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Fingerprinting and tracing the sources of soils and sediments: earth and ocean sciences, geoarchaeological, forensic, and human health applications

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Contents

Abstract

Fine-grained sediment is perhaps the most widespread and pervasive contaminant in aquatic systems reflecting its role in influencing the quality of the water (e.g., turbidity, vector of chemicals and other pollutants) and its detrimental effect on infrastructure (e.g., dams, turbines) and aquatic habitats (e.g., salmonid spawning grounds) through sedimentation. Determining the sources of fine-grained sediment thus represents an important requirement for watershed and coastal management, as well as for understanding landscape and oceanic evolution. Sediment source fingerprinting utilises the diagnostic physical, chemical and biological properties (i.e., tracers) of source materials to enable samples of collected sediment to be apportioned to these sources. This review examines the development of the technique within the earth and ocean sciences, focusing mainly on agricultural landscapes. However, the development of new tracers, such as compound-specific stable isotopes, has allowed the technique to be applied in a growing number of environmental settings including forested (including wildfire-impacted forests), urban and estuarine/coastal settings. This review also describes other applications of the fingerprinting approach such as geoarchaeological (e.g., archaeological site formation), forensic (e.g., identifying the sources of soil/sediment particles in criminal investigations) and human health (e.g., identifying the sources of airborne particulate matter, $PM_{2.5}$) applications. Identifying commonalities in methods and approaches between environments and disciplines should foster collaboration and the exchange of ideas. Furthermore, refinement of the sediment source fingerprinting technique requires that several methodological issues be addressed. These methodological issues range from the initial sampling design through to the interpretation of the final apportionment results. This review also identifies and assesses these methodological concerns.

Keywords: sediment sources; sediment fingerprinting; tracing; sediment properties; fine-grained sediment; sediment provenance

1. Background and history

In recent decades, there has been a rapid growth in the number of studies that have utilized tracing and fingerprinting approaches to investigate the movement of soils and fine sediments in terrestrial and aquatic systems (Koiter et al., 2013a; Walling, 2013; Mabit et al., 2014). This growth is due to the fact that these techniques are able to provide essential information on soil and sediment dynamics that can be used to understand the evolution of landscapes (e.g., Belmont et al., 2007) and assist in river basin management and river restoration (e.g., Owens, 2005, 2008; Evans et al., 2006; Minella et al., 2008, 2014; Walling and Collins, 2008; Gellis and Walling, 2011). In these contexts, the source tracing and fingerprinting techniques have often been part of wider sediment budget investigations (Gellis and Walling, 2011), as the source tracing and fingerprinting techniques alone are sometimes too broad (e.g., topsoil is dominant over channel banks) to enable exact sources (e.g., specific fields, or channel bank reaches) to be determined. Thus, broad classifications of sediment source types can make it difficult to precisely target management strategies intended to control sediment problems. . In addition, most sediment source tracing and fingerprinting results are relative (i.e., expressed as percentages), and sediment transport data are often required to convert values into actual sediment fluxes associated with the sources (e.g., Walling and Woodward, 1995; Smith et al., 2011). Source tracing and fingerprinting techniques used in concert with information on sediment transport and sediment budgeting can offer powerful insights into how landscapes behave and can provide important information on geomorphological processes, which, in turn, can be used to guide river basin and coastal management. Mukundan et al. (2012), for example, have demonstrated how sediment source fingerprinting can be used as a management tool for developing total maximum daily loads (TMDLs) of sediment as part of the TMDL program in the USA.

Early source tracing and fingerprinting studies (e.g., Klages and Hsieh, 1975; Wall and Wilding, 1976) were typically qualitative in nature and concerned with establishing the spatial (e.g., geological) sources of contemporary suspended sediment. These were followed by studies that were more quantitative, again with emphasis on the sources of contemporary sediment (e.g., Peart and Walling, 1986; Walling et al., 1993, 1999; Collins et al., 1997a). Recent developments have seen the technique expanded to include further applications (i.e., new landscape types and research questions, see sections below) and used to determine historical changes in sediment sources using floodplain (e.g., Collins et al., 1997b; Owens and Walling, 2002a; Walling et al., 2003), check dam (e.g., Chen et al., 2016) and lake and reservoir (e.g., Foster and Walling, 1994; Ben Slimane et al., 2013; Pulley et al., 2015) deposits; for a review see D'Haen et al. (2012). The last decade or so has seen an expansion of the types of properties used as tracers and the use of more rigorous statistical approaches and numerical unmixing models.

While there are similarities between approaches concerned with the tracing and fingerprinting of soil and sediment particles in the landscape, there are also some fundamental differences. One useful distinction between the two approaches is that in the case of "tracing" (or "sediment tracing") studies the tracers are pre-selected; in many cases they are applied artificially (e.g., rare-earth elements or fluorescent tracers; Liu et al., 2016). The selection is based on an understanding of the behaviour of that tracer (e.g. fallout radionuclides) and its ability to answer the research questions being investigated. In the case of "source fingerprinting" (or "sediment fingerprinting") studies initially it is unclear what tracers will be selected as fingerprints and samples are analysed for a range of potential tracers and statistical methods are used to identify those that are able to discriminate sources.

The term "source tracing" is a hybrid term often used to refer to the use of tracer properties to identify the source of sediments. Thus, the terms "source tracing" and "source fingerprinting" are often used interchangeably to mean the use of the properties of soils and sediments to infer their origins; for simplicity, in this review we mainly use the term sediment source fingerprinting.

While there have been numerous recent reviews of the sediment source fingerprinting approach (e.g., Walling 2005, 2013; Gellis and Walling, 2011; Mukundan et al., 2012; Guzman et al., 2013; Haddadchi et al., 2013) most of these are concerned with specific aspects of individual approaches and their application. Thus, Walling (2005) provides an overview of the approach using case study examples from primarily agricultural river basins in the UK. Haddadchi et al. (2013) focus on reviewing sediment tracers and mixing models. The reviews by Gellis and Walling (2011) and Mukundan et al. (2012) are mainly concerned with how sediment source fingerprinting approaches can be used as river basin management and restoration tools, while Guzman et al. (2013) focus on the provision of information on soil erosion and redistribution at the scale of hillslopes and small watersheds. Few publications have considered the wider-ranging potential of the approach, especially for applications beyond fluvial geomorphology and landscape evolution. Given the documented increase in the use of sediment source fingerprinting (i.e., Koiter et al., 2013a; Walling 2013), it seems timely to review the applications to date, especially beyond studies focussing on agricultural landscapes, and to consider the wider relevance of the approach in other settings, such as coastal and oceanic, and other uses, such as forensic and human health. A key objective of this review is to encourage interdisciplinary collaboration (i.e. between earth sciences, ocean sciences, hydrology, geomorphology, soil science, atmospheric science, health sciences, archaeology, chemistry, biology) amongst those who use soil and sediment source fingerprinting techniques in disctinctive, but complementary, ways.

It is also pertinent to address some of the research needs to allow the technique to reach its full potential in these new areas, especially given recent developments, such as addressing the non-conservative behaviour of sediment tracer properties (e.g., Parsons and Foster, 2011; Koiter et al., 2013a;

Pulley et al., 2015; Sherriff et al., 2015) and concerns over the impact of correction factors to account for differences in particle size and organic matter content (e.g., Smith and Blake, 2014; Kraushaar et al., 2015; Smith et al., 2015). Many would argue that the approach is at a key stage in its development, and that the research community needs to develop some fundamental principles for its application.

This review: (i) describes the basic principles of sediment source fingerprinting; (ii) synthesises many of the applications of sediment source fingerprinting; and (iii) considers important research needs.

2. **The sediment source fingerprinting approach**

There are numerous soil and sediment properties that can be used to discriminate between the potential sources of the sediment within a river basin or coastal/oceanic environment. This section provides a brief overview of these properties; for comprehensive reviews see Foster and Lees (2000), Collins and Walling (2004), Guzman et al. (2013) and Haddadchi et al. (2013). Many of the main fingerprinting properties are shown in Figure 1 and include physical characteristics (e.g., sediment size, shape, colour), geochemical properties (e.g., trace metals), fallout radionuclides (e.g., ^{7}Be, ^{137}Cs, unsupported ^{210}Pb), mineral magnetic properties (e.g., magnetic susceptibility and isothermal remanence), and organic properties (e.g., compound-specific stable isotopes, microbial communities, pollen). These fingerprint properties have been used individually (e.g., colour; Martinez-Carreras et al., 2010; Brosinsky et al., 2014), within property groups (e.g., fallout radionuclides; Owens et al., 2012; Evrard et al., 2016) or in combination as part of a composite fingerprinting approach (e.g., geochemical, mineral magnetic and fallout radionuclides; Walling and Woodward, 1995; Collins et al., 1997a; Walling et al., 1999) to infer sediment sources.

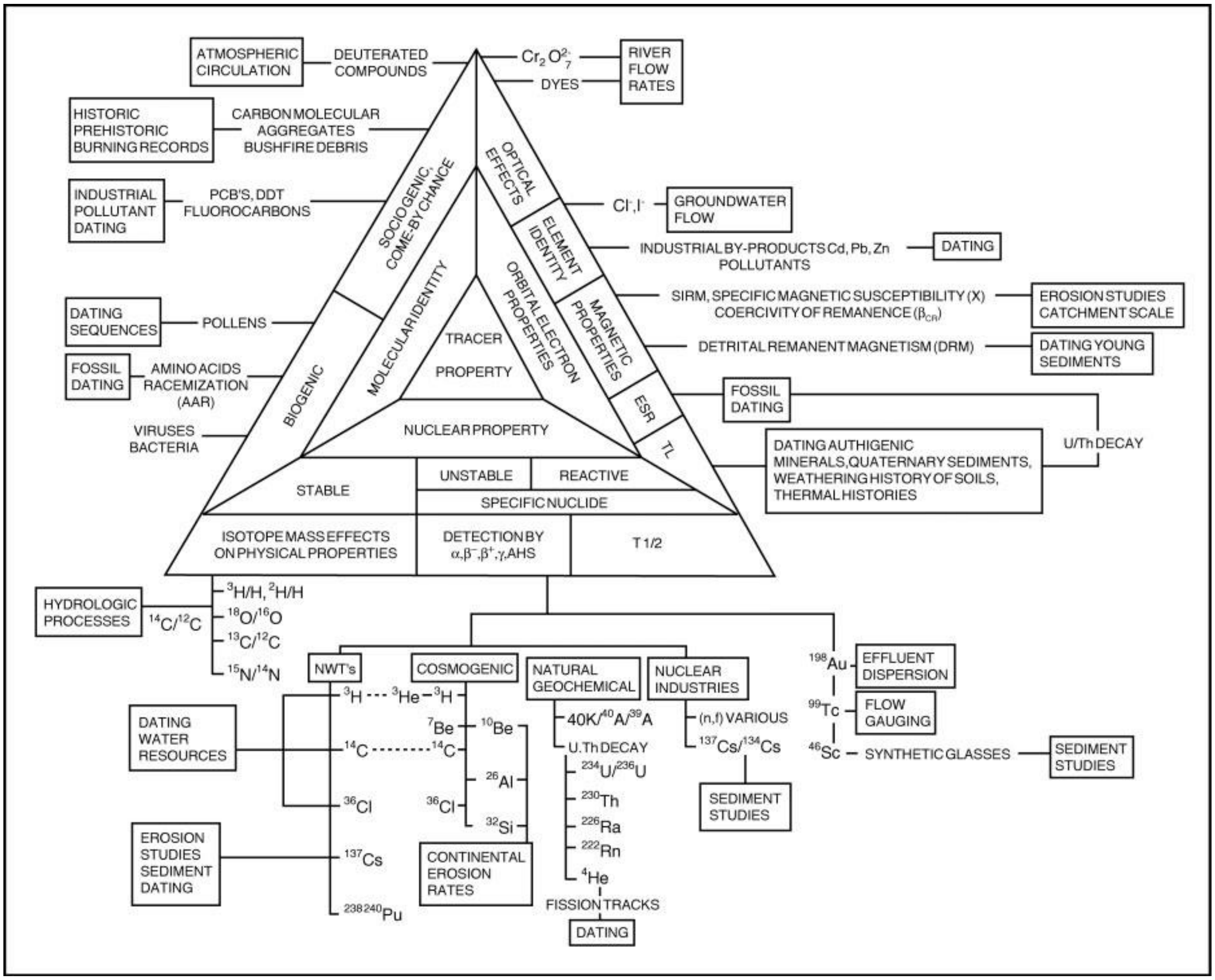

Fig. 1. Properties of earth materials that may be used to date and trace sediment sources (from: Foster and Lees, 2000).

The basic principle behind sediment source fingerprinting is that the sediment properties (e.g., trace element concentrations, radionuclide activities) will reflect their origins. For example, fallout radionuclides label surface soils and thus high activity concentrations in a sediment sample suggests that the sediment was derived from surface soils (i.e., topsoil) as opposed to subsoil material which would be expected to have lower activities (Owens et al., 1996; Wallbrink et al., 1998). Similarly, a sediment sample rich in organic carbon would suggest that it was derived from undisturbed surface soils (e.g., forest, pasture) as opposed to cultivated landscapes or channel bank material (Gellis and Noe, 2013). In most cases, a direct link has been made between the fingerprint properties in the target sediment and those of potential source materials. This assumes that the property used as a fingerprint exhibits a conservative behaviour between source and sink, and that any alterations or transformations are either negligible or can be quantified (Belmont et al., 2014). It is well known, for example, that the particle size distribution of sediment changes as it moves through the landscape (e.g., Walling, 1983; Koiter et al., 2015), and this can influence the concentrations of certain properties, in which case an allowance needs to be made for such changes. In the case of particle size effects, this can be achieved through restricting analysis to a certain size fraction (e.g., < 63 µm) or by using correction factors. However, the use of corrections factors needs to be exercised with caution (Koiter et al., 2013a; Smith and Blake, 2014), and is discussed later (i.e. section 7.4).

The identification of source locations from sediment properties can be achieved both qualitatively and quantitatively. Examples of the former include simple bi-plots of property concentrations for sources and sediments. For example, in the case of plots of ^{137}Cs versus unsupported ^{210}Pb activity, samples from a particular sediment source are required to cluster and to cluster in a different domain space compared to other potential sources (Walling and Woodward, 1992). The relative location of sediment samples on the same plot can be used to infer likely source(s). Thus, in the example shown in Figure 2, the concentrations of ^{137}Cs and unsupported ^{210}Pb in surface soils are significantly higher than those for subsoil/channel bank samples, and the equivalent concentrations for sediment samples fall within the cluster for subsoil/channel bank, thus suggesting that this is the dominant source of sediment in these two watersheds in British Columbia, Canada (Owens et al., 2012).

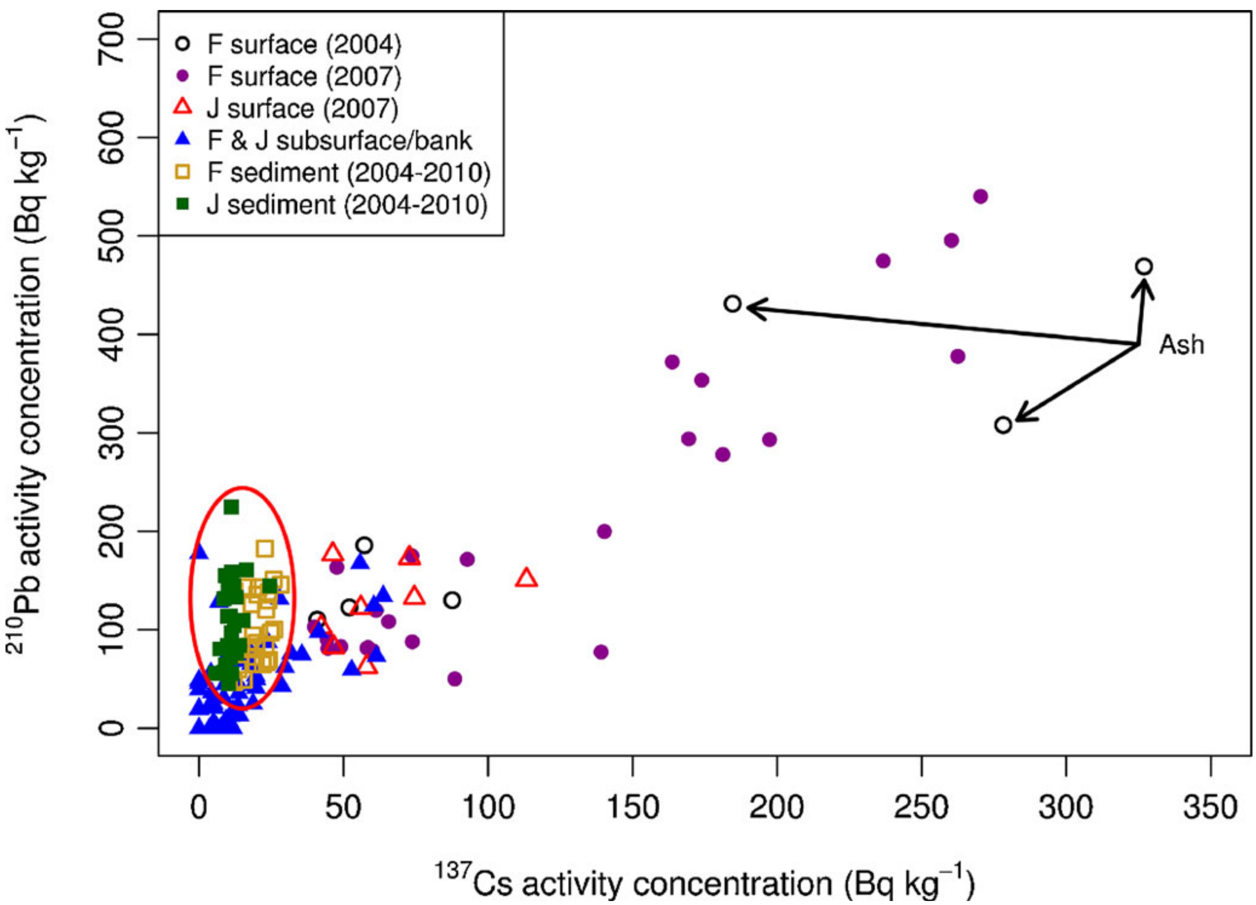

Fig. 2. Bi-plot of ^{137}Cs and unsupported ^{210}Pb activity concentrations for source materials (surface soil and subsoil/channel bank material) and fine-grained sediment samples collected from burnt (Fishtrap Creek, F) and unburnt (Jamieson Creek, J) watersheds in British Columbia, Canada. The plot illustrates the distinction between surface soils and subsoil/channel bank material in both catchments. The sediment samples are enclosed in the envelope (bottom left) (modified from: Owens et al., 2012).

Quantitative source identification is typically achieved through the use of statistical procedures to select fingerprints and the use of multivariate unmixing models to apportion the contribution from each sediment source; sections 7.4 to 7.6 consider these aspects in greater detail. This quantitative approach tends to involve assessment of the uncertainty associated with the results (e.g., via Monte Carlo simulation). The estimation of uncertainty is crucial as it provides a measure of confidence in the results (Rowan et al., 2000). Such information should incorporate the variability associated with *all* aspects of the fingerprinting approach, including field sampling, laboratory analysis and the application of the unmixing model; values of uncertainty based on only one or two of these aspects are likely to be misleading. Understanding, quantifying and reporting these sources of uncertainty – and their implications –needs to be addressed much more comprehensively by the scientific community (Smith et al., 2015).

3. Earth and ocean sciences applications

3.1 Agricultural landscapes

To date, the majority of sediment source fingerprinting studies have been undertaken in agricultural landscapes reflecting the widespread occurrence of agriculture and also concerns associated with soil erosion and off-site effects. Typically, studies have aimed to quantify the relative contribution of agricultural fields to the total sediment budget and to compare such contributions to the sediment delivered from other areas of the basin (e.g., forests, river banks, road ditches, urban areas). The contributions from several case studies, summarized in Table 1, represent only a small fraction of sediment source fingerprinting studies in agricultural watersheds but they highlight the value of the sediment source apportionment approaches. Considering the large range of values estimated for the contribution of agricultural topsoil (i.e., 1-97%), studies prior to and after the implementation of management practices appear relevant in order to focus on problematic areas and to follow the post-change trajectory of sediment dynamics. The great variability of the contributions also highlights the necessity to consider factors other than land cover, which can include agricultural practices, drainage network, slope, weather and antecedent conditions, as these can also influence soil erosion rates. In order to quantify soil erosion rates and to more precisely define the severely eroded areas, it is often necessary to combine sediment source fingerprinting techniques with sediment mass balance estimates. For example, Smith et al. (2014) showed that the increase in the arable area from ~6 % to 36 % for the River Tamar basin (920 km^2) in the south-west of the UK over the period 1990-2007 led to a rise in hillslope sediment yield from 8.8 to 32 kt $year^{-1}$ while the contribution from the erosion of channel banks remained almost constant.

Most studies have used inorganic tracing properties (e.g., fallout radionuclides and geochemistry), but recently organic tracers have also been utilised to provide a more comprehensive and detailed assessment of sediment movement and sources.

