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Sediment Fingerprinting in Fluvial Systems: Review of Tracers, Sediment Sources and Mixing Models

Arman Haddadchi^{1*}, Darren S. Ryder², Olivier Evrard³, Jon Olley⁴

Abstract

Suspended sediments in fluvial systems originate from a myriad of diffuse and point sources, with the relative contribution from each source varying over time and space. The process of sediment fingerprinting focuses on developing methods that enable discrete sediment sources to be identified from a composite sample of suspended material. This review identifies existing methodological steps for sediment fingerprinting including fluvial and source sampling, and critically compares biogeochemical and physical tracers used in fingerprinting studies. Implications of applying different mixing models to the same source data are explored using data from 41 catchments across Europe, Africa, Australia, Asia, and North and South America. The application of seven commonly used mixing models to two case studies from the US (North Fork Broad River watershed) and France (Bléone watershed) with local and global (genetic algorithm) optimization methods identified all outputs remained in the acceptable range of error defined by the original authors. We propose future sediment fingerprinting studies use models that combine the best explanatory parameters provided by the modified Collins (using correction factors) and Hughes (relying on iterations involving all

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1 Introduction

The transport of sediment, and especially the fine sediment particles, can lead to a number of detrimental impacts for stream environments. Suspended sediment loads can lead to a decrease in water quality (Lartiges et al. 2001; Papanicolaou et al. 2003); a reduction of operational capacities in water supply facilities (Morris et al. 1997); an alteration of channel morphology (Wright et al. 1987); an increase in turbidity, restricting light penetration and thereby reducing primary production (Wood et al. 1997); and the smothering of biotic habitats (Richards et al. 1994). Furthermore, fine sediment export may facilitate substantial transfers of carbon and nutrients (Prosser et al. 2001).

Suspended sediments originate from different sources, with the relative contribution from each source varying over time and space as a consequence of different erosional processes. Although several approaches to identify sediment sources exist, many approaches rely on visual estimates (Reid et al. 1996), modeling (Foster 1988), long-term field records (Gellis et al. 2005), or traditional monitoring techniques. The latter employs an indirect approach and involves measurements of erosion activity, including those based on erosion pins to measure the rates of surface lowering (Slattery et al. 1995; Lawler et al. 1999); and erosion plots to document the rates of soil loss from surface sources (Motha et al. 2002). Indirect approaches also face many issues including: a) primary assumptions about the origin of sediment sources, b) difficulty in recording erosion rates due to the spatial variability, and c) inability of these approaches to estimate sediment delivery to the streams (Walling 2005). A thorough review of the direct and indirect approaches to measure sediment mobilization can be found in Collins and Walling (2004). Sediment fingerprinting methods provide a direct approach for quantifying sources of sediment from individual river sections to watershed scale. The procedure involves characterizing the

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4 potential sediment sources by their diagnostic chemical and physical properties, and comparing
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6 these to the properties of transported fluvial material.
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10 Figure 1 identifies the process common to the majority of sediment fingerprinting studies, even
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12 though the methods used for collecting samples (fluvial and source samples), preliminary
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14 analyses, number and type of tracers, statistical parameters to verify different tracers, and models
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16 to determine specific contribution from discrete sources may vary among techniques.
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20 **(Figure 1.)**

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23 This paper builds on reviews of sediment fingerprinting studies from (Collins et al. 2004),
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25 Walling (2005) and Davis et al. (2009) by focusing on: 1) comparison of different fluvial
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27 sampling methods used in sediment tracing studies and their applicability for different
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29 hydrologic and morphologic river conditions; 2) describing the range of sediment properties used
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31 to assign a fingerprint and the potential to quantitatively identify discrete sources of sediment; 3)
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33 comparing the sources of suspended sediment from 41 watersheds around the globe; and 4)
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35 comparing the variability in output from applying a common dataset from two case studies to
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37 seven commonly used mixing models. This is the first study that compares the most prevalent
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39 mixing models (including the application of genetic algorithms) to an actual dataset to quantify
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41 variability in the output depending on the choice of mixing models.
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49 **2 Fluvial and source soil sampling**

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51 Sediment fingerprinting studies rely on the collection of different types of fluvial sediments and
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53 may include river bed sediment (Olley et al. 2000; Dirszowsky 2004; Hughes et al. 2009; Evrard
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55 et al. 2011), dam reservoir samples (Foster et al. 2007; Nosrati et al. 2011), floodplain surface
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57 (Collins et al. 2010) and, most commonly, samples of suspended load (Mizugaki et al. 2008;
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Devereux et al. 2010; Mukundan et al. 2010). In some studies, soil samples were collected from spatially explicit watershed sources: from the top 0.5 cm (Gellis et al. 2009), 2 cm (Walling et al. 1995; Hughes et al. 2009; Collins et al. 2010) or 5 cm (Gruszowski et al. 2003; Minella et al. 2004; Devereux et al. 2010) of the soil surface. Instead of collecting samples from different source types, Motha et al. (2002) and Mizugaki et al. (2008) used a plot for each source type to simulate erosion process inside the plots, and Olley and Caitcheon (2000) used deposited fine-grained sediments as source samples to average out local source area heterogeneity. In a recent study Wilkinson et al. (In press) found that the estimated contributions of spatial source areas within the large study catchments had narrower confidence intervals when source areas were defined using sediment from geologically distinct river tributaries, rather than using soil sampled from geological units in the catchment, since tributary sediment had less-variable geochemistry than catchment soils.

Three primary methods used to collect suspended in-stream sediment samples across watersheds include point samples, time-integrated samples and automated collection of water samples. Based on the type of instruments used, point sampling consists of two approaches; collecting hundreds of liters of stream water and extracting suspended sediment with a continuous flow centrifuge (e.g. Motha et al. 2003; Devereux et al. 2009); and in-situ dewatering techniques using portable centrifuge or filtration systems (e.g., Horowitz et al. 1989). The advantage of the former technique is that it prevents contamination by the successive samples collected.

Time-integrated samplers based on a flow velocity reduction leading to the settling of particles within a trap (Phillips et al. 2000) have been widely adopted in sediment tracing research (Walling et al. 2008; Hatfield et al. 2009; Collins et al. 2010). These collect samples of suspended sediment during flow events, and effectively trap a representative sample of sediment

with an effective particle size of $<63\mu\text{m}$ (Phillips et al. 2000; Russell et al. 2001); they sample through the hydrograph including the rising and falling limbs. Automated water samplers are the more costly method but allow the collection of instantaneous samples, and therefore a better temporal resolution for characterizing suspended sediment flux. Comparisons among sampling strategies are outlined in Table 1, identifying the only two methods that provide data necessary to calculate hysteresis effects are time-integrated and automatic water samplers. Hysteresis impacts on the variation of suspended sediment loading in the falling and rising limb of an event (Williams 1989). Samples from time-integrated and automated water samplers can be representative of the whole watershed area because of their temporal integration of transported sediment during events, but require a longer period of time (>10 days) to collect samples. Point samplers have the benefit of quantifying the effect of discharge on sediment contribution from different sources.

Table 1. Comparing different type of fluvial sampling methods

	Determine Hysteresis effect	Representative sample of whole watershed	Enough quantity of sample	Long sampling time	Instantaneous effect of flood events
Point samples	×	×	×	×	✓
Time-Integrated samples	✓ ^{**}	✓	✓	✓	×
Automated water samples	✓ ^{**}	✓	✓	×	×
Bed load and Flood plain	×	✓	✓	×	×

samples					
Reservoir samples	×	×	✓	×	×

*in in-situ dewatering techniques enough quantity of samples can be collected

**These samplers partially alleviate the hysteresis problem but trapping efficiency of the samplers might also change during the hydrograph, the effect of which has not been quantified.

3 Fingerprint properties (Tracers)

A variety of chemical and physical tracer techniques have been used to investigate the sources of sediment and nutrients to river systems. These tracing techniques all involve measuring of one or more parameters that provide a 'fingerprint' to distinguish one source of sediment from another.

For a parameter to be useful in tracing the source of sediment it needs to be both measurable and conservative such that:

- A tracer signal should be able to distinguish between sediments derived from different source areas;
- For a given source of sediment, which does not change with respect to time, a sediment tracer signal must also be constant in time or vary in a predictable way;
- For a given source of sediment, which does not change with respect to distance along a transport path, a sediment tracer signal must also be constant along this path or vary in a predictable way.

