^e
Evrard et al., 2016	Laos	12	⁷ Be, ²¹⁰ Pb _{ex} , ¹³⁷ Cs	<1000	Runoff samplers	--	Yes
Foucher et al., 2015	France	24	¹³⁷ Cs	<20, 20-50, 50-63, <2000	Th correction, SSA	--	Partial ^e
He and Owens, 1995	United Kingdom		²²⁶ Ra, ²¹⁰ Pb _{ex} , ¹³⁷ Cs	<2000	SSA	--	Yes
Gourdin et al., 2014	Laos	12	⁷ Be, ²¹⁰ Pb _{ex} , ¹³⁷ Cs	<1000	Ratios	--	Partial ^e
Matisoff et al., 2002	United States	70	⁷ Be, ²¹⁰ Pb _{ex} , ¹³⁷ Cs	--	--	--	--
Matisoff et al., 2005	United States	--	⁷ Be, ²¹⁰ Pb _{ex}	--	Ratios	--	--
Murray et al., 1993a	Australia	--	¹³⁷ Cs, ²²⁶ Ra, ²³² Th	<2000	--	--	--
Olley et al., 2013	Australia	47-3842	²¹⁰ Pb _{ex} , ¹³⁷ Cs	<10	--	--	--
Owens et al., 2012	Canada	135, 215	²¹⁰ Pb _{ex} , ¹³⁷ Cs	<63	SSA	--	--
Smith et al., 2011	Australia	1.4	²¹⁰ Pb _{ex} , ¹³⁷ Cs	<63	SSA	--	Yes
Stout et al., 2014	United States	4,300	¹⁰ Be, ²¹⁰ Pb _{ex} , ¹³⁷ Cs	<64, <125 ^a	SSA	--	--
Wallbrink and Murray, 1993	Australia	<0.001	⁷ Be, ²¹⁰ Pb _{ex} , ¹³⁷ Cs	--	--	--	--
Wallbrink et al., 1998	Australia	13500	²¹⁰ Pb _{ex} , ¹³⁷ Cs	<2 ^a	--	--	--
Walling and Woodward, 1992	United Kingdom	12, 46	⁷ Be, ²¹⁰ Pb _{ex} , ¹³⁷ Cs	<63	Source correction	--	--

^a Source samples were sieved to a size fraction that matched sediment samples

^b Fractions: <10, 10-20, 20-40, 40-63, 63-125, 125-250, 250-500, 500-2000µm

^c Sonified before sieving prior to analyses

^d For outwash sediment sample

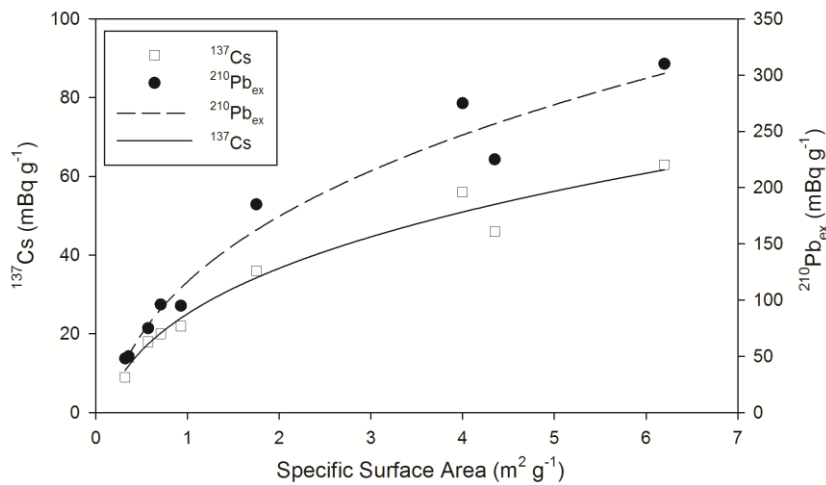
^e Sediments not soils

132 Although they have been used for tracing sediment generated from erosion processes (Wallbrink and
133 Murray, 1996b), beryllium-7 (^7Be , $T_{1/2}= 53$ d) and $^{210}\text{Pb}_{\text{ex}}$ are also increasingly used as chronometers of
134 sediment transfers in riverine systems (Bonniwell et al., 1999; Evrard et al., 2016; Gourdin et al., 2014;
135 Mabit et al., 2014; Smith et al., 2014; Taylor et al., 2013). For example, these radionuclides quantify the
136 relative sediment contribution from 'old' (^7Be -depleted) and 'new' (^7Be -enriched) sources (Evrard et al.,
137 2010; Matisoff et al., 2005). Over longer time scales, additional tracers are capable of providing further
138 chronological information (e.g. ^{10}Be , $T_{1/2}= 1.39 \times 10^6$ y) (Belmont et al., 2011; Stout et al., 2014). In
139 particular, Belmont et al. (2014) combined ^{10}Be , $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs measurements to demonstrate the
140 potential for over-estimating channel source contributions when there is a moderate amount of sediment
141 exchange between the channel and the floodplain in large watersheds over sediment routing timescales.

142 Research has demonstrated that fallout radionuclides are typically enriched in the fine particle size
143 fractions. For example, He and Walling (1996) reported increasing activity concentrations of ^{137}Cs and
144 $^{210}\text{Pb}_{\text{ex}}$ with increasing specific surface area (SSA) (Figure 1). SSA is closely related to particle size (Horowitz,
145 1991) and is reported as the total surface area per unit mass (Rawlins et al., 2010). For example, the SSA
146 of sediment increases with decreasing particle size to the extent that the SSA values for clays may be
147 several orders of magnitude greater than silt and sand (Walling and Moorehead, 1989). As fallout
148 radionuclides are preferentially bound to clay minerals owing to the higher number of potential sorption
149 sites (Fan et al., 2014; Lomenick and Tamura, 1965; Tamura, 1964), activity concentrations typically
150 increase with increasing SSA and decreasing particle size (He and Owens, 1995; Wallbrink et al., 1999).
151 Although less research has documented the relationship between particle size and ^7Be , this fallout
152 radionuclide has also been found to be enriched in fine particle size fractions (Blake et al., 2009; Taylor et
153 al., 2014; Wallbrink and Murray, 1996a). As fallout radionuclides are typically enriched in fine particle size
154 fractions, it may be possible to quantify their relationship with SSA.

155 The challenge is that there are exceptions to the rule. For example, Smith and Blake (2014) observed that
156 ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ were negatively related to SSA in pasture soils ($p < 0.05$ for $^{210}\text{Pb}_{\text{ex}}$ only), whereas they
157 exhibited positive though non-significant relationships with SSA in channel bank and cultivated soils. This
158 highlights the fact that although these fallout radionuclides are generally enriched in the fine particle size
159 fractions, there may be exceptions, and individual sources may behave differently. Potential exceptions
160 may be explained by the presence and amount of HCl-extractable materials, which Singleton et al. (2017)
161 reported to have a stronger control on fallout radionuclides than grain size or mineralogy. Furthermore,
162 the depth-dependent distribution of fallout radionuclides in the soil profile relates to the exposure to

163 fallout and subsequent diffusion and migration processes (Jagercikova et al., 2015). Therefore, fallout
164 radionuclide activity concentrations may decrease with soil depth despite increasing clay content as
165 deeper soil was not exposed to fallout. These exceptions demonstrate the need to understand the
166 relationship between particle size distribution and the tracer property of interest in each study.



167
168 **Figure 1:** Relationship between specific surface area and fallout radionuclide activity concentrations (¹³⁷Cs
169 and ²¹⁰Pb_{ex}) (adaptation of Figure 1 from He and Walling (1996)).

170 2.2 Carbon and Nitrogen Parameters

171 Although they are not as extensively analyzed in sediment tracing research as fallout radionuclides, carbon
172 and nitrogen parameters provide an interesting example into tracer property relationships with particle
173 size. Total organic carbon (TOC) and total nitrogen (TN) often discriminate between sediment derived from
174 surface and subsoil erosion processes as their concentration decreases with depth in the soil profile (Blake
175 et al., 2006; Owens et al., 2006). Carbon stable isotopes ($\delta^{13}\text{C}$) may potentially discriminate between
176 sediment derived from soils with C₃ vegetation (majority of tree or temperate grass species) compared to
177 those covered with C₄ vegetation (grass and cropping species typically under warmer climates) (Fry, 2006;
178 Schimel, 1993). Source discrimination with nitrogen stable isotopes ($\delta^{15}\text{N}$) is more complex. In general, soil
179 $\delta^{15}\text{N}$ increases with depth in soil profiles (Amundson et al., 2003; Natelhoff and Fry, 1988). Other factors
180 such as nitrogen inputs from animal and human waste, along with fertilizers and potentially topographic
181 position, may impact the predictability of $\delta^{15}\text{N}$ in a tracing context. Of note, these parameters trace organic
182 matter (Garzon-Garcia et al., 2017; Olley, 2002) and therefore they may be fractionated by density as well
183 as particle size. Furthermore, these parameters also have the potential for non-conservative behavior due
184 to biological uptake and consumption. Nonetheless, these parameters are increasingly used in sediment

185 tracing research (Fox and Papanicolaou, 2007; Laceby et al., 2015b; Mukundan et al., 2010; Papanicolaou
186 et al., 2003) (Table 2).

187 Similarly to fallout radionuclides, TOC and TN are generally enriched in the fine particle size fraction
188 (Balesdent et al., 1987; Wynn et al., 2005). For example, when normalizing the <2, <10, and <63 μm
189 fractions by the bulk soil (<2 mm) fraction, Laceby et al. (2015b) reported that the <63 μm fraction was
190 significantly different than the <2 and <10 μm fractions for TN, though not for TOC, indicating that these
191 similar properties may behave slightly differently in two Australian catchments (Figure 2). Laceby et al.
192 (2016) also reported significant enrichment for TOC and TN between the bulk soil and the <63 μm fraction
193 for subsoils and cultivated sources, though not for forest source samples in several Japanese catchments
194 (Figure 3).

195 More variability is anticipated in the relationship between particle size and $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ (Balesdent et al.,
196 1987; Bellanger et al., 2004). However, Laceby et al. (2015b) reported that there were not significant
197 differences between particle size fractions for $\delta^{15}\text{N}$, with very limited $\delta^{13}\text{C}$ variation across different
198 particle size fractions (Figure 2). Similarly, Laceby et al. (2016) found little variation between the bulk soil
199 fraction and the <63 μm fraction for $\delta^{13}\text{C}$, with increasing, though not significant, enrichment for $\delta^{15}\text{N}$
200 (Figure 3).

201 Based on the fundamental principles of sediment source fingerprinting, as long as the enrichment is
202 predictable, these organic sediment properties could potentially be effective tracers of erosion processes,
203 particularly in contexts where fallout radionuclides are ineffective. $\delta^{15}\text{N}$ appears more complex and may
204 be enriched or depleted in the different particle size fractions, which requires investigation on a case by
205 case basis. $\delta^{13}\text{C}$ apparently behaves very predictably in a sediment tracing context although there will likely
206 be exceptions. Importantly, the particle size enrichment for TOC, TN and $\delta^{15}\text{N}$ varied for the individual
207 sources (Laceby et al., 2016). Therefore, it is crucial to understand the predictability of particle size effects
208 when tracing sediment sources with these and other carbon and nitrogen properties (e.g. compound
209 specific stable isotopes (Reiffarth et al., 2016)).

210

Table 2: Examples of research using only carbon (C) and nitrogen (N) parameters to trace sediment sources

Reference	Country	Area (km ²)	Parameters	Particle Size (µm)	Corrections or other approaches to address particle size dynamics	Pre-treatment before analysis	Raw Data Available
Fox and Papanicolaou, 2007	United States	0.71	δ ¹³ C, δ ¹⁵ N, C/N	<53	--	Dispersed	Yes
Garzon-Garcia et al., 2017	Australia	2.5, 75, 3076	δ ¹³ C, δ ¹⁵ N, TOC, TN	<10, <63, <500	--	--	Yes
Gibbs, 2008	New Zealand	117	CSSI	<1000	--	--	Yes
Hancock and Reville, 2013	Australia	3860	CSSI	<63	--	--	--
Lacey et al., 2015b	Australia	75, 123, 311	δ ¹³ C, δ ¹⁵ N, TOC, TN	<2, <10, <63, <2000	--	--	--
Lacey et al., 2016	Japan	77, 171, 265	δ ¹³ C, δ ¹⁵ N, TOC, TN	<63, <2000	--	--	Yes
Olley, 2002	Australia	84000	δ ¹³ C, C/N Ratio	<2	--	--	--
Papanicolaou et al., 2003	United States	600	δ ¹³ C, δ ¹⁵ N, C/N	--	--	--	--

211

^a Source samples were dispersed with sodium hexametaphosphate

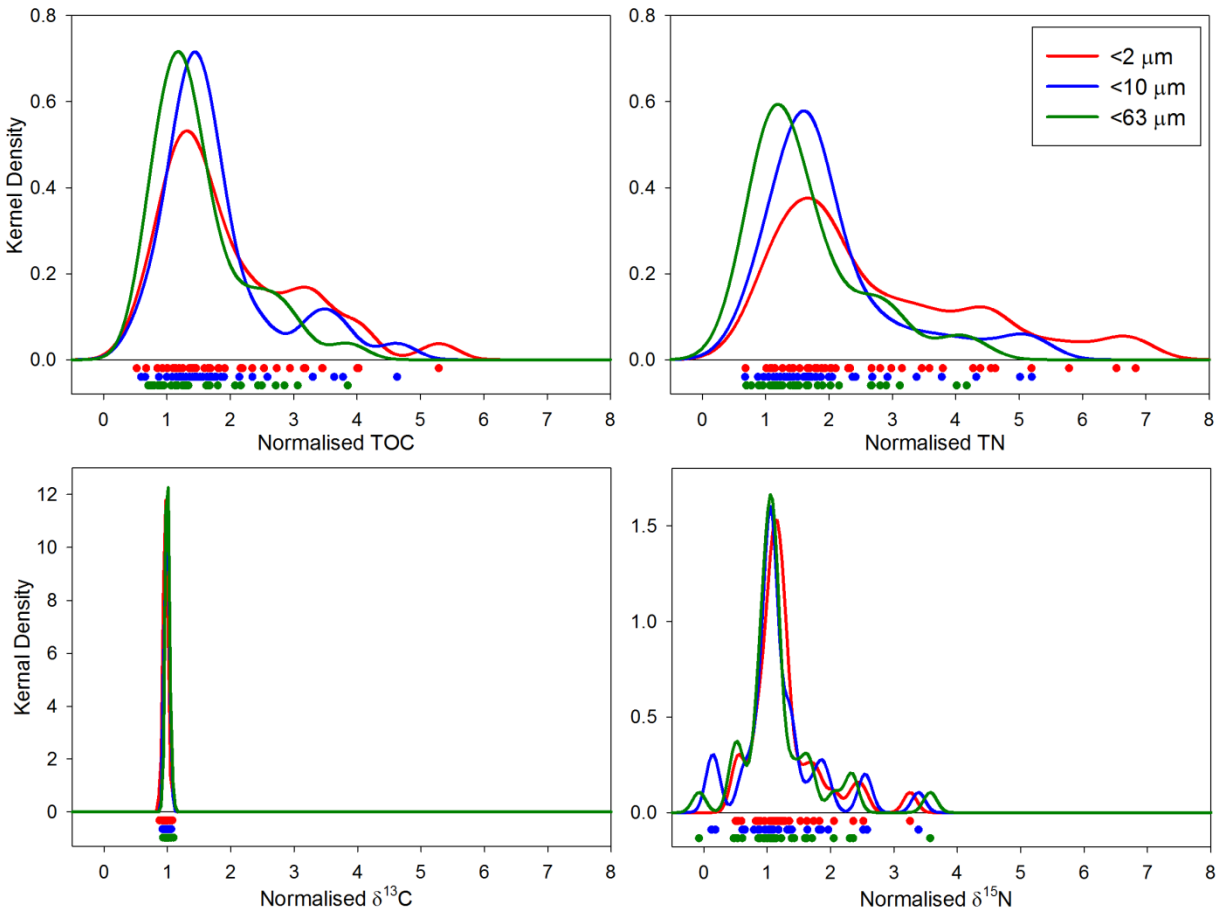


Figure 2: Normalized carbon (C) and nitrogen (N) property distributions for the <2 μm, <10 μm and <63 μm particle size fractions in samples from Knapp Creek and Blackfellow Creek, Australia (modified from Laceby et al., (2015b)). Points under the distributions are the normalized samples color coded to particle size fraction used to derive these distributions with kernel density functions.

