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

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Abstract:

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

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

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1. Introduction

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

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

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

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

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

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

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

2. Sediment Property Predictability

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

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

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

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

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

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

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

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

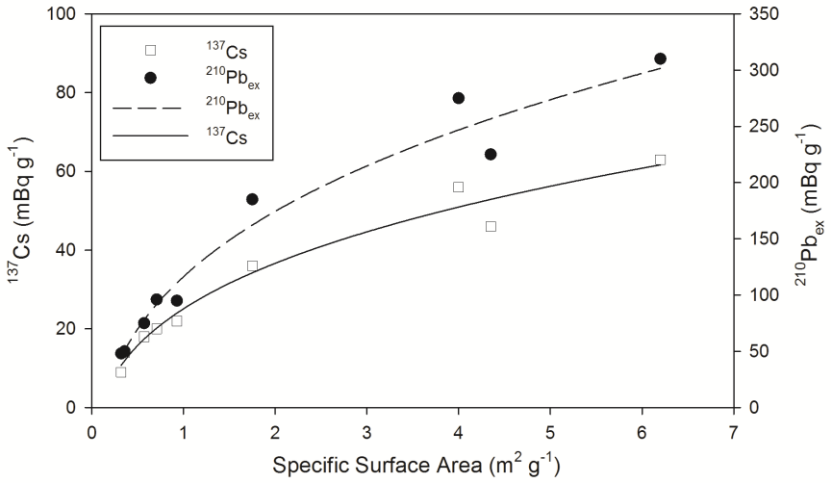

Figure 1: Relationship between specific surface area and fallout radionuclide activity concentrations (^{137}Cs and $^{210}Pb_{ex}$) (adaptation of Figure 1 from He and Walling (1996)).

2.2 Carbon and Nitrogen Parameters

Although they are not as extensively analyzed in sediment tracing research as fallout radionuclides, carbon and nitrogen parameters provide an interesting example into tracer property relationships with particle size. Total organic carbon (TOC) and total nitrogen (TN) often discriminate between sediment derived from surface and subsoil erosion processes as their concentration decreases with depth in the soil profile (Blake et al., 2006; Owens et al., 2006). Carbon stable isotopes ($\delta^{13}C$) may potentially discriminate between sediment derived from soils with C_3 vegetation (majority of tree or temperate grass species) compared to those covered with C_4 vegetation (grass and cropping species typically under warmer climates) (Fry, 2006; Schimel, 1993). Source discrimination with nitrogen stable isotopes ($\delta^{15}N$) is more complex. In general, soil $\delta^{15}N$ increases with depth in soil profiles (Amundson et al., 2003; Natelhoffer and Fry, 1988). Other factors such as nitrogen inputs from animal and human waste, along with fertilizers and potentially topographic position, may impact the predictability of $\delta^{15}N$ in a tracing context. Of note, these parameters trace organic matter (Garzon-Garcia et al., 2017; Olley, 2002) and therefore they may be fractionated by density as well as particle size. Furthermore, these parameters also have the potential for non-conservative behavior due 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 $\delta^{15}N$ and $\delta^{13}C$ (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 $\delta^{15}N$, with very limited $\delta^{13}C$ 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 $\delta^{13}C$, with increasing, though not significant, enrichment for $\delta^{15}N$
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. $\delta^{15}N$ 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. $\delta^{13}C$ 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 $\delta^{15}N$ 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)).

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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	$\delta^{13}C$, $\delta^{15}N$, C/N	<53	--	Dispersed	Yes
Garzon-Garcia et al., 2017	Australia	2.5, 75, 3076	$\delta^{13}C$, $\delta^{15}N$, TOC, TN	<10, <63, <500	--	--	Yes
Gibbs, 2008	New Zealand	117	CSSI	<1000	--	--	Yes
Hancock and Revill, 2013	Australia	3860	CSSI	<63	--	--	--
Laceby et al., 2015b	Australia	75, 123, 311	$\delta^{13}C$, $\delta^{15}N$, TOC, TN	<2, <10, <63, <2000	--	--	--
Laceby et al., 2016	Japan	77, 171, 265	$\delta^{13}C$, $\delta^{15}N$, TOC, TN	<63, <2000	--	--	Yes
Olley, 2002	Australia	84000	$\delta^{13}C$, C/N Ratio	<2	--	--	--
Papanicolaou et al., 2003	United States	600	$\delta^{13}C$, $\delta^{15}N$, C/N	--	--	--	--