Tracers used in sediment fingerprinting studies include sediment color (Grimshaw et al. 1980), color properties (Martínez-Carreras et al. 2010), plant pollen content (Brown 1985), major and trace elemental composition (Jenns et al. 2002; Miller et al. 2005), rare earth elements (Zhang et al. 2008), mineral magnetic characteristics (Hatfield et al. 2009), clay mineralogy (Motha et al. 2003), radionuclide characteristics (Vanden Bygaart et al. 2001; Estrany et al. 2010), organic

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4 matter content (Peart 1993; Walling et al. 1999), carbon and nitrogen stable isotope ratios
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6 (Papanicolaou et al. 2003; Rhoton et al. 2008), Compound Specific Stable Isotope (CSSI)
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8 analysis (Blake et al. 2012) and Diffuse Reflectance Infrared Fourier Transform Spectroscopy
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10 (DRIFTS) (Poulenard et al. 2009; Evrard et al. 2012).
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14 An advantage of physical tracers including color, density and fine sediment dimensions is they
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16 are readily identifiable and easily measurable characteristics (Davis et al. 2009). However, these
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18 tracers can be non-conservative and may change during transport. Grimshaw and Lewin (1980)
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20 and Peart (1993) successfully determined sediment origin using only color as a tracer, whereas
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22 Vanden et al. (2001) unsuccessfully used density as the sole tracer of sediment source due to
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24 large spatial variation in density values. More recently, Krein et al. (2003) demonstrated that
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26 fractal dimension and particle color can provide a fast and easy approach to determine the origin
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28 of sediments and the amount, location and process of sediment storage. Inorganic tracers have
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30 been less successful for attributing specific soil-environmental processes than organic tracers
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32 because of the large number of potential inorganic tracers and processes that may influence the
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34 elemental composition of sediments during transport (Davis et al. 2009).
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41 Sediment geochemistry has been widely used to identify the spatial sources of sediments
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43 delivered to waterways (Olley et al. 2000; Hardy et al. 2010; Weltje et al. 2011). Rock types,
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45 through soil formation and weathering, have a profound influence on the geochemical properties
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47 of their soils and accordingly the geochemical characteristics of their eroded sediments (Klages
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49 et al. 1975; Olley et al. 2001). Different underlying parent rock materials frequently results in
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51 spatial sources with distinct geochemical compositions (Olley et al. 2001; Motha et al. 2002;
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53 Douglas et al. 2009). Sediments eroded from soils derived from a particular rock type often
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55 maintain these distinct geochemical properties during sediment generation and transport
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processes (Hughes et al. 2009). If sediments generated from parental rock types have distinguishable major or trace elemental compositions then sources of transported sediment can be determined (Collins et al. 1996; Collins et al. 1998; Caitcheon et al. 2001) by characterizing and comparing the signature of suspended sediment samples and samples from the source areas (Hughes et al. 2009).

A number of inorganic tracers including rare earth elements (Ce, Eu, La, Lu, Sm, Tb, Yb), trace elements (As, Ba, Co, Cr, Cs, Hf, Sc, Ta, Th, Zn, Ag, Ba, Cd, Cu, Mn, Ni, Pb, Sb, Se, Tl, V), major elements (Fe, K, Na, Al, Ca, Mg, Ti, CaO, Na₂O, K₂O, Al₂O₃, Fe₂O₃, P₂O₅, MgO, SiO₂, TiO₂, Mn₂O₄), total inorganic carbon, nitrogen, phosphorus, and a number of organic tracers including total organic carbon, nitrogen, phosphorus and Loss on Ignition have been applied in sediment fingerprinting studies. Major elements, particularly the relationship between Fe₂O₃ and Al₂O₃, provide useful tracers for discriminating soils with different rock forming minerals (Dyer et al. 1996). The Chemical Index of Alteration (CIA) as proposed by (McLennan 1993) is a useful tracer to identify chemical variations resulting from weathering.

Fallout radionuclide activities are commonly high in surface materials and low or non-existent in subsurface materials (Walling 2005; Caitcheon et al. 2012; Olley et al. 2012), making them useful in distinguish surface and subsurface materials. Furthermore, they frequently distinguish cultivated from uncultivated soils as radionuclides are generally mixed throughout the ploughed layer. In addition, radionuclide tracers are well-suited for use in heterogeneous watersheds since their concentrations are effectively independent of soil type and underlying geology (Walling 2005; Caitcheon et al. 2012; Olley et al. 2012). The most commonly used fallout radionuclides are ¹³⁷Cs, ²¹⁰Pb and ⁷Be.

¹³⁷Cs, which has a half-life of 30.2 yr, is a product of nuclear weapons testing during the 1950s and the 1960s (Loughran et al. 1995) and nuclear accidents (e.g., Chernobyl with significant fallout in Europe; Fukushima with significant fallout in Japan). Global fallout of ¹³⁷Cs peaked in the early 1960s and reached zero in the mid-1980s. The highest concentrations of ¹³⁷Cs are found in undisturbed areas such as forests or where soils were translocated from undisturbed areas and not diluted (Matissoff et al. 2002; Nagle et al. 2004).

Lead-210 (²¹⁰Pb) is a product of atmospheric decay of ²²²Rn gas (fallout ²¹⁰Pb) and in situ decay of ²²⁶Ra, and has a half-life of 22.26 years (Wallbrink et al. 1996). Fallout ²¹⁰Pb in a soil or sediment sample is the excess of ²¹⁰Pb activity over the ²²⁶Ra supported component. This is known as ‘unsupported’ or ‘excess’ ²¹⁰Pb (²¹⁰Pb_{ex}). Like ¹³⁷Cs, ²¹⁰Pb_{ex} generally accumulates in the top 10 cm of soil, but can differ with depth depending on local environmental factors. In addition to fallout radionuclides, Radium-226 (²²⁶Ra) is produced by in situ decay of the uranium series. ²²⁶Ra concentrations are more directly related to rock type (Walling et al. 1995), and can be used as a geogenic radionuclide tracer.

Beryllium-7 is cosmogenic in origin through the spallation of nitrogen and oxygen atoms in the upper atmosphere by cosmic rays. Beryllium-7 (⁷Be) is useful to discriminate surface soils from deeper layers as it is commonly concentrated in the upper 5 mm of the soil profile (Zapata 2003). Unlike ²¹⁰Pb and ¹³⁷Cs, ⁷Be can confirm the relative importance of recently mobilized surface materials due to its very short half-life of 53 days.

Nitrogen and carbon stable isotopes have shown greater potential sensitivity for detecting sediment sources than total elemental composition, and therefore a powerful tool for identifying soil origin (Fox et al. 2008). The stable isotopic signature of nitrogen ($\delta^{15}\text{N}$) is a soil property

proportional to the $^{15}\text{N}/^{14}\text{N}$ isotopic ratio; similarly the carbon stable isotopic signature ($\delta^{13}\text{C}$) is proportional to the $^{13}\text{C}/^{12}\text{C}$ isotopic ratio. The carbon to nitrogen atomic ratio C/N is the ratio of total atomic carbon to nitrogen. The dependence of $\delta^{15}\text{N}$, $\delta^{13}\text{C}$, and C/N on vegetative cover and management, support the argument that the biogeochemical signature of eroded-soil will reflect specific erosion processes (Fox et al. 2007).

The mineral magnetic properties of soils that are related to the underlying geology and soil type include low- and high- frequency magnetic susceptibility (χ_{lf} , χ_{hf}), frequency depended susceptibility (χ_{fd}) anhysteretic remanent magnetization (ARM), isothermal remanent magnetization (IRM), high-field remanent magnetization (HIRM), and saturated isothermal remanent magnetization (SIRM). The advantages of using magnetic tracers to determine discrete sediment sources are: a) the measurement methods are not time- and cost-intensive, b) their potential to discriminate a sample using non-destructive techniques, and c) their high sensitivity to subtle changes in a range of environmental settings (Maher 1998). The disadvantages of magnetic properties is that they are highly particle size-dependent (Hatfield et al. 2009) and are not linearly additive (Lees 1997).

4 Sources of sediment

The development of fingerprinting techniques has enabled discrimination of diverse point and diffuse sources of sediment, including forest roads (Madej 2001; Gruszowski et al. 2003; Minella et al. 2008), graveled roads (Motha et al. 2004), arable lands (Walling et al. 1999; Walling et al. 2001), pasture lands (He et al. 1995; Collins et al. 1997a; Owens et al. 2000), forest floor (Mizugaki et al. 2008), sub-surface areas (Russell et al. 2001; Walling et al. 2008), channel banks (Slattery et al. 2000), landslides (Nelson et al. 2002), gully walls (Krause et al. 2003) and urban sources (Carter et al. 2003).

Pastured lands (grassland topsoils) have been documented as one of the highest contributors to suspended sediment transport in UK (He et al. 1995; Collins et al. 1997a; Owens et al. 2000; Gruszowski et al. 2003; Collins et al. 2010) due to soil deformation and compaction as a result of high livestock densities (Pietola et al. 2005). However, studies in France (Evrard et al. 2011), Australia (Motha et al. 2002) and Iran (Nosrati et al. 2011) show low soil erosion potential from pasturelands as a result of higher vegetative cover that retards both sediment detachment and transport. Site-specific issues such as unvegetated surfaces during high precipitation, increased slope, and reduced soil organic matter content can accelerate erosion processes from cultivated fields.

The importance of roads as sites of sediment origin, deposition and transport has been widely acknowledged (Wemple et al. 2001; Ramos-Scharrón et al. 2007; Sheridan et al. 2008), and their contribution to sediment loads exacerbated by their connectivity within drainage systems (Croke et al. 2001; Motha et al. 2004). A range of sediment tracers have been used to successfully discriminate different types of roads as sediment sources including forest roads (Motha et al. 2002; Mizugaki et al. 2008), street residue (Devereux et al. 2010), farm tracks (Edwards et al. 2008; Collins et al. 2010), unpaved roads or unmetalled roads (Collins et al. 2010; Mukundan et al. 2010) and paved roads or metalled roads (Gruszowski et al. 2003).