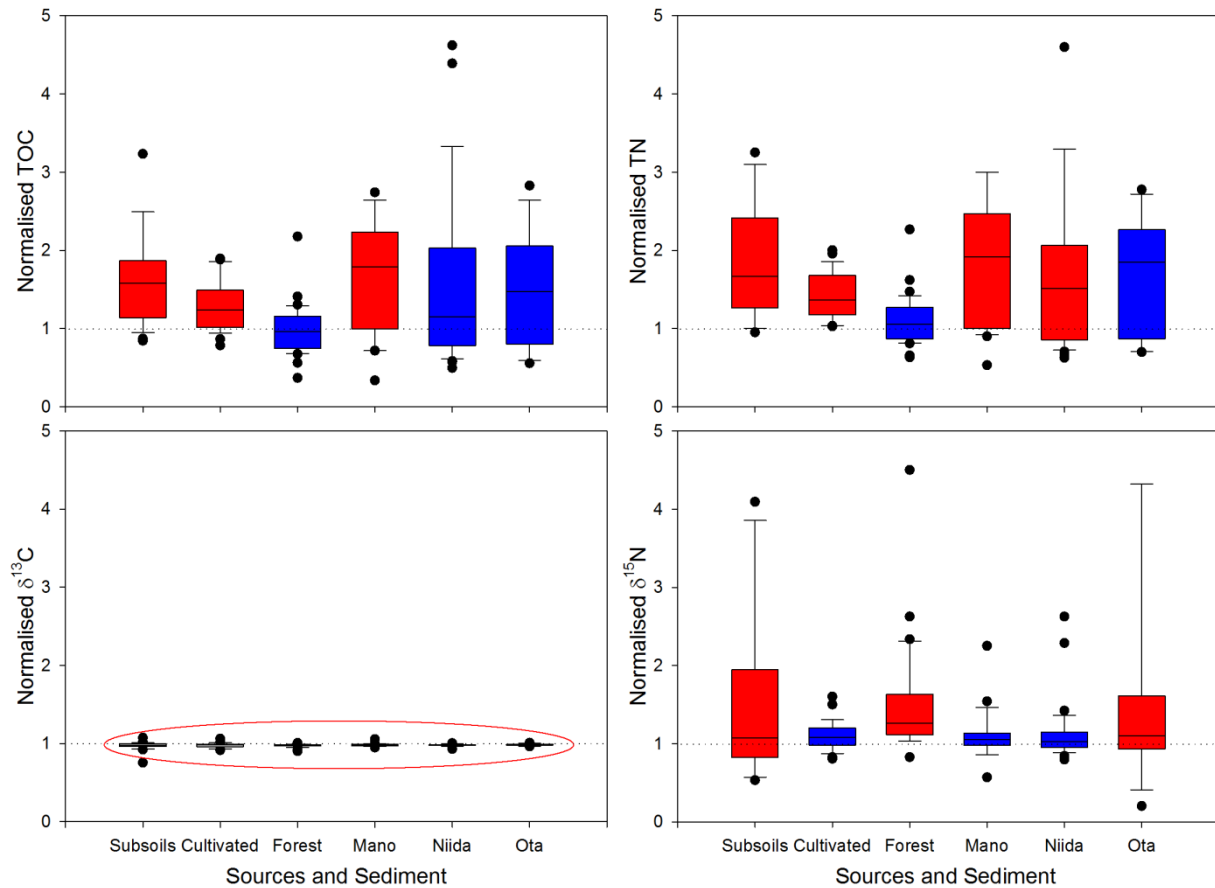


Figure 3: Box plots of the normalized difference between carbon (C) and nitrogen (N) sediment properties in the <63 μm fraction divided by the <2 mm fraction for the three sources and sediment from three Japanese catchments (i.e., Mano, Niida and Ota) with red shading (and the red circle) indicating significant differences between the two fractions and values greater than 1 (the dotted line) being enriched in the <63 μm fraction (modified from Lacey et al., (2016)).

213 **2.3 Elemental Geochemistry**

214 Major and trace elemental geochemistry (including rare earth elements) are often used to identify the
 215 different spatial sources of sediment (Hardy et al., 2010; Lacey and Olley, 2015; Vale et al., 2016) (Table
 216 3). Different parent rock material typically results in sources having distinct elemental geochemistry
 217 (Douglas et al., 2009; Motha et al., 2002; Olley et al., 2001). Eroded sediment often maintain these
 218 geochemical fingerprints, allowing the relative contributions of different sources to be ascertained
 219 (Caitcheon et al., 2006; D'Haen et al., 2013; Hughes et al., 2009). The question is whether these fingerprints
 220 are conservative during sediment generation, transportation and deposition processes.

221 The relationship between particle size enrichment and elemental geochemistry is complex and partly
 222 dependent on the digestion procedure (e.g. acid leached versus total digestion) used to prepare samples

223 for analysis (e.g. inductively coupled plasma mass spectrometry (ICP-MS)). The difficulty is that elemental
224 analyses often provide results for over 40 elements (Table 3) whereas the previous sections examined
225 three fallout radionuclides and four carbon and nitrogen parameters. For each of these 40 plus elements,
226 sediment generation, transport and deposition processes may potentially enrich their elemental
227 concentrations, deplete them, or have a limited impact (e.g. Motha et al., 2002; Russell et al., 2001; Smith
228 and Blake, 2014). The impact of particle size selectivity on elemental geochemistry likely will depend on
229 how elements are incorporated into fine sediment (e.g. within the mineral matrix or adsorbed).

230 Underlying the potential influence of particle size on elemental geochemistry is the effect of sediment
231 source mineralogy. The dominant mineralogy relates directly to particle size where some sources will be
232 enriched in the fine particle size fractions and other sources may be depleted. Thus the geochemical
233 fingerprint likely will change if the <2 mm, the <63 μm , or the <10 μm fraction are sampled and analysed.
234 For example, each of the three different sources in Figure 4 from Gibbs (1967) may have different
235 relationships between elemental concentrations and particle size, which will depend on the particle size
236 range utilized for the sediment source fingerprinting research. Fundamentally, the impact of particle size
237 on all elemental concentrations for each source is difficult to predict and the complexity of the particle
238 size – elemental geochemistry relationship requires more research to comprehensively characterize the
239 predictability of these fingerprints. Accordingly, section 4 of this review provides several
240 recommendations for addressing particle size in sediment source fingerprinting research.

Table 3: Examples of sediment tracing research using only elemental geochemistry (e.g. major, trace and rare earth elements).

Reference	Country	Area (km ²)	Elements Analyzed (n)	Particle Size (µm)	Corrections or other approaches to address particle size dynamics	Pre-treatment before analysis	Raw Data Available
Cooper et al., 2015	United Kingdom	5	11	<63	--	Dispersed ^a	--
D'Haen et al., 2013	Turkey	264	18	<63	--	Dispersed ^b	--
Douglas et al., 2003	Australia	22000	50	<10	--	--	--
Haddadchi et al., 2015	Australia	911	41	<10, 10-63, 63-212	--	--	--
Hardy et al., 2010	Canada	12000	55	63-250	Density separation	--	--
Lacey et al., 2015a	Australia	74	37	<10	--	--	--
Lacey and Olley, 2015	Australia	75, 123, 311	23	<10	--	--	--
Olley and Caitcheon 2000	Australia	650000	10	<10	--	Dispersed ^c	Partial ^d
Vale et al., 2016	New Zealand	5870	44	<63	--	--	--

241

^a Source samples sonified before sieving

242

^b Samples were boiled in distilled water to disperse soil aggregates

243

^c Samples sonified before settling in a water column

244

^d Raw data is only available for sediment core samples

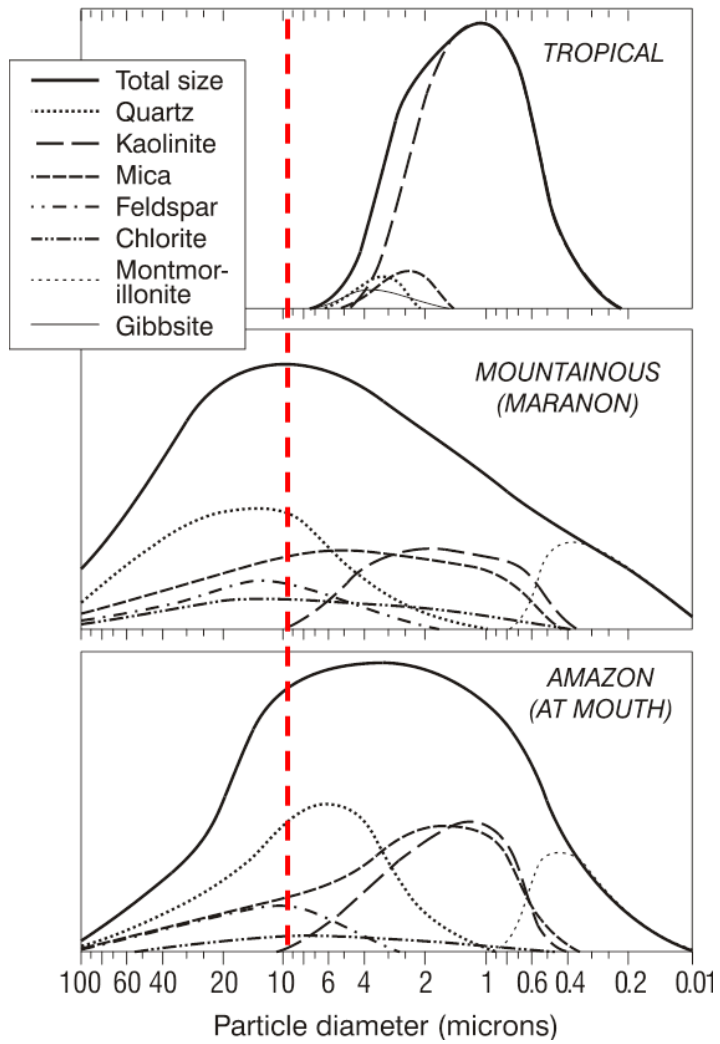


Figure 4: Changes in mineralogy with particle size in sediment from three locations in the Amazon basin from Gibbs (1967) with the red dashed line added to indicate the impact of fractionating the samples at the <math><10 \mu\text{m}</math> particle size.

246 **2.4 Mineral Magnetic Properties**

247 Mineral magnetic properties (e.g. magnetic susceptibility, isothermal remanent magnetisation) have also
248 been widely used to investigate sediment provenance (Blake et al., 2004; Palazón et al., 2015a; Pulley et
249 al., 2015b; Walling et al., 1979) (Table 4). The signatures derived from magnetic minerals may be classified
250 as primary (i.e. from parent material prior to weathering) or secondary (i.e. from chemical processes and
251 other processes and effects) (Hatfield, 2014). As the signature is often derived from parent material,
252 mineral magnetic properties can trace sediment derived from different spatial sources (Caitcheon, 1993).
253 Owing to the potential impact of secondary processes, such as anthropogenic inputs and diagenetic
254 processes, mineral magnetic properties may also provide further source discrimination between different
255 erosion processes (Foster et al., 1998; Pulley et al., 2015b).

256 Magnetic minerals occur in soil and sediment as aggregated concretions, discrete fine grains and particle
257 coatings on very fine grains (Oldfield, 1991; Smith, 1999). Accordingly, mineral magnetic properties may
258 be highly dependent on particle size (Foster et al., 1998; Oldfield et al., 1985). In particular, Hatfield and
259 Maher (2008) demonstrated the importance of characterizing mineral magnetic properties with a particle-
260 size specific approach as different magnetic properties were preferentially associated with different
261 particle size fractions. These authors demonstrated that bacterial magnetosomes formed in lake sediment
262 in the <2 µm and the 2-8 µm fractions. Accordingly, they only quantified source contributions to the 8-31
263 µm and 31-63 µm fractions. In contrast, Pulley et al. (2015b) reported significantly different magnetic
264 properties in the <32 µm and the >32 µm fractions and, for Caitcheon (1998), the 63-125 µm fraction was
265 appropriate for tracing tributary source contributions.

266 The challenge is that the particle size fraction driving the mineral magnetic signature may vary for each
267 catchment. For example, the sand and silt fractions may be the most appropriate particle size in
268 catchments in England to quantify sediment sources with magnetic fingerprinting techniques as these
269 fractions have been found to contribute a significant proportion of the mineral magnetic signature
270 (Hatfield and Maher, 2009). Conversely, high magnetic parameters were found in the clay-size particles in
271 the eastern United States (Oldfield et al., 1985). In South Africa, the impact of particle size on mineral
272 magnetic properties was found to be limited in the >32 µm fraction (Pulley et al., 2015b).

273

Table 4: Examples of sediment source fingerprinting research with only mineral magnetic analyses

Reference	Country	Area (km ²)	Magnetic Parameters (n)	Particle Size (µm)	Corrections or other approaches to address particle size dynamics	Pre-treatment before analysis	Raw Data Available
Blake et al., 2004	Australia	446	9	<10	--	--	Yes
Caitcheon, 1993	Australia	22	2	7 fractions ^a	Density separation	--	--
Foster et al., 1998	United Kingdom	1.5, 12	10	11 fractions ^b	--	Dispersed ^c	--
Hatfield and Mayer, 2009	United Kingdom	240	10	<2, 2-8, 8-31, 31-63, >63	--	Dispersed ^d	--
Oldfield et al., 1985	United States	33	7	10 fractions ^e	--	Dispersed ^f	--
Pulley et al., 2015b	South Africa	148-5751	6	7 fractions ^g	--	Dispersed ^h	--
Slattery et al., 1995	United Kingdom	6	4	<2, 2-16, 16-63	Source correction ⁱ	Dispersed ^j	--
Walling et al., 1979	United Kingdom	12	5	--	--	--	--

^a 7 particle size fractions analyzed: <63, 63-125, 125-250, 250-500, 500-1400, 1400-2000 µm

^b Sediment cores: no fractionation, surface soils: <63 µm, soil cores: <2000 µm, and one bulk sediment samples split into 11 fractions

^c Calgon before dry-sieving to 4 φ and for <4 φ sodium hexametaphosphate and anhydrous Na₂CO₃ for the bulk sample split into 11 fractions.

^d Fractionated samples were dispersed with Calgon prior to sonification

^e 10 fractions analyzed (1-10 φ)

^f Dispersed with Calgon

^g <32, 32-63, 63-125, 125-250, 250-500, 500-1000, 1000-2000µm

^h Dispersed with sonification

ⁱ Corrected to a standard particle size distribution consisting of coarse silt (50%), fine silt (40%) and clay (10%)

^j Dispersal method not provided

275 The challenge for mineral magnetic properties, and other sediment fingerprints, is that these properties
276 are related to particle size and this relationship varies from catchment to catchment, subcatchment to
277 subcatchment, and even potentially from event to event. Further, these sediment properties are not only
278 sensitive to changes in their sources, they are also sensitive to changes in sediment transport processes
279 that may impact the potential abundance and availability of the different particle size fractions over a
280 range of spatial temporal scales (Hatfield, 2014).

281 **2.5 Composite Fingerprinting Approach**

282 Peart and Walling (1986) advocated for multiple parameters to be used when quantifying sediment source
283 dynamics in order to improve the overall consistency and reliability of source ascription results. The
284 combination of multiple parameters creates a composite fingerprint (Walling et al., 1993) that allows for
285 an increased number of sources to be modelled and is theorized to be more representative of the linkages
286 between sediments and their sources, potentially reducing false matches which were hypothesized to
287 potentially occur with individual tracer properties (Collins et al., 1996). Accordingly, a composite
288 fingerprinting approach has been broadly applied in sediment source fingerprinting research combining
289 several or all of the following: fallout radionuclides, carbon and nitrogen parameters, element
290 geochemistry, mineral magnetism and other parameters, thereby providing significant source
291 discrimination (Collins et al., 1996; Evrard et al., 2013; Navratil et al., 2012; Owens et al., 2000) (Table 5).
292 One challenge with the composite fingerprinting approach is that each of the potential complexities within
293 the particle size – tracer parameter relationship described above is integrated into the expanded
294 composite fingerprint. A second challenge is that it is difficult, if not impossible, to link outputs of
295 statistical-based approaches (e.g. composite fingerprinting) back to a process-based understanding of
296 sediment dynamics.

Table 5: Examples of sediment source fingerprinting research using a composite fingerprinting approach incorporating two or more of types of biogeochemical parameters with geochemistry (Geochem), mineral magnetics (Mags), radionuclides (RN), clay mineralogy (Clay min.), diffuse reflectance infrared Fourier transform spectrometry (DRIFTS), X-ray Diffraction (XRD) and other tracers as listed.

