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

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

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59 **Abstract**

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

80

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

83

84 **1. Background and history**

85 In recent decades, there has been a rapid growth in the number of studies that have utilized tracing and
86 fingerprinting approaches to investigate the movement of soils and fine sediments in terrestrial and
87 aquatic systems (Koiter et al., 2013a; Walling, 2013; Mabit et al., 2014). This growth is due to the fact
88 that these techniques are able to provide essential information on soil and sediment dynamics that can
89 be used to understand the evolution of landscapes (e.g., Belmont et al., 2007) and assist in river basin
90 management and river restoration (e.g., Owens, 2005, 2008; Evans et al., 2006; Minella et al., 2008,
91 2014; Walling and Collins, 2008; Gellis and Walling, 2011). In these contexts, the source tracing and
92 fingerprinting techniques have often been part of wider sediment budget investigations (Gellis and
93 Walling, 2011), as the source tracing and fingerprinting techniques alone are sometimes too broad (e.g.,
94 topsoil is dominant over channel banks) to enable exact sources (e.g., specific fields, or channel bank
95 reaches) to be determined. Thus, broad classifications of sediment source types can make it difficult to
96 precisely target management strategies intended to control sediment problems. . In addition, most
97 sediment source tracing and fingerprinting results are relative (i.e., expressed as percentages), and
98 sediment transport data are often required to convert values into actual sediment fluxes associated with
99 the sources (e.g., Walling and Woodward, 1995; Smith et al., 2011). Source tracing and fingerprinting
100 techniques used in concert with information on sediment transport and sediment budgeting can offer
101 powerful insights into how landscapes behave and can provide important information on
102 geomorphological processes, which, in turn, can be used to guide river basin and coastal management.
103 Mukundan et al. (2012), for example, have demonstrated how sediment source fingerprinting can be
104 used as a management tool for developing total maximum daily loads (TMDLs) of sediment as part of the
105 TMDL program in the USA.

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

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

126 The term "source tracing" is a hybrid term often used to refer to the use of tracer properties to identify
127 the source of sediments. Thus, the terms "source tracing" and "source fingerprinting" are often used

128 interchangeably to mean the use of the properties of soils and sediments to infer their origins; for
129 simplicity, in this review we mainly use the term sediment source fingerprinting.

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

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

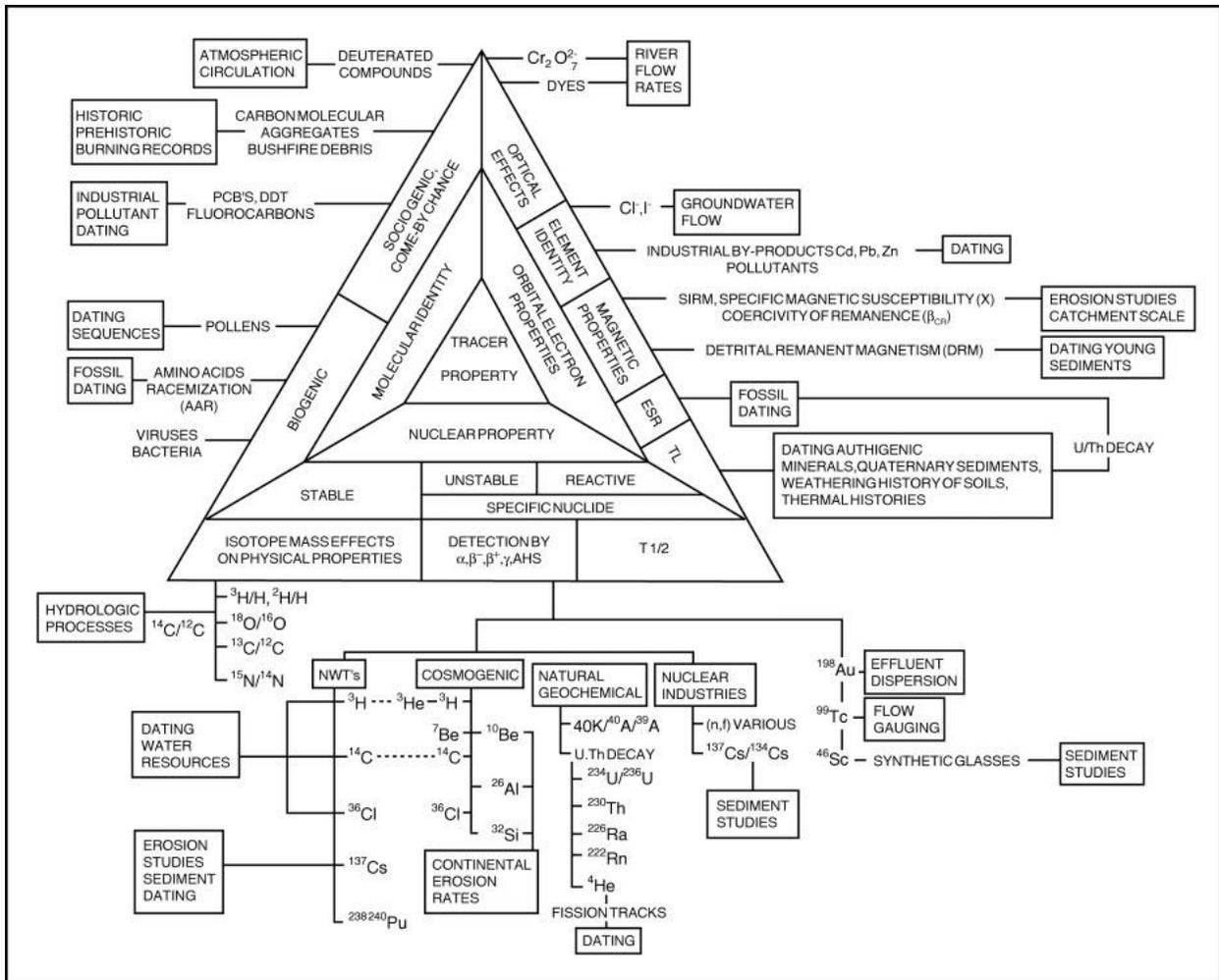
151 Pulley et al., 2015; Sherriff et al., 2015) and concerns over the impact of correction factors to account for
152 differences in particle size and organic matter content (e.g., Smith and Blake, 2014; Kraushaar et al.,
153 2015; Smith et al., 2015). Many would argue that the approach is at a key stage in its development, and
154 that the research community needs to develop some fundamental principles for its application.