[a] Source samples were dispersed with sodium hexametaphosphate

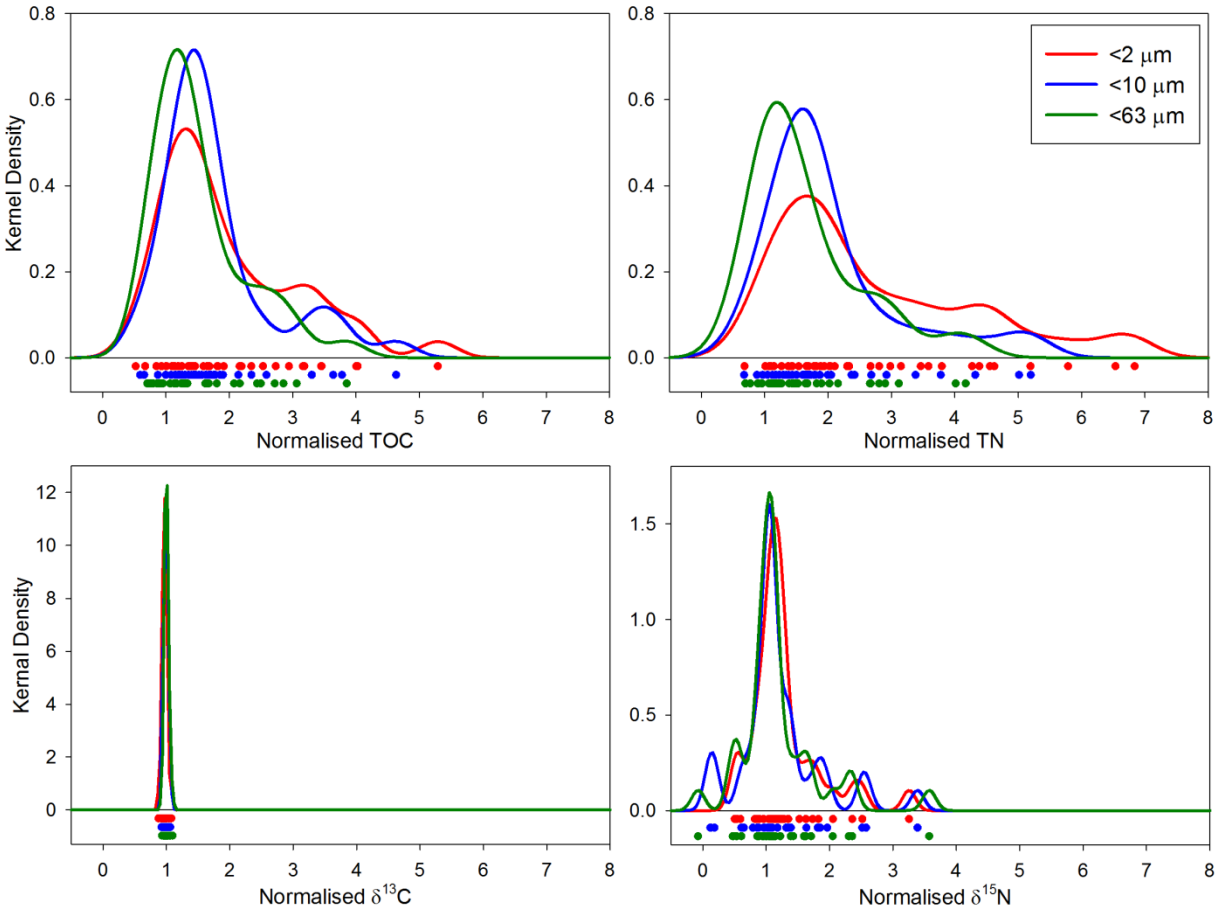

Figure 2: Normalized carbon (C) and nitrogen (N) property distributions for the <2 µm, <10 µm and <63 µm particle size fractions in samples from Knapp Creek and Blackfellow Creek, Australia (modified from Laceby et al., (2015b)). Points under the distributions are the normalized samples color coded to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