The relative importance of channel banks as sediment sources to drainage systems will vary among watersheds due to geology and sediment type, hydrology, channel morphology and dimensions, and riparian land-use pressures (Collins et al. 2010). In south-eastern Australian, channel sources have been documented to contribute up to 90% of the total sediment yield (Olley et al. 1993; Wallbrink et al. 1998; Wasson et al. 1998; Caitcheon et al. 2012; Olley et al. 2012). In the UK, Walling (2005) suggested channel banks typically contributed 50% of transported

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4 sediment load. In contrast, channel bank sources to suspended load have also been found to be
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6 minimal (e.g. Chapman et al. 2001; Russell et al. 2001; Walling et al. 2001), highlighting the
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8 importance of local conditions in regulating channel bank contributions.
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11 A number of fingerprinting studies have developed methods to successfully discriminate
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13 geological sources of sediment rather than sources originating from different land-uses. For
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15 example, Walling and Woodward (1995) categorized the River Calm watershed (UK) into three
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17 dominant rock types including; Cretaceous/Eocene with 20% contribution, Triassic with 42%
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19 and Permian with 26.5%. In Australia, Olley and Caitcheon (2000) found sediments in the
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21 Darling- Barwon watershed were mostly derived from sedimentary and granitic bed rock areas
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23 and less (<5%) from basalt-derived component of cultivated areas, and Wilkinson et al. (2012)
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25 measured sediment source contribution from surface and sub-surface soils of Granitoid, Mafic
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27 and sedimentary rock in 5 river locations and concluded that most of the fine sediment loss in the
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29 study area was derived from subsurface soil sources. Similarly, Evrard et al. (2011), Poulenard et
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31 al. (2012) and Navratil et al. (2012) successfully compared the contribution of four geological
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33 sources to river bed sediment and suspended sediment respectively, within the Bléone watershed
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35 (France).
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44 To summarize the range of tracing techniques, their applicability and success in discriminating
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46 among sources, Table 2 presents data from twenty five published sediment fingerprinting studies
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48 covering 47 watersheds from Europe, Africa, Australia, Asia, and North and South America.
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Table 2. The range of tracing techniques, their applicability and success in discriminating among sources from twenty published sediment fingerprinting studies.

Study	Physical tracers	Organic	Inorganic	Radionuclide	Magnetic tracers	Best tracers	Description of location and sediment sources	Most contributed area (percent of contribution)
Walling et al. (1993)		C, N		^{137}Cs , ^{210}Pb	χ ARM, SIRM, IRM		Jackmoor Brook Basin (UK) six sources: two groups of pastures, three groups of cultivated areas, channel banks	Cultivated areas (57.5%), Pasture surfaces (23.6%), Channel banks (18.9%).
							River Dart Basin four sources: pasture, two groups of cultivated fields, channel banks	Pasture surfaces (48.2%), Cultivated areas (30.8%), Channel bank (21%),
Walling et al. (1995)		C, N		^{137}Cs , $^{210}\text{Pb}_{\text{ex}}$, ^{226}Ra	χ , ARM, SIRM, IRM		River Culm Basin (UK) seven source types: Cretaceus/Eocene pasture, Cretaceus/Eocene cultivated, Triassic pasture, Triassic cultivated, Permian pasture, Permian cultivated, and channel banks	Triassic cultivated (29.5 %), Permian cultivated (19.7), Channel banks (12%)
Slattery et al. (1995)					χ_{lf} , χ_{hf} SIRM, IRM		North Oxfordshire watershed (UK) three sources: Cultivated areas, channel banks, combined surficial soil/channel bank areas	Cultivated areas (38%), Channel banks (34%), combined surficial soil/channel bank areas (28%)
Collins 1997		C, N, P _{tot}	Fe _{pyr} , Fe _{dit} , Al _{pyr} , Al _{dit} , Mn _{pyr} , Fe _{tot} , Al _{tot} , Mn _{tot} , Fe _{oxa} , Mn _{oxa} , Al _{oxa} , Cu, Zn, Pb, Cr, Co, Ni, Na, Mg, Ca, K,	^{137}Cs		Ca, Co, Na, Fe _{dit} , Mn _{oxa} , Ni	The Exe Basin (UK) four sources: woodland, pasture areas, cultivated areas, channel banks	The Exe basin: Pasture areas (71.7%), Cultivated areas (20.4%), Channel banks (5.3%), Woodland (2.6%).
						Fe _{oxa} , Ca, C	The Severn Basin (UK) four sources: woodland, pasture areas, cultivated areas, channel banks	The Severn basin: Pasture areas (65.3%), Cultivated areas (25.4%), Channel banks (7.5%), Woodland (1.8%).
Collins 1997	Absolute particle size	C, N, P _{tot}	Fe _{pyr} , Fe _{dit} , Mn _{pyr} , Mn _{dit} , Al _{pyr} , Al _{dit} , Fe _{tot} , Mn _{tot} , Al _{tot} , Fe _{oxa} , Mn _{oxa} , Al _{oxa} , Cu, Zn, Pb, Cr, Co, Ni, Na, Mg, Ca, K	^{137}Cs , ^{210}pb		Ni, Co, K, P _{tot} , N	The Dart Basin (UK) four sources: woodland, pasture areas, cultivated areas, channel banks	Pasture areas (78%), Cultivated areas (14%), woodland (4.5%), channel banks (3.5%)
						N, Cu, ^{137}Cs	The Plynlimon Basin (Uk) three sources: forest areas, pasture areas, channel banks	Pasture areas (66%), Forest areas (25%), Channel banks (9%)

Study	Physical tracers	Organic	Inorganic	Radionuclide	Magnetic tracers	Best tracers	Description of location and sediment sources	Most contributed area (percent of contribution)
Wallbrink, Murray et al. 1998				^{137}Cs , $^{210}\text{Pb}_{\text{ex}}$		^{137}Cs , $^{210}\text{Pb}_{\text{ex}}$	Murrumbidgee River (Australia) uncultivated areas, cultivated areas, channel banks	Uncultivated areas (78%), Cultivated areas (22%)
Wallling et al. 1999		C, N, P, P_{tot}	Al, Ca, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, Pb, Sr, Zn, total P	^{137}Cs , $^{210}\text{Pb}_{\text{ex}}$, ^{226}Ra	χ , SIRM	N, Total P, Sr, Ni, Zn	Swale River (UK) four sources: woodland, uncultivated areas, cultivated areas, channel banks	Uncultivated areas (42%), Cultivated areas (30%), Channel banks (28%)
						^{226}Ra , ^{137}Cs , $^{210}\text{Pb}_{\text{ex}}$, Fe, Al	Ure River four sources: woodland, uncultivated areas, cultivated areas, channel banks	Uncultivated areas (45%), Channel banks (37%), Cultivated areas (17%)
							Nidd River four sources: woodland, uncultivated areas, cultivated areas, channel banks	Uncultivated areas (75%), Channel banks (15%)
							Ouse River four sources: woodland, uncultivated areas, cultivated areas, channel banks	Cultivated areas (38%), Channel banks (37%), Uncultivated areas (24.6%)
							Wharfe River four sources: woodland, uncultivated areas, cultivated areas, channel banks	Uncultivated areas (69.5%), Channel banks (22.5%)
Nicholls 2001		C, N	Al, Ca, Cr, Co, Cu, Fe, Pb, Mg, Mn, Ni, K, Sr, Na, Zn	^{137}Cs , $^{210}\text{Pb}_{\text{ex}}$, ^{226}Ra		^{226}Ra , Fe, Cr, C, ^{137}Cs , K, N	Upper Torridge watershed (UK) four sources: channel banks, cultivated area, pasture land, woodland	Pasture land (47%), Cultivated area (28%), Channel Banks (23%)
Russell et al. 2001		C, N	Al, Ca, Cr, Co, Cu, Fe, Pb, Mg, Mn, Ni, K, Sr, Na, Zn, As	^{137}Cs , $^{210}\text{Pb}_{\text{ex}}$, ^{226}Ra	χ_{lf} , χ_{fd} , ARM, SIRM, IRM	Land use: Al_p , Fe, Mg, Mn, ^{137}Cs , K, χ_{lf} , ARM, SIRM	Belmont watershed (UK) five sources: pasture areas, arable areas, hopyards, channel banks, field drains	Field drains (55.3%), Arable areas (17.5%), Hopyard (12%), Channel banks (11%)
						Soil type: Al_p , SIRM, ARM, ^{137}Cs , X_{lf} , Pb, Mg, K, Fe, Mn	Belmont watershed (UK) five sources: Bromyard, Middleton, Compton, channel banks, field drains	Field drains (54.5%), Bromyard (12.9%), Channel banks (11.9%), Middleton (11.8%)
						Land use: ^{137}Cs , As, N, ARM, SIRM, Pb, χ_{lf} , C	Jubilee watershed (UK) five sources: pasture areas, arable areas, hopyards, channel banks, field drains	Field drains (47.8%), Arable areas (30.1%), Channel banks (12%), Hopyards (7%)
						Soil type: K, Mg, As, Mn, ^{137}Cs , χ_{lf} , ARM, SIRM	Jubilee watershed (UK) four sources: Bromyard, Middleton, channel banks, field drains	Field drains (54.7%), Middleton (30.5%), Channel banks (11.1%)
Wallling et al. 2001		C, N	Al, As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sb, Sn, Sr, Zn, Ca, K Mg, Na, Al_{dit} , Fe_{dit} , Mn_{dit}	^{137}Cs , $^{210}\text{Pb}_{\text{ex}}$		Ni, K, Cu, Cr, Ca, Total of $\text{Al}_{\text{pyrophosphate}}$ and Al_{dit} , Mn_{dit} , Al_{dit}	Kaleya River Basin (Zambia) four sources: communal cultivation areas,	Cultivated areas (66%), Bush grazing areas (17%), Channel banks and

