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

3

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15

16 **Abstract:**

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

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

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46 **1. Introduction**

47 Accelerated soil erosion impacts land and water quality worldwide. Although sediment is a natural
48 component of fluvial systems that provides fundamental structure to riverine landscapes and is essential
49 in many aquatic ecosystems (Vercruysse et al., 2017; Wohl et al., 2015), the excess supply of fine
50 sediment from accelerated soil erosion often degrades riverine and coastal environments (McCulloch et
51 al., 2003; Owens et al., 2005; Walling and Collins, 2016) and contributes to the downstream transfer of
52 particle-bound contaminants (Gateuille et al., 2014; Yamashiki et al., 2014). Elevated suspended
53 sediment loads may also increase the cost of operating and maintaining water treatment and
54 transportation infrastructure (Clark, 1985). Knowledge of the relative contribution of different sources
55 supplying sediment to riverine, lacustrine and coastal systems is a crucial prerequisite to implementing
56 efficient best practices necessary to limit the off-site impacts of excessive sediment delivery (Belmont et
57 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
62 magnetic parameters, fallout radionuclides, major and trace element geochemistry, and compound
63 specific stable isotopes (CSSI) have all been used to trace sediment sources (Blake et al., 2012; Caitcheon,
64 1993; Evrard 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,
69 properties of the eroded sediment should remain constant through sediment detachment,
70 transportation and deposition processes, or again, vary in a predictable and measurable way (Belmont et
71 al., 2014; Koiter et 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
77 lighter and without bonds binding them to the substrate (Morgan, 2005; Poesen, 1992). Thereafter, the
78 particle size of suspended sediment directly influences settling velocities in aquatic systems (Gibbs et al.,
79 1971) resulting in the transport and deposition of suspended sediment being particle size selective
80 (Viparelli et al., 2013; Walling et al., 2000). Fluvial transport, including overland flow, produces changes
81 in the characteristics of the material being transported in comparison to the original source material. In
82 general, the average size of particles decreases, while the degree of sorting and the average roundness
83 increases, with distance travelled. These changes result from a combination of selective transportation,
84 deposition, and particle abrasion, with these processes acting over the entire landscape (Frings, 2008;
85 Krumbein and 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;
98 Koiter et al., 2013b; Owens et al., 2016; Smith et al., 2013), and a recent emphasis on modelling
99 approaches (Cooper et al., 2014; Haddadchi et al., 2014; Laceby and Olley, 2015; Palazón et al., 2015b;
100 Zhang and Liu, 2016), few studies have investigated the effects of particle size on sediment source
101 signatures (Olley and Murray, 1994; Russell et al., 2001; Smith and Blake, 2014). Here, we review the
102 effects and challenges (section 2), approaches (sections 3 and 4) and opportunities (section 5) of particle
103 size selectivity within the sediment source fingerprinting technique. We mainly focus on riverine
104 environments, specifically particle size selectivity as sediment moves from hillslopes into and through
105 river channels, but the concepts and examples presented are also relevant for similar applications in
106 other aquatic systems such 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
116 properties – but recognise that many of the findings are also likely to be relevant to other
117 biogeochemical properties 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
120 determine the relative contributions of sediment from different erosion processes to waterways (Ben
121 Slimane et al., 2016; IAEA, 2014; Matisoff et al., 2002; Owens et al., 2012; Smith et al., 2011; Wallbrink et
122 al., 1998; Walling and Woodward, 1992) (Table 1). As both ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ are concentrated near the
123 soil surface, as they are atmospheric fallout products, sediments eroded from rill or sheet erosion often
124 have high ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ activity concentrations (Walling, 2005), whereas sediments eroded from
125 subsoil channel bank or gully erosion processes have low ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ activity concentrations
126 (Belmont et al., 2014; Olley et al., 2013; Wallbrink et al., 1999). Comparing ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ in suspended
127 sediments and sediments generated by these different erosion processes generally allows for the
128 relative sediment contributions from these different erosion processes to be ascertained (Ben Slimane et
129 al., 2013; Wallbrink and Murray, 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

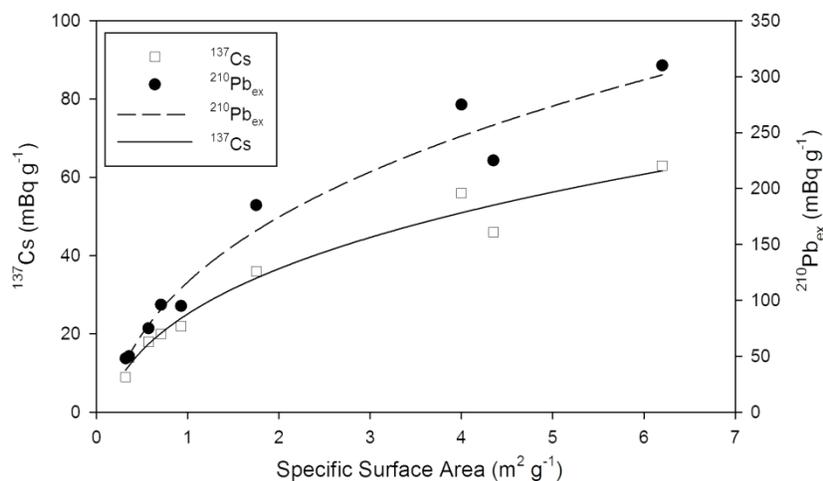
^e Sediments not soils

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
141 sediment exchange between the channel and the floodplain in large watersheds over sediment routing
142 timescales.