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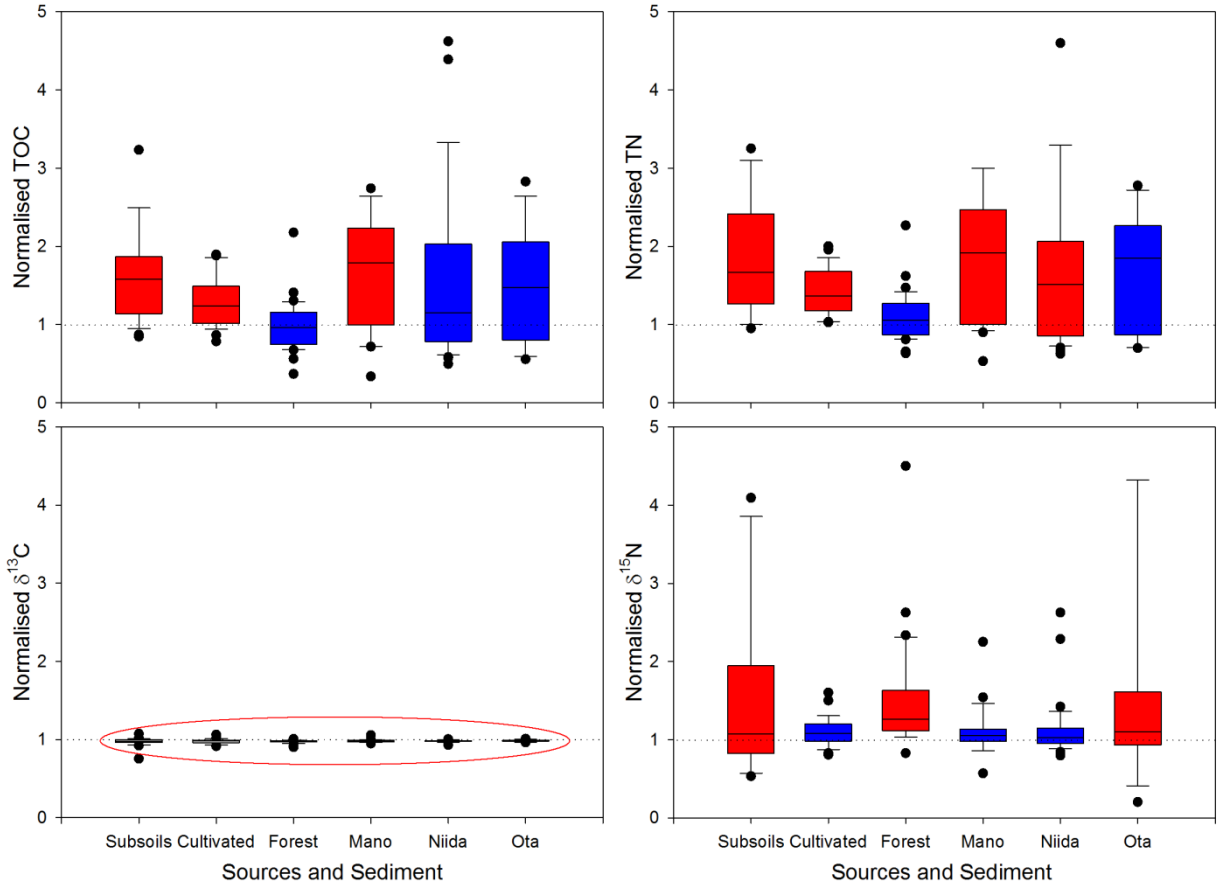