Reference	Country	Area (km ²)	Parameters	Particle Size (µm)	Corrections or other approaches to address particle size dynamics	Pre-treatment before analysis	Raw Data Available
Ben Slimane et al., 2013	Tunisia	2.6	RN, C, N	<2000	SSA	--	--
Ben Slimane et al., 2016	Tunisia	0.6-4	RN, C	<2000	SSA	--	--
Blake et al., 2006	Borneo	<2	Geochem, N	<125	--	--	--
Blake et al., 2012	United Kingdom	1.5	CSSI, Geochem, Mags	<63	SSA ^a	--	--
Caitcheon et al., 2006	Australia	9051	Geochem, RN	<10	--	--	Partial ^b
Collins et al., 1996	United Kingdom	601, 4325	Geochem, RN, C,N	--	SSA	--	--
Devereux et al., 2010	United States	188	Geochem, RN, C	<63	--	--	--
Douglas et al., 2009	Australia	638	Geochem, RN	<10	--	--	--
Douglas et al., 2006a	Australia	144000	Geochem, Clay Min.	<10	--	--	--
Douglas et al., 2006b	Australia	144000	Geochem, Clay Min.	<10	--	--	--
Evrard et al., 2011	France	907	Geochem, RN	<2000	Sc comparison	--	Partial ^c
Evrard et al., 2013	Mexico	3, 9, 12, 630	Geochem, RN, DRIFTS, C, N, δ ¹³ C	<250	d ₅₀ comparison	--	Partial ^c
Hatfield and Mayer, 2008	United Kingdom	240	Geochem, Mags	<2, 2-8, 8-31, 31-63	--	Dispersed ^d	--
Hughes et al., 2009	Australia	6000	Geochem, RN	<10	--	--	--
Koiter et al., 2013b	Canada	74	Geochem, RN	<2000	--	--	--
Le Gall et al., 2016	France	24	Geochem, RN, Sr Isotopes	<63, <2000	Th-correction	--	Yes
Martínez-Carreras et al., 2010	Luxembourg	0.7, 3, 4	Geochem, RN, Colour, C, N, P	<63	--	--	--
Minella et al., 2008	Brazil	1.2	Geochem, C	<150	--	--	--
Motha et al., 2002	Australia	110	Geochem, RN, Mags	<2, 2-20, 20-40, 40-63	Source correction ^e	Dispersed ^f	--
Motha et al., 2003	Australia	65	Geochem, RN	<2, 2-20, 20-40, 40-63	Source correction ^e	Dispersed ^f	--
Mukandan et al., 2010	United States	182	Geochem, RN, δ ¹⁵ N,C,N	<2000	Texture comparison	--	--

Navratil et al., 2012	France	905	Geochem, RN	<63	--	--	Partial ^c
Owens et al., 2000	United Kingdom	4390	Geochem, RN, Mags, C, N	<63	SSA	Dispersed ^d	--
Owens et al., 2006	Canada	135, 215	Geochem, Mags, C, N	<500	--	--	--
Palazon et al., 2015a	Spain	1509	Geochem, Mags, RN, C	<63	--	--	Partial ^c
Poleto et al., 2009	Brazil	0.8	Geochem, C	<63	SSA	--	--
Pulley et al., 2015a	United Kingdom	1634	Geochem, Mags, RN	<63	SSA / None	--	--
Russell et al., 2001	United Kingdom	1.5, 4	Geochem, Mags, RN, C, N	<2, 10, 38, 63	Tracer specific particle size correction factor	--	--
Tiecher et al., 2016	Brazil	1.2	Geochem, DRIFTS, XRD	<63	--	--	--
Sherriff et al., 2015	United Kingdom	11	Geochem, Mags	<125	--	--	--
Smith and Blake 2014	United Kingdom	920	Geochem, RN, C	<63	SSA / None	--	--
Stone et al., 2014	Canada	751	Geochem, C	<63	SSA	--	--
Walling et al., 1993	United Kingdom	12, 46	RN, Mags, C,N	<63	None (Mags) and source correction (FRN)	--	--
Walling et al., 1999	United Kingdom	818, 3315	Geochem, Mags, RN,C,N	<63	SSA	--	--
Zhang and Liu, 2016	United States	15.6	Geochem, C,N	<53	--	--	--

^a SSA correction was only used for the composite fingerprinting approach, not the CSSI tracers

^b Only geochemistry data is available

^c Sediments not sources

^d Dispersed with Calgon and then sonified

^e Fractional mass of each sediment size fraction was multiplied by their corresponding source tracer property and summed

^f Dispersed with sonification

298 **3. Main Approaches to Address Particle Size**

299 Owing to the potential of particle size to affect the values of tracer properties, researchers have adopted
300 two main approaches to predict or mitigate particle size effects on sediment source fingerprints:
301 fractionation and particle size corrections. Tables 1 to 5 summarize the different approaches used by
302 researchers to address particle size for fallout radionuclides (Table 1), carbon and nitrogen parameters
303 (Table 2), elemental geochemistry (Table 3), mineral magnetic properties (Table 4) and combinations of
304 multiple parameters in a composite fingerprinting approach (Table 5).

305 **3.1 Fractionation**

306 To address particle size impacts on sediment fingerprint properties, researchers often fractionate both
307 their sediment and source samples to a specific and comparable particle size fraction using settling based
308 on Stokes' Law or sieving. The objective is to minimize potential sorting-induced differences between
309 source and sediment properties. Conceptually, any particle size fraction can be isolated to attempt to
310 achieve this objective, though it is mainly the <10 µm and <63 µm fractions that have been isolated in
311 sediment source fingerprinting research.

312 The <10 µm fraction is predominantly used in Australia to research the source of very fine silt and clay
313 material. The logic supporting the use of the <10 µm fraction is that it is the dominant size fraction being
314 transported in these river systems and this fraction has the greatest ecological and water quality impact
315 (Douglas et al., 2003; Olley and Caitcheon, 2000). The <10 µm fraction is isolated in settling columns based
316 on Stokes' Law with assumptions of constant temperature, roundness and density of the particles
317 (Fontaine et al., 2000; Walden and Slattery, 1993).

318 The <63 µm fraction is arguably the most adopted sediment tracing particle size fraction (Devereux et al.,
319 2010; Pulley et al., 2015a; Walling et al., 1993). This fraction represents the silt and clay material that is
320 transported preferentially as suspended sediment in riverine, lacustrine, estuarine and coastal systems.
321 The <63 µm fraction is also one of the smallest dry/wet sieve sizes at the fine sand to silt size boundary
322 (i.e. an operationally defined fraction/separation). Research examining the different impacts of wet sieving
323 and dry sieving on biogeochemical properties may be warranted in the sediment source fingerprinting
324 context, along with research on the impact of different pre-treatments to disperse aggregates prior to
325 analyses (Tables 1-5).

326 One often overlooked component of addressing particle size is that the fraction isolated should include
327 the range of fingerprint property values in the potential sources. This is particularly important for
328 elemental geochemistry. For example in Figure 4, it is apparent that isolating the <2 µm, <10 µm and <63
329 µm fractions will likely result in different elemental compositions for the three sources based on the
330 particle size fraction selected. This may be particularly important with mineral magnetic tracer properties
331 where different signatures exist in the <32 and the 32-63 µm soil and sediment fractions, indicating that
332 sieving to <63 µm may be inappropriate in some regions (Pulley et al., 2015b).

333 A second overlooked component is that the particle size fraction isolated for sediment property analyses
334 should directly relate to the research objective. For example, researchers have shown that if the objective
335 is to examine the source of material degrading the Great Barrier Reef near Australia, the <16 µm particle
336 size fraction should likely be targeted (Bartley et al., 2014). Researchers should support their choice of
337 particle size fraction by relating it to the particle size being transported in the stream system (Wallbrink et
338 al., 1999), or the particle size of the sediment-associated contaminants of interest (Olley and Caitcheon,
339 2000). For both approaches, there is a trade-off between fractionating down to the finest particle size (e.g.
340 <2 µm) versus using a broader particle size fraction (e.g. <63 µm) that may require more steps to address
341 discrepancies between source and sediment particle size distributions. In some regions, the abundance of
342 material present in the sediment and sources may even control the particle size selected as there may not
343 be sufficient <10 µm or even <63 µm material available for analysis. Indeed, the results from sediment
344 source fingerprinting research will only relate to the particle size fraction examined. If narrow particle size
345 ranges are fractionated (e.g. <2 µm or <10 µm), the results from the source apportionment modelling will
346 only apply to that fraction and not the entire suspended sediment load more generally (Mukundan et al.,
347 2012).

348 Ultimately, the key to using fractionation to address particle size differences is to ensure that the grain
349 size distribution of the source material is similar to that of the sediment sampled (i.e. Poulencard et al.,
350 2009). For example, Sherriff et al. (2015) found that the 90th percentile of the suspended sediment
351 distribution for select samples was frequently >63 µm and thus these authors sieved samples to <125 µm.
352 Furthermore, it is important to understand the relationship between particles size distribution and event
353 magnitude, and even how this relationship is impacted by seasonality (Bogen, 1992; Lewis, 1996). In this
354 regard, it would be beneficial for fingerprinting studies to report summary statistics for particle size data
355 of both source soils and sediments to present this important comparison. The challenge is that particle
356 size fractionation of samples alone will not necessarily reduce the discrepancy between the fractionated

357 samples (Cooper et al., 2015a; Kersten and Smedes, 2002). If there are significant differences between
358 source and sediment particle size distributions remaining after fractionation, particle size corrections are
359 potentially required.

360 **3.2 Particle Size Corrections**

361 To mitigate differences in the particle size distributions of source soil and sediment, corrections have been
362 applied based on particle size characteristics of source and sediment material (Collins et al., 1996; Slattery
363 et al., 1995; Walling and Woodward, 1992; Walling et al., 1993) (Tables 1, 4, 5). The initial corrections
364 reconstructed particle size distribution and tracer parameters of the source materials to allow for the
365 direct comparison with the sampled sediment (Slattery et al., 1995; Walling and Woodward, 1992; Walling
366 et al., 1993). The objective of these corrections was to reduce the impact of source and sediment particle
367 size distributions on tracer parameters prior to quantifying source contributions.

368 A second approach to particle size corrections is the incorporation of a within-model weighting (Collins et
369 al., 1996) which has been broadly applied (Collins et al., 2012; Poletto et al., 2009; Stone et al., 2014; Walling
370 et al., 1999). The within-model correction incorporates some variant of a particle size weighting, such as
371 the ratio of the mean SSA in sediment to the mean SSA in each source (Collins et al., 1996; Collins et al.,
372 2010). The logic supporting the use of a SSA derived correction is that particle size and specific surface
373 area are closely related (Horowitz, 1991). Surface area is one of the most important controls on sediment
374 trace element concentrations as the majority of these interactions are postulated to be related to surface
375 area chemistry or surface area reactions (Horowitz and Elrick, 1987).

376 Although widely applied, there are acknowledged limitations and challenges with particle size corrections.
377 Russell et al. (2001) reported a large range in SSA between different sources and sediment for a catchment
378 in the UK and found that a linear-based particle size correction may be inappropriate in some cases. Smith
379 and Blake (2014) further demonstrated that the fundamental assumption of some particle size corrections
380 (i.e. positive linearity between particle size and tracer concentration) does not apply to all tracer properties
381 or equally to properties from different sources (Figure 5). These authors reported that this basic
382 assumption of linearity needs to be constantly examined and its dependence on analytical methods (e.g.
383 acid leached versus total digestion for ICP-MS analyses) should also be considered. Particle size
384 corrections, if adopted, probably should be more rigorous than simple SSA ratio model weightings (e.g.
385 Motha et al., 2002; Motha et al., 2003; Russell et al., 2001; Slattery et al., 1995; Walling and Woodward,

386 1992). There may even be potential to incorporate particle size properties and organic matter content
 387 effects simultaneously with a stepwise multiple regression analysis model (Kraushaar et al., 2015).

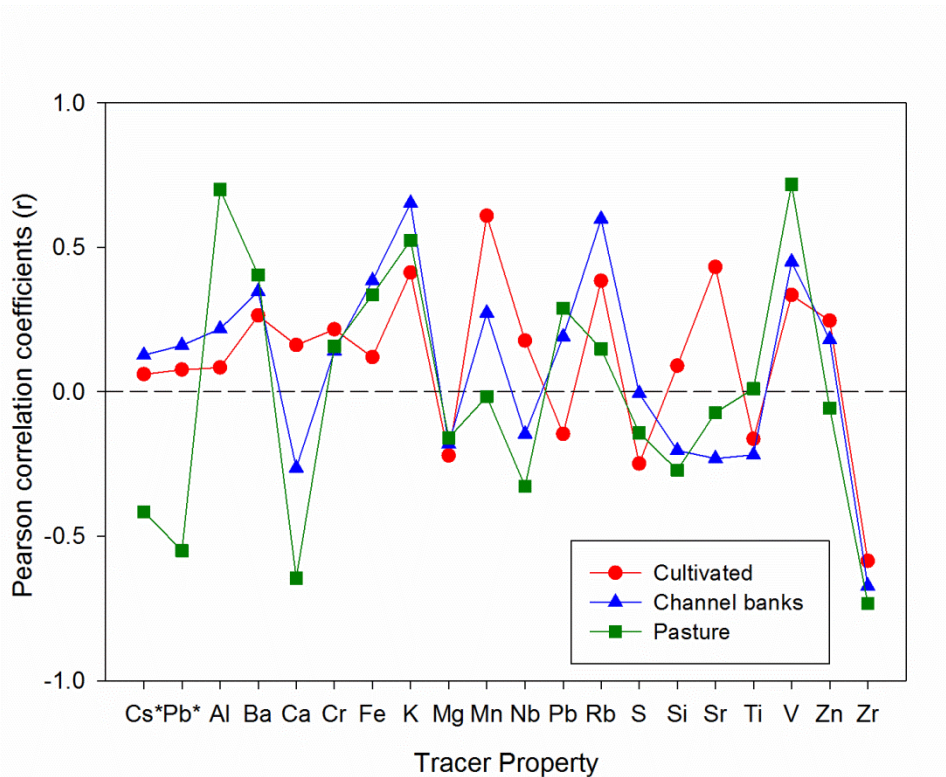


Figure 5: Pearson correlation coefficients (r) between Specific Surface Area (SSA) and tracer property for three sources from Smith and Blake (2014) (data from Table 1) with the * indicating fallout radionuclides (^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$) on the x axis and the remainder being elements.

388

389 The challenge is that there are a variety of non-linear responses between sediment biogeochemistry and
 390 particle size (Motha et al., 2002; Russell et al., 2001; Smith and Blake, 2014). There are assumptions with
 391 particle size measurements that are often not acknowledged (e.g. all particles are spherical and
 392 transported as discrete particles) and there are uncertainties and errors with laser particle size
 393 measurements that are not often reported nor propagated into total modelling uncertainty. The impact
 394 of these errors varies for different particle size distributions, with a notable increase in instrumentation
 395 error with decreasing particle size (Merkus, 2009) (Figure 6). One question for future research is whether
 396 or not potential errors on the SSA analysis could result in a substantially different interpretation of the
 397 results. Further, stable soil aggregates and agglomerated composite particles that are formed by particle
 398 to particle interaction after mobilization (Droppo et al., 2005) may also impact relationships between
 399 source and sediment fingerprints. Given the potential uncertainty of accurate SSA ratio-based corrections,

400 researchers often do not apply particle size corrections and instead rely on physical reasoning of sediment
401 transport processes and/or sample fractionation (e.g. Koiter et al., 2013a; Martinez-Carreras et al., 2010).

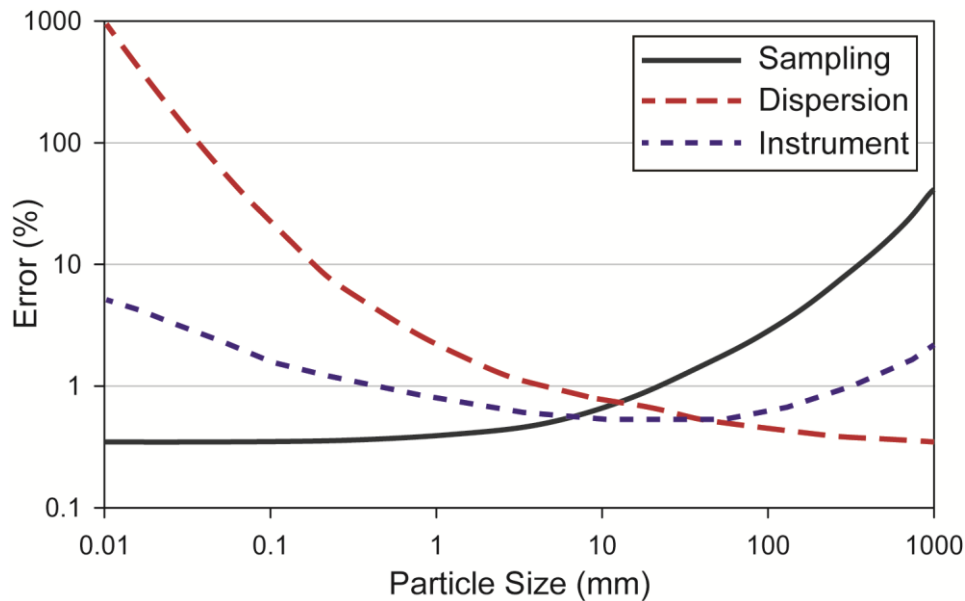


Figure 6: Relationship between potential sources of error and particle size distribution, adapted from Merkus (2009).

402

403 **3.3 Combining Size Fractionation and Corrections Procedures**

404 Moving forward, the first step towards addressing particle size should be to fractionate the source and
405 sediment material according to the research question (Table 6), which in turn requires information on the
406 particle size composition of the sediment and/or contaminants in question. Second, the results of this
407 fractionation should be assessed to determine whether the particle size distributions of the source and
408 sediment material are not significantly different. If they are significantly different, third, the application of
409 corrections to tracer properties is probably required for each source with enough samples to ensure source
410 representativeness. Particle size corrections are likely to be more effective if researchers are working with
411 a wide target fraction that is susceptible to sorting effects. The actual impact of the corrections should also
412 be assessed and reported upon, particularly the impact of the corrections on the original basis for source
413 discrimination. The optimal approach for understanding the impact of particle size corrections involves
414 the comparison of biogeochemical properties from samples fractionated across a variety of particle size
415 fractions (e.g. He and Walling, 1996; Laceby et al., 2015b; Russell et al., 2001). Understanding the impacts
416 of particle size on tracer properties should be one of the fundamental first steps when trialling new tracer
417 properties in sediment source fingerprinting research.