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

156 The challenge is that there are exceptions to the rule. For example, Smith and Blake (2014) observed that
157 ^{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
158 exhibited positive though non-significant relationships with SSA in channel bank and cultivated soils. This
159 highlights the fact that although these fallout radionuclides are generally enriched in the fine particle size
160 fractions, there may be exceptions, and individual sources may behave differently. Potential exceptions
161 may be explained by the presence and amount of HCl-extractable materials, which Singleton et al. (2017)
162 reported to have a stronger control on fallout radionuclides than grain size or mineralogy. Furthermore,

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



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

171 2.2 Carbon and Nitrogen Parameters

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

185 also have the potential for non-conservative behavior due to biological uptake and consumption.
186 Nonetheless, these parameters are increasingly used in sediment tracing research (Fox and
187 Papanicolaou, 2007; Laceby et al., 2015b; Mukundan et al., 2010; Papanicolaou et al., 2003) (Table 2).

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

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

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

211

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 ^a	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 Revill, 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	--	--	--	--

^a Source samples were dispersed with sodium hexametaphosphate

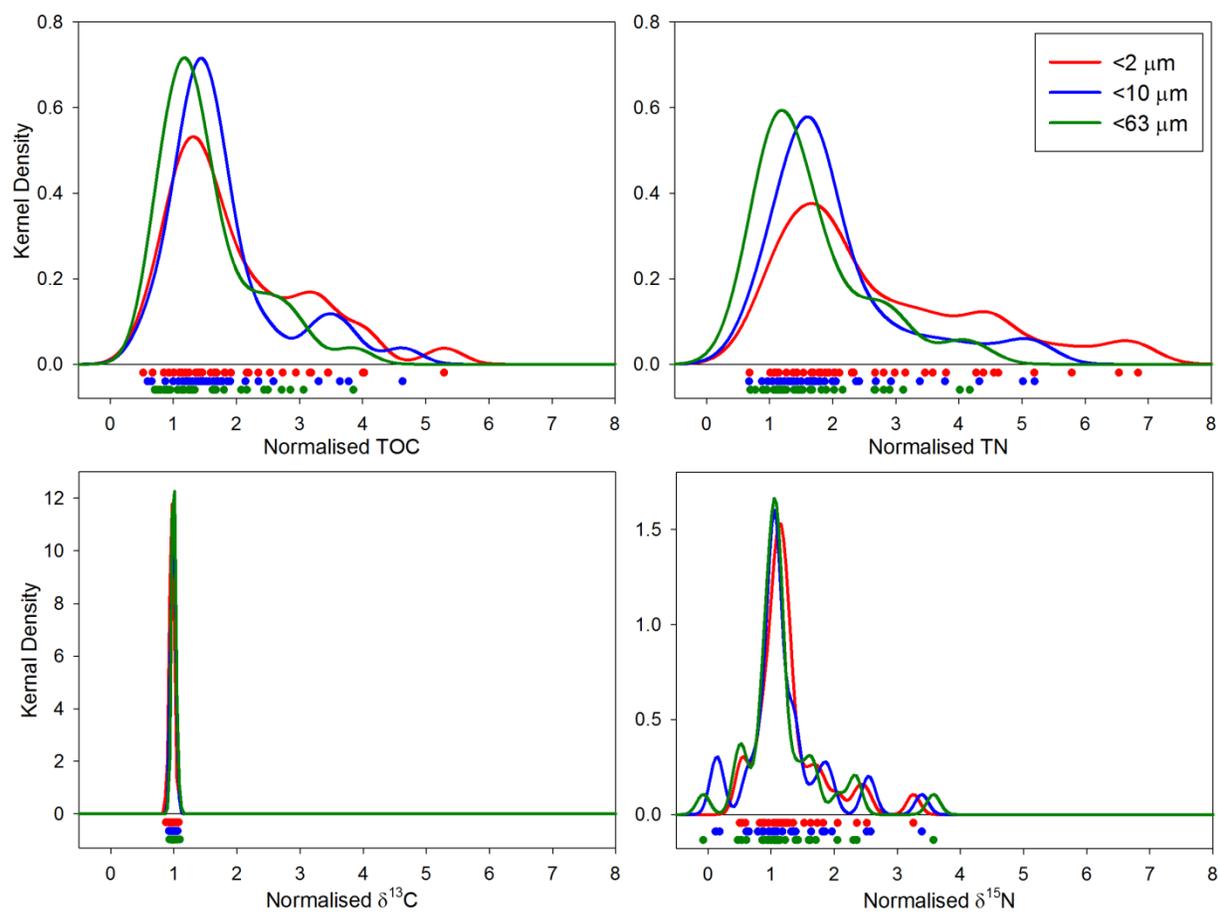


Figure 2: Normalized carbon (C) and nitrogen (N) property distributions for the <2 μm , <10 μm and <63 μm particle size fractions in samples from Knapp Creek and Blackfellow Creek, Australia (modified from Laceby et al., (2015b)). Points under the distributions are the normalized samples color coded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