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

157 **2. The sediment source fingerprinting approach**

158 There are numerous soil and sediment properties that can be used to discriminate between the potential
159 sources of the sediment within a river basin or coastal/oceanic environment. This section provides a brief
160 overview of these properties; for comprehensive reviews see Foster and Lees (2000), Collins and Walling
161 (2004), Guzman et al. (2013) and Haddadchi et al. (2013). Many of the main fingerprinting properties are
162 shown in Figure 1 and include physical characteristics (e.g., sediment size, shape, colour), geochemical
163 properties (e.g., trace metals), fallout radionuclides (e.g., ^7Be , ^{137}Cs , unsupported ^{210}Pb), mineral
164 magnetic properties (e.g., magnetic susceptibility and isothermal remanence), and organic properties
165 (e.g., compound-specific stable isotopes, microbial communities, pollen). These fingerprint properties
166 have been used individually (e.g., colour; Martinez-Carreras et al., 2010; Brosinsky et al., 2014), within
167 property groups (e.g., fallout radionuclides; Owens et al., 2012; Evrard et al., 2016) or in combination as
168 part of a composite fingerprinting approach (e.g., geochemical, mineral magnetic and fallout
169 radionuclides; Walling and Woodward, 1995; Collins et al., 1997a; Walling et al., 1999) to infer sediment
170 sources.