Figure 3: Box plots of the normalized difference between carbon (C) and nitrogen (N) sediment properties in the <63 µm fraction divided by the <2 mm fraction for the three sources and sediment from three Japanese catchments (i.e., Mano, Niida and Ota) with red shading (and the red circle) indicating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

2.3 Elemental Geochemistry

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

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

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

Underlying the potential influence of particle size on elemental geochemistry is the effect of sediment source mineralogy. The dominant mineralogy relates directly to particle size where some sources will be enriched in the fine particle size fractions and other sources may be depleted. Thus the geochemical fingerprint likely will change if the <2 mm, the <63 µm, or the <10 µm fraction are sampled and analysed. For example, each of the three different sources in Figure 4 from Gibbs (1967) may have different relationships between elemental concentrations and particle size, which will depend on the particle size range utilized for the sediment source fingerprinting research. Fundamentally, the impact of particle size on all elemental concentrations for each source is difficult to predict and the complexity of the particle size – elemental geochemistry relationship requires more research to comprehensively characterize the predictability of these fingerprints. Accordingly, section 4 of this review provides several 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	--	--
Laceby et al., 2015a	Australia	74	37	<10	--	--	--
Laceby 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	--	--	--

[a] Source samples sonified before sieving
[b] Samples were boiled in distilled water to disperse soil aggregates
[c] Samples sonified before settling in a water column
[d] Raw data is only available for sediment core samples

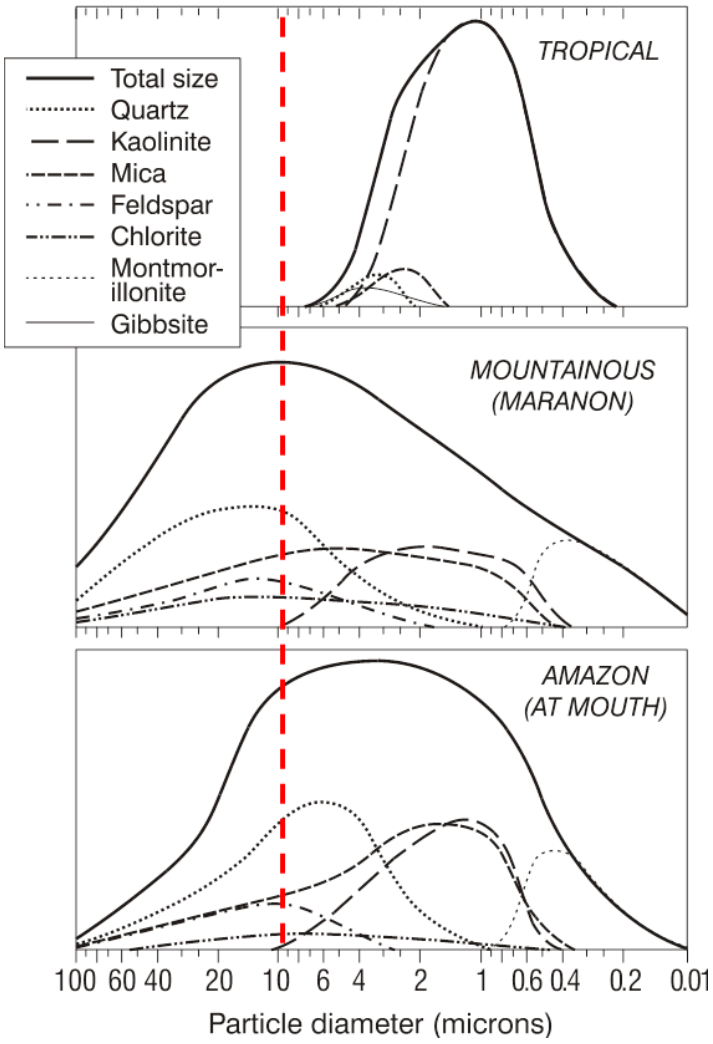

Figure 4: Changes in mineralogy with particle size in sediment from three locations in the Amazon basin from Gibbs (1967) with the red dashed line added to indicate the impact of fractionating the samples at the <10 µm particle size.