3.1.1 Inorganic tracers: identification of erosion processes

The information provided by sediment source fingerprinting studies can be improved by combining several tracer properties that have originated as a result of different processes (e.g., due to weathering of bedrock, atmospheric fallout, or land-use). For example, Gruszowski et al. (2003) used mineral magnetic, geochemical and radionuclide signatures to highlight the role of road ditches as a conveyer of 30 % of sediment eroded from agricultural topsoils in a watershed in the UK. Determining these preferential pathways also helps to prevent counterproductive management practices. For example, subsurface drain installation in agricultural watersheds has often been considered as an efficient way ofpreventing erosion by increasing water infiltration rates in soils. Fingerprinting studies in contrasting watersheds in England, Australia and France (e.g., Russell et al., 2001; Wilkinson et al., 2013; Foucher et al., 2015) have demonstrated that large amounts of fine sediment were derived from eroded sub-surface soils, as the subsurface drainage altered the hydrology of the watershed. Similarly, Walling et al. (2002) reported that between 30 % and 60 % of eroded sediment was transferred through subsurface drains in two small agricultural watersheds in the midlands of the UK.

Sediment source fingerprinting studies have also revealed temporal changes in sediment contributions from different parts of agricultural watersheds, which suggest that there is seasonality in certain erosion processes. For example, recent studies (e.g., Gellis and Noe, 2013; Lamba et al., 2015a) have shown that freeze–thaw processes are likely to increase the contribution of bank erosion in agricultural watersheds. Fingerprinting studies have also revealed unexpected effects of changes in agricultural practices from a management perspective. For example, Minella et al. (2008) reported that the decrease of erosion rates in cropland areas in a watershed in Brazil resulted in an increase in erosion of the channel banks due to changes in the transport capacity and competence of channel flows.

In addition to providing useful information on sediment origins in agricultural watersheds, fingerprinting approaches can also be used to investigate the contribution of sediment-associated nutrients (e.g., nitrogen and phosphorus) and contaminants (e.g., metals, polychlorinated biphenyls) related to agricultural activities (e.g., Walling et al., 2008). Therefore, sediment-associated phosphorus deposited in rivers or lakes can be traced back to cropland or pasture erosion; alternatively, a higher contribution of banks can lead to a dilution of particulate phosphorus by less phosphorus-rich sediment (Lamba et al., 2015b). Nutrient concentrations can also be used as tracers for sediment source fingerprinting (e.g., Ben Slimane et al., 2013), however, the inclusion of such fingerprints are controversial due to the potential of non-conservative behaviour during transport (Koiter et al., 2013a).

To illustrate more fully the potential application of inorganic sediment fingerprinting properties for identifying sediment sources in an agricultural watershed, we present findings for the South Tobacco Creek watershed in Manitoba, Canada.. The downstream impacts of sediments and associated contaminants, phosphorus in particular, in this watershed required that sources of sediment be determined and appropriate management practices be developed and implemented. The watershed has an area of ~75 km^2 and the creek flows east, from the glacial till landscape of the Prairie Pothole Region, over the Cretaceous shale bedrock of the Manitoba Escarpment, across the glaciolacustrine plain of the Red River Valley. The predominant land use is agriculture, largely conventionally cultivated annual crop production. Over several years, samples of suspended and channel-stored fine-grained sediment were collected along the length of the main channel. Potential source materials, including stream bank, bedrock, riparian soil and field soils were collected in association with each sediment sampling location.

Koiter et al. (2013b) used geochemical and radionuclide fingerprinting properties to establish the relative contributions of sediment sources. The suspended sediments in the uppermost reaches were dominated by topsoil sources (64 %–85 %), whereas the suspended sediments exported farther downstream had a higher proportion of sediment coming from streambank (32 %–51 %) and shale bedrock (29 %–40 %) sources. This switch in the dominant sources of sediment between the headwaters and the watershed outlet were due to: (i) changes in sediment storage and connectivity; (ii) a transition in the dominant erosion processes from topsoil to streambank erosion; and (iii) the incision of the stream channel through the shale bedrock as it crosses the Manitoba Escarpment. Additional work by Barthod et al. (2015), using colour parameters (i.e., reflectance spectrometry, VIS-NIR) as fingerprint properties, supported the findings of Koiter at al. (2013b) and showed colour to be an effective tracer of sediment (Figure 3). Following these studies, there has been a shift from soil conservation practices that minimize soil losses by water erosion to management practices that manage the runoff. Although inorganic tracers have proved successful in fingerprinting sediment sources, more spatially detailed investigations are needed as the broad sediment sources identified have limited utility in making management decisions. The use of organic tracers, such as compound-specific stable isotopes, may allow for the identification of more detailed sediment sources (e.g., crop specific) to be identified.

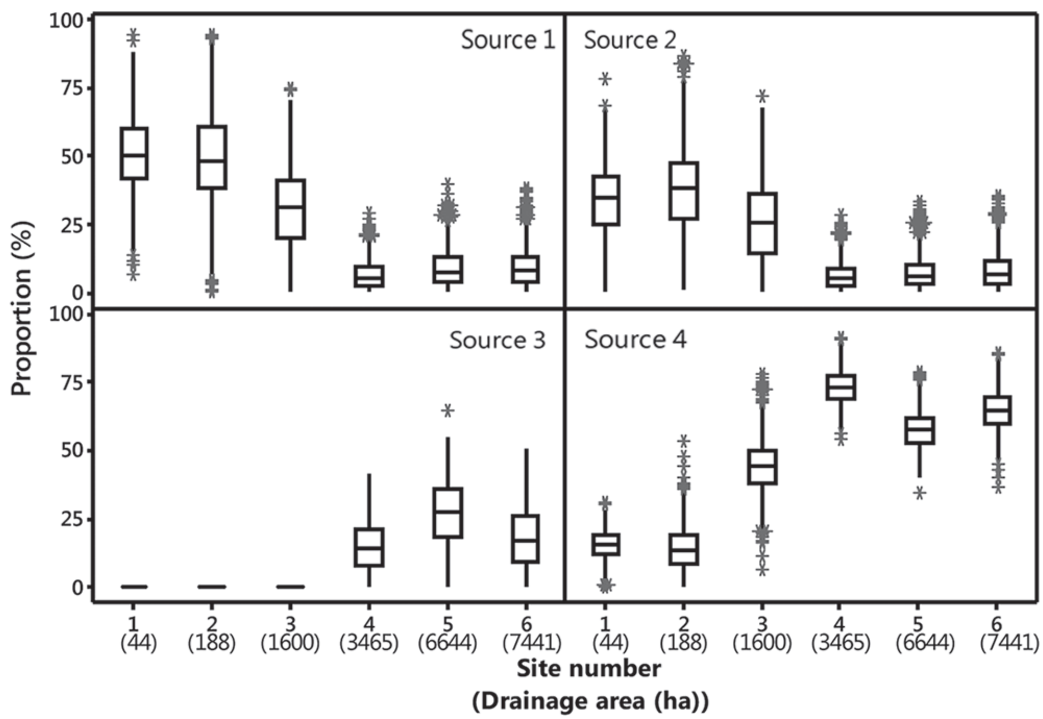

Fig. 3. Box and whisker plots showing the relative contributions of potential sources to suspended sediment collected from monitoring stations in the agricultural South Tobacco Creek watershed, Manitoba, Canada. Source 1: topsoil from agricultural fields, riparian areas, and forest valley walls; source 2: stream bank materials above an escarpment; source 3: stream bank materials within and below an escarpment; and source 4: outcrop shale materials. The lower and upper hinges correspond to the first and the third quartiles, respectively. The whiskers are created using the Tukey method. Potential outliers are plotted as crosses (from: Barthod et al., 2015).

3.1.2 Organic tracers: identification of contributions from different land uses

There has been considerable interest in developing organic tracers for use as sediment source fingerprints (e.g., Granger et al., 2007). Early studies (e.g. Brown, 1985) demonstrated the use of pollen and spores to establish the sources of suspended sediment. Other studies have demonstrated the use of stable isotopes of carbon (C) and nitrogen (N) to infer sources of fine-grained sediment (McConnachie and Petticrew, 2006; Fox and Papanicolaou, 2007; Schindler Wildhaber et al., 2012; Laceby et al., 2015a). Recent developments include: the use of the natural and artificially-applied DNA-markers (e.g., Mahler et al., 1998; Granger et al., 2007) associated with contrasting source materials; and the use of plant-associated fatty-acids and compound-specific stable isotopes (e.g., Reiffarth et al., 2016). Other studies (e.g., Zhang et al., 2016) have also demonstrated the potential of using microbial communities to identify sediment derived from different land use activities.

Plant-derived organic tracers (referred to as biomarkers hereafter) can complement more established inorganic tracers such as fallout radionuclides and geochemical properties. Whereas some traditional tracing techniques may cover large areas of a watershed without exhibiting a high degree of variability, biomarkers reflect input from the vegetation on the surface by deposition onto the soil and entrapment within the sediment. The high recalcitrance and low biodegradability of certain fatty acids (FAs), as well as their relatively high abundance, suggests they offer considerable potential as sediment tracers.

Plants produce FAs that are structurally indistinguishable; for example, all roots produce C22:0 and C24:0 FAs (Pollard et al., 2008; Wiesenberg et al., 2012). Differentiating the source of the FAs based solely on vegetation type may be accomplished by examining the carbon isotopic signature of a particular FA. Carbon isotope values reflect the ratio of ^{13}C:^{12}C, and are often reported using the delta scale ($\delta^{13}C$). The delta value is reported relative to a commonly used standard for that particular element (e.g., Vienna-Pee Dee Belemnite for carbon); values are either negative or positive relative to the standard, which is defined as no shift. Maize and wheat, for example, are C4 and C3 plants, respectively, and, due to the differing methods of CO_2 fixation, should exhibit significant differences between their leaf and root FA $\delta^{13}C$ values (O'Leary, 1988). Average $\delta^{13}C$ values for C4 and C3 plants bulk ($^{13}C_{bulk}$) tissue have been reported as -13‰ and -27 ‰, respectively (Glaser, 2005), with FAs further depleted in ^{13}C, relative to the bulk tissue, by approximately 6-8 ‰ (Ruess and Chamberlain, 2010).

Several researchers (e.g., Gibbs, 2008; Blake et al., 2012; Hancock and Revill, 2013; Cooper et al., 2015; Alewell et al., 2016) have investigated the use of FA biomarkers for soil and sediment tracing. Specifically, the carbon isotope signatures of FAs found in the soil were used, together with $^{13}C_{bulk}$ for soil and sediment, and total organic carbon (TOC), to apportion sediment sources in small watersheds. For the purpose of sediment source fingerprinting, the analysis to determine the isotopic signature of individual organic compounds has been referred to as compound-specific stable isotope (CSSI) analyses; the term CSSI has been adopted by the International Atomic Energy Agency (IAEA) in this context.

To illustrate the application of CSSIs, Blake et al. (2012) investigated a small agricultural watershed (1.45 km^{-2}) in the UK, where the climate is temperate. The sampling site consisted of permanent pasture, ley pasture, maize stubble, winter wheat and woodland. Blake et al. (2012) used CSSI data to determine the sources of sediment at the stream outlet for the watershed after a period of heavy rainfall, and compared the CSSI-based estimated yields from each source to ones calculated based on geochemical data. They found that the geochemical data overestimated the contribution of cultivated topsoil to the watershed sediment yield during the event as compared to the estimates provided by the CSSI-based tracers. The geochemical-based overestimate was attributed to the ley pasture in the watershed, which constituted a significant portion of the area in the watershed and had been previously cultivated. The ley pasture would have a geochemical fingerprint similar to other cultivated fields, and therefore, the two sources would be indistinguishable by geochemical fingerprinting means. Using an unmixing model

(IsoSource; see section 7.6 for more information on unmixing models) and CSSIs, Blake et al. (2012) determined that the major sediment contribution at the watershed outlet came from pasture sources, which represent ~65 % of the watershed area. The results indicated the potential usefulness of the CSSI technique as a sediment source fingerprinting tool, albeit in a small agricultural watershed.

The use of CSSIs as a sediment source fingerprinting tool is still in its infancy. Early results suggest that CSSIs of FAs may help differentiate crop-specific sediment sources and thus complement information derived from more conventional, inorganic tracing properties. More research is needed to address the concerns raised by Blake et al. (2012), Alewell et al. (2016) and others, including sources of variability associated with environmental processes, field sampling and laboratory preparation and analysis (Reiffarth et al., 2016).

3.2 Forested landscapes

3.2.1 Deforestation and forest harvesting

Soil disturbance by forest harvesting operations have been widely observed to increase the sediment loads of receiving streams and river networks (e.g., Leeks and Marks, 1997; Douglas et al., 1999; Stott and Mount, 2004; Walling, 2006; Kreutzweiser et al., 2009). In this context, sediment tracing and fingerprinting approaches are of value in identifying sediment sources and hence the processes leading to increased sediment load to inform Best Management Practices (BMPs) (Wallbrink and Croke, 2002). Soil surface disturbances by forestry operations are wide ranging in spatial scale and impact but generally include the direct impact of forestry operations themselves on soil structure, as well as the construction of: (i) temporary log processing areas; (ii) skid tracks along which logs are hauled for processing; and (iii) local and regional road networks (Douglas et al., 1999; Motha et al., 2003). While the skid track and road infrastructure is considered to be the major factor in generating sediment and delivering it to the stream system, it can be difficult to assess the additional role of tree harvest operations using conventional monitoring approaches (Motha et al., 2003). Tracer applications have utilised fallout radionuclide budgeting approaches and sediment source fingerprinting to address this issue – in a similar way to those studies in agricultural landscapes described above (i.e., section 3.1) – and some examples are reviewed below.

Wallbrink et al. (2002) adopted a fallout radionuclide tracer budget approach to explore sources of erosion, rates of sediment transport and storage, and losses from the system. The tracer budget approach was based on fallout ^{137}Cs distribution across the landscape with a particular challenge of overcoming spatial heterogeneity in fallout due to interception by the forest canopy and leaf litter distribution on the soil surface. A landscape element (or unit) approach was adopted with defined units of: (i) log landing areas; (ii) skid tracks; (iii) general forest harvesting areas; and (iv) filter strips that were retained for wildlife and riparian protection. Once the areal extent of each unit was defined, a large number of soil cores were collected and bulked together in groups of 10 to be counted as a single sample. In this way, the mean inventory of the reference site and each landscape unit could be obtained with an appropriate quantification of standard error to estimate the net losses and gains between landscape units. The budget showed that the skid tracks generated the greatest losses of soil but a large proportion of the mobilised material was captured by the filter strips and retention banks plus the general harvest area, where infiltration capacity reduced overland flow. The results demonstrate the value of adopting a landscape budget approach to sediment source assessment.

The shorter-lived fallout radionuclide ^{7}Be has also been used effectively by Schuller et al. (2006) to quantify net soil losses and retention by buffer features following forest harvest operations. The study was based around a single heavy rainfall event and the fallout radionuclide-based data were compared to soil loss estimated using erosion pins with close agreement. The key challenge noted by the study was ensuring that the distribution of ^{7}Be prior to the event was spatially uniform, especially considering the legacy of prior erosion events, which led to the development of a methodological refinement to apply ^{7}Be over extended time periods (Walling et al., 2009).

The importance of forest roads has also been explored using the geochemical properties of sediment (Motha et al., 2003) and colour tracers of sediment source (Erkine, 2013). Motha et al. (2003) utilised a geochemical fingerprinting approach to distinguish material derived from the harvested soil surface and roads in logged watersheds of south-eastern Australia. They utilised a chemical index of alteration (CIA) to describe the degree of weathering of alumina-silicate minerals. The approach allowed discrimination of highly weathered surface materials from moderately weathered gravel road surfaces. In addition, fallout radionuclides were used to discriminate surface versus subsurface soils. Enrichment ratios for grain size and organic matter were applied to the source data based on quantified relationships between the target tracer properties and these two factors. The results showed that while the undisturbed forest dominated the sediment load of the receiving streams due to 93 % catchment coverage, the disturbed areas, and in particular the roads, made a disproportionately large contribution given their aerial extent. Others have adopted a similar geochemical approach to determine the contribution of material mobilised by logging operations in tropical rainforest watersheds of Malaysian Borneo. Here, the x-ray fluorescence (XRF) geochemistry of a lateral channel bench deposit (cf., Hughes et al., 2010) was used alongside an unsupported ^{210}Pb-based record of accretion (Walsh et al., 2011). While the timing of peaks in accretion coincided with the known history of logging within the watershed, in particular in the steep headwaters , a shift in the geochemical signal indicated a greater contribution from less weathered (i.e., deep sourced) material. This was linked to incision of skid trails and temporary logging roads plus increased landslide events after logging operations during extreme rainfall events (Douglas et al., 1999).

While there are fewer examples of sediment source fingerprinting applications in forested watersheds compared to agricultural systems, the results of work to date consistently highlight the important role of tracks and roads as sediment sources in forested watersheds, information that would be difficult to obtain and contextualise against slope erosion through monitoring programs alone.

3.2.2 Wildfires

Post-wildfire sediment source fingerprinting studies have focused largely on quantifying contributions from hillslope surface and channel bank (subsurface) sources of fine sediment using fallout radionuclide tracers (Table 2; also see review by Smith et al., 2013). These studies have employed ^{137}Cs and unsupported ^{210}Pb for source discrimination because of the pronounced differences in activity concentrations of these radionuclides with soil depth in forest environments (Wallbrink and Murray, 1996; Blake et al., 2009; Owens et al., 2012; also see Fig. 2). Recent work has also used additional radionuclides such as ^{239,240}Pu to discriminate hillslope and channel sources of fine sediment transported by post-fire debris flows (Smith et al., 2012). In burnt forest environments, fallout radionuclides provide the most useful tracer properties for watershed-scale source discrimination, and may be very effectively coupled with process measurements and monitoring (Smith et al., 2013). For example, suspended sediment flux measurements combined with tracer-based estimates of proportional source contributions allow for calculation of source-specific sediment yields (Smith et al., 2011). Such information is important for interpreting post-fire changes in proportional source contributions to sediment exports associated with vegetation and soil recovery.

The discrimination of spatial sources according to burnt and unburnt areas or based on areas burnt at different severities has received less attention. Previous studies have examined the potential for geochemical (Blake et al., 2006a), mineral magnetic (Blake et al., 2006c) and organic compounds (Oros et al., 2002) to discriminate burn-defined spatial sources, but did not attempt to estimate source contributions to downstream sediment fluxes. To date, only Stone et al. (2014) have sought to quantitatively apportion sediment contributions from spatial sources defined according to unburnt, burnt only, and burnt and salvage logged sub-basins. However, this study did not identify any environmental basis for why the selected geochemical tracer properties might discriminate these sources. Difficulties may arise due to natural and burn-related variability affecting the basis for source discrimination. Geochemical and fallout radionuclide tracer concentrations may increase in burnt soils due to the loss of soil organic mass and with inputs of ash from burnt vegetation or decrease due to vaporisation losses during soil heating (Johansen et al., 2003; Blake et al., 2006a, 2006c; Owens et al., 2006, 2012; Perreault et al., 2012). Mineral magnetic properties can be enhanced in surface soils by burning (Longworth et al., 1979; Clement et al., 2011), while heating and post-fire leaching and bio-transformations contribute to changes in organic compounds in burnt surface soils (Oros et al., 2002). Where the ash content of soil accounts for most of the difference in geochemical or fallout radionuclide concentrations between soils burnt at different severities and unburnt areas, the requirement for conservative behaviour of these tracer properties during transport is unlikely to hold. This reflects the potential for density-related differences in the transport behaviour of ash and mineral soil, resulting in the modification of surface soil tracer signatures during downstream transfer. Magnetic properties show the greatest potential for burnt soil spatial source discrimination because burn-related changes are associated with the mineral component of surface soils (Blake et al., 2006b).

The forested highlands of south-eastern Australia have been the focus of most source tracing and fingerprinting work following wildfires. Studies were situated in the Blue Mountains in New South Wales and the forested uplands in north-eastern Victoria, both regions subject to severe wildfires. In the Blue Mountains, a multiple-catchment (17-629 km^2) source tracing study was conducted over a 5 year period following wildfire (Wilkinson et al., 2009). Fallout radionuclides were used to trace contributions from hillslope surface and subsurface (river bank and gully) sources to in-channel sediment deposits. Post-fire erosion resulted in a change in sediment sources from 80 % subsoil prior to the fire up to 86 % surface soil for one of the study watersheds in the first year after burning (Wilkinson et al., 2009). Hillslope surface contributions then declined after the fire. In north-eastern Victoria, hillslope surface erosion was found to decline from approximately 100 % in the first year to 58% in the fourth year after wildfire in an intensively monitored 1.36 km^2 research watershed (Smith et al., 2011). Coupling this information with sediment load data showed that hillslope erosion accounted for 93 % of the total post-fire suspended sediment yield over the study period, while the hillslope contribution in the first year after fire equated to 75 % of the measured fine sediment output (Smith et al., 2011). Despite differences between the studies in terms of watershed size, geology, topography, soil and forest type, hillslope surface sources dominated in the first year after wildfire and showed similar declining trends with post-fire recovery. In contrast, studies in North America have used sediment source fingerprinting to demonstrate the dominance of channel bank sources following wildfire. For example, Owens et al. (2012) showed that channel bank sources contributed ca. 91.5% to the sediment flux exported from the 135 km^2 Fishtrap Creek watershed in British Columbia, Canada, for the period 2004-2010, following a wildfire in 2003. The difference between findings for North America (see also Moody and Martin, 2009) compared to European and Australian landscapes reflect differences in climatic drivers, and vegetation type and recovery (Owens et al., 2013).