Study	Physical tracers	Organic	Inorganic	Radionuclide	Magnetic tracers	Best tracers	Description of location and sediment sources	Most contributed area (percent of contribution)
			Al _{pyr} , Fe _{pyr} , Mn _{pyr} , P _{tot}	²²⁶ Ra		Sr, ¹³⁷ Cs, Co, P _{tot}	commercial cultivation areas, channel banks and gullies, bush grazing areas	gullies (17%)
Gruszowski et al. (2003)			P, Fe, Al, Na, K, Mg, Ca, Cd, Cu, Ni, Mn, Zn	¹³⁷ Cs	χ _{lf} , χ _{hf} , χ _{fd} , χ _{fd%} , χ _{ARM} , S _{ratio} , ARM, IRM ₁₀₀ , IRM ₈₈₀ , HIRM	χ _{lf} , χ _{ARM} , IRM ₈₈₀ , Fe, Al, Na, Cu, ¹³⁷ Cs	River Leadon watershed (UK) five sources: arable areas, grassland areas, sub-soils, channel banks, road sources	Sub-soils (35%), Road sources (30%), Grassland topsoils (13.8%), Arable topsoils (13.6%), Channel banks (8%)
Motha et al. (2004)			Al ₂ O ₃ /Fe ₂ O ₃ , Al ₂ O ₃ /(100-SiO ₂), CIA	¹³⁷ Cs, ²¹⁰ Pb _{ex}	IRM ₈₅₀ /χ	Al ₂ O ₃ /Fe ₂ O ₃ , Al ₂ O ₃ /(100-SiO ₂), CIA, ¹³⁷ Cs, ²¹⁰ Pb _{ex}	East Tarago watershed (Australia) four sources: gravel-surfaced roads, grouped lands (un-graveled roads, pasture and cultivated lands on basalt-derived soils), cultivated lands on granite-derived soils, and forest	Gravel-surfaced roads (41%), Grouped lands (18%), Cultivated lands on granite-derived soils (13%) and Forest(14%)
Minella et al. (2004)		C _{tot}	N _{tot} , P _{tot} , K _{tot} , Ca _{tot} , Na _{tot} , Mg _{tot} , Cu _{tot} , Pb _{tot} , Cr _{tot} , Co _{tot} , Zn _{tot} , Ni _{tot} , Fe _{tot} , Mn _{tot} , Al _{tot} , Fe _{dit} , Fe _{oxa} , Mn _{dit} , Al _{dit} , Al _{oxa}			Fe _{tot} , Fe _{oxa} , Al _{oxa} , Mn _{tot} , Ca, P	Lajeado Ferreira River (Brazil) three sources: field areas, pasture areas, unpaved roads	Pasture areas (77.9%), Unpaved roads (21.3%)
Mizugaki et al. (2008)				¹³⁷ Cs, ²¹⁰ Pb _{ex}			Two watersheds of Tsuzura River (Japan): Hinoki 156 watershed four sources: forest floor, landslide scar, truck trail, channel bank; b) Hinoki 155 watershed two sources: forest floor, landslide.	Hinoki 156 watershed: Forest Floor (46%) Hinoki 155 watershed: Forest Floor (70%)
Bellis et al. (2009)		P, N, C/N, C _{tot} , δ ¹³ C, δ ¹⁵ N		²¹⁰ Pb _{ex}		N, Total C, δ ¹³ C, δ ¹⁵ N, ²¹⁰ Pb _{ex}	Pokomoke River (US) four sources: channel banks, ditch Bed, crop area, forest area	Ditch bed (62%), Crop area (20%), Stream and Ditch banks (14%)
		P, N, C/N, C _{tot} , δ ¹³ C, δ ¹⁵ N		²¹⁰ Pb _{ex}		Total C, C/N, δ ¹⁵ N, δ ¹³ C	Mattawoman Creek (US) four source: banks, construction sites, crop lands, forest area	Forest (34%), Banks (28%), Crop land (19%), Construction sites (19%)
		C, P, N, C/N, δ ¹³ C, δ ¹⁵ N		²¹⁰ Pb _{ex} , ¹³⁷ Cs		Organic C, δ ¹³ C, P	Little Connestoga Creek (US) three sources: channel banks, construction sites, crop land	Cultivated areas (61%), Channel banks (39%)
Mukundan et al. (2010)		C _{tot} , N _{tot} , P _{tot} , S _{tot}	Be, Mg, Al, K, Ca, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Pb, U	¹³⁷ Cs		¹³⁷ Cs, δ ¹⁵ N, Cr and U	North Fork Broad River (US) three sources: channel banks, construction sites and unpaved roads, pastures	Channel banks (60%), Construction sites and unpaved roads (23 to 30%), Pastures (10 to 15%)

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Study	Physical tracers	Organic	Inorganic	Radionuclide	Magnetic tracers	Best tracers	Description of location and sediment sources	Most contributed area (percent of contribution)					
(Collins et al. 2010)			Al, As, Ba, Bi, Cd, Ce, Co, Cr, Cs, Cu, Dy, Er, Fe, Ga, Gd, Ge, Hf, Ho, K, La, Li, Mg, Mn, Mo, Na, Nd, Ni, Pb, Pd, Pr, Rb, Sb, Sc, Sm, Sn, Sr, Tb, Ti, Tl, V, Y, Yb, Zn, Zr, P			South House Sub-catchment: Tb, P, Ge, Tl, Ga, Eu, Ba Little Puddle Sub-catchment: Tb, Ga, Ba, Ge, Mn, Sm, Bi. Briantspuddle: Tb, Pd, Y, Ge, FeGa, Ti, Hf, Mn, Cr, Li.	South House, Little Puddle, Briants Puddle sub-catchments (UK) four source: pasture areas, cultivated areas, farm tracks, channel banks		Pasture areas	Cultivated areas	Farm tracks	Channel banks	
								South House	46	7	1	46	
								Little Puddle	45	16	12	27	
								Briants puddle	44	6	10	40	
(Collins et al. 2010)			Al, As, Ba, Bi, Cd, Ce, Co, Cr, Cs, Cu, Dy, Er, Eu, Fe, Ga, Gd, Ge, Hf, Ho, In, K, La, Li, Mg, Mn, Mo, Na, Nd, Ni, Pb, Pd, Pr, Rb, Sb, Sc, Sm, Sn, Sr, Tb, Ti, Tl, U, V, Y, Yb, Zn, Zr			Brue : Sb, Ti, Fe, As, Mn, V, Ce, Ge Cary : Sb, Ti, Fe, Na, Bi, Zn, In, V, Y, Pd, Cr, Sr Halse Water: Sb, Ti, Cd, Pd, Yb, Co, As, K, Ba Isle : Sb, In, Ti, Fe, Na, Sn, Cu, Cr Tone: Sr, Tl, Sb, Hf, Ti, Ni, Pd, La, Sc, Al, Zr, Yb, Mg, Rb, Na, Sn Upper Parrett: Sb, Ti, Zn, Al, K, Sr, Mg Yeo: Sb, Ti, Na, Fe, Sn, Cu, Al, V, Bi, Co	River Brue, River Cary, River Halse, River Isle, River Tone, Upper Parrett River, Yeo River (UK) five sources: pasture areas, cultivated areas, channel banks/subsurface sources, road verge, sewage treatment works (STW)		Pasture areas	Cultivated	Channel banks	Road verges	STW
								Brue	67	21	10	1	1
								Cary	38	6	43	11	2
								Halse	29	57	12	11	1
								Isle	44	12	30	11	3
								Tone	51	13	22	13	1
								Parrett	60	17	18	3	2
								Yeo	10	30	29	29	2

Study	Physical tracers	Organic	Inorganic	Radionuclide	Magnetic tracers	Best tracers	Description of location and sediment sources	Most contributed area (percent of contribution)
Levereux et al. (2010)		C _{tot} , S _{tot}	SiO ₂ , Al ₂ O ₃ , Fe ₂ O ₃ , MgO, CaO, Na ₂ O, K ₂ O, TiO ₂ , P ₂ O ₅ , MnO, Cr ₂ O ₃ , Ni, Sc, Ba, Be, Co, Cs, Ga, Hf, Nb, Rb, Sn, Sr, Ta, Th, U, V, W, Zr, Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Mo, Cu, Pb, Zn, Ni, As, Cd, Sb, Bi, Ag, Au, Hg, Tl, Sc	¹³⁷ Cs, ⁴⁰ K		Ho, Sr, W	Northeast Branch Anacostia River watershed (US) three sources: channel banks, streets, upland areas	Channel banks (58%), Streets (13%), Upland areas (30%)
Rouhpeima et al. (2010)	Clay mineral; Smaktite, Colorite, Illite, Kaolinite	C,N,P	Na, Mg, Ca, K, Cr, Co		χ_{lf} , χ_{fd}	Amrovan watershed: C, P, Kaolinite, K. Royan watershed: Cholorite, χ_{fd} , N, C	Amrovan watershed (Iran) three geological formations: Quaternary, Hezardareh, Upper Red, and gully erosion	Upper red formation (36%), Hezar dareh formation (28%), Gully erosion (21%)
							Royan watershed five geological formations: Upper Red, Karaj, Lar, Shemshak, Quaternary, and gully erosion	Quaternary units (32%), Karaj formation (33%), Gully erosion (27%)
Nosrati et al. (2011)		C _{tot} , N _{tot}	Al, B, Ba, Bi, Ca, Cd, Co, Cr, Cu, Fe, Ga, K, Li, Mg, Mn, Mo, Na, Ni, P, Pb, Se, Sr, Te, Tl, Zn. Biochemical tracers: ureas, alkaline phosphatase, β -glucosidase, dehydrogenase			Dehydrogenase, B, Total C, Sr, Co, Tl	Hive watershed (Iran) three sources: rangeland areas, orchard areas, channel banks	Streambanks (70%), Pasture areas (19%), Orchard areas (11%)
Wilkinson, Hancock et al., (2012)		C _{tot}	Al, As, Ba, Ca, Cl, Co , Cr, Cu, Dy, Er, Eu, Fe, Ga , Gd , Ge , Hf , Ho ,K ,La ,Mn ,Mo , Na , Nd, Ni, P, Pb, Rb, Sc , Se, Si XRF 0.025 P P P Sm , S ,Sr , Tb , Th , Ti , Tl , Tm , U,V,Y , Yb,Zn , Zr.	¹³⁷ Cs, ²¹⁰ Pb, ⁷ Be, ²²⁸ Ra		¹³⁷ Cs, ²¹⁰ Pb, C _{tot}	Burdekin River Australia Primarily Surface erosion, channel bank erosion	Surface erosion (17%), channel bank erosion (83%)
Collins et al. (2012)			Al, As, Ba, Bi, Cd, Ce, Co, Cr, Cs, Cu, Dy, Er, Eu, Fe, Ga, Gd, Ge, Hf, Ho, K, La, Li, Mg, Mn, Mo, Na, Nd, Ni, Pb, Pd, Pr, Rb, Sb, Sc, Sm, Sn, Sr, Tb, Ti, Tl, U, V, Y, Yb, Zn, Zr.			Mg, U, Pd, Y, As, Pr, Cu, Sr	River Axe watershed (UK) four sources: pasture areas, cultivated areas, channel banks/subsurface sources, road verges.	Pasture areas (38%), road verges (37%), channel banks/subsurface sources (22%), cultivated areas (3%)