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2.4 Mineral Magnetic Properties

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

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

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

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 Na_2CO_3 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

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

2.5 Composite Fingerprinting Approach

Peart and Walling (1986) advocated for multiple parameters to be used when quantifying sediment source dynamics in order to improve the overall consistency and reliability of source ascription results. The combination of multiple parameters creates a composite fingerprint (Walling et al., 1993) that allows for an increased number of sources to be modelled and is theorized to be more representative of the linkages between sediments and their sources, potentially reducing false matches which were hypothesized to potentially occur with individual tracer properties (Collins et al., 1996). Accordingly, a composite fingerprinting approach has been broadly applied in sediment source fingerprinting research combining several or all of the following: fallout radionuclides, carbon and nitrogen parameters, element geochemistry, mineral magnetics and other parameters, thereby providing significant source discrimination (Collins et al., 1996; Evrard et al., 2013; Navratil et al., 2012; Owens et al., 2000) (Table 5). One challenge with the composite fingerprinting approach is that each of the potential complexities within the particle size – tracer parameter relationship described above is integrated into the expanded composite fingerprint. A second challenge is that it is difficult, if not impossible, to link outputs of statistical-based approaches (e.g. composite fingerprinting) back to a process-based understanding of 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, $\delta^{13}C$	<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, $\delta^{15}N$,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

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3. Main Approaches to Address Particle Size

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

3.1 Fractionation

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

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

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

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

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

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

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

3.2 Particle Size Corrections

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

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

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

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

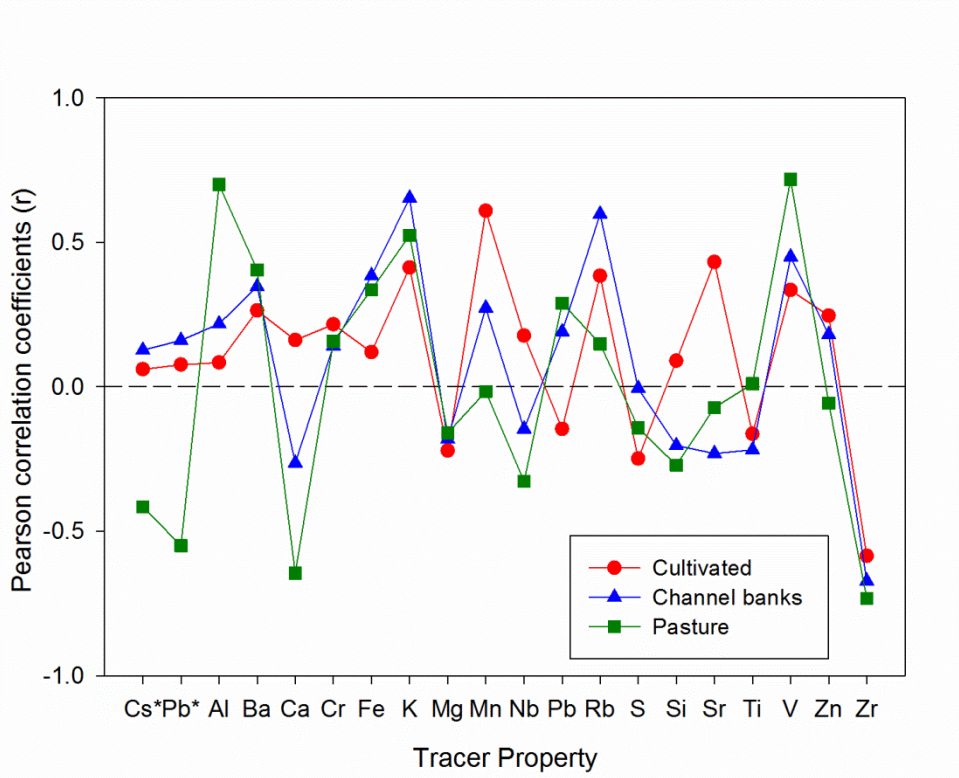

Figure 5: Pearson correlation coefficients (r) between Specific Surface Area (SSA) and tracer property for three sources from Smith and Blake (2014) (data from Table 1) with the * indicating fallout radionuclides (^{137}Cs and $^{210}Pb_{ex}$) on the x axis and the remainder being elements.