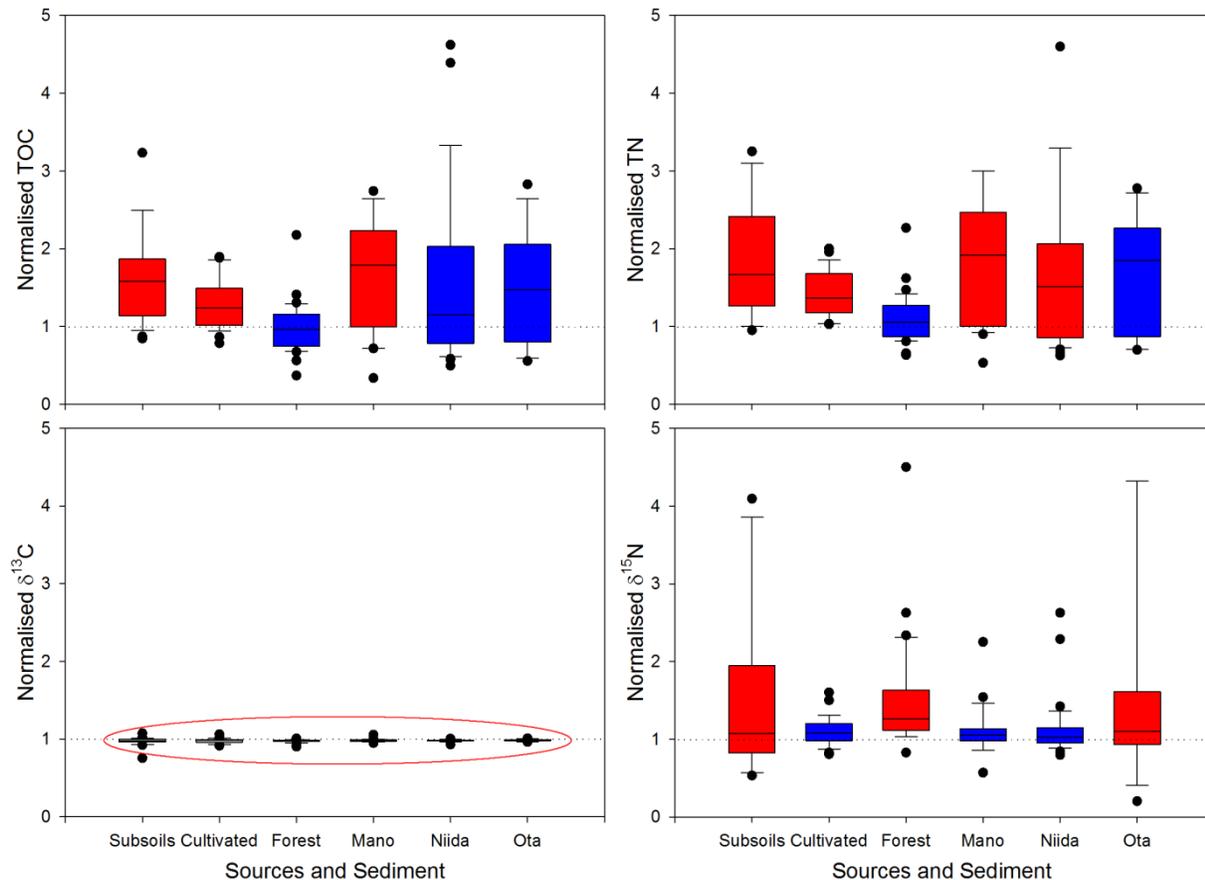


Figure 3: Box plots of the normalized difference between carbon (C) and nitrogen (N) sediment properties in the <63 μm fraction divided by the <2 mm fraction for the three sources and sediment from three Japanese catchments (i.e., Mano, Niida and Ota) with red shading (and the red circle) indicating 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)).

214 **2.3 Elemental Geochemistry**

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

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

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

232 Underlying the potential influence of particle size on elemental geochemistry is the effect of sediment
233 source mineralogy. The dominant mineralogy relates directly to particle size where some sources will be
234 enriched in the fine particle size fractions and other sources may be depleted. Thus the geochemical
235 fingerprint likely will change if the <2 mm, the <63 μm , or the <10 μm fraction are sampled and analysed.
236 For example, each of the three different sources in Figure 4 from Gibbs (1967) may have different
237 relationships between elemental concentrations and particle size, which will depend on the particle size
238 range utilized for the sediment source fingerprinting research. Fundamentally, the impact of particle size
239 on all elemental concentrations for each source is difficult to predict and the complexity of the particle
240 size - elemental geochemistry relationship requires more research to comprehensively characterize the
241 predictability of these fingerprints. Accordingly, section 4 of this review provides several
242 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	--	--	--