418 **4. Alternative Techniques to Address Particle Size**

419 Although particle size fractionation and particle size corrections are the two main approaches used to
420 account for any predictable changes in biogeochemical properties during sediment generation,
421 transportation and deposition processes, the challenges with particle size selectivity have opened up new
422 avenues to explore alternative approaches and research directions.

423 **4.1 Tributary (or Confluence) Tracing**

424 One approach to mitigating potential particle size impacts on sediment source fingerprinting is to
425 incorporate a tributary tracing or confluence tracing research design (Caitcheon, 1993; Hatfield and
426 Maher, 2008; Laceby et al., 2015a; Olley and Caitcheon, 2000; Vale et al., 2016; Walling et al., 1999). The
427 concept of a tributary tracing approach is that researchers sample sediment in the different upstream
428 tributaries and use these samples as a potential source of sediment sampled further downstream (Figure
429 7). The tributary sampling approach models sediment as a source and a sink, thus removing a significant
430 proportion of the impact of potential particle size enrichment on fingerprint properties. This approach has
431 also been recently applied to a lacustrine environment by Le Gall et al. (2016) who modelled the source of
432 material sampled in the downstream section of a pond in France based on the geochemical properties of
433 deposited sediment in the inlets of the two main tributaries.

434 There may be potential particle size enrichment or depletion impacts on fingerprint properties that may
435 occur during transportation and settling processes, although the most significant particle size enrichment
436 typically occurs during the initial stages of mobilization and transportation that often occurs on hillslopes
437 and in ephemeral systems (Stone and Walling, 1997). As material moves into the riverine system, sediment
438 particle size often becomes increasingly uniform and thus fewer differences may be anticipated. Of course,
439 there are always exceptions. For example, Koiter et al. (2015) used a recirculating flume to demonstrate
440 that it was not only distance travelled that impacted particle size, but also channel bed characteristics such
441 as roughness, porosity and inter-gravel flow. Therefore, the tributary tracing technique may have a limited
442 ability to mitigate particle size impacts where tributaries have distinctly different channel bed
443 characteristics and/or where there is a considerable distance between tributary and downstream sampling
444 sites.

Table 6: Overview of particle size implications at different steps in the sediment source fingerprinting research process

Step	Particle Size Implications
------	----------------------------

1. Research Objective	<ul style="list-style-type: none"> • Fractionate source and sediment material according to the research question • The particle size fraction analyzed should relate to the dominant particle size being transported in the system or the dominant particle size of the sediment-associated contaminant of interest
2. Research Design	<ul style="list-style-type: none"> • Consider the potential impacts of particle size on the research design and the different opportunities available to address and investigate particle size impacts on sediment source fingerprinting results • Consider using a tributary tracing sampling design or edge-of-field samplers to mitigate particle size impacts on tracing parameters • Plan and budget particle size analyses for sediment and source materials to understand whether or not there are significant impacts of particle size selectivity on the tracer parameters in the study region
3. Sample Processing, Analysis, Modelling	<p>A) Fractionation</p> <ul style="list-style-type: none"> • Ensure the particle size distribution of the source material is not significantly different than that of the sediment • Consider the potential impact of fractionation (e.g. sieving/settling) on biogeochemical properties and potential challenges with stable soil aggregates and sediment flocculants <p>B) Corrections</p> <ul style="list-style-type: none"> • Consider corrections if fractionation does not remove the differences between source and sediment particle size distributions • Always plot and assess the impact of the corrections on tracer parameters and source discrimination <p>C) Modelling</p> <ul style="list-style-type: none"> • Consider modelling different particle size fractions or comparing results with and without corrections on artificial mixtures to truly understand particle size impacts in the study region
5. Results	<ul style="list-style-type: none"> • Present the relationship between particle size and the tracer properties of interest • Assess and report on the impact of fractionation and corrections in the results with an emphasis on any potential impacts on the original basis of source discrimination • Present summary statistics for particle size distributions to facilitate comparisons between particle size distributions in source and sediment material
6. Discussion	<ul style="list-style-type: none"> • Discuss the impact of particle size in relation to other sediment source fingerprinting studies
7. Supplementary Information	<ul style="list-style-type: none"> • Provide corrected and uncorrected data for parameters used and all particle size data for future use by the research community (including sample coordinates, d_{50}, d_{90}, SSA and pre-treatments used prior to particle size analyses).

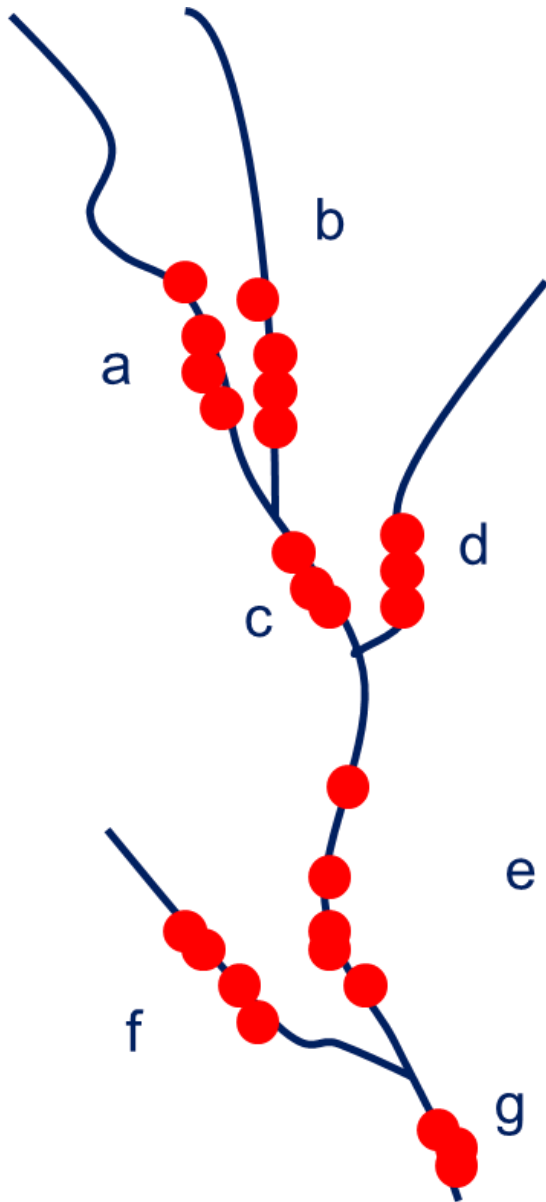


Figure 7: An example of a sampling design with the tributary tracing technique where the letters indicate the different tributaries that could be sampled in this theoretical catchment. In this situation, sediment collected at sites c, e and g is compared to sediment collected where it may be able to infer which tributaries (i.e. a, b, d or f) were the main sediment sources.

447 The tributary tracing technique may also address potential challenges that arise from variations in tracer
448 properties at the sub-catchment or reach scale. For example, it is conceivable that magnetic properties or
449 elemental concentrations may vary significantly at the subcatchment scale. The tributary tracing approach
450 may capitalize on these variations, incorporating them into contrasts between different sources, in this
451 instance, between different tributaries. A question for future research is what should be the benchmark
452 for indicating that the spatial variability in tracer properties has been sufficiently constrained, let alone the
453 heterogeneity in potential fractionation of different particle sizes?

454 Future research needs to examine the potential of tributary tracing research designs to address particle
455 size enrichment impacts on biogeochemical tracer properties, particularly when comparing sediment
456 sampled instream to material deposited in lakes and reservoirs, and tributaries with different channel bed
457 roughness. Indeed, more research is also required to examine the impact of in-stream sampling approach
458 (e.g. lag deposits, time-integrated samplers, instantaneous samplers, and channel bed-material samplers)
459 on sediment characteristics as different in-stream sampling methods may result in particle size biases that
460 may impact source modelling results. A sampling design with sufficient replication of these different in-
461 stream sampling methods in various catchments around the world may indeed provide further
462 understanding regarding the relationship between particle size and sampling methodology and how the
463 sediment sampling design may impact fingerprint properties and even potentially mixing model results.

464 **4.2 Edge-of-Field Samplers**

465 An alternative technique that may limit potential particle size effects on biogeochemical tracers during
466 mobilization and initial transportation phases is the installation of edge-of-field samplers (Panuska et al.,
467 2008; Wallbrink and Murray, 1993). Edge-of-field sampling approaches capture sediment after the initial
468 mobilization and transport processes and are an alternative technique for examining whether particle size
469 corrections are required (Evrard et al., 2016), or if they have been applied correctly (Foucher et al., 2015).
470 Conceptually, samplers installed on USLE plots (Brooks et al., 2014; Wischmeier and Smith, 1978) could
471 opportunistically sample material mobilized from hillslopes. Gerlach troughs (Gerlach, 1967) could sample
472 suspended sediment on hillslopes for further analyses after it has been mobilized and transported
473 downslope. Similarly, V-notch weirs combined with automated sediment samplers may also provide an
474 effective approach for sampling hillslope sediments in ephemeral gullies and other ditch type landscape
475 features (Freebairn and Wockner, 1986).

476 The key is to understand the impact of the initial mobilization and transport processes on both particle
477 size and the fingerprinting parameters of interest. For example, Evrard et al. (2016) demonstrated that
478 there was no significant difference between the fallout radionuclide activity concentrations in surface
479 soils, riverine sediments and sediments sampled with edge-of-field samplers in Laos. Conversely, in a
480 lowland and well-drained agricultural catchment in France, Foucher et al. (2015) sampled material in an
481 ephemeral rill during a rainfall event to confirm both the hyper enrichment of fine particles in overland
482 flow (<2 μm) and also to provide confidence that both the SSA- and Th-based corrections were applied
483 effectively. Further research is required to examine the potential of edge-of-field samplers to improve our
484 understanding of particle size impacts on biogeochemical tracer properties and understand how to apply
485 these samplers for a range of complex source types (e.g. unpaved roads and farm tracks). For example, it
486 would be beneficial to compare the impact of tributary sampling technique, an edge-of-field sampling
487 approach and top soil grab sampling on particle size distributions and to investigate whether or not these
488 sampling approaches mitigate particle size effects on biogeochemical tracers.

489 **4.3 Multiple Fraction Tracing**

490 Another technique to examine differences and potential particle size effects on conservative behavior is
491 tracing different particle size fractions (Caitcheon, 1998; Haddadchi et al., 2015; Laceby et al., 2016).
492 Although there has been a significant amount of research invested into developing particle size corrections
493 (Collins et al., 1996; Russell et al., 2001; Walling et al., 1993), less frequently have the different fractions
494 been traced and the results from tracing different size fractions been examined.

495 Caitcheon (1998) reported that source contributions from a sedimentary rock tributary basin varied only
496 by 13% (i.e. from 63% to 76%) when fingerprinting with mineral magnetic properties from different particle
497 size fractions (<63 μm , 63-125 μm , 125-250 μm and 250 - 500 μm). This author reported that the 63-125
498 μm fraction was most likely representative of the bulk material being transported in this particular
499 catchment. Using elemental geochemistry, Haddadchi et al. (2015) traced the fractionated <10 μm , 10-63
500 μm and 63-212 μm fractions. These authors found that although the maximum difference was high (33%),
501 there was <15% mean absolute difference between modelled source contributions with these fractions
502 (Figure 8). In summary, Haddadchi et al. (2015) indicated that their results highlight the importance of
503 fractionating the particle size most relevant to the management objective of the research.

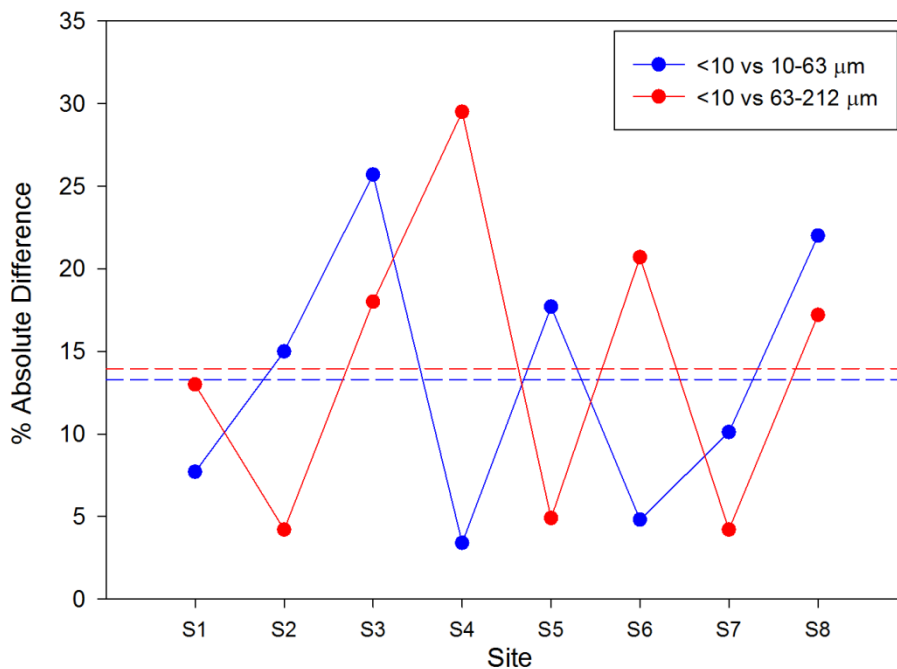


Figure 8: Mean absolute differences (dashed lines) in the source contributions between the <10 μm and the 10 - 63 μm (blue) and between the <10 μm and the 63 - 212 μm fractions (red) for eight sites in Australia plotted with data from Figure 9 in Haddadchi et al. (2015).

504

505 Laceby et al. (2016) also compared the impact of tracing different particle size fractions for three

506 catchments in the Fukushima region in Japan. These authors compared the relative model difference from

507 tracing the <63 μm and the <2 mm fractions with TOC, TN, $\delta^{13}\text{C}$, and $\delta^{15}\text{N}$ for 76 sediment samples taken

508 in three different catchments (Figure 3). The authors reported a maximum modelled average difference

509 between the <2 mm and <63 μm fraction of 14%, 11% and 7% for the three catchments, and a mean

510 relative difference of only 6% (SD 3%) for all possible modelled source contributions. The limited

511 differences between these models was likely driven by the fact that only 19% (SD 13%) of the material in

512 these sediment samples was <63 μm compared to 23% (SD 12%) of the material for the source samples,

513 indicating that ~80% of material was >63 μm for both source and sediment samples. A similar approach

514 should be applied in catchments with finer sediment transiting the system to characterize the potential

515 impact of fine sediment material on modelling results when tracing different fractions.

516 These studies raise interesting questions regarding what are the actual differences in mixing model results

517 arising from the impacts of particle size on tracer properties. Are these impacts greater than analytical and

518 model uncertainty? Is this particle size impact significant for management objectives? These fundamental

519 research questions present an opportunity for sediment source fingerprinting research with a focus on
520 methodological sensitivity to particle size effects in different systems. The challenge moving forward will
521 be for researchers to quantify the solid discharge in each particle size fraction in order to compare and
522 model both the relative source contributions (i.e. unweighted) and the absolute source contributions (i.e.
523 weighted by the abundance in each fraction), which could be significantly different.

524 **5. Particle Size Opportunities**

525 After material is mobilized from sources, sediment fingerprinting researchers often conceptualize riverine
526 systems as a black box, where the processes that occur between source and sediment sampling are not
527 well understood (i.e. Figure 3 in Koiter et al., 2013b). If researchers focus on furthering our understanding
528 of the relationship between particle size and biogeochemical properties, they may simultaneously start to
529 examine processes occurring within this black box. De-convoluting the complex relationship between
530 particle size and biogeochemical properties may provide significant insight into the processes of sediment
531 mobilization, transport and deposition within this black box, including the potential unique behaviour of
532 different sediment source parameters. Accordingly, there are multiple opportunities to advance sediment
533 source fingerprinting research and further our understanding of the complex relationship between particle
534 size and tracer parameter predictability.

535 **5.1 Mineralogy, Particle Size and Elemental Geochemistry**

536 The most important research objective and opportunity regarding particle size is to increase our
537 understanding of sediment tracer predictability. One approach to improving this predictability is
538 connecting particle size, mineralogy and elemental geochemistry. For example, in Figure 4, there is a clear
539 connection between sediment mineralogy and particle size, which will have direct ramifications for the
540 elemental compositions of sediment sources. Significant advances in sediment source fingerprinting
541 research are likely to be made through connecting these three components. More research into sediment
542 mineralogy may result in novel tracing techniques that are grounded in a logical basis for source
543 discrimination (e.g. Afshar et al., 2016; Bainbridge et al., 2016; Gingele and De Deckker, 2004; Tiecher et
544 al., 2016), that also provide significant information on the complex relationship between particle size and
545 elemental concentrations.