3.3 Urban landscapes

Compared to agricultural and forested landscapes, relatively few studies have quantitatively determined the sources of sediment in urban waterheds with studies to date being concentrated in Australia (e.g., Ormerod, 1999; Charlesworth et al., 2000), Brazil (e.g., Poleto et al., 2009; Franz et al., 2014), China (e.g., Yin and Li, 2008), UK (e.g., Charlesworth et al., 2000; Carter et al., 2003) and USA (e.g., Devereux et al., 2010). In part this stems from the inherent hydrological complexities associated with urban areas, alongside multiple diffuse sediment sources, and the potential for transformations in the physical and chemical properties of the sediment (Taylor and Owens, 2009). For example, most urbanized and industrialized rivers receive discharges from sewage treatment works (STWs) and industrial facilities, which contain dissolved materials that interact with fine-grained sediment and change its chemical composition. For example, Owens and Walling (2002b) showed that the phosphorus (P) content of suspended sediment in the River Aire, UK, increased immediately downstream of STWs reflecting inputs of both particulate and dissolved P from the STWs; in the case of the dissolved P this adsorbed onto the passing suspended sediment, so that the P content of the downstream sediment no longer reflected the P content of the original source. Other studies have explored the complex interactions between P and fine-grained sediment in rivers receiving urban and industrial inputs; for a review, see Withers and Jarvie (2008).

Numerous studies have qualitatively inferred the sources of sediment in urban aquatic systems (e.g., rivers, lake/reservoirs and estuaries; Charlesworth et al., 2000; Zhong et al., 2012). Many of these have used multivariate statistical approaches such as principal components analysis (PCA) and cluster analysis (CA) to match the contaminant properties of sediment (e.g., metals, PCBs) to those of potential sources. Typically, the elevated concentrations (i.e., relative to natural baseline, such as Cu from a copper mine) or presence of an element (e.g., artificial substances, such as PCBs from industry) indicate the likely source. One of the most comprehensive studies to use the sediment source fingerprinting approach to quantify the sources of sediment in an urban watershed was by Carter et al. (2003). These authors determined how the sources of the suspended sediment transported in the River Aire, UK, changed in a downstream direction reflecting inputs of new sources from urban areas. In the agricultural (mainly pasture) headwaters of this watershed the main sediment sources were uncultivated topsoil (range for several streams = 16-57 %) and channel bank (43-80 %). However, downstream of the main urban areas (including the cities of Leeds and Bradford) the relative contribution from these source types decreased (combined = 22-40 %) reflecting contributions from additional sources such as cultivated topsoils (20-45 %), solids from STWs (14-18 %) and road-deposited sediment (RDS, 19-22 %). The change in the contribution of channel bank sources also reflects the artificial protection of many channel banks from erosion and lateral migration in urban areas.

The high contribution of urban sources (ca. 40 % from STWs and RDS) illustrates the marked contrast of sediments in urban watersheds to those in non-urbanized watersheds described in previous sections. The results from the River Aire are consistent with those of Yin and Li (2008) who used ^{7}Be and ^{210}Pb as fingerprints to estimate that about 60 % of the suspended sediments at the outlet of a sewer system in Wuhan City, China, was derived from the drainage system (gutter sediments and combined sewer sediments), with about 40 % from RDS. They are also consistent with Poleto et al. (2009) who used a composite fingerprint based on geochemical (e.g., metals) and carbon, to determine that roads (paved and unpaved) and channel banks contributed 69 % and 31 %, respectively, to the suspended sediment load transported in a river near Porto Alegre in Brazil.

One of the important aspects of these studies is that while the contribution from urban sediment sources such as RDS and solids from STWs has been recognised (e.g., Foster and Charlesworth, 1996; Charlesworth et al., 2000; Owens et al., 2011), fingerprinting has enabled such contributions to be quantified, albeit in a relative sense. Furthermore, the work by Carter et al. (2003) also provided important new information on the timing of such contributions, and how this changes during the course of a rainfall event (Fig. 4). In the case of RDS, contributions from this source increased during the event reflecting this increased connectivity of the road network to the channel system as the rainstorm progressed (i.e., more runoff), while the contributions from STWs and other sources decreased during the course of the event because of dilution effects. The complex hydrology and flashy nature of urban catchments can pose significant challenges for sampling design and monitoring efforts.

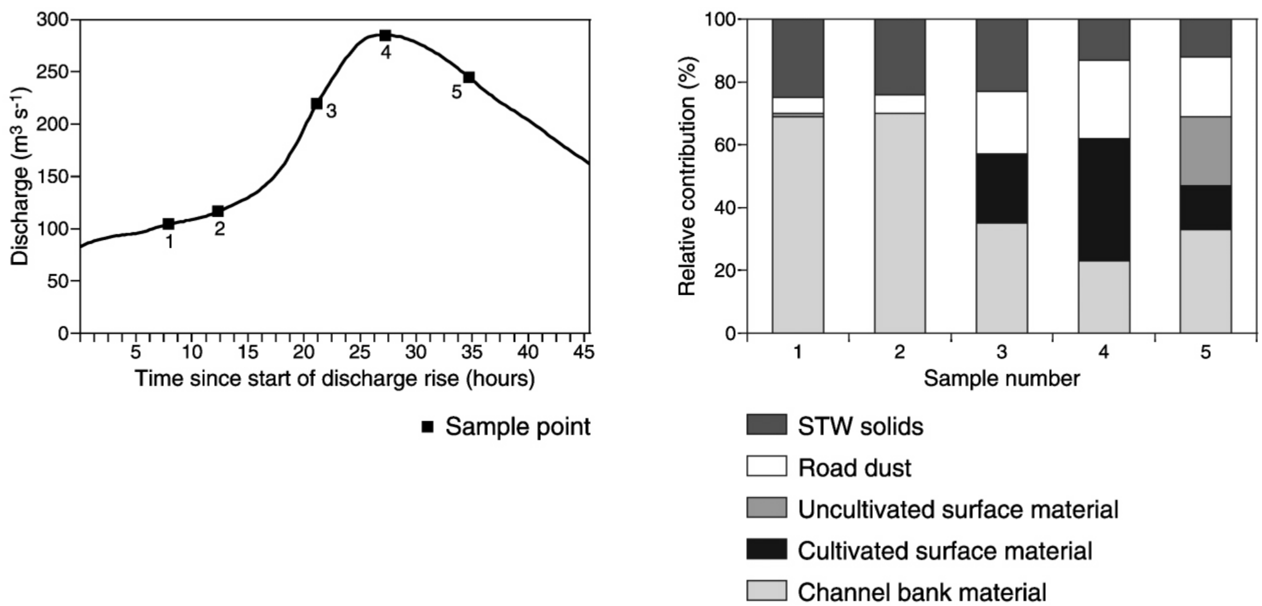

Fig. 4. Variation in the relative contribution from surface material from uncultivated and cultivated areas, channel bank material, road-deposited sediment and solids from sewage treatment works (STWs) for five suspended sediment samples collected from the downstream reaches of the River Aire, UK, during a rainfall-driven high streamflow event during March 1998 (from: Carter et al., 2003).

Other source tracing and fingerprinting studies in urban environments have been concerned with atmospheric sediment (i.e., dust and airborne fine particulate matter). Much of this interest has been driven by the health concerns associated with the impacts of fine particulate material (e.g., $PM_{2.5}$) on the human respiratory system. Thus numerous studies have attempted to fingerprint the sources of fine airborne particulates in urban areas based on the geochemical properties of the sediment (e.g., Park and Kim 2005), and these are reviewed in Section 6.

3.4 Estuarine and coastal landscapes

At the interface between watersheds and the marine environment, estuaries represent critical sink zones for fine sediment and associated nutrients and contaminants. Estuarine morphology and fluvial and tidal currents are key factors in determining the processes that control sediment accretion and, linked to this, knowledge of sediment source is a key part of the management of sediment problems and remediation strategies. Sediment source information is also critical to inform coastal management strategies particularly in the context of management realignment schemes for flood defence and habitat restoration (Rotman et al., 2008). Although limited in number compared to river basin studies (described above), a range of sediment tracing and fingerprinting approaches have been used in estuarine and coastal environments including recent advances in CSSIs (Gibbs, 2008), application of mineral magnetic properties (e.g., Yu and Oldfield, 1989; Jenkins et al., 2002, 2005; Rotman et al., 2008), rare earth elements (REEs) (Zhou et al., 2010) and radionuclide fingerprints (e.g., Hebinck et al., 2007; Yeager et al., 2005, 2006) – with a focus on geogenic uranium and thorium series-based signatures. These tracers are largely controlled by mineralogy and are therefore less likely to be affected by transformations associated with transfers from freshwater to saline conditions. This is an important consideration for acid soluble geochemical properties that have been extensively used in river basin systems (section 3.1).

Siltation problems are a key driver for knowledge of sediment sources and sediment fluxes to estuaries. Gibbs (2008) presents a study where CSSI signatures were used to apportion sediment deposited in estuarine mangroves back to potential upstream source areas. The agricultural watershed feeding into the estuary in northern New Zealand presented a range of land uses from pasture (~70 %) to native forest (~20 %) and plantation pine forest (~8 %) in the steeplands plus ~4 % urban coverage. In addition, outer estuary sediments were sampled as a separate source. The study was driven by notable declines in the abundance of a suspension-feeding bivalve which were hypothesised to result from enhanced suspended solids linked to watershed disturbance with implications for oyster aquaculture sited on the inter tidal flats. The isotopic signature highlighted the input of the pine plantation activities to sediment stored in upper and mid estuary deposits. The lower estuary was dominated by reworked materials from the coastal zone. In addition to identifying the potential of this relatively new sediment tracing and fingerprinting tool (i.e., CSSIs; also see section 3.1.2), the study highlighted the wide variation in source inputs in different zones of the estuary and the importance of comprehensive sampling strategies in sediment sink zones.

As well as being perceived as an environmental problem in estuaries, sediment supply can also be a key factor in the development of coastal zone habitats. Where management strategies are designed to enhance these, or mitigate detrimental effects, knowledge of sediment source can be critical part of the decision-making process. As such, sediment source fingerprinting has been used to identify sources of sediment entering barrier reefs such as those in Australia (e.g. Douglas et al., 2010; Bainbridge et al., 2016) and other sensitive coastal environments. In this context, Rotman et al. (2008) used mineral magnetic signatures to develop knowledge of sediment sources contributing to the restoration of intertidal flats and saltmarsh. A particular focus was to examine how the managed realignment sites interacted with wider geomorphological processes in the estuary. The sampling programme focussed on proximal sources of sediment to include established saltmarsh and intertidal deposits seaward of the study site, plus terrestrial sources. Magnetic signatures were shown to discriminate the sampled sources based largely on different magnetite concentration and magnetic grain size. Apportionment results suggested that a large component of deposited material in the managed realignment site was derived from existing eroding saltmarshes which did not align with the management goal of habitat creation.

Model ‘efficiency’ (often termed goodness of fit) was also explored and lower values in some areas indicated that either a source was missing from the analysis or that signature transformation might be an issue in some locations.

3.5 Oceanic environments and sedimentary basins

Oceanic environments and sedimentary basins are the ultimate sink for sediment along the sediment cascade and the abundant room for storage within these features creates a long geologic record of sedimentation which is useful for the reconstruction and interpretation of the geological history of the Earth. It is interesting to note that while many of the other disciplines and applications covered within this review use the term “sediment source fingerprinting” the oceanography, geology, and sedimentology disciplines typically use the term “sediment provenance” but the ultimate goal remains the same (i.e., identify the sources of sediment). However, the objectives of these types of sediment provenance studies are typically much larger in both temporal (e.g., palaeoclimate; Meyer et al., 2011) and spatial extent (e.g., palaeogeography; Yang et al., 2006) but also investigate contemporary and anthropogenic driven sediment fluxes (e.g., environmental degradation; McCulloch et al., 2003). Furthermore, these studies are not limited to unconsolidated sediment but also include the analysis and interpretation of the sources of sediment within siliciclastic sedimentary rocks (e.g., submarine fan sandstones; Morton et al., 2005). In addition, many sediment provenance studies do not directly sample potential sources but rather draw conclusions about the characteristics of the sources (e.g., sediments source from rocks intermediate between felsic and mafic composition; Armstrong-Altrin et al., 2015).

Weltje and von Eynatten (2004) identified and described three main approaches to characterizing the properties of sediment within provenance studies and include the analysis of: 1) bulk sediment; 2) specific groups of minerals; and 3) single grains. The analysis of bulk sediment is one of the more common approaches used in many of the other fingerprinting applications described in this review (e.g., Section 3.1.1) and includes the determination of the concentration of major, trace, and rare-earth elements (REE), isotopic ratios, and bulk mineralogy. It is interesting to note that REE concentration data in these types of provenance studies, are often presented as normalized to either the average upper continental crust (UCC) concentration (e.g., Padoan et al., 2011) or to chondrite (a stony meteorite) concentration (e.g., Meinhold et al., 2007). This normalization makes it easier to display the values as some elements are orders of magnitude different, and allows for the detection of anomalously high (> 1) or low (<1) concentrations.

The isolation and the characterization of specific mineral groups, typically heavy minerals (> 2.85 g cm^{-3}), allows for greater discriminatory power as the inclusion of the more common light minerals (< 2.85 g cm^{-3}; quartz, feldspar and mica) may not add much additional information on the potential source of sediment (Dill, 1998). While the heavy mineral fraction makes up only a small fraction of the sample (often < 1 %) there are < 30 mineral species that can be identified and used in sediment provenance studies, many of which have a characteristic paragenesis (association of minerals in a particular type of rock) (Morton, 1985). The identification and abundance (i.e., assemblage) of these heavy minerals have been used to provide information about the potential sources of sediment (e.g., Rodríguez et al., 2012). In some case studies, the ratio of stable minerals with similar densities has been used to fingerprint sediment to avoid potential issues of changes in composition due to transport, deposition and diagenetic processes (e.g., garnet/zircon; Morton et al., 2005).

Single grain analysis is typically limited to the coarse-grained sediment (> 63 µm) as the silt and clay grains are too small given the resolution of the analytical equipment (Weltje and von Eynatten, 2004). However, it is important to consider that single grain analysis provides information on the source of the mineral of interest which can result in biased results with respect to the source of the sediment as a whole (Morton and Hallsworth, 1994). There are two main types of single grain analysis - geochemical composition and radiometric dating - and von Eynatten and Dunkl (2012) provide a review of single grain techniques. For geochemical analysis (also known as varietal heavy mineral analysis), the variability in the concentration of major and trace elements of different minerals are used to differentiate between sediment sources (e.g., Tsikouras et al., 2011). One of the more common single grain radiometric dating analysis used to fingerprint sediment is zircon geochronology. The mineral zircon is highly resistant to both chemical and physical weathering and is commonly found in many sedimentary deposits making it ideal to fingerprint. The premise of using geochronology as a fingerprint is to link the sediment to the source of sediment based on the age of the parent rock as the age of the mineral is interpreted to be the crystallization age of a rock (Thomas, 2011).

The large-scale nature of these studies in addition to the unique characteristics of the sources (e.g., potential sources are unknown) and pathways of sediment (e.g., large distance between sources and sink) and the post-deposition environmental conditions (e.g., increased pressure through burial) and processes (e.g., dissolution of less stable minerals) creates a challenging situation which has resulted in the development of different analytical techniques being used to fingerprint sediment. The techniques used to fingerprint sediment are in response to the non-conservative behaviour of sediment properties which is exceptionally important under these circumstances. Therefore, the use of properties of the more durable minerals and the distribution of less mobile elements/isotopes and their ratios as sediment fingerprints are more commonly used (Basu et al. 2016). Table 3 provides a summary of the different sediment properties that have been utilized as diagnostic sediment fingerprints. Furthermore, Thomas (2011) suggests that context, including the stratigraphic, sedimentologic, tectonic, and palaeogeographic setting, is also important to consider when interpreting fingerprinting data, especially in case studies where the sources are non-unique.

Cook et al. (2013) presents an interesting case study using the isotopic ratios of $^{143}Nd/^{144}Nd$ and $^{87}Sr/^{86}Sr$ as sediment fingerprints to identify the sources of sediment in a glaciated environment. In this study, Cook et al. (2013) assessed the vulnerability of the east Antarctic ice sheet (predominately land-based) to warmer temperatures as a result of a changing climate. This was undertaken by investigating sediments that were deposited during an interval in the geological record when the temperature was warmer than present. A core was drilled 310 km offshore (64º24'5'' S 143º53'1'' E; 3,465 m water depth) and the section of the core that was studied was 75 – 125 m (below sea floor) and was deposited during the Pliocene (5.3 - 3.3 million years ago) when the mean annual global temperature was 2 - 3 ºC higher than present. The core section was characterized by two alternating sedimentary layers: diatom-rich silty clay sediment and diatom-poor clayey sediment layers representing warmer and cooler temperature periods, respectively. Within the vicinity of the ice sheet, the isotopic ratios varied between the different rock types and these variations were attributed to differences in both age and lithology and the isotopic ratios provided good discrimination between the potential sources of sediment. The isotopic signatures varied in a systematic manner that paralleled the alternating sedimentary layers suggesting a switch in the dominate source of sediment between the warmer periods (ice retreat) and the cooler periods (ice advance). By estimating the relative contributions from the different sediment sources the patterns of erosion could be inferred and this information provided some direct evidence of the locations of ice margin retreat. Overall, this research provided information that helps predict the future dynamics of the Antarctic ice sheet under a warming climate.

4. Archaeological and geoarchaeological applications

There is a long tradition of geological “provenance” or source fingerprinting research in archaeology and geoarchaeology. Two broad areas of research activity may be identified. The first is aimed at a better understanding cultural processes of raw material exploitation and use, while the second involves a suite of geological and anthropogenic processes associated with the creation of the archaeological record. They may be summarised as studies investigating the procurement of sedimentary material or natural depositional processes.

Procurement involves studies that explore the source of geological raw materials used by past societies in the production of artefacts such as stone tools, pottery, jewellery, statuary, etc. and the built environment (e.g. building stones, mortars, floor and wall plasters). These activities involved the procurement of hard rocks including flint, chert, obsidian or marble, as well as unconsolidated fine-grained sediments. Studies of raw material sources can provide valuable information on how ancient societies interacted with the landscape; they can also provide evidence of past trade and exchange especially when distant sources were involved.

A good deal of research has focussed on the origin of naturally deposited sediments associated with the formation of an archaeological site such as the sediment matrix in a limestone rockshelter or the deposits associated with the burial of ancient settlements on a floodplain. Such deposits are typically the product of natural geomorphological processes but can help to link on-site archaeological records with palaeoenvironmental datasets obtained from the wider landscape. This section will present examples from each of these broad areas of research but will focus on studies designed to identify the source of fine-grained sediments through the use of a fingerprinting approach. This therefore excludes the large body of work on provenancing artefacts made from hard rocks and minerals (e.g. Salgán et al., 2015).

4.1 Fine-grained sediments as raw materials and sourcing clays for pottery

Within the broad field of raw material sourcing there is a very active area of research that has many similarities to the fluvial sediment source fingerprinting described in previous sections. This involves locating the source of fine-grained deposits (clays, silts and/or sands) that, because of particular physical and chemical properties, were deliberately targeted in the production of items such as pottery, wall plasters and building mortars. In the case of pottery production, potters often made use of the same sediment sources over many generations to secure clays and tempers with desirable characteristics. This kind of research allows the exploration of long-term linkages between the people and landscapes of the past (Michelaki et al., 2015). Studies of raw material procurement using a source fingerprinting approach are integral to a better understanding of many aspects of past human behaviour and technology. In the study of ancient buildings, they can shed light on the history of construction phases and even restoration efforts in antiquity; a good deal of work, for example, has focused on the composition of Roman mortars in an attempt to better understand the technology behind the extraordinary durability of Roman building materials (De Luca et al., 2015).

Because the mineralogical and geochemical signatures of the raw materials used in the production of many ceramics (e.g. pottery) are preserved in the finished product, the source of the raw materials can often be identified in the landscape using a source fingerprinting approach. The preparation of materials to be worked into a finished pot can involve mixing sediments collected from quite different and distinctive sources to achieve the optimum combination of fine clay matrix and temper. The temper is normally some kind of angular sand-sized material, with a limited particle size range, that is added in known proportions to the clay to control shrinkage and help the pot withstand high temperatures. It is typically procured from a different location to the source of the clay matrix and therefore provides another dimension to ceramic raw material sourcing projects. The composition of the temper in a pot sherd can commonly be identified in thin section using traditional petrological methods (e.g., Gonzales et al., 2015). Table 4 shows a number of sediment fingerprinting studies and a range of tracer properties applied to the study of ceramic raw material sources.