243

^a Source samples sonified before sieving

244

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

245

^c Samples sonified before settling in a water column

246

^d Raw data is only available for sediment core samples

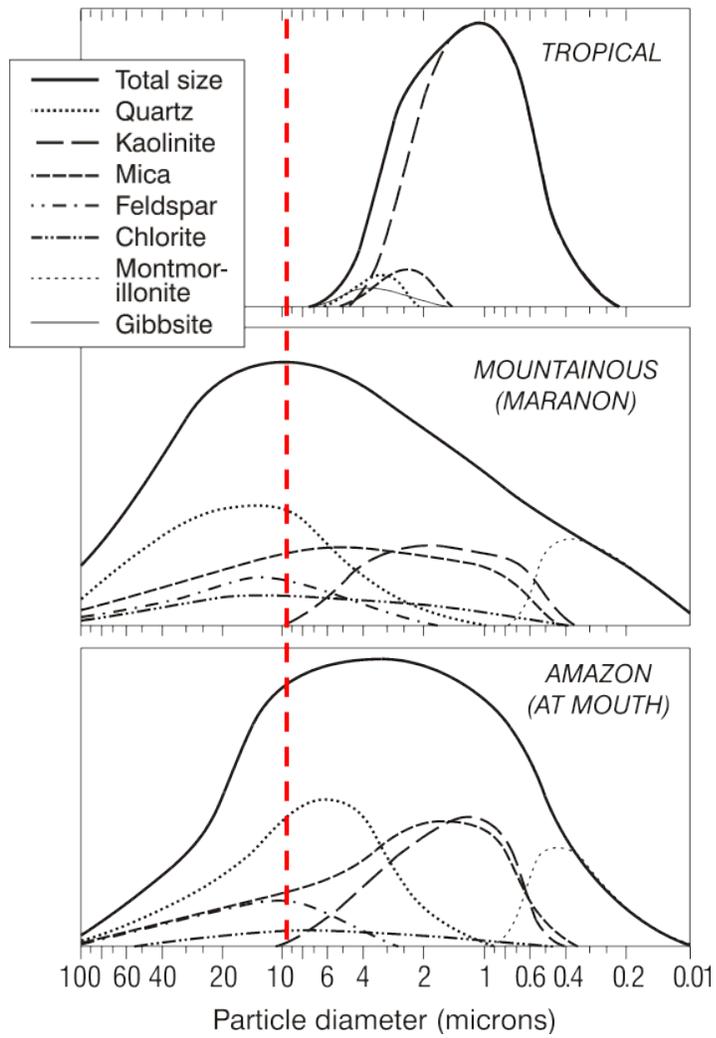


Figure 4: Changes in mineralogy with particle size in sediment from three locations in the Amazon basin from Gibbs (1967) with the red dashed line added to indicate the impact of fractionating the samples at the <10 μm particle size.

248 2.4 Mineral Magnetic Properties

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

258 Magnetic minerals occur in soil and sediment as aggregated concretions, discrete fine grains and particle
259 coatings on very fine grains (Oldfield, 1991; Smith, 1999). Accordingly, mineral magnetic properties may
260 be highly dependent on particle size (Foster et al., 1998; Oldfield et al., 1985). In particular, Hatfield and
261 Maher (2008) demonstrated the importance of characterizing mineral magnetic properties with a
262 particle-size specific approach as different magnetic properties were preferentially associated with
263 different particle size fractions. These authors demonstrated that bacterial magnetosomes formed in
264 lake sediment in the $<2 \mu\text{m}$ and the $2\text{-}8 \mu\text{m}$ fractions. Accordingly, they only quantified source
265 contributions to the $8\text{-}31 \mu\text{m}$ and $31\text{-}63 \mu\text{m}$ fractions. In contrast, Pulley et al. (2015b) reported
266 significantly different magnetic properties in the $<32 \mu\text{m}$ and the $>32 \mu\text{m}$ fractions and, for Caitcheon
267 (1998), the $63\text{-}125 \mu\text{m}$ fraction was appropriate for tracing tributary source contributions.

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

275

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

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

283 **2.5 Composite Fingerprinting Approach**

284 Peart and Walling (1986) advocated for multiple parameters to be used when quantifying sediment
285 source dynamics in order to improve the overall consistency and reliability of source ascription results.
286 The combination of multiple parameters creates a composite fingerprint (Walling et al., 1993) that allows
287 for an increased number of sources to be modelled and is theorized to be more representative of the
288 linkages between sediments and their sources, potentially reducing false matches which were
289 hypothesized to potentially occur with individual tracer properties (Collins et al., 1996). Accordingly, a
290 composite fingerprinting approach has been broadly applied in sediment source fingerprinting research
291 combining several or all of the following: fallout radionuclides, carbon and nitrogen parameters, element
292 geochemistry, mineral magnetism and other parameters, thereby providing significant source
293 discrimination (Collins et al., 1996; Evrard et al., 2013; Navratil et al., 2012; Owens et al., 2000) (Table 5).
294 One challenge with the composite fingerprinting approach is that each of the potential complexities
295 within the particle size – tracer parameter relationship described above is integrated into the expanded
296 composite fingerprint. A second challenge is that it is difficult, if not impossible, to link outputs of
297 statistical-based approaches (e.g. composite fingerprinting) back to a process-based understanding of
298 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