546 Mineralogy was one of the first sediment fingerprinting properties (Klages and Hsieh, 1975; Wall and
547 Wilding, 1975) and returning to incorporate mineralogy more directly into sediment tracing may provide
548 significant insights into particle size impacts on elemental compositions. This may be achieved, although

549 indirectly, through using Diffuse Reflectance Infrared Fourier Transform Spectrometry (DRIFTS) to identify
550 mineralogical groups (e.g. calcite, aluminosilicates, quartz) and discriminate between sediment lithological
551 sources (Poulenard et al., 2012). The use of DRIFTS and other spectroscopy approaches to tracing sediment
552 sources has the potential to develop a strong link to mineralogy and potentially estimate other
553 conventional tracer parameters like texture (Balsam and Deaton, 1996; Ortiz et al., 2009). Other
554 approaches such as clay mineral ratios (e.g. illite/illite + expandable clays) have also shown promise to
555 trace sediment provenance (Bainbridge et al., 2016; Douglas et al., 2006a; Douglas et al., 2006b).
556 Connecting clay mineralogy to element geochemistry and particle size will likely provide complementary
557 information relating to each of these fundamental sediment property characteristics.

558 **5.2 Elemental Ratios and Elemental Normalisation**

559 Moving a step beyond incorporating mineralogy more effectively into sediment tracing research is the
560 potential for incorporating approaches from other disciplines to address particle size effects. For example,
561 in the sediment provenance literature, elemental ratios have long been used to investigate changes in
562 particle size. In particular, the Al/Si ratio is used as a proxy for particle size in sediment provenance
563 research (Figure 9) (Bouchez et al., 2011). Other ratios may indeed be effective, such as the Ca/Al ratio
564 (Chen et al., 2014), along with particle size sensitive elements such as Ti (Bábek et al., 2015), or even simply
565 clay content (Szava-Kovats, 2008). For example, studies reconstructing the evolution of metal enrichment
566 ratios in sediment have also normalized elemental concentrations to Sc, Al, Si, Li or Th to minimize particle
567 size impacts when examining contamination trends (Ayrault et al., 2010; Clark et al., 2014; Grosbois et al.,
568 2012; Kersten and Smedes, 2002; Le Cloarec et al., 2011). Indeed, there are multiple approaches from the
569 sediment provenance field (Armstrong-Altrin et al., 2015; Bábek et al., 2015; Owens et al., 2016; Singh et
570 al., 2005) that present significant opportunities for enhancing the sediment source fingerprinting
571 technique (e.g. Vale et al., 2016).

572 Different elemental and lithogenic radionuclide ratios have been used previously to address particle size
573 and density related enrichment in sediment source fingerprinting research. For example, Olley and Murray
574 (1994) demonstrated that although the concentrations of thorium (Th) isotopes varied with particle size,
575 the $^{230}\text{Th}/^{232}\text{Th}$ ratio remains constant, with sorting by density or particle size producing the same ratio as
576 the bulk soil. Caitcheon (1998) and Murray et al. (1993a) incorporated ratios directly into the mixing model
577 process for mineral magnetics and radionuclides, respectively. Although these modelling approaches used
578 the actual ratio lines to quantify source contributions, they demonstrate the utility of incorporating ratios
579 directly into the mixing model process to address particle size enrichment. More research is required to

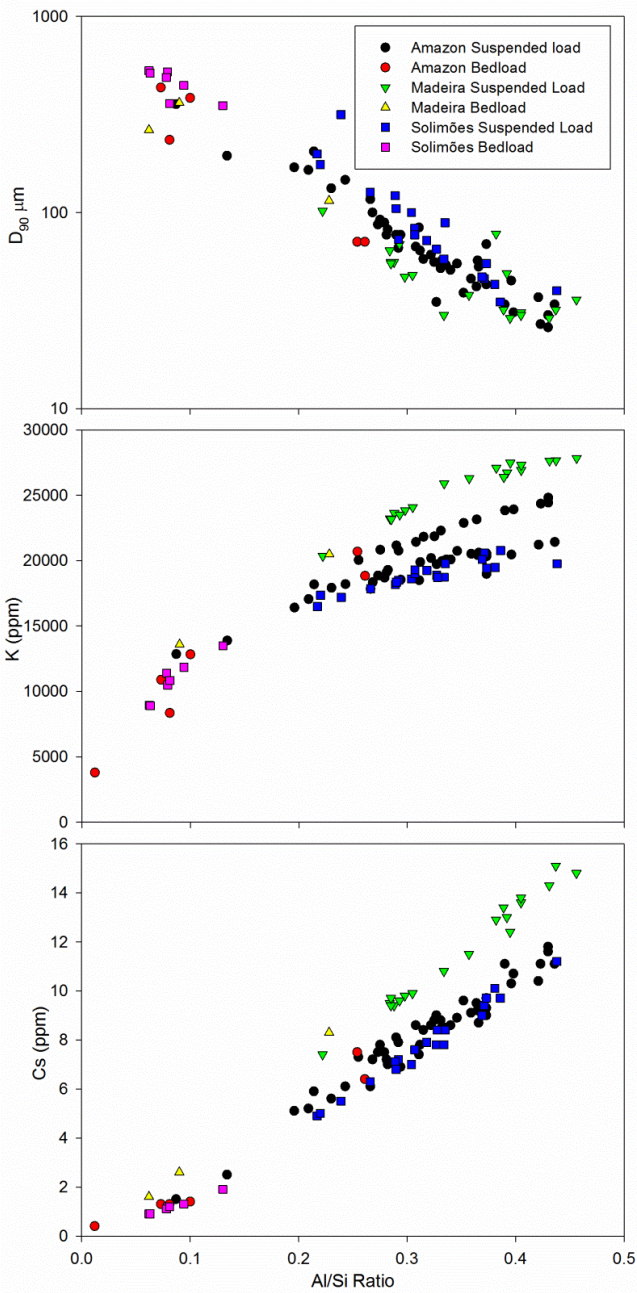
580 examine whether similar approaches may also provide a technique for normalizing particle size effects
581 with frequentist or Bayesian mixing models, or even more insight into the foundation for elemental
582 discrimination between sediment sources. For example, the interrelationships between particle size and
583 biogeochemical tracer properties indicate that particle size may directly contribute to the formation of
584 some contrasts in sediment fingerprints between sources. Accordingly, further advances in understanding
585 these relationships, with experimental designs incorporating different particle size fractions, may provide
586 additional understanding for the basis of discrimination between sediment sources.

587 **5.3 Particle Dynamics**

588 The nature of cohesive sediment transport, and particularly fine sediment transport, is fundamentally
589 complex where the majority of particles are transported as flocs, or composite particles, that are
590 comprised of organic matter, smaller particles and mineral components with a complex structure (Droppo
591 et al., 2005; Walling and Collins, 2016). The presence of these soil aggregates and sediment flocs will have
592 a significant impact on particle size selectivity during sediment mobilization, transportation and deposition
593 processes (Beuselinck et al., 2000). The behaviour of material during these processes is a function of
594 density, aggregate stability, grain shape and grain size (D'Haen et al., 2012).

595 The impact of grain abrasion and disaggregation on ^{137}Cs concentrations was examined by Dyer and Olley
596 (1999) who reported that the fraction produced by abrasion had essentially similar ^{137}Cs concentrations as
597 the corresponding fraction (<40 μm) in non-disturbed soil. Conversely, Crockford and Olley (1998) found
598 that breakage and abrasion processes had a substantial impact on mineral magnetic properties, reducing
599 concentrations in a granitic soil and increasing concentrations in a sedimentary soil. These authors
600 concluded that a tributary or confluence tracing approach should mitigate the impact of these processes
601 on mineral magnetic properties. Although these studies examined the disaggregation of sediments,
602 Droppo et al. (2005) suggested that the actual nature of aggregates and flocs is rarely considered in studies
603 of sediment transport and sources.

604



605 **Figure 9:** Scatter plots of the relationship between the Al/Si ratio and D_{90} , K, and Cs (ppm) from samples
 606 from different tributaries of the Amazon catchment with data from Tables 1 and 2 from Bouchez et al.,
 607 (2011).

608

609 Accordingly, there is an opportunity to improve our understanding of sediment transport processes by
 610 further incorporating the nature of the material being transported more directly into the methodology of
 611 sediment fingerprinting projects. This is important as the sediment fractions isolated by density may be

612 different than those identified by particle size analysis (Gregorich et al., 2006). Different fractionation
613 methods (e.g. wet sieving, dry sieving, grinding) may fragment aggregates which could result in different
614 biogeochemical properties in analyzed aliquots. This may be particularly true with respect to different
615 techniques and conditions for wet and dry sieving, and the use of Stokes' law and settling columns to
616 isolate different particle size fractions. Different source and sediment material (e.g. platy, spherical, and
617 rod-shaped grains) may respond differently to fractionation methods (Droppo et al., 2005; Hatfield, 2014).
618 Different pre-analysis treatments (e.g. sonification, chemical dispersion, physical dispersion) may also
619 impact the biogeochemical fingerprints of interest. Laboratory determined grain size distributions will
620 most likely differ significantly from those in the field owing to the physical and often chemical
621 disaggregation of flocs and aggregates during the laboratory fractionation process (Phillips and Walling,
622 1995; Walling and Collins, 2016).

623 One major research opportunity is whether these alterations between laboratory and field, and the impact
624 of aggregates, are significant to sediment fingerprinting modelling results, or fall within the range of
625 analytical and modelling uncertainty. For example, if soil aggregates and sediment flocs form and/or
626 evolve during sediment mobilization and transportation processes, this may result in a form of non-
627 conservative behaviour between source and downstream sink, adding uncertainty in the form of an
628 'aggregate size' effect on tracer properties. Furthermore, the impact of aggregates directly relates to the
629 approach to sample processing, particularly whether aggregates should be chemically dispersed prior to
630 particle size fractionation or whether samples should not be disaggregated (Koiter et al., 2017). More
631 research is required to characterize the impact of aggregates, and their processing, on sediment source
632 fingerprinting research.

633 Density separation is an intriguing fractionation approach that has not received much attention in the
634 sediment source fingerprinting literature (Hardy et al., 2010). Different clay minerals have different
635 densities that may impact their separation with settling columns in the finer fractions. Furthermore, iron
636 oxides and mineral magnetic properties have been strongly related to density (Hatfield, 2014) and there
637 may be micro aggregates or sediment flocs transported in suspension with the silt or very fine sand fraction
638 because they are lighter. Importantly, these micro-aggregates and sediment flocs will have a high capacity
639 to transport contaminants; however they will have a different resistance (i.e. a lower density) relative to
640 absolute particles of a similar size, predominantly the silt fraction (Droppo et al., 1998).

641 Although density separation is time consuming and expensive, it may be more relevant to fluvial processes
642 compared to sieving the <63 μm fraction or settling the <10 μm fraction. Accordingly, it would be

643 opportunistic to compare density and particle size separation techniques to understand their impacts on
644 sediment fingerprinting modelling results. As researchers apply tracing techniques directly to quantify the
645 relative source contributions of carbon and nitrogen (Cooper et al., 2015b; Garzon-Garcia et al., 2017), and
646 are using different carbon components to model sediment sources (Blake et al., 2012; Gibbs, 2008;
647 Hancock and Revill, 2013; Reiffarth et al., 2016) the utility of density separation may become of increasing
648 importance for tracing different carbon sources in riverine systems.

649 **5.4 Particle Size as a Tracer Property**

650 Situations may arise where particle size may be used as a tracer property in and of itself. The sediment
651 source fingerprinting approach is founded upon the assumptions of conservative behavior and source
652 discrimination. As noted in the introduction and throughout this review, if the sediment source properties,
653 including particle size, are predictable, they could possibly be used in sediment source fingerprinting
654 research.

655 The particle size distribution of source material is dependent on the parent material, weathering and
656 erosion processes. Accordingly, there may be situations where particle size itself may be a useful tracer
657 property, particularly where one sediment source is dominated by sandy quartz material and another by
658 fine clay material. Researchers have effectively utilized particle size to discriminate between pre- and post-
659 dam stratigraphy (Batuca and Jordaan Jr, 2000; Morris and Fan, 1998). Further, the particle size of reservoir
660 sediment has been traced back to the parent rock material (Abraham et al., 1999). If researchers were
661 interested in tracing material in the bed load, or material being transported off shore to oceanic
662 environments, then particle size metrics may be effective tracers in these scenarios, if they are indeed
663 predictable. There may even indeed be situations where particle shape may provide an effective
664 discriminator for determining sediment provenance (Ehrlich et al., 1980).

665 **6. Conclusion**

666 Sediment source fingerprinting researchers have recently tended to avoid in-depth examinations of
667 fundamental topics such as the impact of organic matter on biogeochemical properties, which tracer
668 properties are non-conservative (e.g. soluble, reactive), and what is the impact of particle size on tracer
669 property predictability. One limitation of statistically-oriented approaches to sediment source
670 fingerprinting is that they do not strive to understand the logic of tracer selection, non-conservative
671 tracers, nor particle size impacts. This is particularly limiting, as addressing conservative behaviour is
672 fundamentally more complex than simply acknowledging that sediment samples plot within their source

673 range. Modelling, and the statistical selection of which tracer properties to model, represent only two
674 stages of the sediment source fingerprinting process; although these two steps, at times, appear to
675 dominate the recent literature. Rarely is particle size the focus of research, rather particle size is often
676 simply an acknowledged limitation that must be addressed somehow. After demonstrating that non-
677 conservative tracer properties affect the accuracy of sediment source fingerprinting modelling results,
678 Sherriff et al. (2015) appropriately emphasized that improved strategies to detect non-conservative tracer
679 properties should be a priority of sediment source fingerprinting research.

680 Researchers thus need to start investigating some of these fundamental assumptions and complexity at
681 the core of the sediment source fingerprinting approach. Researchers should strive to understand particle
682 size impacts and the logic of tracer selection, as they are likely to be inherently related. For example, the
683 interrelationships between particle size and biogeochemical tracer properties suggest that in many
684 instances particle size may also contribute to the formation of contrasts in sediment fingerprints between
685 sources. Approaches to address particle size will likely vary from region to region, catchment to catchment,
686 and even from event to event. Accordingly, researchers should always publish particle size and
687 biogeochemical property datasets for important regional and future comparisons, including data from
688 different particle size fractions (Kersten and Smedes, 2002). Of note, from the papers reviewed in this
689 study, only approximately 15% published the raw source and sediment data (Tables 1-5).

690 In summary, particle size dynamics presents both a challenge and an opportunity for sediment source
691 fingerprinting research. The research question, context and objective will determine the approach to
692 addressing particle size and the appropriate fraction for investigation (Table 6). Addressing particle size
693 effects is fundamental to research design, tracer selection, and sampling technique (e.g. edge-of-field
694 samplers). Advances in the understanding of particle size–biogeochemical tracer interactions will likely
695 improve the predictability and therefore accuracy of sediment source fingerprinting.