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Study	Physical tracers	Organic	Inorganic	Radionuclide	Magnetic tracers	Best tracers	Description of location and sediment sources	Most contributed area (percent of contribution)
(Gaitcheon, Olley et al., 2012)				¹³⁷ Cs, ²¹⁰ Pb		¹³⁷ Cs	Daly River (Australia) two sources: Surface erosion, Channel banks erosion	Surface erosion (1%), Channel bank erosion (99%)
							Mitchell River (Australia) Surface erosion, channel bank erosion	Surface erosion (3%), Channel bank erosion (97%)
(Olley, Burton et al 2012)				¹³⁷ Cs, ²¹⁰ Pb		¹³⁷ Cs	Brisbane River Tributaries (Australia) Surface erosion, channel bank erosion	Surface erosion (10%), channel bank erosion (90%)

IRM₈₅₀ = Isothermal remanent magnetization at 850 mT, χ_{lf} =Low frequency magnetic susceptibility, χ_{fd} = Frequency dependent

magnetic susceptibility, tot= total, dit= dithionite, oxa= oxalate, pyr=pyrophosphate

Common themes that emerge from the review presented in Table 2 are:

- Sub-soils, either from rill and gully systems or artificial drainage ditches make a substantial contribution in UK and US watersheds (e.g., 48% and 55% for Jubilee and Belmont Catchment in Russell et al. 2001; 35% for River Leadon in Gruszowski et al. 2003; 62% for Pokomoke River in Gellis et al. 2009).
- Channel banks are a consistent source of suspended sediment (e.g., Northeast Branch Anacostia River watershed in Devereux et al. 2010; Southern Piedmont stream watershed in Mukundan et al. 2010; Hive Watershed in Nosrati et al. 2011). Channel and gully erosion dominates in Australia catchments (Wallbrink et al. 1998; Caitcheon et al. 2012; Olley et al. 2012; Wilkinson et al. In press).
- Upland sub-surface sources (construction sites and roads) can supply a disproportionately high amount of sediment to drainage systems. (e.g., Devereux et al. 2010; Mukundan et al. 2010).
- Magnetic tracers are used in 8 out of 20 studies, and in 6 of these studies they were identified as among the best tracers to differentiate source material. These tracers are used only in studies with a high sub-soil contribution (e.g. Russel et al., 2001; Gruzowski et al., 2001) and not in catchments where the main sediment supply is surface soils (e.g. Walling et al., 1999; Motha et al., 2004).
- Caesium-137 (^{137}Cs), Radium-226 (^{226}Ra) and excess Lead-210 ($^{210}\text{Pb}_{\text{ex}}$) are used as sediment tracers in 16, 6 and 13 studies, respectively. These radionuclide tracers were found to be the best tracers to discriminate sediment sources in 12 studies for ^{137}Cs , 2 studies for ^{226}Ra and 5 studies for $^{210}\text{Pb}_{\text{ex}}$. Fallout radionuclide tracers were able to discriminate sediment sources among different land uses and geologic units. For instance,

¹³⁷Cs was selected to discriminate sub-soil versus surface soil sources in (Walling et al. 1999; Nicholls 2001; Mukundan et al. 2010; Caitcheon et al. 2012)

- In catchments with a high sub-soil contribution (e.g. Nosrati et al., 2011; Devereux et al., 2010) organic tracers were not selected as best tracers, with the exception of Wilkinson Hancock et al., 2012.
- The use of N, C, P, $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ to discriminate between sources among land uses was successful despite their potentially unconservative behavior (e.g. $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$) during transport.
- Achieving discrimination among land use sources based on chemical elements such as REE or metals is poorly studied, and should be urgently addressed in future fingerprinting studies.

Figure 2 summarizes the data from Table 2 and indicates that sub-surface erosion accounts for between 2 to 76%, and typically 15 to 30% of suspended loads. A composite of sources originating from surface erosion processes are the dominant contributor of sediment to drainage systems in all watersheds with values of 70 to 85% commonly estimated (Figure 2). Although the contribution from sub surface erosion (particularly channel banks), changes among systems (as discussed in section 4), their importance as eroded material (sources) and its vicinity to storage (sinks) in catchment budget system makes this the most difficult source to quantify in catchment-scale sediment fingerprinting (see Parsons 2012).

(Figure 2.)

5 Mixing models

In geochemical tracing studies the relative contribution of source material to suspended sediment is usually estimated using a multivariate mixing model. The literature describes many different mathematical forms of mixing models (e.g., Collins et al. 1997a; Rowan et al. 2000; Motha et al. 2003; Evrard et al. 2011). In all mixing models, the objective is to determine the source component proportions (x) in the suspended sediment samples by minimizing the errors (Table 3).

The relative contribution of each source category must satisfy the following constraints:

- a- The fraction of source contributions must lie between 0 and 1:
- b- the percentage source contributions must sum to unity:

Table 3. Commonly used mixing models and their modifications. To make the parameters of each model more comparable, all parameters have been given consistent symbols.

Study	Model	Ref.
Slattery		(Slattery et al. 2000; Gruszowski et al. 2003)
Collins		(Collins et al. 1997a; Olley et al. 2000; Mukundan et al. 2010; Nosrati et al. 2011)
Motha		(Motha et al. 2003; Motha et al. 2004)
Hughes		(Hughes et al. 2009)
Modified Collins		(Collins et al. 2010; Collins et al. 2010)
Landwehr		(Devereux et al. 2010)
Modified Landwehr		(Gellis et al. 2009)

Where:

C_{ij} = concentration of fingerprint property (i) in sediment samples; C_{sj} = concentration of fingerprint property (i) in source category (j); P_j = percentage contribution from source category (j); W_j = particle size correction factor for source category (j); OM_j = organic matter content correction factor for source category (j); W_{ij} = tracer discriminatory weighting or tracer specific weighting; $W_{i,j}$ = weighting representing the within-source variability of fingerprint property (i) in source category (j); S_j = variance of the measured values of tracer i in source area j; n_j = the total number of samples for an individual source; n = number of fingerprint properties; m = number of sediment source categories.

The modified Collins model algorithm (Collins et al. 2010) uses the same approach as the original version (Collins et al. 1997b) to optimize the estimates of the relative contributions from the potential sediment sources, but it includes additional property weightings and a different definition for the W_{ij} parameter. In the modified model, a weighting ($W_{i,j}$) was incorporated to reflect the within-source variability of individual tracer properties and ensure that the fingerprint property values for a particular source characterized by the smallest standard deviation exerted the greatest influence upon the optimized solutions (Collins et al. 2010). The W_{ij} parameter in Collins (1997) is a tracer-specific weighting that can be calculated from the inverse of the root of the variance for each tracer in all sources. The $W_{i,j}$ parameter in the modified Collins is a tracer discriminatory weighting based on the percentage of the source classified correctly using discriminant function analysis.

The Hughes mixing model (Hughes et al. 2009) is modified from Olley and Caitcheon (2000). This model applies a Monte Carlo approach based on replicate samples (not their mean) and runs random iterations to obtain the lowest error. Fundamental differences are evident between the Collins and Hughes models. Firstly, the Collins method uses mean value for each tracer parameter pertaining to each specific source type, whereas the Hughes method uses all individual source samples in the Monte Carlo procedure. Second, correction factors (e.g., particle size) are applied only in the Collins method. The Landwehr model, used by Devereux et al. (2010),

provides a more statistically powerful model as it uses a normalized standard deviation from multiple sources rather than directly relating the values of individual variables. A modified version of the Landwehr model, used by Gellis et al. (2009), model provides additional statistical power by adding a term that divides the variance term in the denominator by m_j (the number of samples in a source area). This is particularly useful when using commonly found elemental tracers that occur in very low concentrations.