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

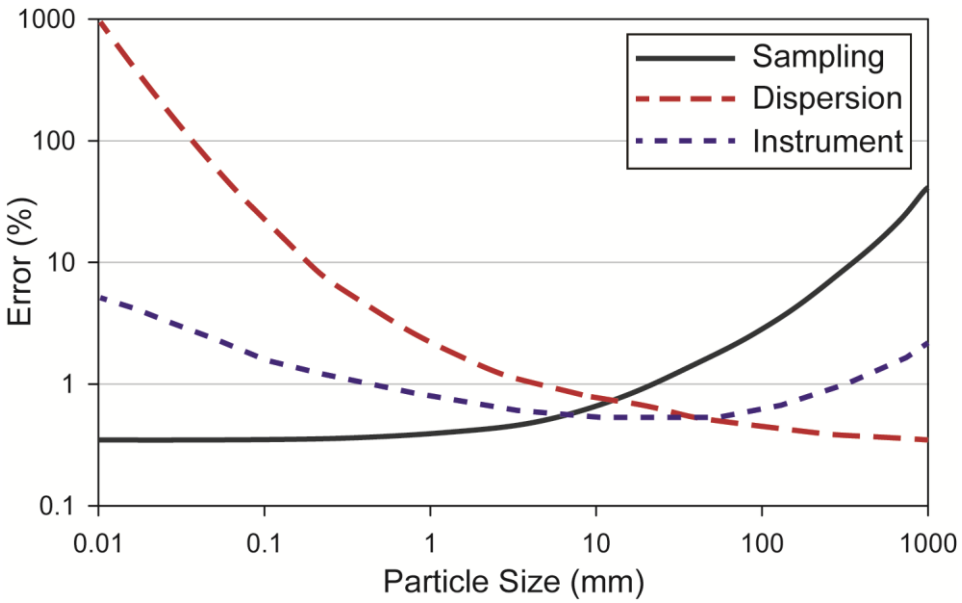

Figure 6: Relationship between potential sources of error and particle size distribution, adapted from Merkus (2009).

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.

4. Alternative Techniques to Address Particle Size

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

4.1 Tributary (or Confluence) Tracing

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

There may be potential particle size enrichment or depletion impacts on fingerprint properties that may occur during transportation and settling processes, although the most significant particle size enrichment typically occurs during the initial stages of mobilization and transportation that often occurs on hillslopes and in ephemeral systems (Stone and Walling, 1997). As material moves into the riverine system, sediment particle size often becomes increasingly uniform and thus fewer differences may be anticipated. Of course, there are always exceptions. For example, Koiter et al. (2015) used a recirculating flume to demonstrate that it was not only distance travelled that impacted particle size, but also channel bed characteristics such as roughness, porosity and inter-gravel flow. Therefore, the tributary tracing technique may have a limited ability to mitigate particle size impacts where tributaries have distinctly different channel bed characteristics and/or where there is a considerable distance between tributary 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	• 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	• 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	A) Fractionation • 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 B) Corrections • 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 C) Modelling • 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	• 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	• Discuss the impact of particle size in relation to other sediment source fingerprinting studies
7. Supplementary Information	• 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).