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

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

307 **3.1 Fractionation**

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

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

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

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

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

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

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

362 **3.2 Particle Size Corrections**

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

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

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

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

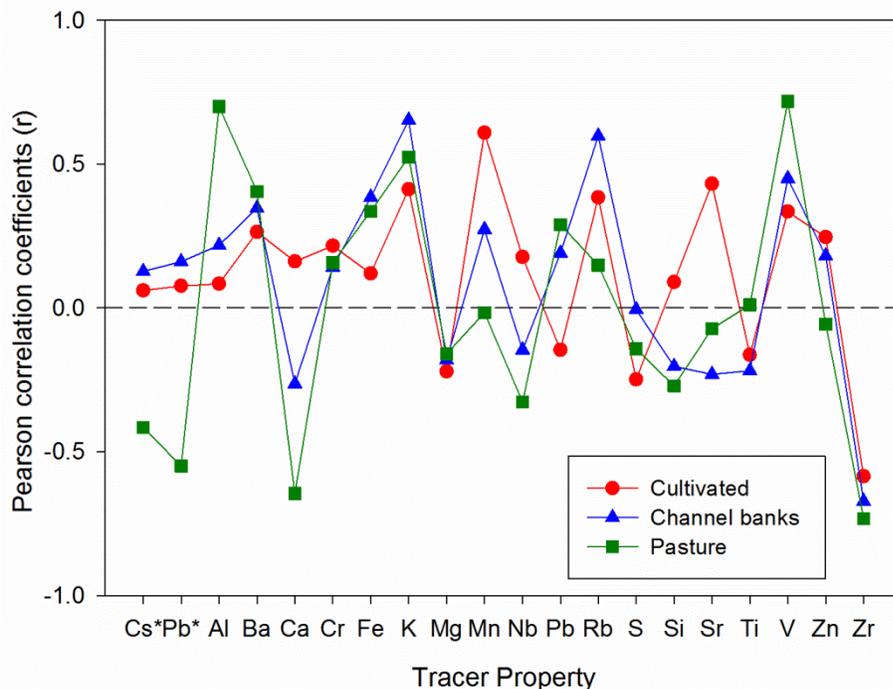


Figure 5: Pearson correlation coefficients (r) between Specific Surface Area (SSA) and tracer property for three sources from Smith and Blake (2014) (data from Table 1) with the * indicating fallout radionuclides (^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$) on the x axis and the remainder being elements.

392

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

403 source and sediment fingerprints. Given the potential uncertainty of accurate SSA ratio-based
404 corrections, researchers often do not apply particle size corrections and instead rely on physical
405 reasoning of sediment transport processes and/or sample fractionation (e.g. Koiter et al., 2013a;
406 Martinez-Carreras et al., 2010).

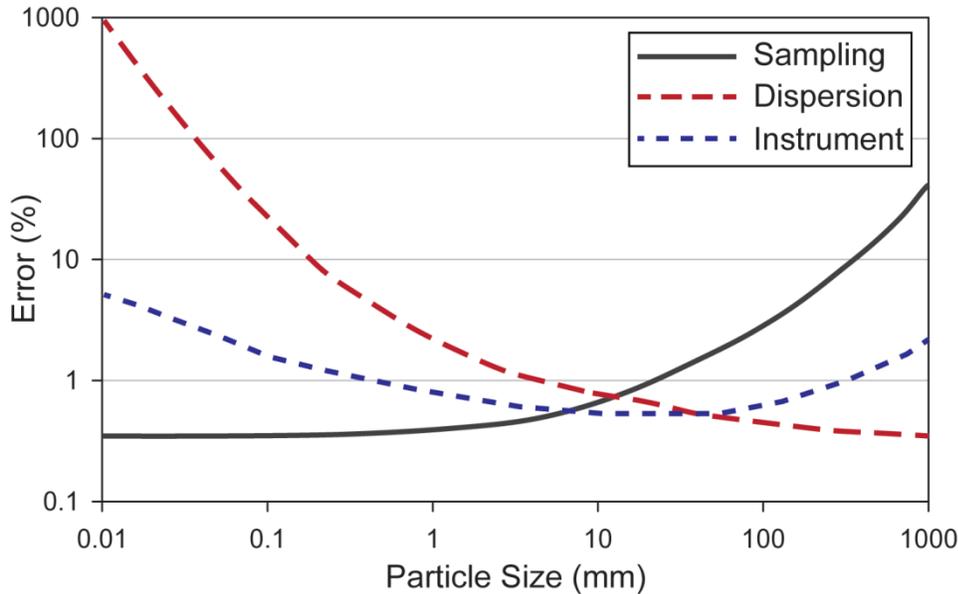


Figure 6: Relationship between potential sources of error and particle size distribution, adapted from Merkus (2009).

407

408 3.3 Combining Size Fractionation and Corrections Procedures

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

421 Russell et al., 2001). Understanding the impacts of particle size on tracer properties should be one of the
422 fundamental first steps when trialling new tracer properties in sediment source fingerprinting research.

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

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

428 **4.1 Tributary (or Confluence) Tracing**

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

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

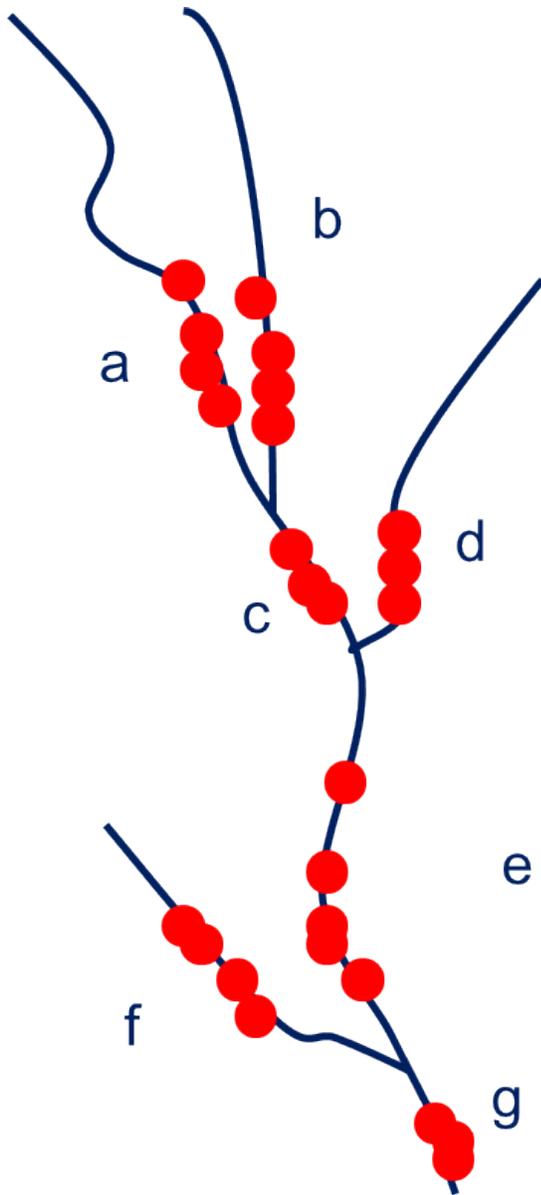


Figure 7: An example of a sampling design with the tributary tracing technique where the letters indicate the different tributaries that could be sampled in this theoretical catchment. In this situation, sediment collected at sites c, e and g is compared to sediment collected where it may be able to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