In a recent study of sediment sources and pottery composition in northern Ghana, Owen et al. (2013) were able to recognise differences in raw material procurement practices between modern and ancient potters using a fingerprinting approach. The potential raw materials were found to cluster into three distinct geographical zones. By combining mineralogical and geochemical data with microstructural observations of pot sherds, they were able to establish linkages between the pottery wares and local sediment sources. Multidimensional scaling (MDS) was used to identify compositional similarities and dissimilarities in the dataset. Mass balance calculations were carried out to establish the extent to which the sediment sources could account for the major and minor element compositions of the pot sherds. It was found that Late Stone Age potters mixed clays from a local escarpment with sediments collected from a local stream bed. The presence of tiny granite clasts and angular granite-derived minerals in some pot sherds pointed to deliberate collection and crushing of granite for use as a temper in pot manufacture (Owen et al. 2013). The source of an aluminous component identified in the pottery could not, however, be identified.

On the Greek island of Aegina in the southern Aegean Sea, Christidis et al. (2014) developed an integrated multi-method approach to establish the source of clays used by Bronze Age potters. They made use of geographical, petrographic, mineralogical, mineral chemistry, and geochemical data to create a substantial database to characterise the composition of the ceramics and to allow effective discrimination between potential sources of clay-rich sediments on the island. Christidis et al. (2014) were able to isolate a distinctive outcrop of Pliocene volcanic clay as the exclusive raw material source for the distinctive Bronze Age ceramics on Aegina. They used the same approach to exclude older Pliocene marls from the same location as a potential raw material source. In common with some of the early quantitative sediment source fingerprinting work in river basins (e.g. Walling and Woodward, 1995), they stressed the importance of using a multi-tracer approach to avoid ambiguous outcomes.

Whether a project is seeking to establish the composition of a pot sherd or a sample of fluvial suspended sediment or use this information to trace the source of the constituents in the wider landscape, the guiding principles are essentially the same. It is also important to appreciate that suspended sediment samples from river basins are complex environmental mixtures that typically include material from multiple diffuse source types. In contrast, ceramics can be produced from raw materials from one or two distinctive point sources. The principles of source identification are essentially the same in each case, however, but this can sometimes make identification of the dominant raw material source both theoretically and practically more straightforward in these archaeological contexts. There is clearly a great deal of scope for collaboration and cross-fertilisation of ideas between these sediment source fingerprinting communities – especially in relation to the statistical treatment of source data and the unmixing of target samples whether these end products result from natural or anthropogenic mixing. In fluvial geomorphology, watershed sediment budget investigations help us to better understand the operation of watershed processes and the evolution of landscapes. In a similar way, linking the composition of a pot to raw materials in the landscape helps us to better understand cultural processes and decision making in the past and directly links archaeological sites and past societies to their wider landscape (Michelaki et al., 2015; Ratto et al., 2015).

4.2 Sediment sources and archaeological site formation

An understanding of sediment sources can also provide valuable information on the formation of depositional records in many kinds of archaeological site (see Goldberg and Macphail, 2006). In the case of rockshelters and caves formed in hard limestone bedrock, the fine-grained components within

Pleistocene and Holocene fills are typically derived from off-site sources since the host rock contains only very small amounts of insoluble clays and silts (Woodward and Bailey, 2000). A range of mechanisms can be involved in the transport of fine sediments from off-site sources into a cave or rockshelter including, most commonly, aeolian, colluvial, and fluvial processes (Farrand, 2001; Woodward and Goldberg, 2001).

The Late Upper Palaeolithic rockshelter site of Boila is located next to the Voidomatis River in Northwest Greece. It contains a sequence of Late-glacial slackwater flood deposits that have been dated using radiocarbon (Figure 5). These slackwater sediments are underlain by coarse-grained fluvial gravels that were deposited by high energy meltwater floods during the last cold stage (Woodward et al., 2008). The contribution to the slackwater deposits from seven potential geological sediment sources in the upstream drainage basin was established via a quantitative sediment source fingerprinting approach that used XRF and magnetic susceptibility (Hamlin et al., 2000; Woodward et al., 2001). The magnetic susceptibility data were helpful in ascertaining the extent of any post-depositional weathering in each of the slackwater flood units. Traditional petrological analysis using thin sections was also employed as an independent test of the fingerprinting results. Such checks can be extremely useful, but they are rarely carried out in sediment source fingerprinting studies. The sediment source data from Boila and the local Pleistocene record showed that the occupation of the rockshelter by Upper Paleolithic hunters only took place following climatic amelioration in the region after the deglaciation of the basin headwaters towards the end of the last cold stage. The sediment source data from the rockshelter showed that a distinctive glacial input to the fine-grained load of the Voidomatis River dominated the cold stage river but was rapidly replaced by non-glacial sediment inputs after about 17 ka (Woodward et al., 2008) (Figure 5). Sediment delivery from gullies on flysch bedrock has dominated the fine sediment load of the Voidomatis River for much of the Late-glacial and all of the Holocene.

In a very different setting, Kourampas et al. (2009) studied the changing sources of the sediment in a Late Pleistocene to early Holocene (>31,000 to c. 7800 years BP) granite rockshelter record in humid tropical southwest Sri Lanka. They developed an allogenic sediment index to quantify the changing input of fine sediments to the rockshelter over time. In a tropical dry environment, Herries (2006) employed mineral magnetic tracers to detect the changing input of very fine-grained sediments derived from weathered soils that were blown into Sibudu Cave in South Africa during colder and dustier phases of the Late Pleistocene. At this location, major changes in sediment sources and the dominant geomorphological processes were related to shifts in monsoon intensity after the Last Glacial Maximum. By quantifying long-term changes in fine sediment sources it is possible to link the formation of such sediment records (and the archaeology associated with them) to changes in the operation of geomorphological processes in the wider landscape (see Woodward and Bailey, 2000).

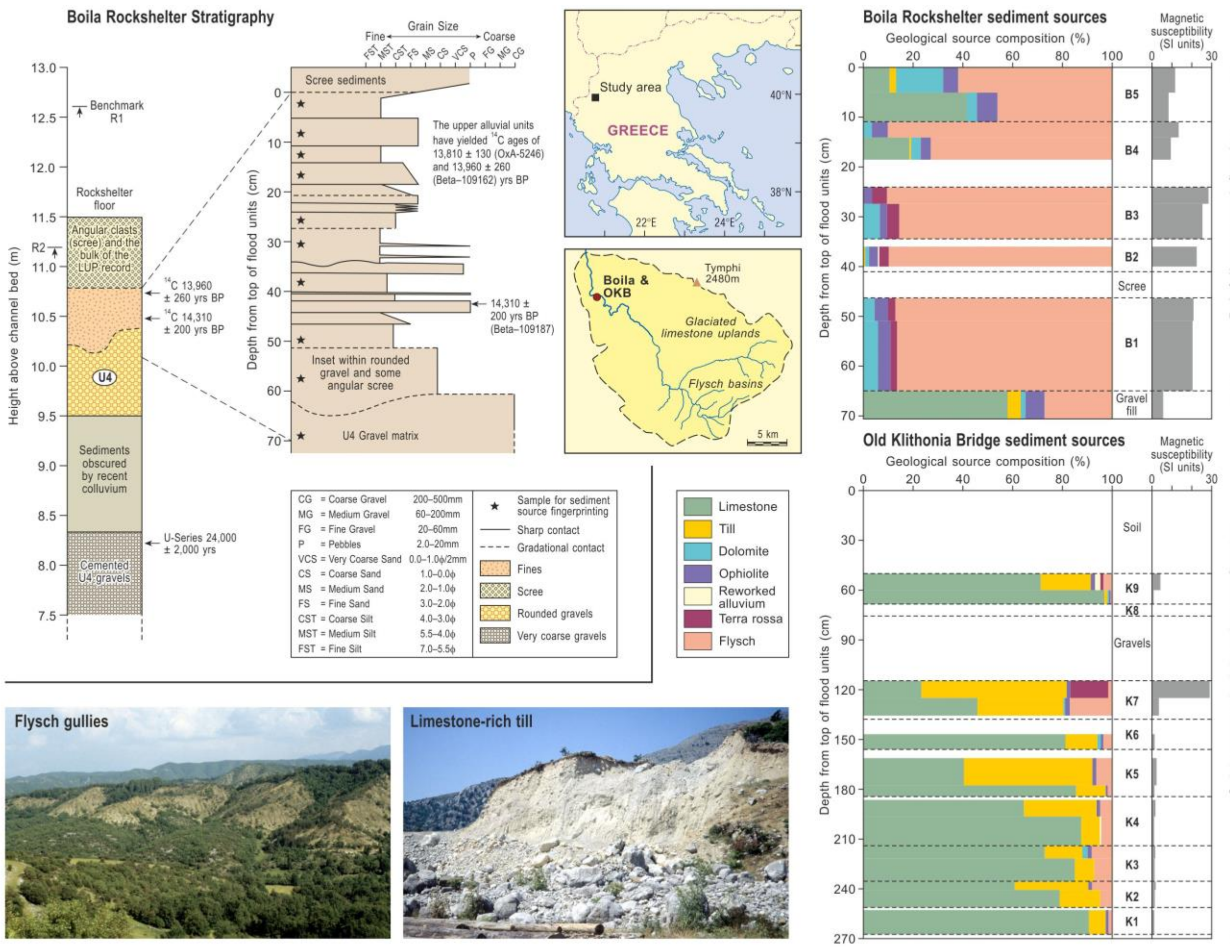

Figure 5. Sediment fingerprinting of fine-grained fluvial slackwater sediments deposited in the lower reaches of the Voidomatis River basin in NW Greece towards the end of the last cold stage. The inset map shows the location of the Boila and Old Klithonia Bridge (OKB) sites on the left bank of the Voidomatis. The stars on the Boila Rockshelter stratigraphy mark where samples were taken for sediment provenance analysis. Samples (n = 52) of potential source materials were collected from across the catchment. See Hamlin et al. (2000) and Woodward et al. (2008) for further details. Fine sediments from gullied lowland flysch landscapes have dominated the suspended sediment load of this river during the Late glacial and Holocene. The suspended load was dominated by glacially-comminuted limestone silts and clays during periods of glaciation on Mount Tymphi (Woodward et al., 1992; Hughes et al.,

2006). A key feature of these Late Pleistocene sediment fingerprinting data is the abrupt end of the meltwater-dominated system during the last deglaciation.

5. Forensic applications

Soil forensics is similar to sediment source fingerprinting as it utilizes the physical and biogoechemical properties of soils (i.e., fingerprints) as trace evidence (Ruffell, 2010). There is typically a two-way exchange of trace evidence (e.g., soil or sediment) when subjects and objects come into contact with each other. This is where soil forensics can establish or refute a link between people and objects and a particular location of interest using soils as trace evidence. Table 5 shows the wide range of scenarios in which soil forensics have been studied, or used, along with the soil properties and analytical techniques utilized to compare soil samples within the context of forensic investigations. There are many different mechanisms by which soil can be transferred away from a crime scene including cadavers, clothing, footwear, vehicles and tools (see Table 5 for a list of references).

Soil forensics draws many parallels with the other applications of sediment source fingerprinting outlined in this review. Similarly, locating and appropriately characterizing source materials (e.g., spatial variability; see Section 7.1) is important and other evidence can be used to narrow the sampling area (e.g., near footprints) (Pye et al., 2006; Dawson and Hillier, 2010). In some cases, the potential sources of the soil or sediment may not be immediately known and the potential source needs to be deduced from the evidence collected (e.g., Lombardi, 1999; see below for a description of the case). The selection of independent measurements (i.e., some soil properties are correlated; see Section 7.2) is important as this provides the strongest case for determining the origins because the use of non-independent properties as supportive evidence may be problematic in a court of law (Morgan and Bull, 2007a). The contamination or mixing of soil/sediment pre-, syn- and post-forensic investigation are important considerations as this may lead to erroneous conclusions (Morgan and Bull, 2007a). This is where the development of protocols in documenting, collecting, preserving, preparing, analyzing and interpreting the data is important (also see Section 7.3). In a similar way to the issues surrounding particle size correction factors (also see Section 7.4), it is important to have an understanding of the mechanism of soil/sediment transfer as the process can be particle-size selective (e.g., clay tends to stick to footwear whereas sand does not) (Dawson and Hillier, 2010).

One of the biggest differences between soil forensics and other fingerprinting applications in earth and environmental sciences is in the philosophy of the science. Soil forensics often seeks to *exclude* potential sources whereas the other applications are more concerned with identifying and confirming potential sources (Morgan and Bull, 2007a). For example, soil samples sharing similar properties cannot be said to have the same origins, but only that it cannot be excluded from having been derived from the same location. Another difference relates to the size of soil samples, for example, Ruffell and Sandiford (2011) only recovered 300 soil particles from an article of clothing, whereas 1 – 10 g of soil/sediment is typically used in earth and environmental science applications. The types (e.g., non-destructive and destructive analysis) and order of soil analysis needs to be carefully selected due to the small masses that are often recovered (Dawson and Hillier, 2010).

Very few cases are reported fully in the literature due to the sensitive nature of criminal investigations. Lomardi (1999) presents an interesting case report in which soil forensics contributed to a murder investigation. On March 16, 1978, the Italian Prime Minister, Aldo Moro, was kidnapped and on May 8, 1978 he was found dead, from gunshot wounds, in the trunk of a parked car near the centre of Rome. Small amounts of sand and soil were collected from the victim's trouser cuffs, shoes and coat pockets as well as from the floor, fenders and tires of the vehicle in which the victim was found. Along with the soil and sand there were also traces of plant material, asphalt, fibres and an assortment of building materials (e.g., brick chips). These samples were used to help identify the origin of the vehicle and the last whereabouts of the victim. A range of analytical techniques was used to characterize both the sand and soil samples including: particle size analysis (sieving), particle morphology (microscopy), mineralogy (x-ray diffraction, scanning electron microscopy and polarized microscopy), soil colour (microscopy), pollen analysis (microscopy) and the identification of micro-fossils (microscopy).

Results from the analysis showed that the sand was well sorted and had a rounded to sub-rounded morphology which is typical of a beach deposit. Based on the colour, mineralogy and the micro-fossil assemblage the area of origin was narrowed down to the Tyrrhenian coast near Rome (~150 km in length). Since there were limited data on the composition of sand along this stretch of coast a systematic sampling of sand was initiated. Samples were collected at beach sites with road access, for a total of 92 sampling locations, with 1 – 3 samples collected at each location. Initial screening, using microscopy, eliminated 22 samples as having clear differences from the collected evidence. The remaining 70 samples were analyzed using polarized microscopy. The analysis narrowed the initial 150 km search area down to a more manageable 11 km stretch of the coast.

The soil samples contained a mixture of halloysite-rich clay, glassy scoriae and an assemblage of volcanic minerals with an overall reddish-brown colour. The soil samples were identified as volcanic in origin; however, this type of soil is very prevalent in the area covering more than 6000 km^2. The vast areal extent of this soil type prevented any meaningful insight to the origin of the soil. Pollen extracted from the soil was identified as coming from Cypress and Hazel trees, which is produced in the winter. This information established that the soil adhered to the vehicle prior to the abduction of the victim. The fingerprinting of the soil and sand evidence found on the victim and car was one line of enquiry that helped to reconstruct a timeline of events leading up to the murder and to corroborate the testimony from potential suspects.

6 Human health applications: fingerprinting airborne particles

Soils and sediments, and the contaminants associated with them, can be a risk to human health (Abrahams, 2002). The main pathways by which soils and sediments and associated contaminants enter humans are: direct ingestion; inhalation through nose and mouth; and adsorption through the skin. Consequently, there has been an interest in determining the origin of soils and sediments known to be a risk to human health. One such application is the identification of the sources of very fine airborne particles, especially in urban areas. In this context, particulate material with a diameter less than 10 µm (i.e. PM_{10}) or 2.5 µm ($PM_{2.5}$) is of greatest concern because when they are inhaled they are able to penetrate deep into the alveoli where they can be deposited and absorbed. The effects include chronic lung disease, lung cancer, influenza, asthma, and increased mortality. In addition to the small size of such particles, they are also detrimental to human health because of the contaminants associated with them, such as metals and polycyclic aromatic hydrocarbons (PAHs). Numerous studies have collected such particles and used geochemical fingerprints to identify sources. The approach is broadly similar to that described above for other applications, and involves establishing diagnostic chemical (e.g., trace elements, organic elements, PAHs) and physical (e.g. particle morphology) fingerprints, and the use of statistical and modelling approaches (e.g. Receptor Modelling, Positive Matrix Factorization, Principal Components Analysis) to quantitatively determine source types or spatial areas (Breed et al., 2002; Dogan et al., 2008; Callén et al., 2014; Huang et al., 2014; Suman et al., 2014). Typically, airborne particles are collected using specialised samplers with filters. Some studies in the urban environment have determined sources of road-deposited sediment (RDS) which is itself a major source of PM_{10} and $PM_{2.5}$ and an important contributor to the fine-grained sediment load of urban rivers (Owens et al., 2011; Karanasiou et al., 2014).

For example, Karanasiou et al. (2014) collected samples of RDS from Madrid, Spain, in 2009. They used chemical analysis (major and trace elements, carbon) of the <10 µm fraction to establish fingerprints and applied Positive Matrix Fractionization to determine its source. Their results (Fig. 6) illustrate the importance of both natural and anthropogenic sources of fine-grained RDS and thus PM_{10}, including sources from the construction industry. Such information was used to provide guidance on street sweeping approaches in Madrid to minimise detrimental impacts on human health. Other studies (e.g. Merefield et al., 1999, 2000) have used similar approaches (including SEM/EDAX and XRD analysis) to determine sources of airborne particles associated with construction, mining and other resource extraction industries.

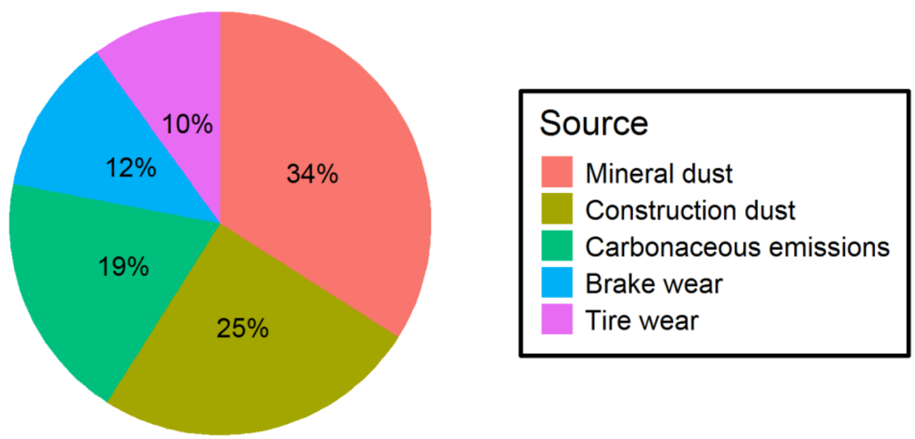

Fig. 6. Sources of road deposited sediment (<10 µm fraction) for samples collected from Madrid, Spain, in 2009 determined using chemical analysis and Positive Matrix Fractionization (modified from: Karanasiou et al., 2014).

7. Methodological considerations and recommendations

While there has been a rapid growth in studies undertaking sediment source fingerprinting in a range of environments and applications, there are still aspects of the approach that warrant further improvement in order to increase its robustness and acceptability. The following sections consider some of these needs; further considerations and background can be found in Walling (2013) and Smith et al. (2015).

7.1 Collection of soil and sediment samples

As with most areas of earth and environmental sciences, field sampling of the materials under investigation (i.e., soils and sediments) represents a key consideration. The design of the sampling programme needs to be flexible to suit the specific environment and research objective (e.g., the various applications described in Sections 3 to 6), since there cannot be a single approach to the sampling of soils and sediments. However, it is important to optimise the sampling of soils and sediments in order to characterize both the spatial and temporal variability of fingerprint properties. For example, in the case of surface soil material, samples of the upper 0-2 cm layer are usually collected and this is assumed to represent the material mobilised by erosion processes and delivered to stream channels. Even with adjustments for particle size effects (see section 7.4) such material is unlikely to be truly representative of the material that is delivered to channels, coastal areas or oceans. Thus, it may be more appropriate to sample eroded soil and sediment that is being actively transported. This could be achieved by sampling during runoff events or installing suitable samplers (i.e., Gerlach troughs). Furthermore, some studies (e.g. Walling et al., 1999; Laceby et al., 2015b) have used suspended sediment samples collected from tributary streams to characterize the sources (type and spatial) of suspended or deposited sediment in downstream waterbodies, thereby reducing the need for correction factors. In addition, there are concerns surrounding the nature of the most appropriate sampling design and how effectively it characterizes a particular source (e.g., random, transect or strategic sampling) or tracer property (Wilkinson et al., 2015). Understanding the geomorphological context in terms of the erosional history of the landscape (at least in a general way) and the dominant water and sediment transport pathways can be helpful in the design of an appropriate sampling framework.

In a similar way, it is important to ensure that the target sediment samples are representative of the materials for which the study is attempting to determine its sources. Typically, actively transported fluvial sediment or atmospheric particles are sampled, or it is the fine sediment stored in the upper layers of the channel bed, or estuarine/coastal zone that is collected. In other situations, when the focus of the study is on reconstructing past sediment sources, rockshelter, floodplain, wetland, lake or ocean sediments are collected. In all cases it is important to consider if the samples are temporally and spatially representative. For example, Koiter et al. (2013b) and Wethered et al. (2015) have demonstrated that the location of the sampling point (i.e., headwaters or downstream basin outlet) can influence which sediment sources dominate due to issues of scale and hydro-geomorphological connectivity; this links back to the reason for the study and to the research objectives. In terms of the temporal representativeness of suspended sediment, Walling et al. (1999) suggest that samples be collected over a range of flow conditions, and that the data be flow-weighted (as opposed to simple averages) so that the final source apportionment results account for variations in sediment fluxes.