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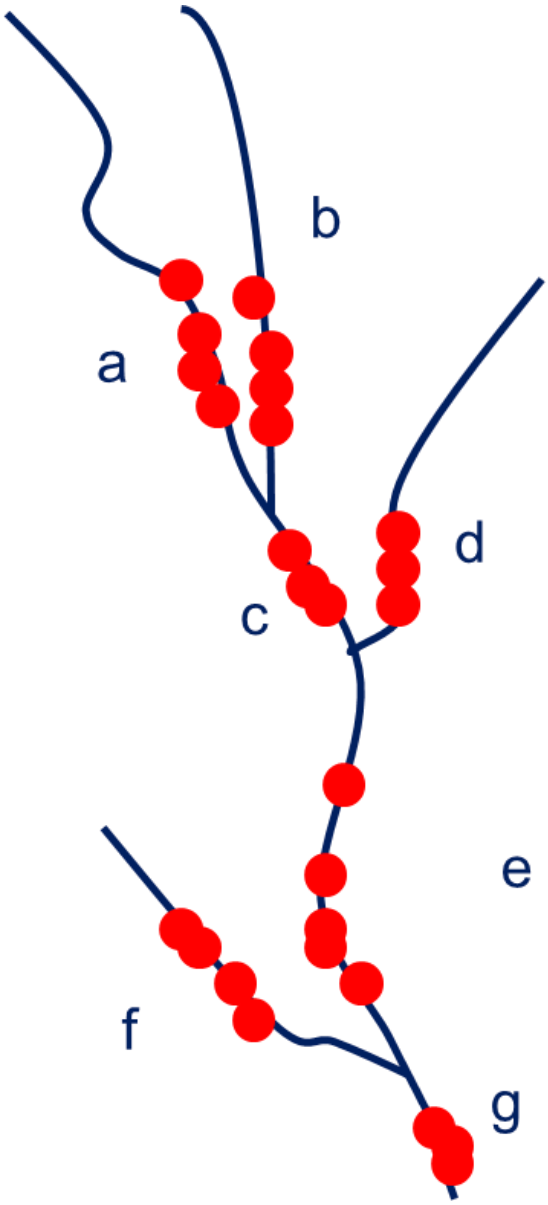

Figure 7: An example of a sampling design with the tributary tracing technique where the letters indicate the different tributaries that could be sampled in this theoretical catchment. In this situation, sediment collected at sites c, e and g is compared to sediment collected where it may be able to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

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

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

4.2 Edge-of-Field Samplers

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

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

4.3 Multiple Fraction Tracing

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

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

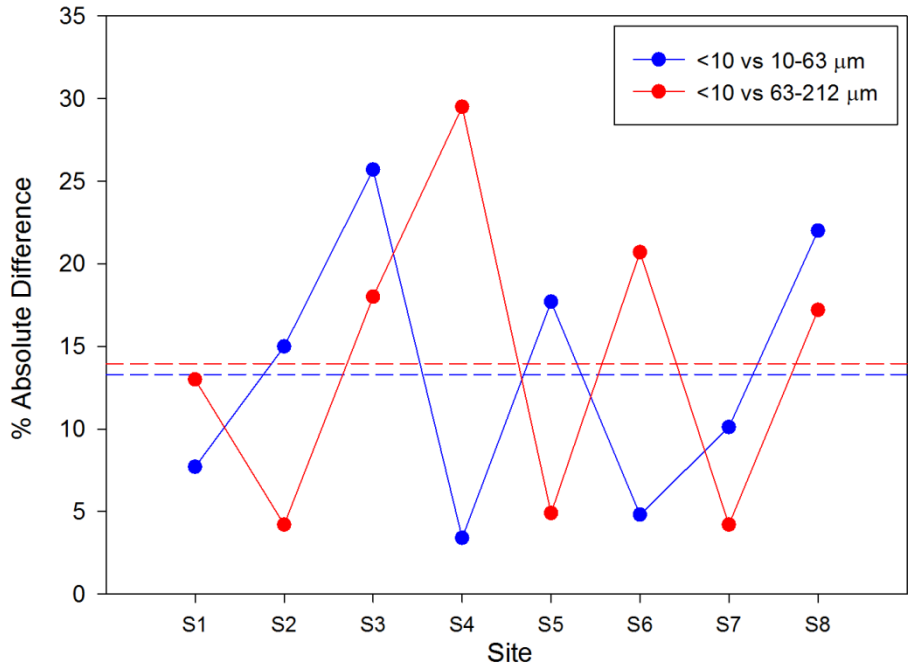


Figure 8: Mean absolute differences (dashed lines) in the source contributions between the <10 µm and the 10 - 63 µm (blue) and between the <10 µm and the 63 - 212 µm fractions (red) for eight sites in Australia plotted with data from Figure 9 in Haddadchi et al. (2015).