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173

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

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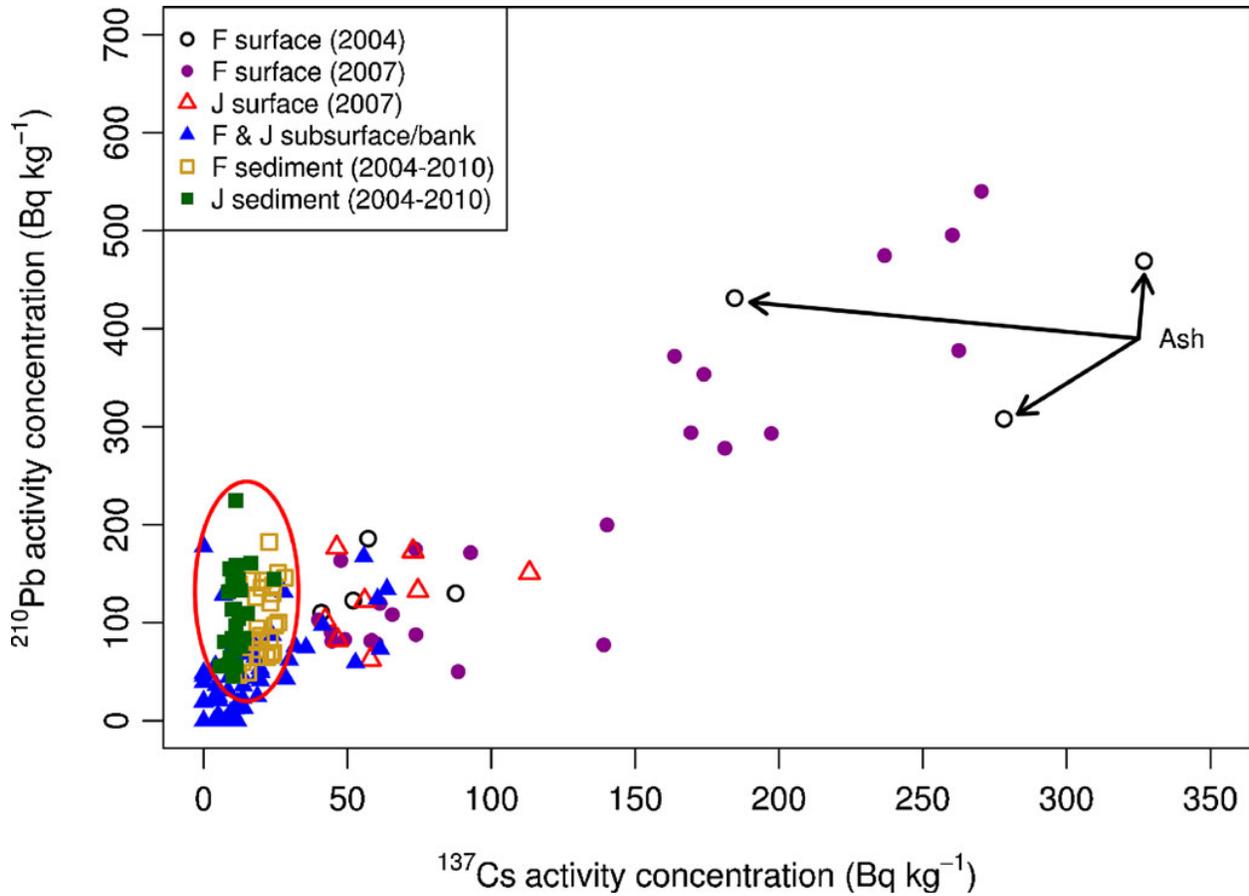
177 The basic principle behind sediment source fingerprinting is that the sediment properties (e.g., trace
 178 element concentrations, radionuclide activities) will reflect their origins. For example, fallout
 179 radionuclides label surface soils and thus high activity concentrations in a sediment sample suggests that
 180 the sediment was derived from surface soils (i.e., topsoil) as opposed to subsoil material which would be

181 expected to have lower activities (Owens et al., 1996; Wallbrink et al., 1998). Similarly, a sediment
182 sample rich in organic carbon would suggest that it was derived from undisturbed surface soils (e.g.,
183 forest, pasture) as opposed to cultivated landscapes or channel bank material (Gellis and Noe, 2013). In
184 most cases, a direct link has been made between the fingerprint properties in the target sediment and
185 those of potential source materials. This assumes that the property used as a fingerprint exhibits a
186 conservative behaviour between source and sink, and that any alterations or transformations are either
187 negligible or can be quantified (Belmont et al., 2014). It is well known, for example, that the particle size
188 distribution of sediment changes as it moves through the landscape (e.g., Walling, 1983; Koiter et al.,
189 2015), and this can influence the concentrations of certain properties, in which case an allowance needs
190 to be made for such changes. In the case of particle size effects, this can be achieved through restricting
191 analysis to a certain size fraction (e.g., $< 63 \mu\text{m}$) or by using correction factors. However, the use of
192 correction factors needs to be exercised with caution (Koiter et al., 2013a; Smith and Blake, 2014), and
193 is discussed later (i.e. section 7.4).