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

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

470 **4.2 Edge-of-Field Samplers**

471 An alternative technique that may limit potential particle size effects on biogeochemical tracers during
472 mobilization and initial transportation phases is the installation of edge-of-field samplers (Panuska et al.,
473 2008; Wallbrink and Murray, 1993). Edge-of-field sampling approaches capture sediment after the initial
474 mobilization and transport processes and are an alternative technique for examining whether particle
475 size corrections are required (Evrard et al., 2016), or if they have been applied correctly (Foucher et al.,
476 2015). Conceptually, samplers installed on USLE plots (Brooks et al., 2014; Wischmeier and Smith, 1978)
477 could opportunistically sample material mobilized from hillslopes. Gerlach troughs (Gerlach, 1967) could
478 sample suspended sediment on hillslopes for further analyses after it has been mobilized and
479 transported downslope. Similarly, V-notch weirs combined with automated sediment samplers may also
480 provide an effective approach for sampling hillslope sediments in ephemeral gullies and other ditch type
481 landscape features (Freebairn and Wockner, 1986).

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

495 **4.3 Multiple Fraction Tracing**

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

501 Caitcheon (1998) reported that source contributions from a sedimentary rock tributary basin varied only
502 by 13% (i.e. from 63% to 76%) when fingerprinting with mineral magnetic properties from different
503 particle size fractions (<63 μm , 63-125 μm , 125-250 μm and 250 - 500 μm). This author reported that the
504 63-125 μm fraction was most likely representative of the bulk material being transported in this
505 particular catchment. Using elemental geochemistry, Haddadchi et al. (2015) traced the fractionated <10
506 μm , 10-63 μm and 63-212 μm fractions. These authors found that although the maximum difference was
507 high (33%), there was <15% mean absolute difference between modelled source contributions with
508 these fractions (Figure 8). In summary, Haddadchi et al. (2015) indicated that their results highlight the
509 importance of fractionating the particle size most relevant to the management objective of the research.

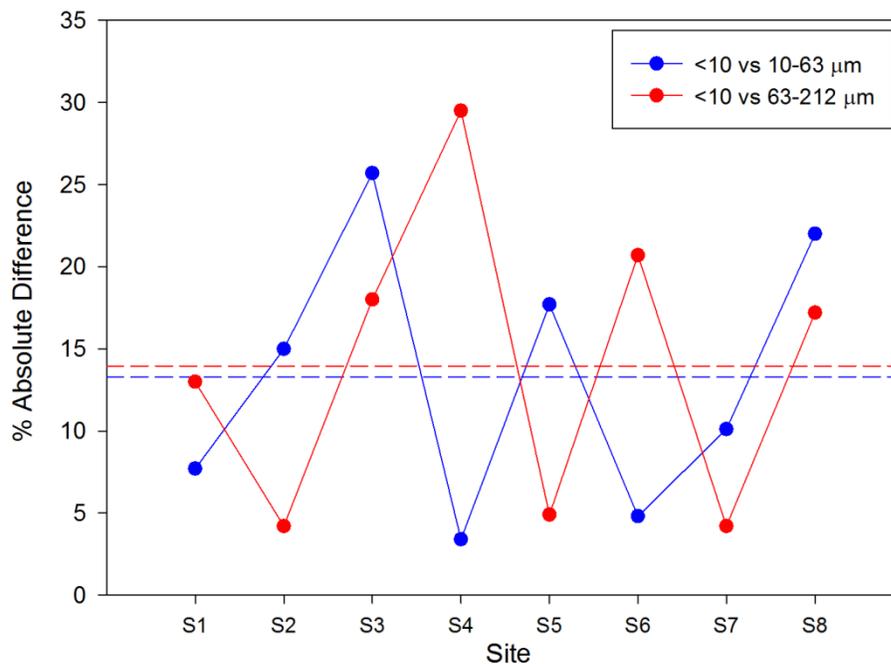


Figure 8: Mean absolute differences (dashed lines) in the source contributions between the <10 μm and the 10 - 63 μm (blue) and between the <10 μm and the 63 - 212 μm fractions (red) for eight sites in Australia plotted with data from Figure 9 in Haddadchi et al. (2015).

510

511 Laceby et al. (2016) also compared the impact of tracing different particle size fractions for three
 512 catchments in the Fukushima region in Japan. These authors compared the relative model difference
 513 from 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
 514 taken in three different catchments (Figure 3). The authors reported a maximum modelled average
 515 difference between the <2 mm and <63 μm fraction of 14%, 11% and 7% for the three catchments, and a
 516 mean relative difference of only 6% (SD 3%) for all possible modelled source contributions. The limited
 517 differences between these models was likely driven by the fact that only 19% (SD 13%) of the material in
 518 these sediment samples was <63 μm compared to 23% (SD 12%) of the material for the source samples,
 519 indicating that ~80% of material was >63 μm for both source and sediment samples. A similar approach
 520 should be applied in catchments with finer sediment transiting the system to characterize the potential
 521 impact of fine sediment material on modelling results when tracing different fractions.