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, $\delta^{13}C$, and $\delta^{15}N$ 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

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

5. Particle Size Opportunities

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

5.1 Mineralogy, Particle Size and Elemental Geochemistry

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

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

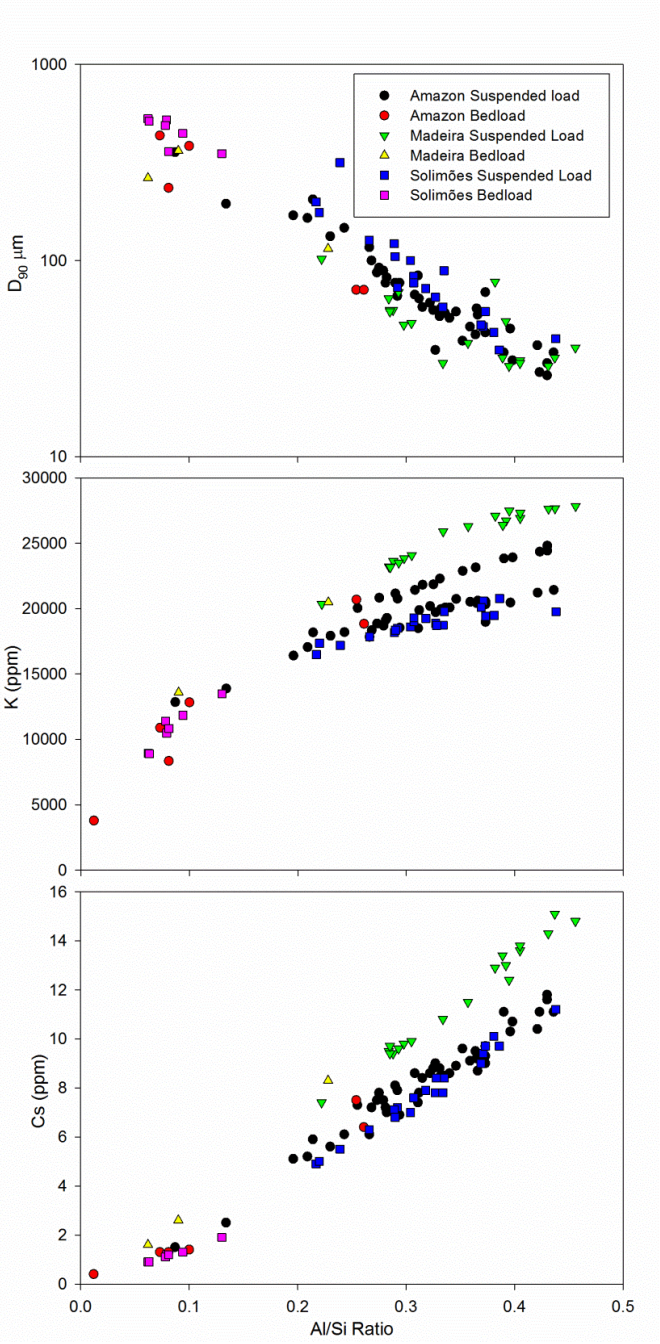
572 Different elemental and lithogenic radionuclide ratios have been used previously to address particle size
573 and density related enrichment in sediment source fingerprinting research. For example, Olley and Murray
574 (1994) demonstrated that although the concentrations of thorium (Th) isotopes varied with particle size,
575 the $^{230}Th/^{232}Th$ 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

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

5.3 Particle Dynamics

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

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



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

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

5.4 Particle Size as a Tracer Property

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

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

6. Conclusion

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

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

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

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

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