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698 **References**

- 699 Abraham, J., Allen, P., Dunbar, J. and Dworkin, S., 1999. Sediment type distribution in reservoirs: sediment
700 source versus morphometry. *Environmental Geology* 38, 101-110.
- 701 Afshar, S., Saghafian, B. and Vosoughifar, H., 2016. Source apportionment of the sediments entering dam
702 using lithological and mineralogical studies. *Global Journal of Environmental Science and*
703 *Management* 2, 265-274.
- 704 Amundson, R., Austin, A.T., Schuur, E.A.G., Yoo, K., Matzek, V., Kendall, C., Uebersax, A., Brenner, D. and
705 Baisden, W.T., 2003. Global patterns of the isotopic composition of soil and plant nitrogen. *Global*
706 *Biogeochemical Cycles* 17, 1031.
- 707 Armstrong-Altrin, J.S., Machain-Castillo, M.L., Rosales-Hoz, L., Carranza-Edwards, A., Sanchez-Cabeza, J.-A.
708 and Ruíz-Fernández, A.C., 2015. Provenance and depositional history of continental slope
709 sediments in the Southwestern Gulf of Mexico unraveled by geochemical analysis. *Continental*
710 *Shelf Research* 95, 15-26.
- 711 Ayrault, S., Priadi, C.R., Evrard, O., Lefèvre, I. and Bonté, P., 2010. Silver and thallium historical trends in
712 the Seine River basin. *Journal of Environmental Monitoring* 12, 2177-2185.
- 713 Bábek, O., Grygar, T.M., Faměra, M., Hron, K., Nováková, T. and Sedláček, J., 2015. Geochemical
714 background in polluted river sediments: How to separate the effects of sediment provenance and
715 grain size with statistical rigour? *CATENA* 135, 240-253.
- 716 Bainbridge, Z., Lewis, S., Smithers, S., Wilkinson, S., Douglas, G., Hillier, S. and Brodie, J., 2016. Clay mineral
717 source tracing and characterisation of Burdekin River (NE Australia) and flood plume fine
718 sediment. *Journal of Soils and Sediments* 16, 687-706.
- 719 Balesdent, J., Mariotti, A. and Guillet, B., 1987. Natural ¹³C abundance as a tracer for studies of soil organic
720 matter dynamics. *Soil Biology and Biochemistry* 19, 25-30.
- 721 Balsam, W.L. and Deaton, B.C., 1996. Determining the composition of late Quaternary marine sediments
722 from NUV, VIS, and NIR diffuse reflectance spectra. *Marine Geology* 134, 31-55.
- 723 Bartley, R., Bainbridge, Z.T., Lewis, S.E., Kroon, F.J., Wilkinson, S.N., Brodie, J.E. and Silburn, D.M., 2014.
724 Relating sediment impacts on coral reefs to watershed sources, processes and management: A
725 review. *Science of The Total Environment* 468–469, 1138-1153.
- 726 Batuca, D.G. and Jordaan Jr, J., 2000. *Silting and Desilting of Reservoirs*. A.A. Balkema Publishers,
727 Rotterdam, 357 pp.
- 728 Bellanger, B., Huon, S., Velasquez, F., Valles, V., Girardin, C. and Mariotti, A., 2004. Monitoring soil organic
729 carbon erosion with $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ on experimental field plots in the Venezuelan Andes. *CATENA*
730 58, 125-150.
- 731 Belmont, P., Gran, K.B., Schottler, S.P., Wilcock, P.R., Day, S.S., Jennings, C., Lauer, J.W., Viparelli, E.,
732 Willenbring, J.K., Engstrom, D.R. and Parker, G., 2011. Large shift in source of fine sediment in the
733 Upper Mississippi River. *Environmental Science and Technology* 45, 8804–8810.
- 734 Belmont, P., Willenbring, J., Schottler, S., Marquard, J., Kumarasamy, K. and Hemmis, J., 2014. Toward
735 generalizable sediment fingerprinting with tracers that are conservative and nonconservative over
736 sediment routing timescales. *Journal of Soils and Sediments* 14, 1479-1492.
- 737 Ben Slimane, A., Raclot, D., Evrard, O., Sanaa, M., Lefèvre, I., Ahmadi, M., Tounsi, M., Rumpel, C., Mammou,
738 A.B. and Le Bissonnais, Y., 2013. Fingerprinting sediment sources in the outlet reservoir of a hilly
739 cultivated catchment in Tunisia. *Journal of Soils and Sediments* 13, 801-815.
- 740 Ben Slimane, A., Raclot, D., Evrard, O., Sanaa, M., Lefevre, I. and Le Bissonnais, Y., 2016. Relative
741 contribution of rill/interrill and gully/channel erosion to small reservoir siltation in Mediterranean
742 environments. *Land Degradation and Development* 27, 785-797.
- 743 Beuselinck, L., Govers, G., Steegen, A. and Poesen, J., 2000. Impact of soil aggregates on the size selectivity
744 of the sediment deposition process. In: M. Stone (Editor), *The Role of Erosion and Sediment*

745 Transport in Nutrient and Contamination Transfer, IAHS Publication No. 263, IAHS Press,
746 Wallingford, 99-107.

747 Blake, W., Walsh, R., Sayer, A. and Bidin, K., 2006. Quantifying fine-sediment sources in primary and
748 selectively logged rainforest catchments using geochemical tracers. *Water, Air, & Soil Pollution:*
749 *Focus 6*, 615-623.

750 Blake, W.H., Ficken, K.J., Taylor, P., Russell, M.A. and Walling, D.E., 2012. Tracing crop-specific sediment
751 sources in agricultural catchments. *Geomorphology* 139–140, 322-329.

752 Blake, W.H., Wallbrink, P.J., Doerr, S.H., Shakesby, R.A. and Humphreys, G.S., 2004. Sediment
753 redistribution following wildfire in the Sydney region, Australia: a mineral magnetic tracing
754 approach. In: V. Golosov, V.R. Belyaev and D.E. Walling (Editors), *Sediment Transfer through the*
755 *Fluvial System*, IAHS Publication No. 288, IAHS Press, Wallingford, 52-59.

756 Blake, W.H., Wallbrink, P.J., Wilkinson, S.N., Humphreys, G.S., Doerr, S.H., Shakesby, R.A. and Tomkins,
757 K.M., 2009. Deriving hillslope sediment budgets in wildfire-affected forests using fallout
758 radionuclide tracers. *Geomorphology* 104, 105-116.

759 Bogen, J., 1992. Monitoring grain size of suspended sediments in rivers. In: J. Bogen, D.E. Walling and T.
760 Day (Editors), *Erosion and Sediment Transport Monitoring Programmes in River Basins*, IAHS
761 Publication No. 210, IAHS Press, Wallingford, 183-190.

762 Bonniwell, E.C., Matisoff, G. and Whiting, P.J., 1999. Determining the times and distances of particle transit
763 in a mountain stream using fallout radionuclides. *Geomorphology* 27, 75-92.

764 Bouchez, J., Gaillardet, J., France-Lanord, C., Maurice, L. and Dutra-Maia, P., 2011. Grain size control of
765 river suspended sediment geochemistry: Clues from Amazon River depth profiles. *Geochemistry,*
766 *Geophysics, Geosystems* 12, 1-24.

767 Bradford, J., Truman, C. and Huang, C., 1992. Comparison of three measures of resistance of soil surface
768 seals to raindrop splash. *Soil Technology* 5, 47-56.

769 Brooks, A., Spencer, J., Borombovits, D., Pietsch, T. and Olley, J., 2014. Measured hillslope erosion rates in
770 the wet-dry tropics of Cape York, northern Australia: Part 2, RUSLE-based modeling significantly
771 over-predicts hillslope sediment production. *CATENA* 122, 1-17.

772 Caitcheon, G.G., 1993. Sediment Source Tracing Using Environmental Magnetism - a New Approach with
773 Examples from Australia. *Hydrological Processes* 7, 349-358.

774 Caitcheon, G.G., 1998. The significance of various sediment magnetic mineral fractions for tracing
775 sediment sources in Killimicat Creek. *CATENA* 32, 131-142.

776 Caitcheon, G.G., Douglas, G. and Palmer, M., 2006. Sediment Source Tracing in the Lake Burragorang
777 Catchment. *CSIRO Land and Water*, Canberra, 28.

778 Chen, J.B., Gaillardet, J., Bouchez, J., Louvat, P. and Wang, Y.N., 2014. Anthropophile elements in river
779 sediments: Overview from the Seine River, France. *Geochemistry, Geophysics, Geosystems* 15,
780 4526-4546.

781 Clark, D.E., Vogels, M.F., van der Perk, M., Owens, P.N. and Petticrew, E.L., 2014. Effects of a small-scale,
782 abandoned gold mine on the geochemistry of fine stream-bed and floodplain sediments in the
783 Horsefly River watershed, British Columbia, Canada. *Mineralogical Magazine* 78, 1491-1504.

784 Clark, E.H., 1985. The off-site costs of soil erosion. *Journal of Soil and Water Conservation* 40, 19-22.

785 Collins, A.L. and Walling, D.E., 2002. Selecting fingerprint properties for discriminating potential suspended
786 sediment sources in river basins. *Journal of Hydrology* 261, 218-244.

787 Collins, A.L., Walling, D.E. and Leeks, G.J.L., 1996. Composite fingerprinting of the spatial source of fluvial
788 suspended sediment: a case study of the Exe and Severn river basins, United Kingdom.
789 *Géomorphologie: Relief, Processus, Environnement* 2, 41-53.

790 Collins, A.L., Walling, D.E., Webb, L. and King, P., 2010. Apportioning catchment scale sediment sources
791 using a modified composite fingerprinting technique incorporating property weightings and prior
792 information. *Geoderma* 155, 249-261.

793 Collins, A.L., Zhang, Y., McChesney, D., Walling, D.E., Haley, S.M. and Smith, P., 2012. Sediment source
794 tracing in a lowland agricultural catchment in southern England using a modified procedure
795 combining statistical analysis and numerical modelling. *Science of The Total Environment* 414,
796 301-317.

797 Cooper, R.J., Krueger, T., Hiscock, K.M. and Rawlins, B.G., 2014. Sensitivity of fluvial sediment source
798 apportionment to mixing model assumptions: A Bayesian model comparison. *Water Resources*
799 *Research* 50, 9031-9047.

800 Cooper, R.J., Krueger, T., Hiscock, K.M. and Rawlins, B.G., 2015a. High-temporal resolution fluvial sediment
801 source fingerprinting with uncertainty: a Bayesian approach. *Earth Surface Processes and*
802 *Landforms* 40, 78-92.

803 Cooper, R.J., Pedentchouk, N., Hiscock, K.M., Disdle, P., Krueger, T. and Rawlins, B.G., 2015b. Apportioning
804 sources of organic matter in streambed sediments: An integrated molecular and compound-
805 specific stable isotope approach. *Science of The Total Environment* 520, 187-197.

806 Crockford, R. and Olley, J., 1998. The effects of particle breakage and abrasion on the magnetic properties
807 of two soils. *Hydrological Processes* 12, 1495-1505.

808 D'Haen, K., Verstraeten, G., Duser, B., Degryse, P., Haex, J. and Waelkens, M., 2013. Unravelling changing
809 sediment sources in a Mediterranean mountain catchment: a Bayesian fingerprinting approach.
810 *Hydrological Processes* 27, 896-910.

811 D'Haen, K., Verstraeten, G. and Degryse, P., 2012. Fingerprinting historical fluvial sediment fluxes. *Progress*
812 *in Physical Geography* 36, 154-186.

813 Davis, C.M. and Fox, J.F., 2009. Sediment fingerprinting: Review of the method and future improvements
814 for allocating nonpoint source pollution. *Journal of Environmental Engineering* 135, 490-504.

815 Devereux, O.H., Prestegard, K.L., Needelman, B.A. and Gellis, A.C., 2010. Suspended-sediment sources in
816 an urban watershed, Northeast Branch Anacostia River, Maryland. *Hydrological Processes* 24,
817 1391-1403.

818 Douglas, G., Caitcheon, G. and Palmer, M., 2009. Sediment source identification and residence times in
819 the Maroochy River estuary, southeast Queensland, Australia. *Environmental Geology* 57, 629-
820 639.

821 Douglas, G., Ford, P., Palmer, M., Noble, R. and Packett, R., 2006a. Fitzroy River Basin, Queensland,
822 Australia. I. Identification of sediment sources in impoundments and flood events. *Environmental*
823 *Chemistry* 3, 364-376.

824 Douglas, G., Ford, P., Palmer, M., Noble, R. and Packett, R., 2006b. Fitzroy River, Queensland, Australia. II.
825 Identification of sources of estuary bottom sediments. *Environmental Chemistry* 3, 377-385.

826 Douglas, G., Palmer, M. and Caitcheon, G., 2003. The provenance of sediments in Moreton Bay, Australia:
827 a synthesis of major, trace element and Sr-Nd-Pb isotopic geochemistry, modelling and landscape
828 analysis. *Hydrobiologia* 494, 145-152.

829 Droppo, I., Nackaerts, K., Walling, D. and Williams, N., 2005. Can flocs and water stable soil aggregates be
830 differentiated within fluvial systems? *CATENA* 60, 1-18.

831 Droppo, I., Walling, D. and Ongley, E., 1998. Suspended sediment structure: implications for sediment and
832 contaminant transport modelling. In: W. Summer, E. Klaghofer and W. Zhang (Editors), *Modelling*
833 *Soil Erosion, Sediment Transport and Closely Related Hydrological Processes*, IAHS Publication No.
834 249, IAHS Press, Wallingford, 437-444.

835 Dyer, F.J. and Olley, J.M., 1999. The effects of grain abrasion and disaggregation on ¹³⁷Cs concentrations in
836 different size fractions of soils developed on three different rock types. *CATENA* 36, 143-151.

837 Ehrlich, R., Brown, P.J., Yarus, J.M. and Przygocki, R.S., 1980. The origin of shape frequency distributions
838 and the relationship between size and shape. *Journal of Sedimentary Research* 50, 475-483.

839 Evrard, O., Lacey, J.P., Huon, S., Lefèvre, I., Sengtaheuanghoung, O. and Ribolzi, O., 2016. Combining
840 multiple fallout radionuclides (^{137}Cs , ^7Be , $^{210}\text{Pb}_{\text{xs}}$) to investigate temporal sediment source
841 dynamics in tropical, ephemeral riverine systems. *Journal of Soils and Sediments* 16, 1130-1144.
842 Evrard, O., Navratil, O., Ayrault, S., Ahmadi, M., Némery, J., Legout, C., Lefèvre, I., Poirel, A., Bonté, P. and
843 Esteves, M., 2011. Combining suspended sediment monitoring and fingerprinting to determine
844 the spatial origin of fine sediment in a mountainous river catchment. *Earth Surface Processes and
845 Landforms* 36, 1072-1089.
846 Evrard, O., Némery, J., Gratiot, N., Duvert, C., Ayrault, S., Lefèvre, I., Poulenard, J., Prat, C., Bonté, P. and
847 Esteves, M., 2010. Sediment dynamics during the rainy season in tropical highland catchments of
848 central Mexico using fallout radionuclides. *Geomorphology* 124, 42-54.
849 Evrard, O., Poulenard, J., Némery, J., Ayrault, S., Gratiot, N., Duvert, C., Prat, C., Lefèvre, I., Bonté, P. and
850 Esteves, M., 2013. Tracing sediment sources in a tropical highland catchment of central Mexico by
851 using conventional and alternative fingerprinting methods. *Hydrological Processes* 27, 911-922.
852 Fan, Q., Yamaguchi, N., Tanaka, M., Tsukada, H. and Takahashi, Y., 2014. Relationship between the
853 adsorption species of cesium and radiocesium interception potential in soils and minerals: an
854 EXAFS study. *Journal of Environmental Radioactivity* 138, 92-100.
855 Fontaine, T.A., Moore, T.D. and Burgoa, B., 2000. Distributions of contaminant concentration and particle
856 size in fluvial sediment. *Water Research* 34, 3473-3477.
857 Foster, I.D.L., Lees, J.A., Owens, P.N. and Walling, D.E., 1998. Mineral magnetic characterization of
858 sediment sources from an analysis of lake and floodplain sediments in the catchments of the Old
859 Mill Reservoir and Slapton Ley, South Devon, UK. *Earth Surface Processes and Landforms* 23, 685-
860 703.
861 Foucher, A., Lacey, J.P., Salvador-Blanes, S., Evrard, O., Le Gall, M., Lefèvre, I., Cerdan, O., Rajkumar, V.
862 and Desmet, M., 2015. Quantifying the dominant sources of sediment in a drained lowland
863 agricultural catchment: The application of a thorium-based particle size correction in sediment
864 fingerprinting. *Geomorphology* 250, 271-281.
865 Fox, J.F. and Papanicolaou, A.N., 2007. The use of carbon and nitrogen isotopes to study watershed erosion
866 processes. *Journal of the American Water Resources Association* 43, 1047-1064.
867 Freebairn, D. and Wockner, G., 1986. A study of soil erosion on vertisols of the eastern Darling Downs,
868 Queensland .II. The effect of soil, rainfall, and flow conditions on suspended sediment losses. *Soil
869 Research* 24, 159-172.
870 Frings, R.M., 2008. Downstream fining in large sand-bed rivers. *Earth-Science Reviews* 87, 39-60.
871 Fry, B., 2006. *Stable Isotope Ecology*. Springer. New York., 308 pp.
872 Garzon-Garcia, A., Lacey, J.P., Olley, J.M. and Bunn, S.E., 2017. Differentiating the sources of fine
873 sediment, organic matter and nitrogen in a subtropical Australian catchment. *Science of The Total
874 Environment* 575, 1384–1394.
875 Gateuille, D., Evrard, O., Lefevre, I., Moreau-Guigon, E., Alliot, F., Chevreuil, M. and Mouchel, J.-M., 2014.
876 Mass balance and decontamination times of Polycyclic Aromatic Hydrocarbons in rural nested
877 catchments of an early industrialized region (Seine River basin, France). *Science of The Total
878 Environment* 470, 608-617.
879 Gerlach, T., 1967. Hillslope troughs for measuring sediment movement. *Revue de Geomorphologie
880 Dynamique* 17, 173.
881 Gibbs, M., 2008. Identifying source soils in contemporary estuarine sediments: A new compound-specific
882 isotope method. *Estuaries and Coasts* 31, 344-359.
883 Gibbs, R.J., 1967. Amazon River: Environmental Factors That Control Its Dissolved and Suspended Load.
884 *Science* 156, 1734-1737.
885 Gibbs, R.J., Matthews, M.D. and Link, D.A., 1971. The relationship between sphere size and settling
886 velocity. *Journal of Sedimentary Research* 41, 7-18.

887 Gingele, F. and De Deckker, P., 2004. Fingerprinting Australia's rivers with clay minerals and the application
888 for the marine record of climate change. *Australian Journal of Earth Sciences* 51, 339-348.

889 Gourdin, E., Evrard, O., Huon, S., Reyss, J.-L., Ribolzi, O., Bariac, T., Sengtaheuanghoung, O., Ayrault, S. and
890 A.L., C., 2014. Spatial and temporal variability of ^7Be and ^{210}Pb wet deposition during four
891 successive monsoon storms in a catchment of northern Laos. *Journal of Environmental*
892 *Radioactivity* 136, 195-205.