522 These studies raise interesting questions regarding what are the actual differences in mixing model
 523 results arising from the impacts of particle size on tracer properties. Are these impacts greater than
 524 analytical and model uncertainty? Is this particle size impact significant for management objectives?

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

531 **5. Particle Size Opportunities**

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

542 **5.1 Mineralogy, Particle Size and Elemental Geochemistry**

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

553 Mineralogy was one of the first sediment fingerprinting properties (Klages and Hsieh, 1975; Wall and
554 Wilding, 1975) and returning to incorporate mineralogy more directly into sediment tracing may provide

555 significant insights into particle size impacts on elemental compositions. This may be achieved, although
556 indirectly, through using Diffuse Reflectance Infrared Fourier Transform Spectrometry (DRIFTS) to
557 identify mineralogical groups (e.g. calcite, aluminosilicates, quartz) and discriminate between sediment
558 lithological sources (Poulenard et al., 2012). The use of DRIFTS and other spectroscopy approaches to
559 tracing sediment sources has the potential to develop a strong link to mineralogy and potentially
560 estimate other conventional tracer parameters like texture (Balsam and Deaton, 1996; Ortiz et al., 2009).
561 Other approaches such as clay mineral ratios (e.g. illite/illite + expandable clays) have also shown
562 promise to trace sediment provenance (Bainbridge et al., 2016; Douglas et al., 2006a; Douglas et al.,
563 2006b). Connecting clay mineralogy to element geochemistry and particle size will likely provide
564 complementary information relating to each of these fundamental sediment property characteristics.

565 **5.2 Elemental Ratios and Elemental Normalisation**

566 Moving a step beyond incorporating mineralogy more effectively into sediment tracing research is the
567 potential for incorporating approaches from other disciplines to address particle size effects. For
568 example, in the sediment provenance literature, elemental ratios have long been used to investigate
569 changes in particle size. In particular, the Al/Si ratio is used as a proxy for particle size in sediment
570 provenance research (Figure 9) (Bouchez et al., 2011). Other ratios may indeed be effective, such as the
571 Ca/Al ratio (Chen et al., 2014), along with particle size sensitive elements such as Ti (Bábek et al., 2015),
572 or even simply clay content (Szava-Kovats, 2008). For example, studies reconstructing the evolution of
573 metal enrichment ratios in sediment have also normalized elemental concentrations to Sc, Al, Si, Li or Th
574 to minimize particle size impacts when examining contamination trends (Ayrault et al., 2010; Clark et al.,
575 2014; Grosbois et al., 2012; Kersten and Smedes, 2002; Le Cloarec et al., 2011). Indeed, there are
576 multiple approaches from the sediment provenance field (Armstrong-Altrin et al., 2015; Bábek et al.,
577 2015; Owens et al., 2016; Singh et al., 2005) that present significant opportunities for enhancing the
578 sediment source fingerprinting technique (e.g. Vale et al., 2016).

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

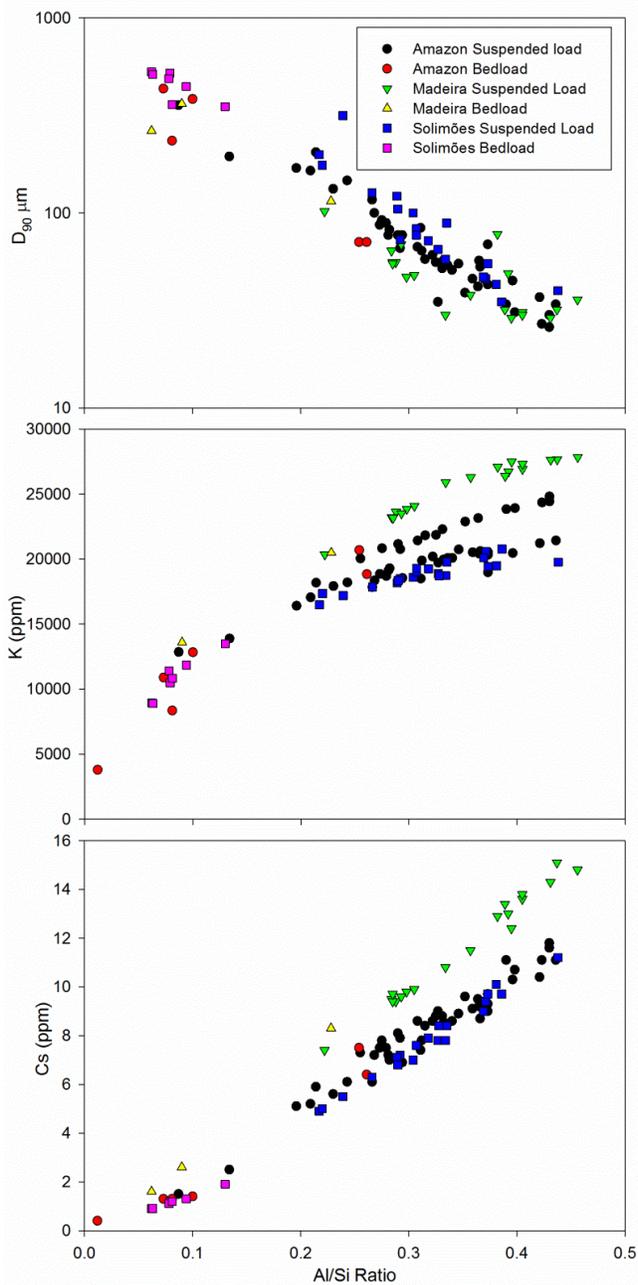
586 utility of incorporating ratios directly into the mixing model process to address particle size enrichment.
587 More research is required to examine whether similar approaches may also provide a technique for
588 normalizing particle size effects with frequentist or Bayesian mixing models, or even more insight into
589 the foundation for elemental discrimination between sediment sources. For example, the
590 interrelationships between particle size and biogeochemical tracer properties indicate that particle size
591 may directly contribute to the formation of some contrasts in sediment fingerprints between sources.
592 Accordingly, further advances in understanding these relationships, with experimental designs
593 incorporating different particle size fractions, may provide additional understanding for the basis of
594 discrimination between sediment sources.