5.1 Genetic algorithms and mixing models

It has been suggested that local optimization tools (e.g. Excel solver) are not appropriate to represent global solutions (Collins et al. 2010; Collins et al. 2012). In sediment fingerprinting studies, these methods are not able to find the best optimum sediment contribution minimizing mixing model errors. To overcome this problem, (Collins et al. 2012) proposed a revised modeling approach comparing the results of both local and global (genetic algorithm) optimization tools to determine the uncertainties with the following goodness of fit (GOF) equation:

$$- \frac{\sum_{j=1}^n (C_j - \hat{C}_j)^2}{n-1}$$

Genetic algorithms (GA) were developed as a stochastic search technique based on biological processes of natural selection and the survival of the fittest. The advantages of GA as one of the most powerful optimization methods are its applicability to non-convex, highly non-linear and complex problems (Goldberg 1989), its ability to generate more than one optimum solution, and its independency from restrictive assumptions.

Advantages and differences of global optimization (Genetic Algorithms) compared to local optimization methods can be listed as follows: a) unlike local methods, the GA uses the objective

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4 itself, not the derivative information; b) the inherent random property of GA helps avoid local
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6 optima; c) when there are multiple solution points, it is impossible for local optimization
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8 methods to find the solution because they cannot jump over to a global solution; and d) through
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10 numerous variables global optimization is possible. Collins et al. (2010) compared the
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12 performance of both local and global (genetic algorithm) optimization techniques, demonstrating
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14 that GA based on random initial values minimized the objective functions compared to local
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16 searching techniques.
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21 To explore the output differences from the application of GA to the datasets in this study, we
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23 used the GAtool in MATLAB to compute sediment contribution of mixing models as objective
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25 functions. GA parameters were set up as follows: population size = 50, cross over ratio = 0.5,
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27 mutation rate = 0.1, number of iterations = 10,000 and the use of a single point cross over
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29 function along with a uniform selection procedure. Chromosome set-ups were computed based
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31 on the number of sources (i.e. three and four sources for North Fork Broad River catchment and
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33 Bléone catchments, respectively). As described in Collins et al. (2012) different values can be
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35 extracted from iterations of GAs including mean and median of all iterations using (i)
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37 conventional random repeat sampling as applied in this study or (ii) Latin hypercube sampling
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39 (LHS) method.
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46 47 **5.2 Comparison of mixing models**

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49 In this section, we use data from two sediment fingerprinting case studies in the North Fork
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51 Broad River (NFBR, USA) watershed (Mukundan et al. 2010) and Bléone River watershed in
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53 France (Evrard et al. 2011) to compare differences in relative contribution of sediment sources
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55 generated by applying the seven mixing models listed in Table 3. There are some fundamental
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57 differences between these two studies; fluvial sampling sites in the NFBR watershed were
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located at the end of the system, whereas sampling sites in the Bléone watershed were distributed as a continuum along the Bléone River and Bès River, resulting in sampling location as an important parameter. Sampling design was also influenced by differing objectives; discriminating sediment sources based on land-use in the NFBR watershed, whereas in the Bléone watershed the objective was to discriminate geologic soil types.

5.2.1 North Fork Broad River watershed

North Fork Broad River (NFBR) is located in the Piedmont region of Georgia (USA) and drains an area of 182 km². A total of 99 soil samples from three different land-uses were collected, consisting of 37 samples from potentially erodible bank faces; 32 samples from construction sites and unpaved roads; and 30 samples from pasture areas. Sediment samples were also collected from six different storm events (see Figure 3). Mukundan et al. (2010) analyzed 21 tracers including 15 trace elements (Be, Mg, Al, K, Ca, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Pb, and U), four total organic and inorganic elements (C, N, O, and S), stable isotope of N ($\delta^{15}\text{N}$), and a radionuclide isotope (^{137}Cs). Using discriminant function analysis (DFA) and removing non-conservative tracers based on their concentrations in stream sediment, four sediment fingerprint properties (^{137}Cs , $\delta^{15}\text{N}$, Cr, U) were selected as inputs into the mixing models (Table 4).

Table 4. Mean and standard deviation of the optimum fingerprint properties and their trace discriminatory weighting from DFA in NFBR watershed.

Fingerprint property selected	Mean	Standard Deviation	Wilks' Lambda	% source type samples classified correctly	Tracer Discriminatory weighting
$\delta^{15}\text{N}$	4.67 (‰)	4.7	0.444	65.7	1.5
Cr	54.21 (mg kg ⁻¹)	51.5	0.336	57.6	1.3
^{137}Cs	9.75 (Bq kg ⁻¹)	17.3	0.291	49.5	1.1
U	4.1 (mg kg ⁻¹)	2.8	0.289	43.3	1.0

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8 One of the aims of this review is to compare the variability in outputs from applying a common
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10 dataset to seven widely used mixing models. Figure 3 provides clear evidence that the
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12 application of different mixing models to the same dataset will produce dramatically different
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14 results. However, the contribution of sources in sediment transport, using local optimization
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16 methods (simple bars) are more similar to each other than using global optimization methods that
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18 has reduced variability within, but not among individual models. For example, on March 16 with
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20 2.1 m³/s water discharge and turbidity of 38NTU, local optimization methods identified the
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22 contribution of channel banks ranged between 55% with the Slattery model and 88% with the
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24 Hughes model. Differences in the contribution of channel banks among models using GA are
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26 much more variable between the modified Collins model showing that 96% of sediment
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28 originated from this source, and only 1% of material provided by this source according to
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30 Landwehr and modified Landwehr mixing models.
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38 The influence of discharge on the selection of model and optimization method is evidenced
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40 during the highest discharge event (Q=32.5m³/s) on January 7th. Using local optimization
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42 produces consistency in results among the 7 models compared with global optimization. For
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44 example, channel banks contributed between 82% with Landwehr model and 93% with Slattery
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46 and Motha models using local optimization. Applying GA techniques to the dataset produces a
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48 range of source contribution from channel banks from 91% with modified Collins to 0% with
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50 Landwehr model.
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56 In total, channel banks are the main sediment supply in all sampling events and GA-based
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58 mixing models, except for Landwehr and modified Landwehr mixing models in which pasture
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areas were shown as dominant. Using local optimization methods, channel banks remained the dominant source of sediment in all mixing models. Furthermore, the results of the Motha model based on the root mean square of relative errors, and Slattery model based on the sum of squares of errors are identical in both global and local optimization methods. Although the modified Landwehr model divides the number of samples in a source area by the variance, the percentage source sediment contribution is identical in both Landwehr and modified Landwehr models. This phenomenon is also observed for Collins and modified Collins models when local optimization methods alone are considered.

5.2.2 Bléone watershed

The Bléone watershed is a 907 km² mountainous subalpine watershed located in the Durance River district in south-eastern France. A total of 18 soil samples from four different geologic units were collected, consisting of 8 samples from Black marl; 6 from Marl-limestone sites; 2 from Quaternary deposits and 2 from Conglomerate. Riverbed sediment was collected from three sites along the Bléone River, and at two sites along the Bès River and their origin was calculated using the seven mixing models listed in Table 3.

Table 5. Mean and standard deviation of the best fingerprint properties and their tracer discriminatory weighting from DFA in Bleon watershed.

Fingerprint property selected	Mean	Standard Deviation	Wilks' Lambda	% source type samples classified correctly	Tracer Discriminatory weighting
Ra-226	23.5	7.9	0.0405	38.9	1.2
Al	4.7	1.6	0.0076	77.8	2.3
Ni	40.2	12	0.0024	33.3	1
V	75.3	24	0.0001	66.7	2
Cu	15.5	5.2	0.000515	44.4	1.3
Ag	0.2	0.08	0.000253	38.9	1.2

Forty fingerprint properties including radionuclide elements (^{137}Cs , $^{210}\text{Pb}_{\text{ex}}$, ^{40}K , ^{226}Ra , ^{228}Ra , ^{228}Th , ^{234}Th), rare earth elements (Ce, Eu, La, Lu, Sm, Tb, Yb), major elements (Fe, K, Na, Al, Ca, Mg, Ti) and trace elements (As, Ba, Co, Cr, Cs, Hf, Sc, Ta, Th, Zn, Ag, Co, Cr, Cs, Hf, Sc, Ta, Th, Zn) were analyzed in both surface soil and sediment samples. The ability of these tracers to discriminate between potential sediment sources was investigated by conducting the Kruskal-Wallis H-test and discriminant function analysis (DFA). Finally, one geogenic radionuclide (^{226}Ra) and five metal (Al, Ni, V, Cu, Ag) tracers were selected as the best tracers using DFA (Table 5).

(Figure 4.)

Contrary to the NFBR watershed, we cannot assess the stability of each mixing model in Bléone watershed as the sampling locations change along both Bès and Bléone Rivers. All mixing models generate different percentages of contributions using both local optimization and genetic algorithm optimization methods in the Bléone watershed (as also reported in NFBR). The use of GA optimization produces a wider range of sediment source contributions than using local methods. For example, at site BE7 of the Bès River (light grey), Black marl and Quaternary deposits are identified as the main sediment supply using local optimization methods. In contrast, almost all suspended sediments are identified as originating from Marl-limestone sources when using the modified Collins and Landwehr models with GA optimization, with the Collins, Hughes, Motha and Slattery mixing models recording both the quaternary deposit and black marl as the dominant sediment sources.

In both the NFBR and Bléon watersheds, the Motha and Slattery mixing models provide similar results for the relative contribution of source sediments using both local and global optimization.