893 Gregorich, E., Beare, M., McKim, U. and Skjemstad, J., 2006. Chemical and biological characteristics of
894 physically uncomplexed organic matter. *Soil Science Society of America Journal* 70, 975-985.

895 Grosbois, C., Meybeck, M., Lestel, L., Lefèvre, I. and Moatar, F., 2012. Severe and contrasted polymetallic
896 contamination patterns (1900–2009) in the Loire River sediments (France). *Science of The Total*
897 *Environment* 435, 290-305.

898 Guzmán, G., Quinton, J., Nearing, M., Mabit, L. and Gómez, J., 2013. Sediment tracers in water erosion
899 studies: current approaches and challenges. *Journal of Soils and Sediments* 13, 816-833.

900 Haddadchi, A., Olley, J. and Laceby, J.P., 2014. Accuracy of mixing models in predicting sediment source
901 contributions. *Science of The Total Environment* 497–498, 139-152.

902 Haddadchi, A., Olley, J. and Pietsch, T., 2015. Quantifying sources of suspended sediment in three size
903 fractions. *Journal of Soils and Sediments* 15, 2086-2100.

904 Haddadchi, A., Ryder, D.S., Evrard, O. and Olley, J., 2013. Sediment fingerprinting in fluvial systems: review
905 of tracers, sediment sources and mixing models. *International Journal of Sediment Research* 28,
906 560-578.

907 Hancock, G.J. and Revill, A.T., 2013. Erosion source discrimination in a rural Australian catchment using
908 compound-specific isotope analysis (CSIA). *Hydrological Processes* 27, 923-932.

909 Hardy, F., Bariteau, L., Lorrain, S., Theriault, I., Gagnon, G., Messier, D. and Rougerie, J.F., 2010.
910 Geochemical tracing and spatial evolution of the sediment bed load of the Romaine River, Quebec,
911 Canada. *CATENA* 81, 66-76.

912 Hatfield, R.G., 2014. Particle size-specific magnetic measurements as a tool for enhancing our
913 understanding of the bulk magnetic properties of sediments. *Minerals* 4, 758-787.

914 Hatfield, R.G. and Maher, B.A., 2008. Suspended sediment characterization and tracing using a magnetic
915 fingerprinting technique: Bassenthwaite Lake, Cumbria, UK. *The Holocene* 18, 105-115.

916 Hatfield, R.G. and Maher, B.A., 2009. Fingerprinting upland sediment sources: particle size-specific
917 magnetic linkages between soils, lake sediments and suspended sediments. *Earth Surface*
918 *Processes and Landforms* 34, 1359-1373.

919 He, Q. and Owens, P.N., 1995. Determination of suspended sediment provenance using caesium-137,
920 unsupported lead-210 and radium-226: a numerical mixing model approach. In: I. Foster, A.
921 Gurnell and B. Webb (Editors), *Sediment and Water Quality in River Catchments*, Wiley,
922 Chichester, 207-227.

923 He, Q. and Walling, D.E., 1996. Interpreting particle size effects in the adsorption of ^{137}Cs and unsupported
924 ^{210}Pb by mineral soils and sediments. *Journal of Environmental Radioactivity* 30, 117-137.

925 Horowitz, A.J., 1991. A primer on sediment-trace element chemistry, 2nd edition. Open-File Report 91-76,
926 United States Geological Survey; Books and Open-File Reports Section, Denver, Colorado, 136 pp.

927 Horowitz, A.J. and Elrick, K.A., 1987. The relation of stream sediment surface area, grain size and
928 composition to trace element chemistry. *Applied Geochemistry* 2, 437-451.

929 Hughes, A.O., Olley, J.M., Croke, J.C. and McKergow, L.A., 2009. Sediment source changes over the last 250
930 years in a dry-tropical catchment, central Queensland, Australia. *Geomorphology* 104, 262-275.

931 IAEA, 2014. Guidelines for Using Fallout Radionuclides to Assess Erosion and Effectiveness of Soil
932 Conservation Strategies, IAEA (International Atomic Energy Agency) publication, IAEA-TECDOC-
933 1741. Vienna, Austria, 213 pp.

934 Jagercikova, M., Cornu, S., Le Bas, C. and Evrard, O., 2015. Vertical distributions of ¹³⁷Cs in soils: a meta-
935 analysis. *Journal of Soils and Sediments* 15, 81-95.

936 Keil, R.G. and Mayer, L.M., 2014. 12.12 - Mineral Matrices and Organic Matter A2 - Holland, Heinrich D. In:
937 K.K. Turekian (Editor), *Treatise on Geochemistry* (Second Edition). Elsevier, Oxford, 337-359.

938 Kersten, M. and Smedes, F., 2002. Normalization procedures for sediment contaminants in spatial and
939 temporal trend monitoring. *Journal of Environmental Monitoring* 4, 109-115.

940 Klages, M.G. and Hsieh, Y.P., 1975. Suspended solids carried by the Gallatin River of southwestern
941 Montana: II. Using minerology for inferring sources. *Journal of Environmental Quality* 4, 68-73.

942 Koiter, A., Lobb, D., Owens, P., Petticrew, E., Tiessen, K.D. and Li, S., 2013a. Investigating the role of
943 connectivity and scale in assessing the sources of sediment in an agricultural watershed in the
944 Canadian prairies using sediment source fingerprinting. *Journal of Soils and Sediments* 13, 1676-
945 1691.

946 Koiter, A.J., Owens, P.N., Petticrew, E.L. and Lobb, D.A., 2013b. The behavioural characteristics of sediment
947 properties and their implications for sediment fingerprinting as an approach for identifying
948 sediment sources in river basins. *Earth-Science Reviews* 125, 24-42.

949 Koiter, A.J., Owens, P.N., Petticrew, E.L. and Lobb, D.A., 2015. The role of gravel channel beds on the
950 particle size and organic matter selectivity of transported fine-grained sediment: implications for
951 sediment fingerprinting and biogeochemical flux research. *Journal of Soils and Sediments* 15,
952 2174-2188.

953 Koiter, A.J., Owens, P.N., Petticrew, E.L. and Lobb, D.A., 2017. The role of soil surface properties on the
954 particle size and carbon selectivity of interrill erosion in agricultural landscapes. *CATENA* 153, 194-
955 206.

956 Kraushaar, S., Schumann, T., Ollesch, G., Schubert, M., Vogel, H.-J. and Siebert, C., 2015. Sediment
957 fingerprinting in northern Jordan: element-specific correction factors in a carbonatic setting.
958 *Journal of Soils and Sediments* 15, 2155-2173.

959 Krumbein, W.C. and Sloss, L.L., 1951. Stratigraphy and sedimentation. *Soil Science* 71, 401.

960 Laceby, J.P., Huon, S., Onda, Y. and Evrard, O., 2016. Do forests represent a long-term source of
961 contaminated particulate matter in the Fukushima Prefecture? *Journal of Environmental*
962 *Management* 183, 742–753.

963 Laceby, J.P., McMahon, J., Evrard, O. and Olley, J., 2015a. A comparison of geological and statistical
964 approaches to element selection for sediment fingerprinting. *Journal of Soils and Sediments* 15,
965 2117-2131.

966 Laceby, J.P. and Olley, J., 2015. An examination of geochemical modelling approaches to tracing sediment
967 sources incorporating distribution mixing and elemental correlations. *Hydrological Processes* 29,
968 1669-1685

969 Laceby, J.P., Olley, J., Pietsch, T.J., Sheldon, F. and Bunn, S.E., 2015b. Identifying subsoil sediment sources
970 with carbon and nitrogen stable isotope ratios. *Hydrological Processes* 29, 1956-1971.

971 Le Cloarec, M.F., Bonte, P.H., Lestel, L., Lefèvre, I. and Ayrault, S., 2011. Sedimentary record of metal
972 contamination in the Seine River during the last century. *Physics and Chemistry of the Earth, Parts*
973 *A/B/C* 36, 515-529.

974 Le Gall, M., Evrard, O., Foucher, A., Laceby, J.P., Salvador-Blanes, S., Thil, F., Dapoigny, A., Lefèvre, I.,
975 Cerdan, O. and Ayrault, S., 2016. Quantifying sediment sources in a lowland agricultural catchment
976 pond using ¹³⁷Cs activities and radiogenic ⁸⁷Sr/⁸⁶Sr ratios. *Science of The Total Environment* 566-
977 567, 968–980.

978 Le, M.H., Cordier, S., Lucas, C. and Cerdan, O., 2015. A faster numerical scheme for a coupled system
979 modeling soil erosion and sediment transport. *Water Resources Research* 51, 987-1005.

980 Lewis, J., 1996. Turbidity-controlled suspended sediment sampling for runoff-event load estimation. *Water*
981 *Resources Research* 32, 2299-2310.

982 Lomenick, T.F. and Tamura, T., 1965. Naturally Occurring Fixation of Cesium-137 on Sediments of
983 Lacustrine Origin. *Soil Science Society of America Journal* 29, 383-387.

984 Mabit, L., Benmansour, M., Abril, J.M., Walling, D.E., Meusburger, K., Iurian, A.R., Bernard, C., Tarján, S.,
985 Owens, P.N. and Blake, W.H., 2014. Fallout ^{210}Pb as a soil and sediment tracer in catchment
986 sediment budget investigations: a review. *Earth-Science Reviews* 138, 335-351.

987 Martinez-Carreras, N., Udelhoven, T., Krein, A., Gallart, F., Iffly, J.F., Ziebel, J., Hoffmann, L., Pfister, L. and
988 Walling, D.E., 2010. The use of sediment colour measured by diffuse reflectance spectrometry to
989 determine sediment sources: Application to the Atert River catchment (Luxembourg). *Journal of*
990 *Hydrology* 382, p 49-63.

991 Matisoff, G., Bonniwell, E.C. and Whiting, P.J., 2002. Soil erosion and sediment sources in an Ohio
992 watershed using Beryllium-7, Cesium-137, and Lead-210. *Journal of Environmental Quality* 31, 54-
993 61.

994 Matisoff, G., Wilson, C.G. and Whiting, P.J., 2005. The $^7\text{Be}/^{210}\text{Pb}_{\text{xs}}$ ratio as an indicator of suspended
995 sediment age or fraction new sediment in suspension. *Earth Surface Processes and Landforms* 30,
996 1191-1201.

997 Mayer, L., 1999. Extent of coverage of mineral surfaces by organic matter in marine sediments. *Geochimica*
998 *et Cosmochimica Acta* 63, 207-215.

999 McCulloch, M., Fallon, S., Wyndham, T., Hendy, E., Lough, J. and Barnes, D., 2003. Coral record of increased
1000 sediment flux to the inner Great Barrier Reef since European settlement. *Nature* 421, 727-730.

1001 McLaren, P. and Bowles, D., 1985. The effects of sediment transport on grain-size distributions. *Journal of*
1002 *Sedimentary Research* 55, 457-470.

1003 Merkus, H.G., 2009. *Particle Size Measurements: Fundamentals, Practice, Quality*, 17. Springer Science &
1004 Business Media, 532 pp.

1005 Morgan, R., 2005. *Soil Erosion and Conservation*, Third Edition. Blackwell Publishing, Oxford, 303 pp.

1006 Morris, G.L. and Fan, J., 1998. *Reservoir Sedimentation Handbook: Design and Management of Dams,*
1007 *Reservoirs, and Watersheds for Sustainable Use.* McGraw Hill Professional, New York, 848 pp.

1008 Moss, A. and Walker, P., 1978. Particle transport by continental water flows in relation to erosion,
1009 deposition, soils, and human activities. *Sedimentary Geology* 20, 81-139.

1010 Motha, J.A., Wallbrink, P.J., Hairsine, P.B. and Grayson, R.B., 2002. Tracer properties of eroded sediment
1011 and source material. *Hydrological Processes* 16, 1983-2000.

1012 Motha, J.A., Wallbrink, P.J., Hairsine, P.B. and Grayson, R.B., 2003. Determining the sources of suspended
1013 sediment in a forested catchment in southeastern Australia. *Water Resources Research* 39, 1056.

1014 Mukundan, R., Radcliffe, D.E., Ritchie, J.C., Risse, L.M. and Mckinley, R.A., 2010. Sediment fingerprinting
1015 to determine the source of suspended sediment in a southern piedmont stream. *Journal of*
1016 *Environmental Quality* 39, 1328-1337.

1017 Mukundan, R., Walling, D.E., Gellis, A.C., Slattery, M.C. and Radcliffe, D.E., 2012. Sediment Source
1018 Fingerprinting: Transforming From a Research Tool to a Management Tool. *Journal of the*
1019 *American Water Resources Association* 48, 1241-1257.

1020 Murray, A., Stanton, R., Olley, J. and Morton, R., 1993a. Determining the origins and history of
1021 sedimentation in an underground river system using natural and fallout radionuclides. *Journal of*
1022 *Hydrology* 146, 341-359.

1023 Murray, A.S., Olive, L.J., Olley, J.M., Caitcheon, G.G., Wasson, R.J. and Wallbrink, P.J., 1993b. Tracing the
1024 Source of Suspended Sediment in the Murrumbidgee River, Australia. In: N.E. Peters, E. Hoehn, C.
1025 Leibundgut, N. Tase and D.E. Walling (Editors), *Tracers in Hydrology*, IAHS Publication No. 215,
1026 IAHS Press, Wallingford, 293-302.

1027 Natelhoffer, K.J. and Fry, B., 1988. Controls on natural nitrogen-15 and carbon-13 abundances in forest
1028 soil organic matter. *Soil Science Society of America Journal* 52, 1633-1640.

- 1029 Navratil, O., Evrard, O., Esteves, M., Legout, C., Ayrault, S., Némery, J., Mate-Marin, A., Ahmadi, M.,
1030 Lefèvre, I. and Poirel, A., 2012. Temporal variability of suspended sediment sources in an alpine
1031 catchment combining river/rainfall monitoring and sediment fingerprinting. *Earth Surface*
1032 *Processes and Landforms* 37, 828-846.
- 1033 Oldfield, F., 1991. Environmental magnetism — A personal perspective. *Quaternary Science Reviews* 10,
1034 73-85.
- 1035 Oldfield, F., Maher, B., Donoghue, J. and Pierce, J., 1985. Particle-size related, mineral magnetic source
1036 sediment linkages in the Rhode River catchment, Maryland, USA. *Journal of the Geological Society*
1037 142, 1035-1046.
- 1038 Olley, J., Burton, J., Smolders, K., Pantus, F. and Pietsch, T., 2013. The application of fallout radionuclides
1039 to determine the dominant erosion process in water supply catchments of subtropical South-east
1040 Queensland, Australia. *Hydrological Processes* 27, 885-895.
- 1041 Olley, J. and Caitcheon, G., 2000. Major element chemistry of sediments from the Darling-Barwon river
1042 and its tributaries: implications for sediment and phosphorus sources. *Hydrological Processes* 14,
1043 1159-1175.
- 1044 Olley, J.M., 2002. Organic carbon supply to a large lowland river and implications for aquatic ecosystems.
1045 In: F.J. Dyer, M.C. Thoms and J.M. Olley (Editors), *The Structure Function and Management*
1046 *Implications of Fluvial Sedimentary Systems*, IAHS Publication No. 276, IAHS Press, Wallingford,
1047 27-33.
- 1048 Olley, J.M., Caitcheon, G.G., Hancock, G. and Wallbrink, P.J., 2001. *Tracing and Dating Techniques for*
1049 *Sediment and Associated Substances*. CSIRO Land and Water, Canberra.
- 1050 Olley, J.M. and Murray, A.S., 1994. Origins of variability in the $^{230}\text{Th}/^{232}\text{Th}$ ratio in sediments. In: D.E. Walling
1051 and Q. He (Editors), *Variability in Stream Erosion and Sediment Transport*, IAHS Publication No.
1052 224, IAHS Press, Wallingford, 65-70.
- 1053 Ortiz, J.D., Polyak, L., Grebmeier, J.M., Darby, D., Eberl, D.D., Naidu, S. and Nof, D., 2009. Provenance of
1054 Holocene sediment on the Chukchi-Alaskan margin based on combined diffuse spectral
1055 reflectance and quantitative X-Ray Diffraction analysis. *Global and Planetary Change* 68, 73-84.
- 1056 Owens, P.N., Batalla, R.J., Collins, A.J., Gomez, B., Hicks, D.M., Horowitz, A.J., Kondolf, G.M., Marden, M.,
1057 Page, M.J., Peacock, D.H., Petticrew, E.L., Salomons, W. and Trustrum, N.A., 2005. Fine-grained
1058 sediment in river systems: environmental significance and management issues. *River Research and*
1059 *Applications* 21, 693-717.
- 1060 Owens, P.N., Blake, W.H., Gaspar, L., Gateuille, D., Koiter, A.J., Lobb, D.A., Petticrew, E.L., Raiffarth, D.,
1061 Smith, H.G. and Woodward, J.C., 2016. Fingerprinting and tracing the sources of soils and
1062 sediments: earth and ocean science, geoarchaeological, forensic, and human health applications.
1063 *Earth-Science Reviews* 162, 1-23.
- 1064 Owens, P.N., Blake, W.H., Giles, T.R. and Williams, N.D., 2012. Determining the effects of wildfire on
1065 sediment sources using ^{137}Cs and unsupported ^{210}Pb : the role of landscape disturbances and
1066 driving forces. *Journal of Soils and Sediments* 12, 982-994.
- 1067 Owens, P.N., Blake, W.H. and Petticrew, E.L., 2006. Changes in sediment sources following wildfire in
1068 mountainous terrain: A paired-catchment approach, British Columbia, Canada. In: B. Kronvang, J.
1069 Faganeli and N. Ogrinc (Editors), *The Interactions Between Sediments and Water*. Springer
1070 Netherlands, 273-281.
- 1071 Owens, P.N., Walling, D.E. and Leeks, G.J.L., 2000. Tracing fluvial suspended sediment sources in the
1072 catchment of the River Tweed, Scotland, using composite fingerprints and a numerical mixing
1073 model. In: I. Foster (Editor), *Tracers in Geomorphology*. Wiley, Chichester, 291-308.
- 1074 Palazón, L., Gaspar, L., Latorre, B., Blake, W. and Navas, A., 2015a. Identifying sediment sources by applying
1075 a fingerprinting mixing model in a Pyrenean drainage catchment. *Journal of Soils and Sediments*
1076 15, 2067-2085.