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

204

205



206

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

212

213

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

225

226 **3. Earth and ocean sciences applications**

227 **3.1 Agricultural landscapes**

228 To date, the majority of sediment source fingerprinting studies have been undertaken in agricultural
229 landscapes reflecting the widespread occurrence of agriculture and also concerns associated with soil
230 erosion and off-site effects. Typically, studies have aimed to quantify the relative contribution of
231 agricultural fields to the total sediment budget and to compare such contributions to the sediment
232 delivered from other areas of the basin (e.g., forests, river banks, road ditches, urban areas). The
233 contributions from several case studies, summarized in Table 1, represent only a small fraction of
234 sediment source fingerprinting studies in agricultural watersheds but they highlight the value of the
235 sediment source apportionment approaches. Considering the large range of values estimated for the
236 contribution of agricultural topsoil (i.e., 1-97%), studies prior to and after the implementation of

237 management practices appear relevant in order to focus on problematic areas and to follow the post-
238 change trajectory of sediment dynamics. The great variability of the contributions also highlights the
239 necessity to consider factors other than land cover, which can include agricultural practices, drainage
240 network, slope, weather and antecedent conditions, as these can also influence soil erosion rates. In
241 order to quantify soil erosion rates and to more precisely define the severely eroded areas, it is often
242 necessary to combine sediment source fingerprinting techniques with sediment mass balance estimates.
243 For example, Smith et al. (2014) showed that the increase in the arable area from ~6 % to 36 % for the
244 River Tamar basin (920 km²) in the south-west of the UK over the period 1990-2007 led to a rise in
245 hillslope sediment yield from 8.8 to 32 kt year⁻¹ while the contribution from the erosion of channel banks
246 remained almost constant.

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

250

251 **3.1.1 Inorganic tracers: identification of erosion processes**

252 The information provided by sediment source fingerprinting studies can be improved by combining
253 several tracer properties that have originated as a result of different processes (e.g., due to weathering
254 of bedrock, atmospheric fallout, or land-use). For example, Gruszowski et al. (2003) used mineral
255 magnetic, geochemical and radionuclide signatures to highlight the role of road ditches as a conveyer of
256 30 % of sediment eroded from agricultural topsoils in a watershed in the UK. Determining these
257 preferential pathways also helps to prevent counterproductive management practices. For example,
258 subsurface drain installation in agricultural watersheds has often been considered as an efficient way
259 of preventing erosion by increasing water infiltration rates in soils. Fingerprinting studies in contrasting

260 watersheds in England, Australia and France (e.g., Russell et al., 2001; Wilkinson et al., 2013; Foucher et
261 al., 2015) have demonstrated that large amounts of fine sediment were derived from eroded sub-surface
262 soils, as the subsurface drainage altered the hydrology of the watershed. Similarly, Walling et al. (2002)
263 reported that between 30 % and 60 % of eroded sediment was transferred through subsurface drains in
264 two small agricultural watersheds in the midlands of the UK.

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

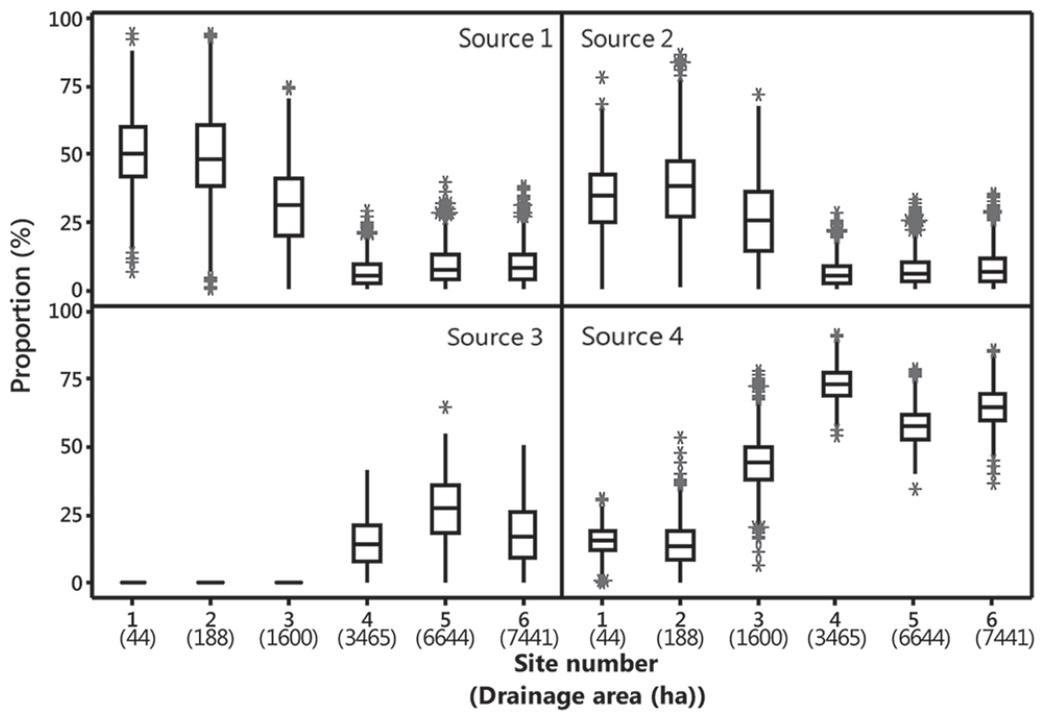
273 In addition to providing useful information on sediment origins in agricultural watersheds, fingerprinting
274 approaches can also be used to investigate the contribution of sediment-associated nutrients (e.g.,
275 nitrogen and phosphorus) and contaminants (e.g., metals, polychlorinated biphenyls) related to
276 agricultural activities (e.g., Walling et al., 2008). Therefore, sediment-associated phosphorus deposited
277 in rivers or lakes can be traced back to cropland or pasture erosion; alternatively, a higher contribution of
278 banks can lead to a dilution of particulate phosphorus by less phosphorus-rich sediment (Lamba et al.,
279 2015b). Nutrient concentrations can also be used as tracers for sediment source fingerprinting (e.g., Ben
280 Slimane et al., 2013), however, the inclusion of such fingerprints are controversial due to the potential of
281 non-conservative behaviour during transport (Koiter et al., 2013a).