In the Bléone watershed, the use of GA and local optimization methods with the Landwehr and modified Landwehr models were not able to predict similar source contributions for sediments, whereas these models gave identical results using both GA and local optimization in the NFBR watershed.

5.2.3 Goodness of fit results

The accuracy of source contribution values resulting from the application of 7 mixing models and two optimization methods can be tested with goodness-of-fit (GOF) values (Table 6).

Table 6. GOF values of seven mixing model and two optimisations

Mixing models	Optimization method	GOF (%)					
		Bléone catchment			NFBR catchment		
		Min	Mean	Max	Min	Mean	Max
Collins	GA	53	75.5	90	13.3	15.4	16.8
	Local	62.2	76.8	89.2	30.3	54	79
Modified Collins	GA	43.4	60.5	70	22.3	61	73.7
	Local	60.8	72.5	87.8	18	55.7	75.3
Hughes	GA	61.6	76.7	88.5	1	21.7	78
	Local	63	77	88.6	35.7	60.3	75.4
Landwehr	GA	48	63.7	74.7	<0	<0	<0
	Local	59.7	75.6	88	25.5	48	67.3
Modified Landwehr	GA	56	70.4	85	<0	<0	<0
	Local	59.7	75.5	87.3	22.6	50.6	73.2
Motha	GA	64.4	76.4	88	68.4	31	73.8
	Local	64.4	76.3	88.8	48.7	23.3	77
Slattery	GA	64.7	76.1	89	69.3	30.6	75
	Local	62.8	76.3	88.8	67.5	28.2	77

Improved accuracy in both catchments was obtained when applying the original Collins model using a local optimisation method than using a modified Collins mixing model. The use of GA in the modified Collins mixing model, improved accuracy to 61% within the catchment with more source samples (NFBR with 99 source samples in 3 sources), compared with local optimisation with a 55.7% goodness-of-fit. In the catchment with fewer sources (Bleon with 18 source samples in 4 sources), local optimization was the more powerful method for calculating

source contributions (GOF=72.5%). In the Hughes model that uses the actual values rather than statistic parameters, local optimization produced a higher goodness-of-fit of 77% and 60.3% in Bleon and NFBR catchments respectively..

Comparing the application of all mixing models in each catchment, the Hughes mixing model appears a more robust method in Bléone catchment using local optimization method (GOF=77%), and the modified Collins in NFBR catchment using GA optimization (GOF= 61%).

6 Conclusion

Suspended sediments in fluvial systems can lead to a number of detrimental environmental and operational impacts. Sediment fingerprinting techniques have been applied to fluvial systems to identify sources of sediment; however the selection of model and optimization method can have profound effect on the output of sediment fingerprinting analyses. This is the first review that has compared the most prevalent mixing models (including the application of genetic algorithms) to an actual dataset to quantify variability in the output depending on the application of mixing model.

All sediment fingerprinting studies must decide on the choice of field sampling methods, and selection of tracers as well as mixing models. Allowing for time and budget constraints, the study objective should drive the field sampling method. For example, fluvial sampling is the preferred method to determine the origin of sediment deposited in a dam, whereas point sampling is the most appropriate method to monitor sediment contribution in a flood event. Budget will also drive the selection of tracers used as sediment fingerprint properties. Physical tracers are less expensive and can be measured easily, but they are not conservative and may lead to ambiguity in interpretation of results. Geochemical tracers are favored due to large number of

elements available for sediment fingerprint measurements. Radionuclide tracers are the most powerful tracers to distinguish soils from different land uses, but need expensive instruments.

Our review of 25 sediment fingerprinting studies identified land-use and geology as the most prevalent discriminators of sediment sources. The relative importance of sediment sources to drainage systems should vary among different catchments due to the contrasts in geology, watershed morphology, hydrology, connectivity of river systems, human interference and many more factors. This inherent variability translates to a reliance on the final step of all sediment fingerprinting studies; computing the contribution of different sediment sources via mixing models. Using a common dataset, we have shown that different mixing models can identify different relative contributions of sediment sources, but that the range of values among models are within an acceptable range of errors (i.e. relative error, mean squared error etc.) in objective functions reported by the original authors. Based on GOF, the modified Collins and Hughes mixing models are the most powerful models to estimate the source contribution to transported sediments. Also, global optimization methods must be carefully applied when using the Hughes mixing model. We suggest the use of a model that combines the best explanatory parameters from modified Collins (it uses correction factors) and Hughes (it uses iterations of all data not mean values) with optimization based on genetic algorithms would best predict the relative contribution of sediment sources to fluvial systems.

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Figure 1. The process required for sediment fingerprinting in fluvial systems, including sample collection, tracer selection and analyses, mixing model selection to determine sediment source contribution.

Figure 2. Frequency distributions for the contribution of channel bank/Sub-surface and surface sources of sediment from the 47 watersheds reviewed in Table 2.

Figure 3. Percent relative contribution of three sediment sources (channel banks, construction sites, pastures) based on seven mixing models and seven flood event in the NFBR watershed. Q is flow discharge in m^3/s and T is turbidity in NTU (nephelometric turbidity unit).

Figure 4. Percentage of relative contribution of four geologic sources to sediment (Black marl, Marl-limestone, Quaternary deposit, Conglomerate) for seven mixing models and three sediment samples along the Bléone River, and two sediment samples along the Bes River.

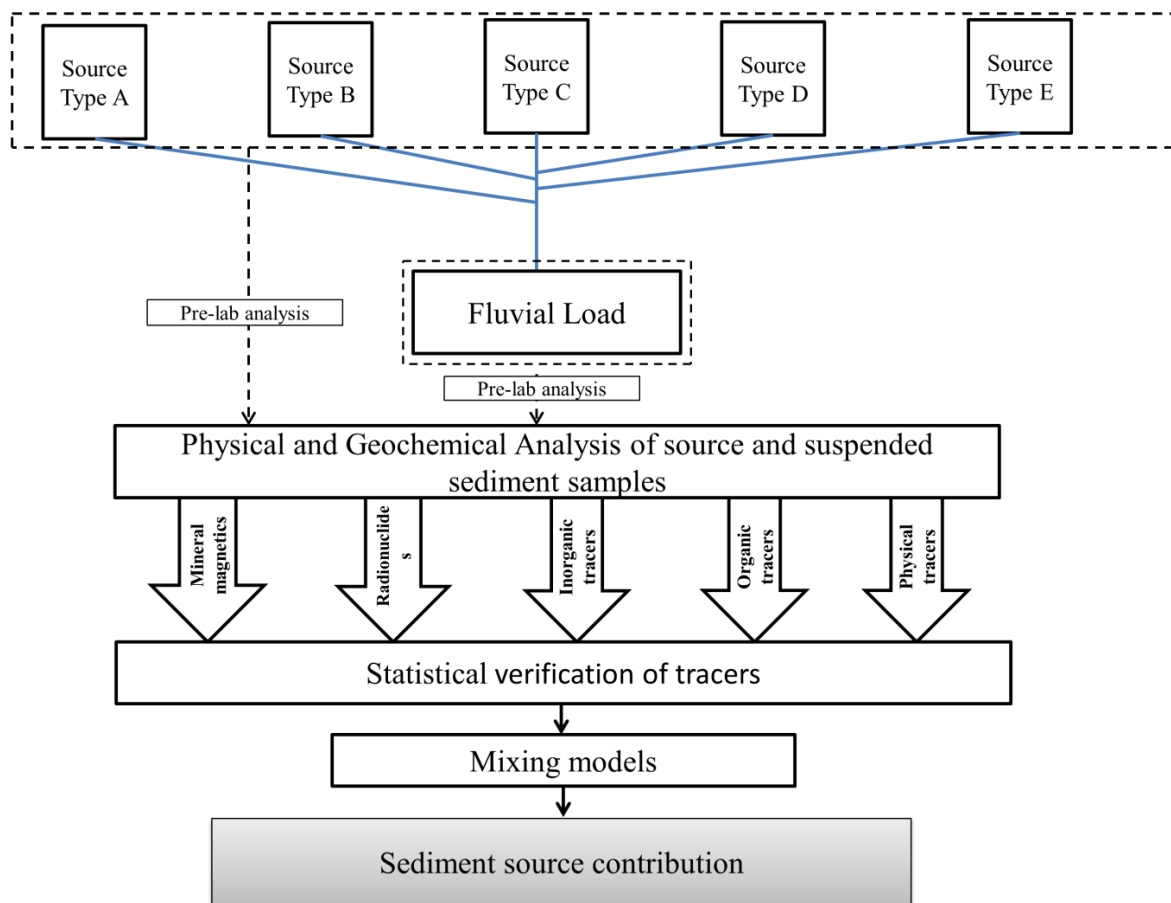


Figure 1.