It is also necessary when developing a source material and sediment sampling protocol to consider the requirements of the statistical and numerical approaches that are used to convert fingerprinting property data into quantitative estimates of sediment sources. For example, in the case of source materials, there is a need consider the number of samples to be collected so as to provide a meaningful representation of the fingerprinting properties of that source. The number of samples to be collected should be guided be the spatial variability of the fingerprint properties in that source, which is likely to vary for different properties (e.g. fallout radionuclides, geochemical properties, colour parameters, CSSIs) and different sources (e.g. cultivated fields, forest, channel banks). In addition, the spatial variability of source materials is likely to increase with larger study areas. Furthermore, while the concentrations of some properties may be essentially random within sources, others (e.g. fallout radionuclides) may exhibit some spatial structure due to erosion and deposition processes or environmental gradients (e.g. Wilkinson et al., 2015) and may require a stratified sampling protocol (e.g. Wilkinson et al., 2015) and/or the collection of samples along transects or environmental gradients (e.g. Koiter et al., 2013b). Thus, while some studies have provided estimates of the number of samples to be collected (typically of the order of 30 composite samples per source type), in reality each study area is different and requires site-specific considerations.

7.2 Selection of fingerprint properties

While it may be possible to measure soil and sediment samples for hundreds of different physical, chemical and biological properties, of paramount importance is the selection of properties that make sense in terms of how fingerprints have developed (Collins and Walling, 2002). This is required for three reasons: (i) so that the source and sediment samples are collected in the correct way (e.g., soils and sediment to the correct depth, as in the case of fallout products like ^{7}Be or artificial amendments like fertilisers); (ii) so that laboratory analysis is cost-effective (i.e., avoid unnecessary analysis); and (iii) so that the user can understand and interpret the data. Again, this will depend on the study site and the purpose of the investigation. In most cases the reason for the study can help to inform the selection of suitable fingerprints. Thus, if the aim is to investigate the relative contributions of topsoil compared to channel bank sources, then properties that either label surface materials (e.g., some organic properties, fallout radionuclides and other fallout products, and agricultural amendments) or are enriched in subsoil materials (i.e., geochemical properties that reflect bedrock or surficial materials) are likely to be worth considering.

Once soil and sediment properties have been selected and measured, an addition step prior to further statistical and numerical analysis is to plot the data (i.e. bi-plots; Figure 2) and consider if the data make sense in the context of any prior knowledge concerning the environmental distribution of a given fingerprint property. For example, if fallout radionuclides are elevated in subsoil or channel bank material compared to topsoil, then either the property is not appropriate as a fingerprint and/or may have been compromised (e.g., soil disturbance) or there are additional processes that are occurring which warrant further investigation. For example, Owens et al. (2012) measured some subsoil samples with high ^{137}Cs and unsupported ^{210}Pb values in a wildfire-affected watershed in British Columbia, Canada (see Fig. 2). In this case, the complete combustion of individual trees had exposed subsoil materials to new fallout of unsupported ^{210}Pb and some redistribution of old ^{137}Cs-enriched surface soil. For the fingerprinting technique to be robust and repeatable it is crucial that the processes controlling their behaviour within the particular landscape or river basin are well understood and can be predicted. Furthermore, fingerprints that fall below the detection limit in one or more samples are typically discarded without consideration as to whether the fingerprint has the potential to provide good discrimination. There are statistical procedures available to deal with left-censored data but their uses in sediment source fingerprinting studies have not been fully explored.

7.3. Laboratory analysis and data reporting

While there has been a reasonable amount of debate on appropriate field sampling and statistical procedures (e.g. Walling, 2005, 2013; Koiter et al., 2013a; Smith et al., 2015), there has been much less discussion of laboratory analysis. In this respect, appropriate sample storage and preparation prior to analysis represent fundamental requirements, particularly as the requirements are likely to vary for different analyses. This is likely to be especially so for organic tracers, and to some extent for colour analyses due to issues associated with geochemical stabilization. As sample analysis can be destructive for some determinants (e.g., particle size, organic matter via loss-on-ignition, some geochemistry) there is also a need to consider the order of analysis if a sample is being analysed for a range of determinants (i.e., particle size, colour, geochemical, radionuclide, CSSI properties). This is especially relevant for low-mass samples.

Once samples have been analysed, then there should be a detailed and complete reporting on analysis, including the reporting of any deviations from standard protocols. This should include the reporting of fingerprint data in publications. At a minimum, this should take the form of summary table(s) containing source and sediment fingerprint data without any corrections or modifications. It could also extend to the inclusion of complete datasets tabulated in supplementary material. Such a step would then allow readers to run the same procedures described in the paper to confirm the outcomes of the data processing and source unmixing. This enhanced level of transparency would be likewise supported by reporting of specific statistical programmes or R packages used in the analysis, along with inclusion of any bespoke code in the supplementary material (see section 7.6). Such steps could make significant progress towards reducing ambiguities and ensuring reproducibility in data treatment and analysis between sediment source fingerprinting studies.

7.4. Particle size and organic matter correction factors

One of the most controversial aspects of the sediment source fingerprinting technique is the use of correction factors to account for differences in some properties between sources and sediments. The two main correction factors relate to the particle size distribution and organic matter content, as many fingerprint properties are often related to these two parameters (Horowitz, 1991). In many studies (e.g., Walling and Woodward, 1995; Collins et al., 1997a; Walling et al., 1999; Owens et al. 2000) relatively simple correction factors are determined based on the ratio of measures of the particle size distribution (i.e., median particle size or specific surface area) and/or organic matter content (i.e., OM%) of the source and sediment samples. It is typically assumed that the relation between tracer property concentration and particle size or organic matter content is linear. However, some studies (e.g., Foster et al., 1998; Russell et al., 2001; Smith and Blake, 2014; Taylor et al., 2014) have demonstrated that such relations can be more complex, and may be tracer- and site-specific. Other studies (e.g., Stone and Walling, 1997) have shown that the particle size composition and organic matter content of sediment can change as it moves from source to sink. For example, Koiter et al. (2015) used a recirculating flume to simulate river channel conditions, and documented that both particle size and organic matter content changed over time (i.e. distance travelled) and that the degree of change was influenced by the depth and porosity of the channel bed sediment. Indeed, some studies (e.g., Martinez-Carreras 2010; Dutton et al., 2013; Koiter et al., 2013a; Smith and Blake, 2014; Palazón et al., 2015a) have stated that particle size and organic matter correction factors are inappropriate. Smith and Blake (2014) have demonstrated that such manipulation of the raw data can significantly change source apportionment results. Thus it is important to consider if such corrections factors are appropriate, and clearly further research is required to address this concern.

Other studies have attempted to overcome problems of particle size and/or organic matter influences by focussing on a specific size fraction. While most studies restrict analysis of source materials and sediment samples to the <63 μm fraction, other studies have fractionated source and sediment samples prior to analysis. For example, Haddadchi et al (2015) separated samples into fine sand (63-212 μm), silt (10-63 μm) and fine silt and clay (<10 μm). The selection of finer size fractions can remove some of the confounding influences of changing particle size composition between source and sink, but it may also limit the representativeness of the results, as most suspended sediment in rivers is >2 µm (Walling et al., 2000).

The choice of particle size fraction for analysis can influence the source apportionment results (Haddadchi et al., 2015). Hence, it is important that studies explain the choice of size fraction for analysis in relation to the research context and objective. Previously the choice of the <63 µm fraction was justified because it was considered representative of sediments largely transported in suspension that were of most concern in terms of water quality impacts and the transport of particle-bound contaminants (e.g. Walling et al., 1999). However, such a choice may be unsuitable in environments dominated by sandy materials or where the research objective is unrelated to water quality. Alternatively, a focus on a finer fraction such as <10 µm may be justified by concerns related to the specific impacts of these finer-grained sediments on particular ecosystems, such as coral reefs (Bainbridge et al., 2016) and oceanic environments. It may also be justified from a process perspective, for example, in the study of sediment source contributions to post-fire debris flows, where fine sediment supply is an important factor in debris flow initiation from burnt hillslopes (Smith et al., 2012).

7.5 Conservative behaviour of soil and sediment properties

One of the fundamental assumptions underpinning the source tracing and fingerprinting technique is that the properties of the collected sediment samples are directly comparable to those in the potential source materials. In some cases, allowance is made for differences in the particle size and organic matter content between the two types of materials (see section 7.4). However, Koiter et al. (2013a) have recently reviewed the broader environmental literature on the range of soil/sediment properties used as fingerprints and demonstrated that for several properties there exists the potential for considerable modification in concentrations as sediments move through river basins; i.e., from hillslopes through riparian zones, into and through river channels, and into deposition in floodplain, lake/reservoir, estuarine and marine environments. Several properties, such as phosphorus, are known to exhibit non-conservative behaviour during fluvial transport and short-term storage in river channels (e.g., Withers and Jarvie, 2008), while other properties are known to undergo transformations in medium- to long-term storage elements such as floodplains, lakes, wetlands and oceans due to changes in redox, pH, salinity and other environmental conditions (Hudson-Edwards et al., 1998; Owens et al., 1999; Foster et al., 2006; Pulley et al., 2015).

While it may be possible to account for such behaviour, most sediment source fingerprinting studies do not do this in a rigorous or standardized way. The conservative behaviour of tracers can be assessed through experimentation. For example, Poulenard et al. (2012) assessed the conservative behaviour of Diffuse Reflectance Infrared Fourier Transform Spectrometry (DRIFTS;) including colour) properties by placing samples in micro-porous bags and submerging them in the river for up to two weeks. They found that the DRIFTS signature was sufficiently conservative to be used as a potential fingerprint property. In addition, Legout et al. (2013) assessed the conservative behaviour of the colourimetric properties of source materials in a similar experiment for up to 63 days, and found comparable results.

A common procedure to help identify if fingerprint properties are behaving conservatively is to compare the property values (mean, median or range) of the sources to those of the collected target sediment, sometimes called the “range test”, with the idea that if the sediment values for a particular property fall outside of the values for the potential sources that either the property is exhibiting non-conservative behaviour or that not all sources have been identified or fully characterised. Properties that fail this test are often discounted as potential fingerprints and removed from further analysis. This approach represents a useful test of property behaviour, although it represents only part of the solution. The range test cannot definitively identify all tracers that are behaving non-conservatively. Rather it could also reveal the existence of an un-sampled source or a tracer property may have been altered by non-conservative behaviour during mobilization and deposition but remains within the source range. This could affect the source apportionment but would not be captured by the application of the range test alone. Others (e.g. Koiter et al., 2013b; Kraushaar et al., 2015; Laceby et al., 2015b) advocate that tracer selection should not be based purely on statistical procedures and that knowledge of the hydrological, geomorphological and geochemical processes controlling tracer behaviour should also be used to help select appropriate soil and sediment properties for sediment fingerprinting. Koiter et al. (2013a) make some more general recommendations to help identify, and reduce, problems associated with non-conservative behaviour of fingerprinting properties, but further research is needed to understand such behaviour and incorporate such understanding into the sediment source fingerprinting technique.

7.6 Statistical and unmixing model approaches and incorporation of uncertainties

Statistical tests and unmixing models are crucial for enabling raw data on property concentrations to be converted to meaningful values of source contributions. There are several approaches available for both the statistical procedures (e.g., Walling et al., 1993; Yu and Oldfield 1993; Walling and Woodward, 1995; Collins et al., 1997a) and unmixing models used. Others (e.g., Collins et al., 2010, 2012a, b) have suggested some recent developments to the statistical framework. In the case of unmixing models, earlier frequentist-based approaches used optimization to minimise residuals between sources and sediment mixtures to estimate the unknown proportional source contributions (e.g., Collins et al., 1997a; Walling et al., 1999). These approaches are now increasingly being superseded by Bayesian models drawn from studies of diet within ecology (i.e., IsoSource, SIAR, MixSIAR; Parnell et al., 2013) which are showing much promise, and consequently are recommended by organisations such as IAEA and are increasingly used in recent studies, especially in earth sciences (e.g., Dutton et al., 2013; Koiter et al., 2013b; Barthod et al., 2015; Cooper et al., 2015a, b). Bayesian unmixing models have several advantages over frequentist-based models, including: (i) better incorporation of prior information; and (ii) incorporation and reporting of uncertainties. Indeed, unlike many previous unmixing models, Bayesian models can explicitly capture many of the sources of uncertainty presently associated with the sediment source fingerprinting approach including: spatial variability in fingerprint properties across the study area, uncertainties associated with instrumental precision, covariance between fingerprint properties, and residual model errors (Cooper et al., 2015b). Furthermore, several of the Bayesian models are open-source with standard operating procedures (SOPs), often with graphical user interfaces (GUIs) and operated using R (e.g. MixSIAR), which has increased their usage in a variety of settings. Such an approach provides the opportunity for some level of standardisation and should enable researchers to reproduce and compare results.

While studies in earth sciences have tended to use unmixing models (either frequentist- or Bayesian-based), other fields have used different approaches, such as Positive Matrix Factorization models, although often the underlying concepts are similar. In several cases the sources are inferred using principal components analysis or similar statistical procedures (e.g. studies of airborne particulates within human health: section 6), or through the use of elemental ratios (e.g. provenance studies in ocean sciences: section 3.5).

The limitations of unmixing models and the fingerprinting approach as a whole, including estimation of uncertainties, are not well quantified and can be difficult to assess. Haddadchi et al. (2014b) attempted to assess the accuracy of several different unmixing models, and found that some perform better than others. Furthermore, weightings factors have been widely used in the frequentist optimization-based models to account for the differing ability of tracers to discriminate between sources or to enable tracers with lower within-source variability to exert a greater influence on source apportionment results (Collins et al., 2010). However, work by Laceby and Olley (2015a) and Haddadchi et al. (2014b) showed that the inclusion of such weightings actually reduced the accuracy of source contribution predictions compared to known artificial mixtures. It follows that the inclusion of any such parameters requires explicit justification and support based on the evaluation of model outputs compared to experimental or synthetic datasets. Indeed, the use of synthetic or virtual sediment mixture data is an efficient way of evaluating the performance of different unmixing model structures or data treatments (Palazón et al, 2015b), and it is recommended that this practice be more widely adopted by sediment fingerprinting studies alongside measured datasets.

7.7 Linking source fingerprinting to sediment budgets

Most sediment source fingerprinting studies have determined the *relative* contributions from different sources and not absolute mass contributions. In most cases, the former information is adequate. However, when determining sediment sources over time (e.g., events, seasons, years, millenia) it is more important to either determine mass contributions from the sources and/or consider sediment flux and storage information. For example, it may be possible for the relative contribution from two sources to remain constant from year to year but the absolute mass flux to change. Thus some studies (e.g., Smith et al., 2011) have linked sediment source data to sediment flux data to determine if sediment mass contributions from different sources have changed.

In a similar way, changes in upstream storage could mask changes in sediment sources (e.g., Trimble, 1983). For example, an increase in hillslope erosion rates in headwater areas could be offset by concomitant increased sediment storage on floodplains, such that no net change in relative sources is determined at a downstream sediment collection site if all other sources remain constant. Incorporating sediment source information within wider sediment budget investigations should help to ensure that an overall picture of sediment dynamics within a river basin or marine environment is obtained. Thus, sediment source fingerprinting represents one tool and is most effective when it is utilised with other tools (e.g. sediment budgets, monitoring, remote sensing).

8. Conclusion and perspective

Sediment source fingerprinting has emerged within the last few decades as an important tool that can be used for a range of applications, including those in earth and ocean sciences, (geo)archaeology, forensics and human health. While applications in forensic and health sciences may have longer histories, there has been a dramatic increase within earth sciences, particularly since the 1990s, reflecting the fact that the approach is able to provide useful information on landscape and watershed processes and, especially, as it can be used to inform management decisions. To date, within the earth sciences it has mainly been used within agricultural landscapes but its use within other settings (e.g., forested, urban, estuarine, marine) is increasing. This partly reflects the development of new tracers, such as CSSIs, REEs and clay mineralogy, which can help to tackle new research questions and be used in new environments.

It is apparent from this review that there is considerable commonality between the approaches of the various groups using fingerprinting to identify the sources of airborne and aquatic sediments. These include: (i) the need to collect representative samples of source materials and airborne or aquatic sediments; (ii) the selection of soil/sediment/dust properties that can effectively distinguish between potential sources; and (iii) the broad use of statistical and numerical approaches that are able to quantitatively apportion sediments to sources. Despite this commonality, there are often differences between the approach used which include: (i) the use of different tracer properties and combinations of properties; (ii) differences in the approaches used to account for issues of particle size and conservativeness; and (iii) differences in statistical and numerical approaches, and how to deal with uncertainty. As such, each discipline can learn from the others, and there exists the potential for cross-fertilisation. Indeed, the recent up-take by fluvial geomorphologists and other earth scientists of organic tracers such as stable isotopes, CSSIs and DNA – many of which were developed by biologists and ecologists for other applications (e.g., food webs) – shows that the process is underway. There are also some useful approaches to the selection of appropriate fingerprinting properties and statistical procedures used within other disciplines that could benefit earth science applications. These include procedures to select less mobile elements as fingerprint properties and the use of procedures to normalise data so as to remove some of the confounding influences associated with changes in particle size composition and organic matter content, as used in sediment provenance studies in marine environments (i.e., section 3.5).

One recommendation is that there is a need for comparison between the various tracer groups; for example, comparison between geochemical elements, fallout radionuclides, colour properties and CSSIs (e.g. Blake et al., 2012; Verhayen et al., 2014; Barthod et al., 2015). While each tracer type may give different results, reflecting the sources that they are able to distinguish – topsoil vs subsoil in the case of radionuclides, different crop types in the case of CSSIs – there should be internal consistency in the results. For example, if CSSIs identify that most of the organic component of the sediment is coming from the surface of cropland, then if other tracer groups identify that subsurface soils dominate (e.g., due to gully erosion) then there needs to be further investigation to assess if the difference is real (i.e., organic and mineral component derive from different sources) or if one tracer group is exhibiting non-conservative behaviour.

Similarly, there is a need to compare source fingerprinting results to other, independent lines of evidence. Such an approach is typical in geoarchaeological (section 4) and forensic (section 5) applications, where is it important to both place source fingerprinting or provenance findings within a broader context and to determine the validity of the findings; as such, earth science applications could learn from these disciplines.. Thus, if fingerprinting results suggest that channel bank erosion is the main sediment source yet ground-truthing (e.g., monitoring) and aerial photographs show no evidence for such erosion then there is a need for further investigation to either determine which approach is correct, or to identify the cause of the difference (e.g., intermediate storage between source and sediment collection). In both of these cases (i.e., internal comparisons and comparisons with independent approaches) there is a considerable amount to be learnt about both the techniques themselves and how landscapes function. Sediment source fingerprinting should be viewed as only one component of investigations into sediment (airborne, terrestrial and aquatic) processes and dynamics, and it should be part of a more holistic assessment of landscape and watershed behaviour and functioning.

The sediment source fingerprinting approach offers many advantages over more conventional techniques, such as landscape and watershed monitoring, given that information can be assembled relatively quickly and cheaply; although this in part depends on the actual approach adopted. In reality, the fingerprinting approach should be viewed as a complementary approach to these more conventional approaches. Realisation of the potential of the fingerprinting approach has seen its use expand into new applications as well as a growing interest and recognition of similar approaches in other disciplines. This review has highlighted some of these new applications, reviewed developments in cognate disciplines, and illustrated some of the areas requiring further work if the potential of the technique is to be fully realised.

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References

Abrahams, P.W., 2002. Soils: their implications to human health. Sci.Total Environ. 291, 1-32.

Alewell, C., Birkholz, A., Meusburger K., Schindler Wildhaber Y., Mabit, M., 2016. Quantitative sediment source attribution with compound-specific isotope analysis in a C3-plant dominated catchment (central Switzerland). Biogeosciences 13, 1587-1597.

Anderson, S.L., 2016. A clay source provenance survey in Northwest Alaska: Late Holocene ceramic production in the Arctic. J. Field Archaeol. 41, 1-17.

Armstrong-Altrin, J.S., Machain-Castillo, M.L., Rosales-Hoz, L., Carranza-Edwards, A., Sanchez-Cabeza, J.-A., Ruíz-Fernández, A.C., 2015. Provenance and depositional history of continental slope sediments in the Southwestern Gulf of Mexico unraveled by geochemical analysis. Continental Shelf Res. 95, 15–26.

Bailey, M.J., Morgan, R.M., Comini, P., Calusi, S., Bull, P.A., 2012. Evaluation of particle-induced X-ray emission and particle-Induced γ-ray Emission of quartz grains for forensic trace sediment analysis. Anal. Chem. 84, 2260–2267.

Bainbridge, Z., Lewis, S., Smithers, S., Wilkinson, S., Douglas, G., Hillier, S., Brodie, J., 2016. Clay mineral tracing and characterisation of Burdekin River and flood plume sediment. J. Soils Sediments 16, 687-706.