282 To illustrate more fully the potential application of inorganic sediment fingerprinting properties for
283 identifying sediment sources in an agricultural watershed, we present findings for the South Tobacco
284 Creek watershed in Manitoba, Canada.. The downstream impacts of sediments and associated
285 contaminants, phosphorus in particular, in this watershed required that sources of sediment be
286 determined and appropriate management practices be developed and implemented. The watershed has
287 an area of ~75 km² and the creek flows east, from the glacial till landscape of the Prairie Pothole Region,
288 over the Cretaceous shale bedrock of the Manitoba Escarpment, across the glaciolacustrine plain of the
289 Red River Valley. The predominant land use is agriculture, largely conventionally cultivated annual crop
290 production. Over several years, samples of suspended and channel-stored fine-grained sediment were
291 collected along the length of the main channel. Potential source materials, including stream bank,
292 bedrock, riparian soil and field soils were collected in association with each sediment sampling location.

293 Koiter et al. (2013b) used geochemical and radionuclide fingerprinting properties to establish the relative
294 contributions of sediment sources. The suspended sediments in the uppermost reaches were dominated
295 by topsoil sources (64 %–85 %), whereas the suspended sediments exported farther downstream had a
296 higher proportion of sediment coming from streambank (32 %–51 %) and shale bedrock (29 %–40 %)
297 sources. This switch in the dominant sources of sediment between the headwaters and the watershed
298 outlet were due to: (i) changes in sediment storage and connectivity; (ii) a transition in the dominant
299 erosion processes from topsoil to streambank erosion; and (iii) the incision of the stream channel
300 through the shale bedrock as it crosses the Manitoba Escarpment. Additional work by Barthod et al.
301 (2015), using colour parameters (i.e., reflectance spectrometry, VIS-NIR) as fingerprint properties,
302 supported the findings of Koiter at al. (2013b) and showed colour to be an effective tracer of sediment
303 (Figure 3). Following these studies, there has been a shift from soil conservation practices that minimize
304 soil losses by water erosion to management practices that manage the runoff. Although inorganic tracers
305 have proved successful in fingerprinting sediment sources, more spatially detailed investigations are

306 needed as the broad sediment sources identified have limited utility in making management decisions.
 307 The use of organic tracers, such as compound-specific stable isotopes, may allow for the identification of
 308 more detailed sediment sources (e.g., crop specific) to be identified.