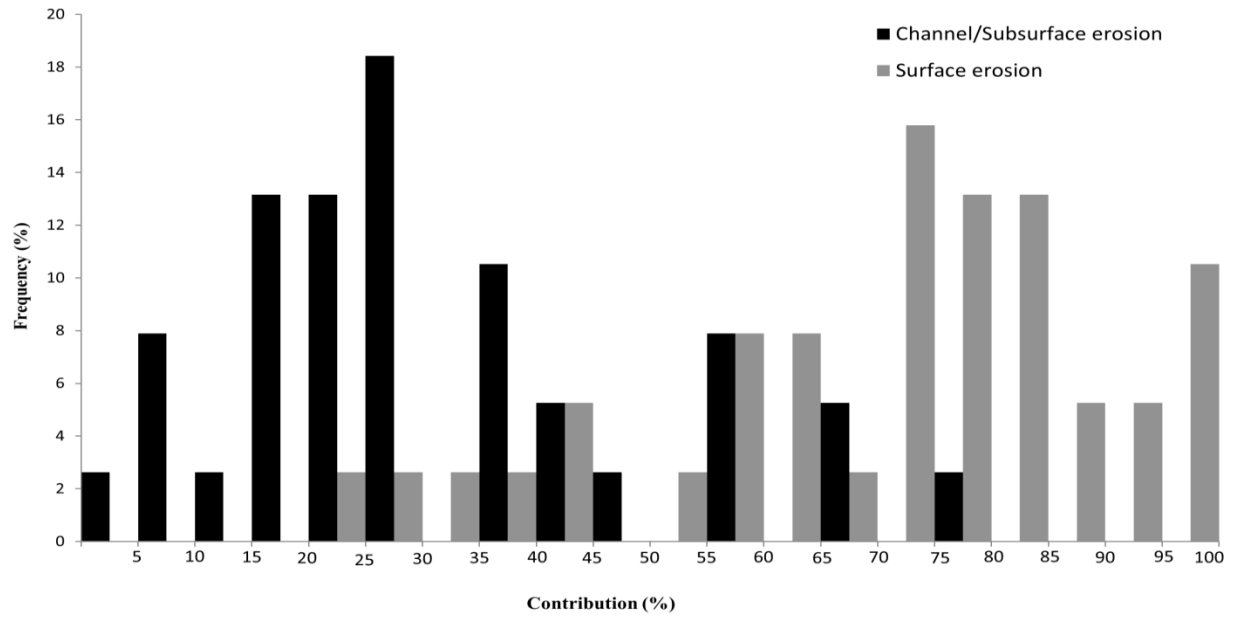


Figure 2.

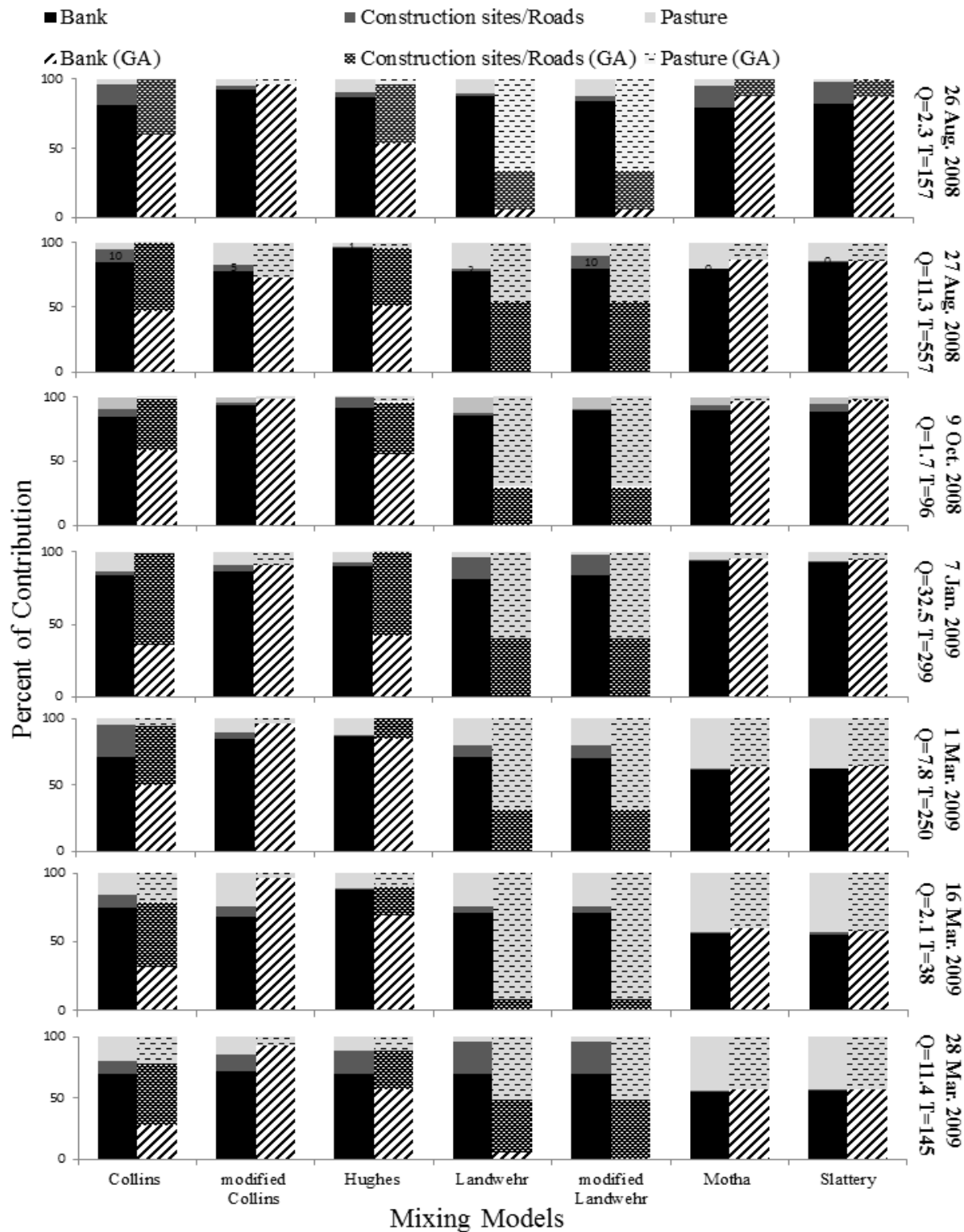


Figure 3.

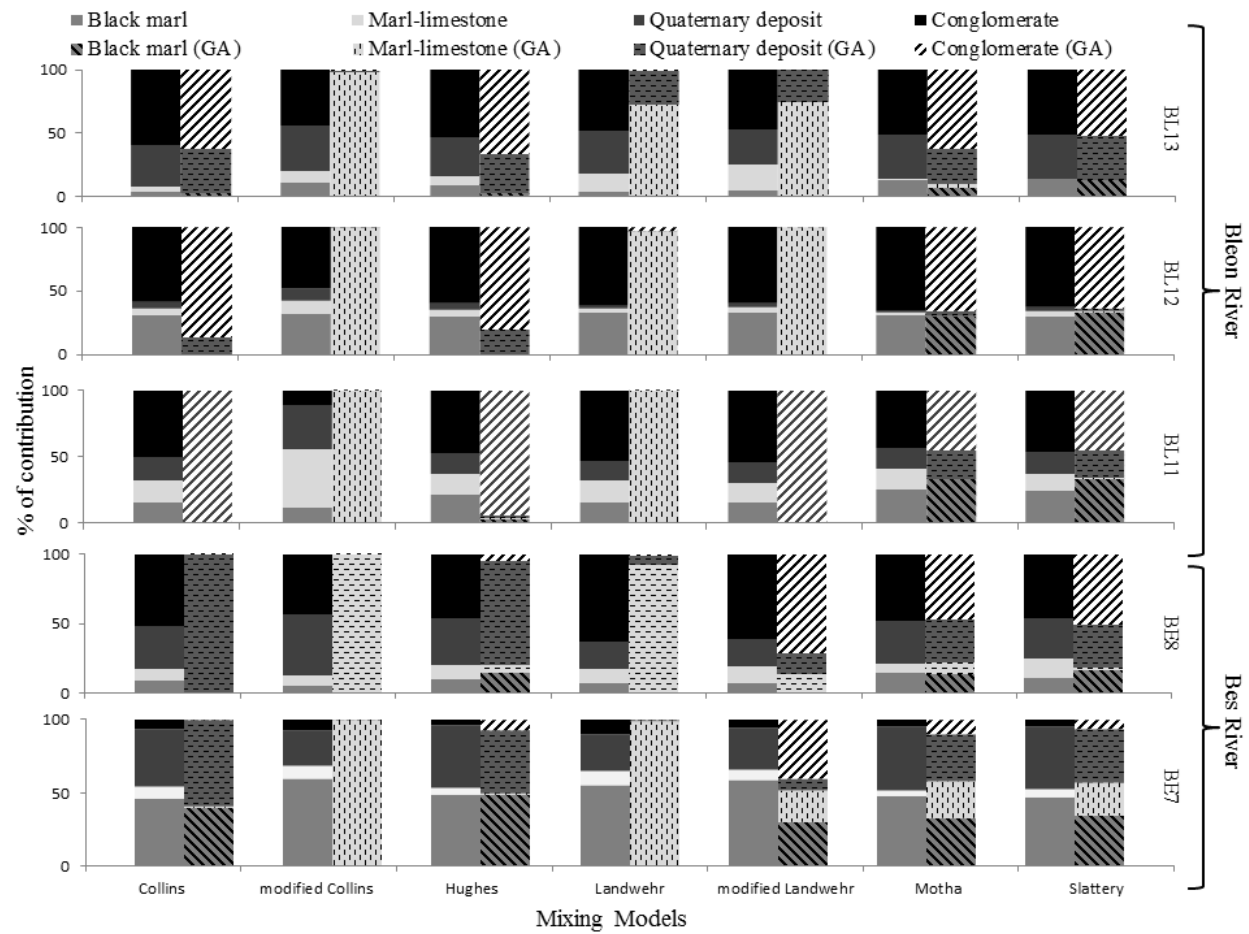


Figure 4.