1077 Palazón, L., Latorre, B., Gaspar, L., Blake, W.H., Smith, H.G. and Navas, A., 2015b. Comparing catchment
1078 sediment fingerprinting procedures using an auto-evaluation approach with virtual sample
1079 mixtures. *Science of The Total Environment* 532, 456-466.

1080 Panuska, J.C., Karthikeyan, K.G. and Miller, P.S., 2008. Impact of surface roughness and crusting on particle
1081 size distribution of edge-of-field sediments. *Geoderma* 145, 315-324.

1082 Papanicolaou, A.N., Fox, J.F. and Marshall, J., 2003. Soil fingerprinting in the Palouse Basin, USA using
1083 stable carbon and nitrogen isotopes. *International Journal of Sediment Research* 18, 278-284.

1084 Peart, M.R. and Walling, D.E. (Editors), 1986. Fingerprinting sediment source: The example of a drainage
1085 basin in Devon, UK. *Drainage Basin Sediment Delivery*, IAHS Publication No. 159, IAHS Press,
1086 Wallingford, 41-55 pp.

1087 Phillips, J. and Walling, D., 1995. An assessment of the effects of sample collection, storage and
1088 resuspension on the representativeness of measurements of the effective particle size distribution
1089 of fluvial suspended sediment. *Water Research* 29, 2498-2508.

1090 Poesen, J., 1992. Mechanisms of overland-flow generation and sediment production on loamy and sandy
1091 soils with and without rock fragments. In: A.J. Parsons and A.D. Abrahams (Editors), *Overland Flow*
1092 *Hydraulics and Erosion Mechanics*. Routledge, London, 275-305.

1093 Poleto, C., Merten, G.H. and Minella, J.P., 2009. The identification of sediment sources in a small urban
1094 watershed in southern Brazil: An application of sediment fingerprinting. *Environmental*
1095 *Technology* 30, 1145-1153.

1096 Poulénard, J., Legout, C., Némery, J., Bramorski, J., Navratil, O., Douchin, A., Fanget, B., Perrette, Y., Evrard,
1097 O. and Esteves, M., 2012. Tracing sediment sources during floods using Diffuse Reflectance
1098 Infrared Fourier Transform Spectrometry (DRIFTS): A case study in a highly erosive mountainous
1099 catchment (Southern French Alps). *Journal of Hydrology* 414-415, 452-462.

1100 Poulénard, J., Perrette, Y., Fanget, B., Quetin, P., Trevisan, D. and Dorioz, J.M., 2009. Infrared spectroscopy
1101 tracing of sediment sources in a small rural watershed (French Alps). *Science of The Total*
1102 *Environment* 407, 2808-2819.

1103 Pulley, S., Foster, I. and Antunes, P., 2015a. The application of sediment fingerprinting to floodplain and
1104 lake sediment cores: assumptions and uncertainties evaluated through case studies in the Nene
1105 Basin, UK. *Journal of Soils and Sediments*, 1-23.

1106 Pulley, S., Rowntree, K. and Foster, I., 2015b. Conservatism of mineral magnetic signatures in farm dam
1107 sediments in the South African Karoo: the potential effects of particle size and post-depositional
1108 diagenesis. *Journal of Soils and Sediments* 15, 2387-2397.

1109 Rawlins, B.G., Turner, G., Mounteney, I. and Wildman, G., 2010. Estimating specific surface area of fine
1110 stream bed sediments from geochemistry. *Applied Geochemistry* 25, 1291-1300.

1111 Reiffarth, D.G., Petticrew, E.L., Owens, P.N. and Lobb, D.A., 2016. Sources of variability in fatty acid (FA)
1112 biomarkers in the application of compound-specific stable isotopes (CSSIs) to soil and sediment
1113 fingerprinting and tracing: A review. *Science of The Total Environment* 565, 8-27.

1114 Russell, M.A., Walling, D.E. and Hodgkinson, R.A., 2001. Suspended sediment sources in two small lowland
1115 agricultural catchments in the UK. *Journal of Hydrology* 252, 1-24.

1116 Schimel, D.S., 1993. *Theory and Application of Tracers. Isotopic Techniques in Plant, Soil and Aquatic*
1117 *Biology*. Academic Press, Inc., San Diego., 119 pp.

1118 Sherriff, S.C., Franks, S.W., Rowan, J.S., Fenton, O. and Ó'hUallacháin, D., 2015. Uncertainty-based
1119 assessment of tracer selection, tracer non-conservativeness and multiple solutions in sediment
1120 fingerprinting using synthetic and field data. *Journal of Soils and Sediments* 10, 2101-2116.

1121 Singh, M., Sharma, M. and Tobschall, H.J., 2005. Weathering of the Ganga alluvial plain, northern India:
1122 implications from fluvial geochemistry of the Gomati River. *Applied Geochemistry* 20, 1-21.

1123 Singleton, A.A., Schmidt, A.H., Bierman, P.R., Rood, D.H., Neilson, T.B., Greene, E.S., Bower, J.A. and
1124 Perdrial, N., 2017. Effects of grain size, mineralogy, and acid-extractable grain coatings on the

- 1125 distribution of the fallout radionuclides ^7Be , ^{10}Be , ^{137}Cs , and ^{210}Pb in river sediment.
1126 *Geochimica et Cosmochimica Acta* 197, 71-86.
- 1127 Slattery, M.C., Burt, T.P. and Walden, J., 1995. The application of mineral magnetic measurements to
1128 quantify within-storm variations in suspended sediment sources. In: C. Leibundgut (Editor), *Tracer*
1129 *Technologies for Hydrological Systems*, IAHS Publication No. 229, IAHS Press, Wallingford, 143-
1130 152.
- 1131 Smith, H.G. and Blake, W.H., 2014. Sediment fingerprinting in agricultural catchments: A critical re-
1132 examination of source discrimination and data corrections. *Geomorphology* 204, 177-191.
- 1133 Smith, H.G., Blake, W.H. and Owens, P.N., 2013. Discriminating fine sediment sources and the application
1134 of sediment tracers in burned catchments: a review. *Hydrological Processes* 27, 943-958.
- 1135 Smith, H.G., Blake, W.H. and Taylor, A., 2014. Modelling particle residence times in agricultural river basins
1136 using a sediment budget model and fallout radionuclide tracers. *Earth Surface Processes and*
1137 *Landforms* 39, 1944-1959.
- 1138 Smith, H.G., Sheridan, G.J., Lane, P.N., Noske, P.J. and Heijnis, H., 2011. Changes to sediment sources
1139 following wildfire in a forested upland catchment, southeastern Australia. *Hydrological Processes*
1140 25, 2878-2889.
- 1141 Smith, J., 1999. An introduction to the magnetic properties of natural materials. *Environmental*
1142 *Magnetism: A Practical Guide*. Technical Guide 6, 5-34.
- 1143 Stone, M., Collins, A., Silins, U., Emelko, M. and Zhang, Y., 2014. The use of composite fingerprints to
1144 quantify sediment sources in a wildfire impacted landscape, Alberta, Canada. *Science of The Total*
1145 *Environment* 473, 642-650.
- 1146 Stone, P. and Walling, D., 1997. Particle size selectivity considerations in suspended sediment budget
1147 investigations. In: R.D. Evans, J. Wisniewski and J.R. Wisniewski (Editors), *The Interactions Between*
1148 *Sediments and Water*, Springer, Netherlands, 63-70.
- 1149 Stout, J.C., Belmont, P., Schottler, S.P. and Willenbring, J.K., 2014. Identifying sediment sources and sinks
1150 in the Root River, Southeastern Minnesota. *Annals of the Association of American Geographers*
1151 104, 20-39.
- 1152 Stumm, W. and Morgan, J.J., 2012. *Aquatic Chemistry: Chemical Equilibria and rates in Natural Waters*.
1153 John Wiley & Sons, Chichester, 1040 pp.
- 1154 Szava-Kovats, R.C., 2008. Grain-size normalization as a tool to assess contamination in marine sediments:
1155 Is the $< 63\mu\text{m}$ fraction fine enough? *Marine Pollution Bulletin* 56, 629-632.
- 1156 Tamura, T., 1964. Consequences of activity release: selective sorption reactions of cesium with soil
1157 minerals. *Nuclear Safety* 5, 262-268.
- 1158 Taylor, A., Blake, W.H. and Keith-Roach, M.J., 2014. Estimating Be-7 association with soil particle size
1159 fractions for erosion and deposition modelling. *Journal of Soils and Sediments* 14, 1886-1893.
- 1160 Taylor, A., Blake, W.H., Smith, H.G., Mabit, L. and Keith-Roach, M.J., 2013. Assumptions and challenges in
1161 the use of fallout beryllium-7 as a soil and sediment tracer in river basins. *Earth-Science Reviews*
1162 126, 85-95.
- 1163 Tiecher, T., Caner, L., Minella, J.P.G., Bender, M.A. and dos Santos, D.R., 2016. Tracing sediment sources in
1164 a subtropical rural catchment of southern Brazil by using geochemical tracers and near-infrared
1165 spectroscopy. *Soil and Tillage Research* 155, 478-491.
- 1166 Vale, S.S., Fuller, I.C., Procter, J.N., Basher, L.R. and Smith, I.E., 2016. Characterization and quantification
1167 of suspended sediment sources to the Manawatu River, New Zealand. *Science of The Total*
1168 *Environment* 543, 171-186.
- 1169 Vercruyssen, K., Grabowski, R.C. and Rickson, R.J., 2017. Suspended sediment transport dynamics in rivers:
1170 Multi-scale drivers of temporal variation. *Earth-Science Reviews* 166, 38-52.
- 1171 Viparelli, E., Wesley Lauer, J., Belmont, P. and Parker, G., 2013. A numerical model to develop long-term
1172 sediment budgets using isotopic sediment fingerprints. *Computers & Geosciences* 53, 114-122.

- 1173 Walden, J. and Slattery, M.C., 1993. Verification of a simple gravity technique for separation of particle
1174 size fractions suitable for mineral magnetic analysis. *Earth Surface Processes and Landforms* 18,
1175 829-833.
- 1176 Wall, G.J. and Wilding, L.P., 1975. Minerology and Related Parameters of Fluvial Suspended Sediments in
1177 Northwestern Ohio. *Journal of Environmental Quality* 5, 168-173.
- 1178 Wallbrink, P. and Murray, A., 1996a. Distribution and variability of ⁷Be in soils under different surface
1179 cover conditions and its potential for describing soil redistribution processes. *Water Resources*
1180 *Research* 32, 467-476.
- 1181 Wallbrink, P.J. and Murray, A.S., 1993. Use of fallout radionuclides as indicators of erosion processes.
1182 *Hydrological Processes* 7, 297-304.
- 1183 Wallbrink, P.J. and Murray, A.S., 1996b. Determining Soil Loss Using The Inventory Ratio Of Excess-Lead
1184 ²¹⁰Pb To Cesium-137. *Soil Science Society of America Journal* 60, 1201-1208.
- 1185 Wallbrink, P.J., Murray, A.S., Olley, J.M. and Olive, L.J., 1998. Determining sources and transit times of
1186 suspended sediment in the Murrumbidgee River, New South Wales, Australia, using fallout ¹³⁷Cs
1187 and ²¹⁰Pb. *Water Resources Research* 34, 879-887.
- 1188 Wallbrink, P.J., Olley, J.M. and Murray, A.S., 1999. Relating suspended sediment to its original soil depth
1189 using fallout radionuclides. *Soil Science Society of America Journal* 63, 369-378.
- 1190 Walling, D. and Moorehead, P., 1989. The particle size characteristics of fluvial suspended sediment: an
1191 overview. *Hydrobiologia* 176, 125-149.
- 1192 Walling, D., Peart, M., Oldfield, F. and Thompson, R., 1979. Suspended sediment sources identified by
1193 magnetic measurements. *Nature* 281, 110-113.
- 1194 Walling, D.E., 2003. Using environmental radionuclides as tracers in sediment budget investigations. In:
1195 D.E. Walling and T. Fergus (Editors), *Erosion and Sediment Transport Measurement in Rivers:*
1196 *Technological and Methodological Advances*, IAHS Publication No. 283, IAHS Press, Wallingford,
1197 57-78.
- 1198 Walling, D.E., 2005. Tracing suspended sediment sources in catchments and river systems. *Science of The*
1199 *Total Environment* 344, 159-184.
- 1200 Walling, D.E. and Collins, A.L., 2016. Fine sediment transport and management. In: D.J. Gilvear, M.T.
1201 Greenwood, M.C. Thoms and P.J. Wood (Editors), *River Science: Research and Management for*
1202 *the 21st Century*, John Wiley & Sons, Chichester, 37-60.
- 1203 Walling, D.E. and Kane, P., 1984. Suspended sediment properties and their geomorphological significance.
1204 In: T.P. Burt and D.E. Walling (Editors), *Catchment Experiments in Fluvial Geomorphology*, Geo
1205 Books, Norwich, 311-334.
- 1206 Walling, D.E., Owens, P.N. and Leeks, G.J.L., 1999. Fingerprinting suspended sediment sources in the
1207 catchment of the River Ouse, Yorkshire, UK. *Hydrological Processes* 13, 955-975.
- 1208 Walling, D.E., Owens, P.N., Waterfall, B.D., Leeks, G.J. and Wass, P.D., 2000. The particle size characteristics
1209 of fluvial suspended sediment in the Humber and Tweed catchments, UK. *Science of The Total*
1210 *Environment* 251, 205-222.
- 1211 Walling, D.E. and Woodward, J.C., 1992. Use of radiometric fingerprints to derive information on
1212 suspended sediment sources In: J. Bogen, D.E. Walling and T. Day (Editors), *Erosion and Sediment*
1213 *Transport Monitoring Programmes in River Basins*, IAHS Publication No. 210, IAHS Press,
1214 Wallingford, 153-164.
- 1215 Walling, D.E., Woodward, J.C. and Nicholas, A.P., 1993. A multi-parameter approach to fingerprinting
1216 suspended-sediment sources. In: N.E. Peters, E. Hoehn, C. Leibundgut, N. Tase and D.E. Walling
1217 (Editors), *Tracers in Hydrology*, IAHS Publication No. 215, IAHS Press, Wallingford, 329-338.
- 1218 Wischmeier, W.H. and Smith, D.D., 1978. Predicting Rainfall Erosion Losses-A guide to Conservation
1219 Planning. *Agricultural Handbook Number 537*, United States Department of Agriculture, Science
1220 and Education Administration, Hyattsville, Maryland, 58 pp.

- 1221 Wohl, E., Bledsoe, B.P., Jacobson, R.B., Poff, N.L., Rathburn, S.L., Walters, D.M. and Wilcox, A.C., 2015. The
1222 Natural Sediment Regime in Rivers: Broadening the Foundation for Ecosystem Management.
1223 *BioScience* 65, 358-371.
- 1224 Wynn, J.G., Bird, M.I. and Wong, V.N.L., 2005. Rayleigh distillation and the depth profile of $^{13}\text{C}/^{12}\text{C}$ ratios
1225 of soil organic carbon from soils of disparate texture in Iron Range National Park, Far North
1226 Queensland, Australia. *Geochimica et Cosmochimica Acta* 69, 1961-1973.
- 1227 Yamashiki, Y., Onda, Y., Smith, H.G., Blake, W.H., Wakahara, T., Igarashi, Y., Matsuura, Y. and Yoshimura,
1228 K., 2014. Initial flux of sediment-associated radiocesium to the ocean from the largest river
1229 impacted by Fukushima Daiichi Nuclear Power Plant. *Scientific Reports* 4.
- 1230 Zhang, X.J. and Liu, B., 2016. Using multiple composite fingerprints to quantify fine sediment source
1231 contributions: A new direction. *Geoderma* 268, 108-118.
- 1232