Barthod, L.R.M., Liu, K., Lobb, D.A., Owens, P.N., Martinez-Carreras, N., Koiter, A.J., Petticrew, E.L., McCullough, G.K., Liu, C., Gaspar, L., 2015. Selecting color-based tracers and classifying sediment sources in the assessment of sediment dynamics using sediment source fingerprinting. J. Environ. Qual. 44, 1605-1616.

Basu, A., Bickford, M.E., Deasy, R., 2016. Inferring tectonic provenance of siliciclastic rocks from their chemical compositions: A dissent. Sediment. Geol. 336, 26–35.

Belfiore, C. M., La Russa, M. F., Barca, D., Galli, G., Pezzino, A., Ruffolo, S. A., Viccaro, M., Fichera, G. V., 2014. A trace element study for the provenance attribution of ceramic artefacts: the case of Dressel 1 amphorae from a late-Republican ship. J. Archaeol. Sci. 43, 91-104.

Belmont, P., Pazzaglia, F.J., Gosse, J.C., 2007. Cosmogenic ^{10}Be as a tracer for hillslope and channel sediment dynamics in the Clearwater River, western Washington State. Earth Planet. Sci. Lett. 264,123–135.

Belmont, P., Willenbring, J.K., Schottler, S.P., Marquard, J., Kumarasamy, K., Hemmis, J.M., 2014. Towards generalized sediment fingerprinting with tracers that are conservative and nonconservative over sediment routing timescales. J. Soils Sediments 14, 1479-1492

Ben Slimane, A., Raclot, D., Evrard, O., Sanaa, M., Lefèvre, I., Ahmadi, M., Tounsi, M., Rumpel, C., Ben Mammou, A., Le Bissonnais, Y., 2013. Fingerprinting sediment sources in the outlet reservoir of a hilly cultivated catchment in Tunisia. J.Soils Sediments 13, 801–815.

Blake, W.H., Wallbrink, P.J., Doerr, S.H., Shakesby, R.A., Humphreys, G.S., English, P., Wilkinson, S., 2006a. Using geochemical stratigraphy to indicate post-fire sediment and nutrient fluxes into a water supply reservoir, Sydney, Australia. In Sediment dynamics and the hydromorphology of fluvial systems, Rowan JS, Duck RW, Werritty A (eds). IAHS Publication No. 306, IAHS Press: Wallingford; 363-370.

Blake, W.H., Walsh, R.P.D., Sayer, A.M., Bidin, K., 2006b. Quantifying fine-sediment sources in primary and selectively-logged rainforest catchments using geochemical tracers. Wat Air Soil Poll: Focus 6, 615–623.

Blake, W.H., Wallbrink, P.J., Doerr, S.H., Shakesby, R.A., Humphreys, G.S., 2006c. Magnetic enhancement in wildfire-affected soil and its potential for sediment-source ascription. Earth Surf. Process. Landforms 31, 249-264.

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

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

Boulay, S., Colin, C., Trentesaux, A., Pluquet, F., Bertaux, J., Blamart, D., Buehring, C., Wang, P., 2003. Mineralogy and sedimentology of Pleistocene sediment in the South China Sea (ODP Site 1144). In: Prell W., Wang P, Blum P, Rea D., Clemens S. (eds) Proceedings of the Ocean Drilling Program, Scientific Results. 184:1–21.

Brachfeld, S., Pinzon, J., Darley, J., Sagnotti, L., Kuhn, G., Florindo, F., Wilson, G., Ohneiser, C., Monien, D., Joseph, L., 2013. Iron oxide tracers of ice sheet extent and sediment provenance in the ANDRILL AND-1B drill core, Ross Sea, Antarctica. Global Planet. Change 110, 420–433.

Breed, C.A., Arocena, J.M., Sutherland, D., 2002. Possible sources of PM_{10} in Prince George (Canada) as revealed by morphology and in situ chemical composition of particulate. Atmos. Environ. 36, 1721-1731.

Brosinsky, A., Foerster, S., Segl, K., Kaufmann, K., 2014. Spectral fingerprinting: sediment source discrimination and contribution modelling of artificial mixtures based on VNIR-SWIR spectral properties. J Soils Sediments 14, 1949-1964.

Brown, A.G., 1985. The potential use of pollen in the identification of suspended sediment sources. Earth Surf. Process. Landforms 10, 27-32.

Brown, A.G., 2006. The use of forensic botany and geology in war crimes investigations in NE Bosnia. Forensic Sci. Int. 163, 204–210.

Bull, P.A., Parker, A., Morgan, R.M., 2006. The forensic analysis of soils and sediment taken from the cast of a footprint. Forensic Science Int. 162, 6–12.

Callén, M.S., Iturmendi A., López, J.M., 2014. Source apportionment of atmospheric PM2.5-bound polycyclic aromatic hydrocarbons by a PMF receptor model. Assessment of potential risk for human health. Environ. Poll. 195, 167-177.

Carter, J., Owens, P.N., Walling, D.E., Leeks, G.J.L., 2003. Fingerprinting suspended sediment sources in an urban river. Sci. Total Environ. 314-316, 513-534.

Cengiz, S., Cengiz Karaca, A., Çakır, İ., Bülent Üner, H., Sevindik, A., 2004. SEM–EDS analysis and discrimination of forensic soil. Forensic Science Int. 141, 33–37.

Charlesworth, S.M., Ormerod, L.M., Lees, J.A., 2000. Tracing sediment within urban catchments using heavy metal, mineral magnetic and radionuclide signatures. In: Foster, I.D.L. (Ed.), Tracers in Geomorphology. Wiley, Chichester, UK, pp. 345-368.

Chen, F., Zhang, F., Fang N., Shi, Z., 2016. Sediment source analysis using the fingerprinting method in a small catchment of the Loess Plateau, China. J. Soils Sediments 16, 1655-1669.

Christidis, G.E., Shriner, C.M., Murray, H.H., 2014. An integrated methodological approach for source-clay determination of ancient ceramics: The Case of Aegina Island, Greece. Clays Clay Minerals, 62, 447-469.

Clement, B.M., Javier, J., Say, J.P., Ross, M.S., 2011. The effects of wildfires on the magnetic properties of soils in the Everglades. Earth Surf. Process. Landforms 36, 460-466.

Collins, A.L., Walling, D.E., 2002. Selecting fingerprint properties for discriminating potential suspended sediment sources in river basins. J. Hydrol. 261, 218–244.

Collins, A.L., Walling, D.E., 2004. Documenting catchment suspended sediment sources: problems, approaches and prospects. Prog. Phys. Geog. 28, 159–196.

Collins A.L., Walling, D.E., Leeks, G.J.L., 1997a. Source type ascription for fluvial suspended sediment based on a quantitative fingerprinting technique. Catena 29, 1-27.

Collins, A.L., Walling, D.E., Leeks, G.J.L., 1997b. Use of the geochemical record preserved in floodplain deposits to reconstruct recent changes in river basin sediment sources. Geomorphology 19, 151–167.

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

Collins, A.L., Zhang, Y., McChesney, D., Walling, D.E., Haley, S.M., Smith, P., 2012a. Sediment source tracing in a lowland agricultural catchment in southern England using a modified procedure combining statistical analysis and numerical modelling. Sci.Total Environ. 414, 301–317.

Collins, A.L., Zhang, Y., Walling, D.E., Grenfell, S.E., Smith, P., Grischeff, J., Locke, A., Sweetapple, A., Brogden, D., 2012b. Quantifying fine-grained sediment sources in the River Axe catchment, southwest England: application of a Monte Carlo numerical modelling framework incorporating local and genetic algorithm optimisation. Hydrol. Process. 26, 1962–1983.

Concheri, G., Bertoldi, D., Polone, E., Otto, S., Larcher, R., Squartini, A., 2011. Chemical elemental distribution and soil DNA fingerprints provide the critical evidence in murder case investigation. PLoS ONE 6:e20222.

Cook, C.P., van de Flierdt, T., Williams, T., Hemming, S.R., Iwai, M., Kobayashi, M., Jimenez-Espejo, F.J., Escutia, C., González, J.J., Khim, B.-K., McKay, R.M., Passchier, S., Bohaty, S.M., Riesselman, C.R., Tauxe, L., Sugisaki, S., Galindo, A.L., Patterson, M.O., Sangiorgi, F., Pierce, E.L., Brinkhuis, H., Klaus, A., Fehr, A., Bendle, J.A.P., Bijl, P.K., Carr, S.A., Dunbar, R.B., Flores, J.A., Hayden, T.G., Katsuki, K., Kong, G.S., Nakai, M., Olney, M.P., Pekar, S.F., Pross, J., Röhl, U., Sakai, T., Shrivastava, P.K., Stickley, C.E., Tuo, S., Welsh, K., Yamane, M., 2013. Dynamic behaviour of the East Antarctic ice sheet during Pliocene warmth. Nature Geosci. 6, 765–769.

Cooper, R.J., Pedentchouk, N., Hiscock, K.M., Disdle, P., Krueger, T., Rawlins, B.G., 2015a. Apportioning sources of organic matter in streambank sediment: an integrated molecular and compound specific stable isotope approach. Sci.Total Environ. 520, 187-197.

Cooper, R.J., Krueger, T., Hiscock, K.M., Rawlins, B.G., 2015b. High temporal resolution fluvial sediment source fingerprinting with uncertainty: a Bayesian approach. Earth Surf. Process. Landforms 40, 78-92.

Cox, R., Peterson, H., Young, J., Cusik, C., Espinoza, E., 2000. The forensic analysis of soil organic by FTIR. Forensic Science Int. 108, 107–116.

Dawson, L.A., Hillier, S., 2010. Measurement of soil characteristics for forensic applications. Surf. Interface Anal. 42, 363–377.

De Luca, R., Miriello, D., Pecci, A., Domínguez-Bella, S., Bernal-Casasola, D., Cottica, D., Bloise, A., Crisci, G.M., 2015. Archaeometric study of mortars from the Garum Shop at Pompeii, Campania, Italy. Geoarchaeology 30, 330–351.

Devereux, O.H., Prestegaard, K.I., Needelman, B.A., Gellis, A.C., 2010. Suspended-sediment source in an urban watershed, northeast branch Anacostia River, Maryland. Hydrol. Process. 24, 1391-1403.

D'Haen, K., Verstraeten, G., Degryse, P., 2012. Fingerprinting historical fluvial sediment fluxes. Prog. Phys. Geog. 36, 154-186.

Dill, H., 1998. A review of heavy minerals in clastic sediments with case studies from the alluvial-fan through the nearshore-marine environments. Earth Sci. Rev. 45, 103–132.

Doğan, G., Güllü, G., Tuncel, G., 2008. Sources and source regions effecting the aerosol composition of the Eastern Mediterranean. Microchem. J. 88, 142-149.

Douglas, I., Bidin, K., Balamurugan, G., Chappell, N.A., Walsh, R.P.D., Greer, T., Sinun, W., l 1999. The role of extreme events in the impacts of selective tropical forestry on erosion during harvesting and recovery phases at Danum Valley, Sabah. Phil. Trans.Royal Soc. London, Series B – Biol. Sci. 354, 1749-1761.

Douglas, G.B., Kuhren, M., Radke, L.C., Hancock, G., Brooke, B., Palmer, M.R., Pietsch, T., Ford, P.W., Trefry, M.G., Packett, R., 2010. Delineation of sediment sources to a coastal wetland in the Great Barrier Reef catchment: influence of climate variability and land clearing since European arrival. Environ. Chem. 7, 190-226.

Dutton, C., Ainsfield, A.C., Ernstburger, H., 2013. A novel sediment fingerprinting method using filtration: application to the Mara River, East Africa. J Soils Sediments 13, 1708-1723.

Erkine, W., 2013. Soil colour as a tracer of sediment dispersion from erosion of forest roads in Chichester State Forest, NSW, Australia. Hydrol. Process. 27, 933–942.

Evans, D.J., Gibson, C.E., Rossell, R.S., 2006. Sediment loads and sources in heavily modified Irish catchments: a move towards informed management strategies. Geomorphology 79, 93-113.

Evrard, O., Poulenard, J., Némery, J., Ayrault, S., Gratiot, N., Duvert, C., Prat, C., Lefèvre, I., Bonté, P., Esteves, M., 2013. Tracing sediment sources in a tropical highland catchment of central Mexico by using conventional and alternative fingerprinting methods. Hydrol. Process. 27, 911–922.

Evrard, O., Laceby, P.J., Huon, S., Lefèvre, I., Sengtaheuanghoung, O., Ribolzi, O., 2016. Combining multiple fallout radionuclides (^{137}Cs, ^{7}Be, $^{210}Pb_{ex}$) to investigate temporal sediment source dynamics in tropical ephemeral river systems. J. Soils Sediments 16, 1130-1144.

Farrand, W.R., 2001. Sediments and stratigraphy in rockshelters and caves: A personal perspective on principles and pragmatics. Geoarchaeology 16, 537–557.

Foster, I.D.L., Charlesworth, S.M., 1996. Heavy metals in the hydrological cycle: trends and explanation. Hydrol. Process. 10, 227-261.

Foster, I.D.L., Lees, J.A., 2000. Tracers in geomorphology: theory and applications in tracing fine particulate sediments. In: Foster, I.D.L. (Ed.), Tracers in Geomorphology. Wiley, Chichester, UK, pp. 3-20.

Foster, I.D.L., Walling, D.E., 1994. Using reservoir deposits to reconstruct changing sediment yields and sources in the catchment of the Old Mill Reservoir, South Devon, UK, over the past 50 years. Hydrol. Sci. J. 39, 247-268.

Foster, I.D.L., Lees, J.A., Owens, P.N., Walling, D.E., 1998. Mineral magnetic characterization of sediment sources from an analysis of lake and floodplain sediments in the catchments of Old Mill Reservoir and Slapton Ley, South Devon, UK. Earth Surf. Process. Landforms 23, 685-703.

Foster, I.D.L., Mighall, T.M., Proffitt, H., Walling, D.E., Owens, P.N., 2006. Post-depositional ^{137}Cs mobility in the sediments of three shallow coastal lagoons, SW England. J. Palaeolimnology 35, 881-895.

Foucher, A., Laceby, P. J., Salvador-Blanes, S., Evrard, O., Le Gall, M., Lefèvre, I., Cerdan, O., Rajkumar, V., Desmet, M., 2015. Quantifying the dominant sources of sediment in a drained lowland agricultural catchment: The application of a thorium-based particle size correction in sediment fingerprinting. Geomorphology 250, 271-281.

Fox, J.F., Papanicolaou, A.N., 2007. The use of carbon and nitrogen isotopes to study watershed erosion processes. J. Am. Water Resour. Assoc. 43, 1047–1064.

Franz, C., Makeschin, F., Weiß, H., Lorz, C., 2014. Sediments in urban river basins: Identification of sediment sources within the Lago Paranoá catchment, Brasilia DF, Brazil – using the fingerprint approach. Sci. Total Environ. 466-467, 513–523.

Gellis, A.C., Noe, G.B., 2013. Sediment source analysis in the Linganore Creek watershed, Maryland, USA, using the sediment fingerprinting approach: 2008 to 2010. J. Soils Sediments 13, 1735–1753.

Gellis, A.C., Walling, D.E., 2011. Sediment-source fingerprinting (tracing) and sediment budgets as tools in targeting river and watershed restoration programs. In: Stream Restoration in Dynamic Fluvial Systems: Scientific Approaches, Analyses, and Tools. Simon, A., Bennett, S. and Castro, J.M. (Eds). American geophysical Union Monograph Series 194, Washington, D.C., USA, 263-291.

Gibbs, M., 2008. Identifying source soils in contemporary estuarine sediments: A new compound-specific isotope method. Estuaries Coasts 31, 344–359.

Glaser, B., 2005. Compound-specific stable-isotope ($\delta^{13}C$) analysis in soil science. J. Plant Nutr. Soil Sci. 168, 633–648.

Goldberg, P., Macphail, R.I., 2006. Practical and Theoretical Geoarchaeology. Blackwell Publishing, Oxford, UK.

Gonzales, D. A., Arakawa, F., Koenig, A., 2015. Petrographic and geochemical constraints on the provenance of sanidine-bearing temper in ceramic potsherds, Four Corners Region, Southwest USA. Geoarchaeology 30, 59–73.

Granger, S.J., Bol, R., Butler, P.J., Haygarth, P.M., Naden, P., Old, G., Owens, P.N., Smith, B.P.G., 2007. Processes affecting transfer of sediment and colloids, with associated phosphorus, from intensively farmed grasslands: tracing sediment and organic matter. Hydrol. Process. 21, 417-422.

Gruszowski, K.E., Foster, I.D.L., Lees, J.A., Charlesworth, S.M., 2003. Sediment sources and transport pathways in a rural catchment, Herefordshire, UK. Hydrol. Process. 17, 2665–2681.

Guedes, A., Ribeiro, H., Valentim, B., Noronha, F., 2009 Quantitative colour analysis of beach and dune sediments for forensic applications: A Portuguese example. Forensic Science Int. 190, 42–51.

Guzmán, G., Quinton, J.N., Nearing, M.A., Mabit, L., Gómez, J.A., 2013. Sediment tracers in water erosion studies: current approaches and challenges. J Soils Sediments 13, 816–833.

Haddadchi, A., Ryder, D.S., Evrard, O., Olley, J., 2013. Sediment fingerprinting in fluvial systems: review of tracers, sediment sources and mixing models. Int. J. Sediment Res. 28, 560-578.

Haddadchi, A., Nosrati, K., Ahmadi, F., 2014a. Differences between the source contribution of bed material and suspended sediments in a mountainous agricultural catchment of western Iran. Catena 116, 105–113.

Haddadchi, A., Olley, J., Laceby, P., 2014b. Accuracy of mixing models in predicting sediment source contributions. Sci. Total Environ. 497-498, 139-152.

Haddachi, A., Olley, J., Pietsch, T., 2015. Quantifying sources of suspended sediment in three size fractions. J. Soils Sediments 15, 2086-2100.

Hamlin, R.H.B., Woodward, J.C., Black, S., Macklin, M.G., 2000. Sediment fingerprinting as a tool for interpreting long-term river activity: the Voidomatis basin, NW Greece. In: Foster, I.D.L. (Ed.) Tracers in Geomorphology. John Wiley and Sons, Chichester, pp. 473–501.

Hancock, G.J., Revill, A.T., 2013. Erosion source discrimination in a rural Australian catchment using compound-specific isotope analysis (CSIA). Hydrol. Process. 27, 923–932.

Hebinck, K., Middelkoop, H., van Diepen, N., van der Graaf, E.R., de Meijer, R.J., 2007. Radiometric fingerprinting of fluvial sediments in the Rhine-Meuse delta, the Netherlands - a feasibility test. Netherlands Journal of Geosciences-Geologie en Mijnbouw 86, 229-240.

Herries, A.I., 2006. Archaeomagnetic evidence for climate change at Sibudu Cave. Southern African Humanities 18, 131-147.

Horowitz, A.J. 1991. A Primer in Sediment-trace Element Chemistry. Lewis Publishers, Michigan, USA

Huang, X.H.H., Bian, Q., Ng, W.M., Louie, P.K.K., Yu, J.Z., 2014. Characterization of PM2.5 major components and source investigation in suburban Hong Kong: A one year monitoring study. Aerosol Air Qual. Res. 14-237-250.

Hudson-Edwards, K.A., Macklin, M.G., Curtis, C.D., Vaughan, D.J., 1998. Chemical mobilization of contaminated metals within floodplains sediment in an incising river system: implications for dating and chemostratigraphy. Earth Surf. Process. Landforms 32, 671-684.

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

Hughes, A.O., Croke, J.C., Pietsch, T.J., Olley, J.M., 2010. Changes in the rates of floodplain and in-channel bench accretion in response to catchment disturbance, central Queensland, Australia. Geomorphology 114, 338-347.

Hughes, P.D., Woodward, J.C., Gibbard, P.L., Macklin, M.G., Gilmour, M.A., Smith, G.R., 2006. The glacial history of the Pindus Mountains, Greece. J. Geol. 114, 413-434.

Jenkins, P.A., Duck, R.W., Rowan, J.S., Walden, J., 2002. Fingerprinting of bed sediment in the Tay Estuary, Scotland: an environmental magnetism approach. Hydrol. Earth Syst. Sci. 6, 1007-1016.

Jenkins, P.A., Duck, R.W., Rowan, J., 2005. Fluvial contribution to the sediment budget of the Tay Estuary, Scotland, assessed using mineral magnetic fingerprinting In: Walling, DE and Horowitz, AJ (eds) Sediment Budgets 1, IAHS Publication 291, IAHS Press, Wallingford, UK, pp. 134-140.

Johansen, M.P., Hakonson, T.E., Whicker, F.W., Breshears, D.D., 2003. Pulsed redistribution of a contaminant following forest fire: cesium-137 in runoff. J. Environ. Qual. 32, 2150-2157.

Karanasiou, A., Amato F., Moreno, T., Lumbreras, J., Borge, R., Linares, C., Boldo, E., Alastuey, A., Querol, X., 2014. Road dust emission sources and assessment of street washing effect. Aerosol Air Qual. Res. 14, 734-743.

Khodakova, A.S., Smith, R.J., Burgoyne, L., Abarno, D., Linacre, A., 2014. Random whole metagenomic sequencing for forensic discrimination of soils. PLoS ONE 9:e104996.