595 **5.3 Particle Dynamics**

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

603 The impact of grain abrasion and disaggregation on ¹³⁷Cs concentrations was examined by Dyer and Olley
604 (1999) who reported that the fraction produced by abrasion had essentially similar ¹³⁷Cs concentrations
605 as the corresponding fraction (<40 µm) in non-disturbed soil. Conversely, Crockford and Olley (1998)
606 found that breakage and abrasion processes had a substantial impact on mineral magnetic properties,
607 reducing concentrations in a granitic soil and increasing concentrations in a sedimentary soil. These
608 authors concluded that a tributary or confluence tracing approach should mitigate the impact of these
609 processes on mineral magnetic properties. Although these studies examined the disaggregation of
610 sediments, Droppo et al. (2005) suggested that the actual nature of aggregates and flocs is rarely
611 considered in studies of sediment transport and sources.

612



613 **Figure 9:** Scatter plots of the relationship between the Al/Si ratio and D_{90} , K, and Cs (ppm) from samples
 614 from different tributaries of the Amazon catchment with data from Tables 1 and 2 from Bouchez et al.,
 615 (2011).

616

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

620 different than those identified by particle size analysis (Gregorich et al., 2006). Different fractionation
621 methods (e.g. wet sieving, dry sieving, grinding) may fragment aggregates which could result in different
622 biogeochemical properties in analyzed aliquots. This may be particularly true with respect to different
623 techniques and conditions for wet and dry sieving, and the use of Stokes' law and settling columns to
624 isolate different particle size fractions. Different source and sediment material (e.g. platy, spherical, and
625 rod-shaped grains) may respond differently to fractionation methods (Droppo et al., 2005; Hatfield,
626 2014). Different pre-analysis treatments (e.g. sonification, chemical dispersion, physical dispersion) may
627 also impact the biogeochemical fingerprints of interest. Laboratory determined grain size distributions
628 will most likely differ significantly from those in the field owing to the physical and often chemical
629 disaggregation of flocs and aggregates during the laboratory fractionation process (Phillips and Walling,
630 1995; Walling and Collins, 2016).

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

641 Density separation is an intriguing fractionation approach that has not received much attention in the
642 sediment source fingerprinting literature (Hardy et al., 2010). Different clay minerals have different
643 densities that may impact their separation with settling columns in the finer fractions. Furthermore, iron
644 oxides and mineral magnetic properties have been strongly related to density (Hatfield, 2014) and there
645 may be micro aggregates or sediment flocs transported in suspension with the silt or very fine sand
646 fraction because they are lighter. Importantly, these micro-aggregates and sediment flocs will have a
647 high capacity to transport contaminants; however they will have a different resistance (i.e. a lower
648 density) relative to absolute particles of a similar size, predominantly the silt fraction (Droppo et al.,
649 1998).

650 Although density separation is time consuming and expensive, it may be more relevant to fluvial
651 processes compared to sieving the <63 μm fraction or settling the <10 μm fraction. Accordingly, it would
652 be opportunistic to compare density and particle size separation techniques to understand their impacts
653 on sediment fingerprinting modelling results. As researchers apply tracing techniques directly to quantify
654 the relative source contributions of carbon and nitrogen (Cooper et al., 2015b; Garzon-Garcia et al.,
655 2017), and are using different carbon components to model sediment sources (Blake et al., 2012; Gibbs,
656 2008; Hancock and Revill, 2013; Reiffarth et al., 2016) the utility of density separation may become of
657 increasing importance for tracing different carbon sources in riverine systems.

658 **5.4 Particle Size as a Tracer Property**

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

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

674 **6. Conclusion**

675 Sediment source fingerprinting researchers have recently tended to avoid in-depth examinations of
676 fundamental topics such as the impact of organic matter on biogeochemical properties, which tracer
677 properties are non-conservative (e.g. soluble, reactive), and what is the impact of particle size on tracer
678 property predictability. One limitation of statistically-oriented approaches to sediment source
679 fingerprinting is that they do not strive to understand the logic of tracer selection, non-conservative

680 tracers, nor particle size impacts. This is particularly limiting, as addressing conservative behaviour is
681 fundamentally more complex than simply acknowledging that sediment samples plot within their source
682 range. Modelling, and the statistical selection of which tracer properties to model, represent only two
683 stages of the sediment source fingerprinting process; although these two steps, at times, appear to
684 dominate the recent literature. Rarely is particle size the focus of research, rather particle size is often
685 simply an acknowledged limitation that must be addressed somehow. After demonstrating that non-
686 conservative tracer properties affect the accuracy of sediment source fingerprinting modelling results,
687 Sherriff et al. (2015) appropriately emphasized that improved strategies to detect non-conservative
688 tracer properties should be a priority of sediment source fingerprinting research.

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

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

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