Klages, M.G., Hsieh, Y.P., 1975. Suspended solids carried by the Galatin River of Southwestern Montana: II Using mineralogy for inferring sources. J. Environ. Qual. 4, 68-73.

Koiter, A.J., Owens, P.N., Petticrew, E.L., Lobb, D.A., 2013a. The behavioural characteristics of sediment properties and their implications for sediment fingerprinting as an approach for identifying sediment sources in river basins. Earth Sci. Rev. 125, 24–42.

Koiter, A.J., Lobb, D.A., Owens, P.N., Petticrew, E.L., Tiessen, K., Li, S., 2013b. Investigation the role of scale and connectivity in assessing the sources of sediment in an agricultural watershed in the Canadian prairies using sediment source fingerprinting. J. Soils Sediments 13, 1676–1691.

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

Kourampas, N., Simpson, I. A., Perera, N., Deraniyagala, S. U., Wijeyapala, W.H., 2009. Rockshelter sedimentation in a dynamic tropical landscape: Late Pleistocene–Early Holocene archaeological deposits in Kitulgala Beli-lena, southwestern Sri Lanka. Geoarchaeology 24, 677–714.

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

Kreutzweiser, D., Capell, S., Good, K., Holmes, S., 2009. Sediment deposition in streams adjacent to upland clearcuts and partially harvested riparian buffers in boreal forest catchments. Forest Ecol. Manag. 258, 1578–1585.

Laceby, J.P., Olley, J., Pietsch, T.J., Sheldon, F., Bunn, S.E., 2015a. Identifying subsoil sediment sources with carbon and nitrogen stable isotope ratios. Hydrol. Process. 29, 1956-1971.

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

Lamba, J., Karthikeyan, K.G., Thompson, A.M., 2015a. Apportionment of suspended sediment sources in an agricultural watershed using sediment fingerprinting. Geoderma 239-240, 25–33.

Lamba, J., Karthikeyan, K.G., Thompson, A.M., 2015b. Using radiometric fingerprinting and phosphorus to elucidate sediment transport dynamics in an agricultural watershed. Hydrol. Process. 29, 2681–2693.

Leeks, G.J.L., Marks, S.D., 1997. Dynamics of river sediments in forested headwater streams: Plynlimon, Hydrol. Earth Syst. Sci. 1, 483-497.

Legout, C., Poulenard, J., Nemery, J., Navratil, O., Grangeon, T., Evrard, O., Esteves, M., 2013. Quantifying suspended sediment sources during runoff events in headwater catchments using spectrocolorimetry. J. Soils Sediments 13, 1478-1492.

Liu, G., Xiao, H., Liu, P., Zhang, Q., Zhang, J., 2016. An improved method for tracing soil erosion using rare earth elements. J. Soils Sediments 16, 1670-1679.

Lombardi, G., 1999. The contribution of forensic geology and other trace evidence analysis to the investigation of the killing of Italian Prime Minister Aldo Moro. J. Forensic Sci. 44, 634–642.

Longworth, G., Becker, L.W., Thompson, R., Oldfield, F., Dearing, J.A., Rummery, T.A., 1979. Mossbauer and magnetic studies of secondary iron oxides in soil. J. Soil Sci. 30, 93–110.

Mabit, L., Benmansour, M., Abril, J.M., Walling, D.E., Meusburger, K., Iurian, A.R., Bernard, C., Tarján, S., Owens, P.N., Blake, W.H., Alewell, C., 2014. Fallout 210Pb as a soil and sediment tracer in catchment sediment budget investigations: A review. Earth Sci. Rev. 138, 335–351.

Mahler, B.J., Winkler, M., Bennett, P., Hillis, D.M., 1998. DNA-labelled clay: a sensitive new method for tracing particle transport. Geology 26, 831–834.

Martínez-Carreras, N., Krein, A., Gallart, F., Iffly, J.F., Pfister, L., Hoffmann, L., Owens, P.N., 2010. Assessment of different colour parameters for discriminating potential suspended sediment sources and provenance: a multi-scale study in Luxembourg. Geomorphology 118, 118-129.

McConnachie, J.L., Petticrew, E.L., 2006. Tracing organic matter sources in riverine suspended sediments: implications for fine sediment transfers. Geomorphology 79, 13–26.

McCulloch, M., Pailles, C., Moody, P., Martin, C.E., 2003. Tracing the source of sediment and phosphorus into the Great Barrier Reef lagoon. Earth Planet. Sci. Lett. 210, 249–258.

Meinhold, G., Kostopoulos, D., Reischmann, T., 2007. Geochemical constraints on the provenance and depositional setting of sedimentary rocks from the islands of Chios, Inousses and Psara, Aegean Sea, Greece: implications for the evolution of Palaeotethys. J. Geol. Soc. 164, 1145–1163.

Merefield, J.T., Stone, I., Barron, J., Jones, J., 1999. Techniques for tracing fugitive mineral dusts for nuisance control and health risk. Trans. Inst. Mining Metallurgy (Sect A) 108, 77-81.

Merefield, J.R., Stone, I.M., Roberts, J., Jones, J., Barron, J., Dean, A., 2000. Fingerprinting airborne particles for identifying provenance. In: Foster, I.D.L. (Ed.), Tracers in Geomorphology. Wiley, Chichester, UK, pp. 85-100.

Meyer, I., Davies, G.R., Stuut, J.-B.W., 2011. Grain size control on Sr-Nd isotope provenance studies and impact on paleoclimate reconstructions: An example from deep-sea sediments offshore NW Africa. Geochem. Geophys. Geosyst. 12, Q03005.

Michelaki, K., Braun, G.V., Hancock, R.G., 2015. Local clay sources as histories of human–landscape interactions: a ceramic taskscape perspective. J. Arch. Method Theory 22, 1-45.

Minc, L.D., Sherman, R.J., Elson, C., Winter, M., Redmond, E.M., Spencer, C.S., 2016. Ceramic provenance and the regional organization of pottery production during the later Formative periods in the Valley of Oaxaca, Mexico: Results of trace-element and mineralogical analyses. J. Archaeol. Sci.: Rep. 8, 28-46.

Minella, J.P.G., Walling, D.E., Merten, G.H. 2008. Combining sediment source tracing techniques with traditional monitoring to assess the impact of improved land management on catchment sediment yields. J. Hydrol. 348, 546-563.

Minella, J.P.G., Walling, D.E., Merten, G.H., 2014. Establishing a sediment budget for a small agricultural catchment in southern Brazil, to support the development of effective sediment management strategies. J. Hydrol. 519, 2189–2201.

Moody, J.A., Martin, D.A., 2009. Synthesis of sediment yields after wildland fire in different rainfall regimes in the western United States. Int. J. Wildland Fire 18, 96–115

Moore, T.E., O'Sullivan, P.B., Potter, C.J., Donelick, R.A., 2015. Provenance and detrital zircon geochronologic evolution of lower Brookian foreland basin deposits of the western Brooks Range, Alaska, and implications for early Brookian tectonism. Geosphere 11, 93–122.

Morgan, R.M., Bull, P.A., 2007a. The philosophy, nature and practice of forensic sediment analysis. Prog. Phys. Geog. 31, 43–58.

Morgan, R.M., Bull, P.A., 2007b. The use of grain size distribution analysis of sediments and soils in forensic enquiry. Sci. Justice 47, 125–135.

Morgan, R.M., Robertson, J., Lennard, C., Hubbard, K., Bull, P.A., 2010. Quartz grain surface textures of soils and sediments from Canberra, Australia: A forensic reconstruction tool. Aust. J. Forensic Sci. 42, 169–179.

Morton, A.C., 1985. Heavy Minerals in Provenance Studies. In: Zuffa GG (ed) Provenance of Arenites. Springer Netherlands, pp 249–277.

Morton, A.C., Hallsworth, C., 1994. Identifying provenance-specific features of detrital heavy mineral assemblages in sandstones. Sediment. Geol. 90, 241–256.

Morton, A.C., Whitham, A.G., Fanning, C.M., 2005. Provenance of Late Cretaceous to Paleocene submarine fan sandstones in the Norwegian Sea: Integration of heavy mineral, mineral chemical and zircon age data. Sediment. Geol. 182, 3–28.

Motha, J.A., Wallbrink, P.J., Hairsine, P.B., Grayson, R.B., 2003. Determining the sources of suspended sediment in a forested catchment in southeastern Australia. Wat. Resourc. Res. 39, DOI: 10.1029/2001WR000794.

Mukundan, R., Walling, D.E., Gellis, A.C., Slattery, M.C., Radcliffe, D.E., 2012. Sediment source fingerprinting: transforming from a research tool to a management tool. J. Amer. Wat. Resourc. Assoc.. Doi: 10.1111/j.1752-1688.2012.00685.x

Nakai, I., Furuya, S., Bong, W., Abe, Y., Osaka, K., Matsumoto, T., Itou, M., Ohta, A., Ninomiya, T., 2014. Quantitative analysis of heavy elements and semi-quantitative evaluation of heavy mineral compositions of sediments in Japan for construction of a forensic soil database using synchrotron radiation X-ray analyses: Forensic soil database using synchrotron radiation X-ray analyses. X-Ray Spectrometry 43, 38–48.

O'Leary, M.H., 1988. Carbon isotopes in photosynthesis. BioSci. 38, 328–336.

Ormerod, L.M., 1999. Estimating sedimentation rates and sources in a partially urbanized catchment using caesium-137. Hydrol. Process. 12, 1009-1020.

Oros, D.R., Mazurek, M.A., Baham, J.E., Simoneit, B.R.T., 2002. Organic tracers from wildfire residues in soils and rain/river wash-out. Wat. Air Soil Poll. 137, 203-233.

Owen, J. V., Casey, J. L., Greenough, J. D., Godfrey-Smith, D., 2013. Mineralogical and geochemical constraints on the sediment sources of Late Stone Age pottery from the Birimi Site, Northern Ghana. Geoarchaeology 28, 394–411.

Owens, P.N., 2005. Conceptual models and budgets for sediment management at the river basin scale. J. Soils Sediments 5, 201–212.

Owens, P.N., 2008. Sediment behaviour, functions and management. In: Sustainable Management of Sediment Resources: Sediment Management at the River Basin Scale edited by P.N. Owens. Elsevier, Amsterdam, 1-29.

Owens, P.N., Walling, D.E., 2002a. Changes in sediment sources and floodplain deposition rates in the catchment of the River Tweed, Scotland, over the last 100 years: the impact of climate and land use change. Earth Surf. Process. Landforms 27, 403-423.

Owens, P.N., Walling, D.E., 2002b. The phosphorus content of fluvial sediment in rural and industrialized river basins. Wat. Res. 36, 685-701

Owens, P.N., Walling, D.E., He, Q., 1996. The behaviour of bomb-derived caesium-137 fallout in catchment soils. J. Environ. Radioact. 32, 169-191.

Owens, P.N., Walling, D.E., Leeks, G.J.L., 1999. Use of floodplain sediment cores to investigate recent historical changes in overbank sedimentation rates and sediment sources in the catchment of the River Ouse, Yorkshire, UK. Catena 36, 21-47.

Owens, P.N., Blake, W.H., Petticrew, E.L., 2006. Changes in sediment sources following wildfire in mountainous terrain: a paired-catchment approach, British Columbia, Canada. Wat. Air Soil Poll.: Focus 6, 637-645.

Owens, P.N., Walling, D.E., Leeks, G.J.L., 2000. Tracing fluvial suspended sediment sources in the catchment of the River Tweed, Scotland, using composite fingerprints and a numerical mixing model. In: Foster, I.D.L. (Ed.), Tracers in Geomorphology, Wiley, Chichester, pp. 291-308.

Owens, P.N., Caley, K.A., Campbell, S., Koiter, A.J., Droppo, I.G., Taylor, K.G., 2011. Total and size-fractionated mass of road-deposited sediment in the city of Prince George, British Columbia, Canada: implications for air and water quality in an urban environment. J. Soils Sediments 11, 1040-1051.

Owens, P.N., Blake, W.H., Giles, T.R., Williams, N.D., 2012. Determining the effects of wildfire on sediment sources using ^{137}Cs and unsupported ^{210}Pb: the role of natural landscape disturbance and driving forces. J. Soils Sediments 12, 982-994.

Owens, P.N., Giles, T.R., Petticrew, E.L., Leggat, M., Moore, R.D., Eaton, B.C., 2013. Muted responses of streamflow and suspended sediment flux in a wildfire-affected watershed. Geomorphology 202, 128-139.

Padoan, M., Garzanti, E., Harlavan, Y., Villa, I.M., 2011. Tracing Nile sediment sources by Sr and Nd isotope signatures (Uganda, Ethiopia, Sudan). Geochimica et Cosmochimica Acta 75, 3627–3644.

Palazón, L., Gaspar, L., Latorre, B., Blake, W.H., Navas, A., 2015a. Identifying sediment sources by applying a fingerprinting mixing model in a Pyrenean drainage catchment. J. Soils Sediments 15, 2067-2085.

Palazón, L., Latorre, B., Gaspar, L., Blake, W.H., Smith, H.G., Navas, A., 2015b. Comparing catchment sediment fingerprinting procedures using an auto-evaluation approach with virtual sample mixtures. Sci. Total Environ. 532, 456-466.

Park, S.S., Kim, Y.J., 2005. Source contributions to fine particulate matter in an urban atmosphere. Chemosphere 59, 217-226.

Parnell, A. C., Phillips, D. L., Bearhop, S., Semmens, B. X., Ward, E. J., Moore, J. W., Jackson, A. L., Grey, J., Kelley, D. J., Inger, R., 2013. Bayesian stable isotope mixing models. Environmetrics 24, 387-399.

Parson, A.J., Foster, I.D.L., 2011. What can we learn about soil erosion from the use of ^{137}Cs? Earth Sci. Rev. 108, 101-113.

Peart, M.R., Walling, D.E., 1986. Techniques for establishing suspended sediment sources in two drainage basins in Devon, UK: a comparative assessment. In: Sediment Budgets. IAHS Pub 174, IAHS Press, Wallingford, UK, pp. 269-279.

Perreault, L.M., Yager, E.M., Aalto, R., 2012. Application of $^{210}Pb_{ex}$ inventories to measure net hillslope erosion at burned sites. Earth Surf. Process. Landforms 38, 133-145.

Pollard, M., Beisson, F., Li, Y., Ohlrogge, J.B., 2008. Building lipid barriers: biosynthesis of cutin and suberin. Trends Plant Sci. 13, 236–246.

Poleto, C., Merten, C.H., Minella, J.P., 2009. The identification of sediment sources in a small urban watershed in southern Brazil: an application of sediment fingerprinting. Environ. Tech. 30, 1145-1153.

Poulenard, J., Legout, C., Némery, J., Bramorski, J., Navratil, O., Douchin, A., Fanget, B., Perrette, Y., Evrard, O., Esteves, M., 2012. Tracing sediment sources during floods using Diffuse Reflectance Infrared Fourier Transform Spectrometry (DRIFTS): A case study in a highly erosive mountainous catchment (Southern French Alps). J. Hydrol. 414-145, 452–462.

Pulley S., Foster, I., Antunes, P., 2015. The application of sediment fingerprinting to floodplain and lake sediment cores: assumptions and uncertainties evaluated through case studies in the Nene Basin, UK. J. Soils Sediments 15, 2132-2154.

Pye, K., Blott, S.J., Croft, D.J., Carter, J.F., 2006. Forensic comparison of soil samples: Assessment of small-scale spatial variability in elemental composition, carbon and nitrogen isotope ratios, colour, and particle size distribution. Forensic Sci. Int. 163, 59–80.

Ratto, N., Gogni, V., Escobar, M. B., Plá, R., 2015. Mud-clay banks and regional geochemistry: The provenance of ceramic raw materials (Department Tinogasta, catamarca, Argentina). Quaternary Int. 375, 13-26.

Reiffarth, D., Petticrew, E.L., Owens, P.N., Lobb, D.A., 2016. Identification of sources of variability in fatty acid (FA) biomarkers in the application of compound-specific stable isotopes (CSSIs) to soil and sediment fingerprinting and tracing: a review. Sci. Total Environ. 565, 8-27.

Renson, V., Jacobs, A., Coenaerts, J., Mattielli, N., Nys, K., Claeys, P. 2013. Using lead isotopes to determine pottery provenance in Cyprus: clay source signatures and comparison with Late Bronze Age Cypriote pottery. Geoarchaeol. 28, 517-530.

Rodríguez, M.P., Lincoñir, L.P., Encinas, A., 2012. Cenozoic erosion in the Andean forearc in Central Chile (33°–34°S): Sediment provenance inferred by heavy mineral studies. Geol. Soc. Amer. Special Pap. 487, 141–162.

Roelofse, F., Horstmann, U.E., 2008. A case study on the application of isotope ratio mass spectrometry (IRMS) in determining the provenance of a rock used in an alleged nickel switching incident. Forensic Sci. Int. 174, 64–67.

Rotman, R., Naylor, L., McDonnell, R., MacNiocaill, C., 2008. Sediment transport on the Freiston Shore managed realignment site: An investigation using environmental magnetism. Geomorphology 100, 241-255.

Rowan, J.S., Goodwill, P., Franks, S.W., 2000. Uncertainty estimation in fingerprinting suspended sediment sources. In: Fister, I.D.L., (Ed.) Tracers in Geomorphology, Wiley, pp. 279-290.

Ruffell, A., 2010. Forensic pedology, forensic geology, forensic geoscience, geoforensics and soil forensics. Forensic Sci. Int. 202, 9–12.

Ruffell, A., Sandiford, A., 2011. Maximising trace soil evidence: An improved recovery method developed during investigation of a $26 million bank robbery. Forensic Sci. Int. 209, 1-7.

Ruess, L., Chamberlain, P.M., 2010. The fat that matters: Soil food web analysis using fatty acids and their carbon stable isotope signature. Soil Biol. Biochem. 42, 1898–1910.

Russell, M.A., Walling, D.E., Hodgkinson, R.A., 2001. Suspended sediment sources in two small lowland catchments in the UK. J. Hydrol. 252, 1-24.

Salgán, L., Garvey, R., Neme, G., Gil, A., Giesso, M., Glascock, M.D., Durán, V., 2015. Las Cargas: characterization and prehistoric use of a Southern Andean obsidian source. Geoarchaeology 30, 139–150.

Schindler Wildhaber, Y., Liechti, R., Alewell, C., 2012. Organic matter dynamics and stable isotope signature as tracers of the sources of suspended sediment. Biogeosciences 9, 1985–1996.

Schuller, P., Iroumé, A., Walling, D.E., Mancilla, H.B., Castillo, A., Trumper, R.E., 2006. Use of Beryllium-7 to document soil redistribution following forest harvest operations. J.Environ. Qual. 35, 1756-1763.

Scott, K.R., Morgan, R.M., Jones, V.J., Cameron, N.G., 2014. The transferability of diatoms to clothing and the methods appropriate for their collection and analysis in forensic geoscience. Forensic Sci. Int. 241, 127–137.

Sherriff, S.C., Franks, S.W., Rowan, J.S., Fenton, O., Ó'hUallacháin D., 2015. Uncertainty- based assessment of tracer selection, tracer non-conservativeness and multiple solutions in sediment fingerprinting using synthetic and field data. J. Soils Sediments 15, 2101-2116.

Smith, H.G., Blake, W.H., 2014. Sediment fingerprinting in agricultural catchments: a critical re-evaluation of source discrimination and data corrections. Geomorphology 204, 177-191.

Smith, H.G., Sheridan, G.J., Lane, P.N.J., Noske, P., Heijnis, H., 2011. Changes to sediment sources following wildfire in a forested upland catchment, southeastern Australia. Hydrol. Process. 25, 2878-2889.

Smith, H.G., Sheridan, G.J., Nyman, P., Child, D.P., Lane, P.N.J., Hotchkis, M.A.C., Jacobsen, G.E., 2012. Quantifying sources of fine sediment supplied to post-fire debris flows using fallout radionuclide tracers. Geomorphology 139-140, 403-415.

Smith, H.G., Blake, W.H., Owens, P.N., 2013. Discriminating fine sediment sources and the application of sediment tracers in burned catchments: a review. Hydrol. Process. 27, 943-958.

Smith, H.G., Blake, W.H., Taylor, A., 2014. Modelling particle residence times in agricultural river basins using a sediment budget model and fallout radionuclide tracers. Earth Surf. Process. Landforms 39, 1944–1959.

Smith, H.G., Evrard, O., Blake, W.H., Owens, P.N., 2015. Preface - Addressing challenges to advance sediment fingerprinting research. J. Soils Sediments 15, 2033-2037.

Stewart, H.A., Massoudieh, A., Gellis, A., 2015. Sediment source apportionment in Laurel Hill Creek, PA, using Bayesian chemical mass balance and isotope fingerprinting. Hydrol. Process. 29, 2545–2560.

Stone P, Walling, D.E., 1997. Particle size selectivity considerations in sediment budget investigations. Wat. Air Soil Poll. 99, 63-70.

Stone, M., Collins, A.L., Silins, U., Emelko, M.B., Zhang, Y.S., 2014. The use of composite fingerprints to quantify sediment sources in a wildfire impacted landscape, Alberta, Canada. Sci. Total Environ. 473-474, 642-650.

Stott, T., Mount, N., 2004. Plantation forestry impacts on sediment yields and downstream channel dynamics in the UK: a review. Prog. Phys. Geog. 28, 197-240.

Suman, Singh, G., Pal, A.K., 2014. Source apportionment of respirable particulate matter using principal component analysis – a case study from India. Rep. Opinion 6, 26-32.

Taylor, K.G., Owens, P.N., 2009. Sediments in urban river basins: a review of sediment–contaminant dynamics in an environmental system conditioned by human activities. J. Soils Sediments 9, 281-303.

Taylor, A., Blake, W.H., Keith-Roach, M.J., 2014. Estimating Be-7 association with soil particle size fractions for erosion and deposition modelling. J. Soils Sediments 14, 1886-1893.

Thomas, W.A., 2011. Detrital-zircon geochronology and sedimentary provenance. Lithosphere 3, 304–308.

Trimble, S.W., 1983. A sediment budget for Coon Creek in the Driftless Area, Wisconsin, 1853–1977. Am. J. Sci. 283, 454–474.

Tsikouras, B., Pe-Piper, G., Piper, D.J.W., Schaffer, M., 2011. Varietal heavy mineral analysis of sediment provenance, Lower Cretaceous Scotian Basin, eastern Canada. Sediment. Geol. 237, 150–165.

Verheyen, D., Diels, J., Kissi, E., Poesen, J., 2014. The use of visible and near-infrared reflectance measurements for identifying the source of suspended sediment in rivers and comparison with geochemical fingerprints. J. Soils Sediments 14, 1869-1885.

Vital, H., Stattegger, K., 2000. Major and trace elements of stream sediments from the lowermost Amazon River. Chem. Geol. 168, 151–168.

von Eynatten, H., Dunkl, I., 2012. Assessing the sediment factory: The role of single grain analysis. Earth Sci. Rev. 115, 97–120.

Wall, G.J., Wilding, L.P., 1976. Mineralogy and related parameters of fluvial suspended sediments in Northwestern Ohio. J. Environ. Qual. 5, 168-173.

Wallbrink P.J., Croke J., 2002. A combined rainfall simulator and tracer approach to assess the role of Best Management Practices in minimising sediment redistribution and loss in forests after harvesting. Forest Ecol. Manag. 170, 217-232

Wallbrink, P.J., Murray, A.S., 1996. Determining soil loss using the inventory ratio of excess lead-210 and cesium-137. Soil Sci. Soc. Amer. J. 60, 1201-1208.

Wallbrink, P.J., Murray, A.S., Olley, J.M., Olive, L.J., 1998. Determining sources and transit times of suspended sediment in the Murrumbidgee River, New South Wales, Australia, using fallout ^{137}Cs and ^{210}Pb. Water Resourc. Res. 34, 879–887.

Wallbrink, P.J., Roddy, B.P., Olley, J.M., 2002. A tracer budget quantifying soil redistribution on hillslopes after forest harvesting. Catena 47, 179–201.

Walling, D.E., 1983. The sediment delivery problem. J. Hydrol. 65, 209-237.

Walling, D.E., 2006. Human impact on land-ocean sediment transfer by the world's rivers. Geomorphology 79, 192-216.

Walling, D.E., 2005. Tracing suspended sediment sources in catchments and river systems. Sci. Total Environ. 344, 159-184.

Walling, D.E., 2013. The evolution of sediment source fingerprinting investigations in fluvial systems. J. Soils Sediments 13, 1658-1675.

Walling, D.E., Collins, A.J., 2008. The catchment sediment budget as a management tool. Environ. Sci. Policy 11, 136-143.

Walling, D.E., Woodward, J.C., 1992. Use of radiometric fingerprints to derive information on suspended sediment sources. In: Bogen, J., Walling, D.E., Day, T.J. (Eds) Erosion and sediment transport monitoring programmes in river basins. IAHS Publ. 210, pp 153-164.

Walling, D.E., Woodward J.C., 1995. Tracing suspended sediment sources in river basins: a case study of the River Culm, Devon, UK. Marine Freshwater Res. 46, 327–336.

Walling, D.E., Woodward, J.C., Nicholas A.P., 1993. A multi-parameter approack to fingerprinting suspended-sediment sources. I: Tracers in Hydrology. IAHS Publ 215, IAHS Press, Wallingford, UK. Pp. 329-338.

Walling, D.E., Owens, P.N., Leeks, G.J.L., 1999. Fingerprinting suspended sediment sources in the catchment of the River Ouse, Yorkshire, UK. Hydrol. Process. 13, 955-975.

Walling, D.E., Owens, P.N., Waterfall, B.D., Leeks, G.J.L., Wass, P.D., 2000. The particle size characteristics of fluvial suspended sediment in the Humber and Tweed catchments, UK. Sci. Total Environ. 251-252, 205-222.

Walling, D.E., Owens, P.N., Foster, I.D.L., Lees, J.A., 2003. Changes in the fine sediment dynamics of the Ouse and Tweed basins in the UK, over the last 100-150 years. Hydrol. Process. 17, 3245-3269.

Walling, D.E., Collins, A.L., Stroud, R.W., 2008. Tracing suspended sediment and particulate phosphorus sources in catchments. J. Hydrol. 350, 274–289.

Walling, D.E., Russell, M.A., Hodgkinson, R.A., Zhang, Y., 2002. Establishing sediment budgets for two small lowland agricultural catchments in the UK. Catena 47, 323–353.

Walling, D.E., Schuller, P., Zhang, Y., Iroumé, A., 2009. Extending the timescale for using beryllium 7 measurements to document soil redistribution by erosion. Wat. Resourc. Res. 45, Article W02418.

Walsh, R.P.D., Bidin, K., Blake, W.H., Chappell, N.A., Clarke, M.A., Douglas, I., Ghazali, R., Sayer, A.M., Suhaimi, J., Tych, W., Annammala, K.V., 2011. Long-term responses of rainforest erosional systems at different spatial scales to selective logging and climatic change. Phil. Trans. Royal Soc. London, Series B – Biol. Sci. 366, 3340-3353.

Weltje, G.J., von Eynatten, H., 2004. Quantitative provenance analysis of sediments: review and outlook. Sediment. Geol. 171, 1–11.

Wethered, A.S., Ralph, T.J., Smith, H.G., Fryirs, K.A., Heijnis, H., 2015. Quantifying fluvial (dis)connectivity in an agricultural catchment using a geomorphic approach and sediment source tracing. J Soils Sediments 15, 2052-2066.

White, L.F., Bailey, I., Foster, G.L., Allen, G., Kelley, S.P., Andrews, J.T., Hogan, K., Dowdeswell, J.A., Storey, C.D., 2016. Tracking the provenance of Greenland-sourced, Holocene aged, individual sand-sized ice-rafted debris using the Pb-isotope compositions of feldspars and 40Ar/39Ar ages of hornblendes. Earth Planet. Sci. Lett. 433, 192–203.

Wiesenberg, G.L.B., Schneckenberger, K., Schwark, L., Kuzyakov, Y., 2012. Use of molecular ratios to identify changes in fatty acid composition of Miscanthus × giganteus (Greef et Deu.) plant tissue, rhizosphere and root-free soil during a laboratory experiment. Org. Geochem. 46, 1–11.

Wilkinson, S.N., Wallbrink, P.J., Hancock, G.J., Blake, W.H., Shakesby, R.A., Doerr, S.H., 2009. Fallout radionuclide tracers identify a switch in sediment sources and transport-limited sediment yield following wildfire in a eucalypt forest. Geomorphology 110: 140-151.

Wilkinson, S.N., Hancock, G.J., Bartley, R., Hawdon, A.A., Keen, R.J., 2013. Using sediment tracing to assess processes and spatial patterns of erosion in grazed rangelands, Burdekin River basin, Australia. Agric. Ecosyst. Environ. 180, 90–102.

Wilkinson, S.N., Olley, J.M., Furuichi, T., Burton, J., Kinsey-Henderson, A.E., 2015. Sediment source tracing with stratified sampling and weighting based on spatial gradients in soil erosion. J. Soil Sediments 15, 2038-2051.

Withers, P.J.A., Jarvie, H.P., 2008. Delivery and cycling of phosphorus in rivers: a review. Sci. Total Environ. 400, 379-395.

Woodward, J.C., Bailey, G.N., 2000. Sediment sources and terminal Pleistocene geomorphological processes recorded in rockshelter sequences in northwest Greece. In: Foster, I.D.L. (Ed.), Tracers in Geomorphology. John Wiley, Chichester, pp. 521–551.

Woodward, J.C., Goldberg, P., 2001. The sedimentary records in Mediterranean rockshelters and caves: Archives of environmental change. Geoarchaeology 16, 327-354.

Woodward, J.C., Lewin, J., Macklin, M.G., 1992. Alluvial sediment sources in a glaciated catchment: the Voidomatis basin, northwest Greece. Earth Surf. Process. Landforms 16, 205-216.

Woodward, J.C., Hamlin, R.H.B., Macklin, M.G., Karkanas, P., Kotjabopoulou, E., 2001. Quantitative sourcing of slackwater deposits at Boila rockshelter: A record of lateglacial flooding and Paleolithic settlement in the Pindus Mountains, Northwest Greece. Geoarchaeology 16, 501–536.

Woodward, J.C., Hamlin, R.H.B., Macklin, M.G., Hughes, P.D., Lewin, J., 2008. Glacial activity and catchment dynamics in northwest Greece: long-term river behaviour and the slackwater sediment record for the last glacial to interglacial transition. Geomorphology 101, 44-67.

Yang, J., Wu, F., Shao, J., Wilde, S., Xie, L., Liu, X., 2006. Constraints on the timing of uplift of the Yanshan Fold and Thrust Belt, North China. Earth Planet. Sci. Lett. 246, 336–352.

Yeager, K.M., Santschi, P.H., Phillips, J.D., Herbert, B.E., 2005. Suspended sediment sources and tributary effects in the lower reaches of a coastal plain stream as indicated by radionuclides, Loco Bayou, Texas. Environ. Geol. 47, 382-395.

Yeager, K.M., Santschi, P.H., Schindler, K.J., Andres, M.J., Weaver, E.A., 2006. The relative importance of terrestrial versus marine sediment sources to the Nueces-Corpus Christi Estuary, Texas: An isotopic approach. Estuaries Coasts 29, 443-454.

Yin, C., Li L., 2008. An investigation on suspended sediment sources in urban stormwater runoff using ^{7}Be and ^{210}Pb as tracers. Wat. Sci. Technol. 57, 1945-1950.

Yu, L., Oldfield, F., 1989. A multivariate mixing model for identifying sediment source from magnetic measurements. Quatern. Res. 32, 168-181.

Yu, L., Oldfield, F., 1993. Quantitative sediment source ascription using mineral magnetic measurements in a reservoir-catchment system near Nijar, SE Spain. Earth Surf. Process. Landforms 18, 441-454.

Zhang, X., Gu, Q., Long, X., Li, Z., Liu, D, Ye, D., He, C., Liu, X., Väänänen, K., Chen, X., 2016. Anthropogenic activities drive the microbial community and its function in urban river sediment. J. Soils Sediments 16, 716-725.

Zhong, L., Li, J., Yan, W., Tu, X., Huang, W., Zhang, X., 2012. Platinum-group and other traffic-related heavy metal contamination in road sediment, Guangzhou, China. J. Soils Sediments 12, 942-951.

Zhou, X.J, Li, A.C., Jiang, F.Q., Meng, Q.Y., 2010. A preliminary study on fingerprinting approach in marine sediment dynamics with the rare earth elements. Acta Oceanologica Sinica 29, 62-77.

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Table1

Examples of the relative contributions of agricultural land (cultivated and pasture/grassland) determined using sediment source fingerprinting approaches to the total sediment budget of river basins. Values in parentheses correspond to the percentage of land in the river basin.

Country (River Basin)	Basin area (km^2)	Contribution from agricultural land	Other main sources	Sediment tracers	Study
USA (Pleasant Valley Creek, WI)	50	45-97% (34%)	Channel banks 3-47%	Geochemical elements	Lamba et al. (2015a)
USA (Laurel Hill Creek, PA)	324	50-95% (27%)	Steam banks 5-50%	Geochemical elements, $\delta^{13}C$, $\delta^{15}N$	Stewart et al. (2015)
England (River Culm)	276	88% (Pasture 28%, Cultivated 60%)	Channel banks 12%	Fallout radionuclides, mineral magnetics, carbon and nitrogen	Walling and Woodward (1995)
England (River Kennet)	214	4% (84%)	Farm track 55%	Geochemical elements	Collins et al. (2012a)
England (River Aire)	1932	20-45% (NA)	Road dust 19-22%	Geochemical elements, TP*	Carter et al. (2003)
Scotland (River Tweed)	4390	54% (87%)	Channel banks/subsoil 39%; woodland 7%	Geochemical elements, ^{137}Cs, mineral magnetics, TP, TOC**	Owens et al. (2000)

Tunisia (Kamech)	2.63	80% (70%)	Gully/Banks 20%	^{137}Cs, TOC	Ben Slimane et al. (2013)
Brazil (Lago Paranoá)	950	1-9% (8%)	Urban areas 81-89% (34%)	Geochemical elements	Franz et al. (2014)
Iran (Taleghani)	26	24% (88%)	Channel banks 72%	Geochemical elements	Haddadchi et al. (2014a)
Australia (Theresa Creek, QLD)	6000	64% (74%)	Channel banks 30%	Geochemical elements, ^{137}Cs, ^{210}Pb	Hughes et al. (2009)

* Total phosphorus; ** Total organic carbon

Table 2

Studies using sediment source fingerprinting to determine sediment sources following wildfire arranged according to river basin size (modified from Smith et al., 2013).

Reference	Location	Watershed area (km^2)[a]	Proportion of the watershed burned (%)[b]	Method[c]	Post-fire measurement period	Proportional hillslope source contributions (%)[d]	
						First year	Subsequent years
Smith et al. (2012)	Dry eucalypt forest, Victoria, Australia	0.07 0.23	100	FRN (^{137}Cs, $^{210}Pb_{ex}$ $^{239,240}Pu$)	Debris flow events	22-69 32-74	n/a n/a
Smith et al. (2011)	Wet eucalypt mountain forest, Victoria, Australia	1.36	99	FRN (^{137}Cs, $^{210}Pb_{ex}$)	3.5 years	96-100	58-76
Stone et al. (2014)	Conifer forest, Alberta, Canada	B: 5-34 U: 161	8-94	Geochemical and organic C	7 years	n/a	n/a[e]
Owens et al. (2012)	Conifer forest, British Columbia, Canada	B: 158 UB: 215	62	FRN (^{137}Cs, $^{210}Pb_{ex}$)	7 years	B: 7 UB: 0	B: 10 UB: 0
Wilkinson et al. (2009)	Blue Mountains, New South Wales, Australia	17	31	FRN (^{137}Cs, $^{210}Pb_{ex}$)	5 years	10	n/a
		183	99			86	55-68
		446	57			45	29-51
		629	69			71	21-63

[a]Burned (B); Unburned (UB)

[b]Not available (n/a)

[c]Fallout radionuclide (FRN) tracers measure sources of fine sediment (either <10 or <63 µm), whereas field survey and erosion measurement techniques capture the full range of particle size fractions

[d]Channel sources constitute the remaining percentage contribution for each of the studies.

[e]This study did not estimate hillslope vs channel bank sources but instead determined relative contributions from burnt and unburnt sections of the watersheds

Table 3. Summary of geologic settings and sediment properties used in sediment provenance studies

Geologic setting	Sediment properties	Reference
Continental slope, Southwestern Gulf of Mexico	Chemical index of alteration Index of chemical maturity Elemental ratios Standardized composite indices	Armstrong-Altrin et al. (2015)
Submarine fan sandstones, Norwegian Sea	Heavy mineral analysis (ratios) Mineral varietal analysis Zircon geochonology (U–Pb)	Morton et al. (2005)
Ice Shelf, Antarctica	Fe-oxide Magnetic susceptibility	Brachfeld et al. (2013)
Amazon River, Brazil	Index of chemical maturity Index of textural maturity Geochemistry	Vital and Stattegger (2000)
Great Barrier Reef lagoon, Australia	$^{143}Nd/^{144}Nd$ $^{87}Sr/^{86}Sr$	McCulloch et al. (2003)
Northern margin of the South China Sea	Elemental ratios Clay mineralogy	Boulay et al. (2003)
Ice-rafted debris, Greenland	Feldspar Pb-isotopes Hornblende geochronology ($^{40}Ar/^{39}Ar$)	White et al. (2016)
Brookian foreland basin, Alaska	Petrography Zircon geochronology (U-Pb and fission-track)	Moore et al. (2015)

Table 4. A selection of recent studies employing sediment fingerprinting methods to investigate the origin of fine-grained sediments used in the production of ceramics for various ancient cultures in a range of geological settings.

Study region	Archaeological period	Ceramics of interest	Fingerprint properties	Potential source materials sampled	Reference
Cyprus	Late Bronze Age c. 1600-1000 BC	Fine pottery ware sherds from three archaeological sites (*n* = 35)	Pb isotope ratios: $^{208}Pb/^{204}Pb$ $^{207}Pb/^{204}Pb$, $^{206}Pb/^{204}Pb$	Clay samples from various geological sources including marls, mudstones, weathered lavas, and Holocene alluvium and colluvium (*n* = 65)	Renson et al. (2013)
Northwest Alaska	Early Alaskan ceramic technology c. 2800-1500 BP	Vessels and some clay lamps (*n* = 360 ceramic samples)	Geochemical analysis using Instrumental Neutron Activation Analysis (INAA)	Regional survey of clay (*n* = 31) and temper (*n* = 28) sources including glacial, alluvial, and beach materials. The survey was informed by ethnographic data on clay and temper sources.	Anderson (2016)
Oaxaca Valley in southern Mexico	Late Middle to Terminal Formative times (c. 700 to 200 BC) of the Zapotec civilization	Shards from vessels (*n* = 500) and daub (*n* = 4) representing four dominant wares from Formative production sites.	Trace elements and mineralogy determined by INAA and optical petrography of ceramic thin sections.	Sampling of field clays (*n* = 320) throughout the central valley from geological materials of various ages.	Minc et al. (2016)
Central Italy and	Late Republican	Amphorae recovered from	Thin section observations	Data on the composition of	

the **Tyrrhenian Sea**	(Roman) era between the 2nd and first half of the 1st century BC.	a shipwreck between the islands of Ponza and Palmarola (*n* = 13)	followed by SEM-EDS and trace element analysis (using LA-ICP-MS) of clinopyroxene crystals within volcanic inclusions in amphorae	clinopyroxenes from sources rocks in the main volcanic provinces of western and southern Italy compiled from the literature.	Belfiore et al. (2014)
Gambaga Escarpment of Northern Ghana	Late Stone Age c. 3500 to 3000 BP	Sherds (*n* = 15) from various types of Kintampo pottery	Mineralogy and bulk chemical composition (major and minor elements) determined SEM-EDS and ICP-MS.	Fine sediment samples collected from active clay pits and the White Volta and Morago rivers (*n* = 15)	Owen et al. (2013)

Table 5.
Summary of applications, fingerprint properties and analytical techniques used in soil forensics.

Scenario investigated	Fingerprint properties – analytical techniques	Transfer mechanism	Reference
Simulated crime scene	Particle morphology – scanning electron microscope	NA	Morgan et al. (2010)
Simulated crime scene	Particle morphology – scanning electron microscopy	NA	Bailey et al. (2012)
Wildlife crime Murder	Trace element mapping – particle-induced x-ray and γ-ray emission	Shovel Vehicle	Bailey et al. (2012)
Wildlife crime Hit and run Murder	Particle size – laser granulometer Particle size – scanning electron microscope	Shovel Vehicle Victim body	Morgan and Bull (2007b)
Murder	Mineralogy – binocular microscopy Particle size – laser granulometer Geochemistry – atomic absorption spectrophotometry Carbon/nitrogen ratio – method not specified Pollen grain identification – binocular microscopy	Footwear	Bull et al. (2006)
Murder	Geochemistry – inductively coupled plasma mass and optical emission spectrometry Bacterial community – amplified ribosomal DNA restriction analysis	Vehicle	Concheri et al. (2011)
War crimes	Mineralogy – x-ray diffraction Pollen grain identification – binocular microscopy	Reburial of human remains	Brown (2006)
Theft/security breach	Isotopic analysis – isotope ratio mass spectrometry	Shipping container	Roelofse and Horstmann (2008)
Bank robbery/kidnapping	Soil colour, particle shape, mineralogy – visual comparisons	Footwear/clothing	Ruffell and Sandiford (2011)
Experimental	Geochemistry – scanning electron microscope and energy dispersive X-ray spectrometer	NA	Cengiz et al. (2004)

Experimental	Soil organic matter – Fourier transform infrared spectroscopy Soil colour – Munsell color chart	NA	Cox et al. (2000)
Experimental	Soil colour – spectrophotometer	NA	Guedes et al. (2009)
Experimental	Soil DNA – random whole metagenomic sequencing	NA	Khodakova et al. (2014)
Experimental	Diatoms – binocular and scanning electron microscope	Clothing	Scott et al. (2014)
Forensic soil database	Geochemistry/mineralogy – synchrotron radiation X-ray analysis	NA	Nakai et al. (2014)