309



310

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

318

319 3.1.2 Organic tracers: identification of contributions from different land uses

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

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

336 Plants produce FAs that are structurally indistinguishable; for example, all roots produce C22:0 and C24:0
337 FAs (Pollard et al., 2008; Wiesenberg et al., 2012). Differentiating the source of the FAs based solely on
338 vegetation type may be accomplished by examining the carbon isotopic signature of a particular FA.
339 Carbon isotope values reflect the ratio of $^{13}\text{C}:^{12}\text{C}$, and are often reported using the delta scale ($\delta^{13}\text{C}$). The
340 delta value is reported relative to a commonly used standard for that particular element (e.g., Vienna-
341 Pee Dee Belemnite for carbon); values are either negative or positive relative to the standard, which is

342 defined as no shift. Maize and wheat, for example, are C4 and C3 plants, respectively, and, due to the
343 differing methods of CO₂ fixation, should exhibit significant differences between their leaf and root FA
344 δ¹³C values (O'Leary, 1988). Average δ¹³C values for C4 and C3 plants bulk (¹³C_{bulk}) tissue have been
345 reported as -13‰ and -27 ‰, respectively (Glaser, 2005), with FAs further depleted in ¹³C, relative to the
346 bulk tissue, by approximately 6-8 ‰ (Ruess and Chamberlain, 2010).

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

354 To illustrate the application of CSSIs, Blake et al. (2012) investigated a small agricultural watershed (1.45
355 km²) in the UK, where the climate is temperate. The sampling site consisted of permanent pasture, ley
356 pasture, maize stubble, winter wheat and woodland. Blake et al. (2012) used CSSI data to determine the
357 sources of sediment at the stream outlet for the watershed after a period of heavy rainfall, and
358 compared the CSSI-based estimated yields from each source to ones calculated based on geochemical
359 data. They found that the geochemical data overestimated the contribution of cultivated topsoil to the
360 watershed sediment yield during the event as compared to the estimates provided by the CSSI-based
361 tracers. The geochemical-based overestimate was attributed to the ley pasture in the watershed, which
362 constituted a significant portion of the area in the watershed and had been previously cultivated. The ley
363 pasture would have a geochemical fingerprint similar to other cultivated fields, and therefore, the two
364 sources would be indistinguishable by geochemical fingerprinting means. Using an unmixing model

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

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

375

376 **3.2 Forested landscapes**

377 **3.2.1 Deforestation and forest harvesting**

378 Soil disturbance by forest harvesting operations have been widely observed to increase the sediment
379 loads of receiving streams and river networks (e.g., Leeks and Marks, 1997; Douglas et al., 1999; Stott
380 and Mount, 2004; Walling, 2006; Kreuzweiser et al., 2009). In this context, sediment tracing and
381 fingerprinting approaches are of value in identifying sediment sources and hence the processes leading
382 to increased sediment load to inform Best Management Practices (BMPs) (Wallbrink and Croke, 2002).

383 Soil surface disturbances by forestry operations are wide ranging in spatial scale and impact but
384 generally include the direct impact of forestry operations themselves on soil structure, as well as the
385 construction of: (i) temporary log processing areas; (ii) skid tracks along which logs are hauled for
386 processing; and (iii) local and regional road networks (Douglas et al., 1999; Motha et al., 2003). While the

387 skid track and road infrastructure is considered to be the major factor in generating sediment and
388 delivering it to the stream system, it can be difficult to assess the additional role of tree harvest
389 operations using conventional monitoring approaches (Motha et al., 2003). Tracer applications have
390 utilised fallout radionuclide budgeting approaches and sediment source fingerprinting to address this
391 issue – in a similar way to those studies in agricultural landscapes described above (i.e., section 3.1) –
392 and some examples are reviewed below.

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

407 The shorter-lived fallout radionuclide ^7Be has also been used effectively by Schuller et al. (2006) to
408 quantify net soil losses and retention by buffer features following forest harvest operations. The study
409 was based around a single heavy rainfall event and the fallout radionuclide-based data were compared to

410 soil loss estimated using erosion pins with close agreement. The key challenge noted by the study was
411 ensuring that the distribution of ^7Be prior to the event was spatially uniform, especially considering the
412 legacy of prior erosion events, which led to the development of a methodological refinement to apply
413 ^7Be over extended time periods (Walling et al., 2009).

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

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

437 **3.2.2 Wildfires**

438 Post-wildfire sediment source fingerprinting studies have focused largely on quantifying contributions
439 from hillslope surface and channel bank (subsurface) sources of fine sediment using fallout radionuclide
440 tracers (Table 2; also see review by Smith et al., 2013). These studies have employed ^{137}Cs and
441 unsupported ^{210}Pb for source discrimination because of the pronounced differences in activity
442 concentrations of these radionuclides with soil depth in forest environments (Wallbrink and Murray,
443 1996; Blake et al., 2009; Owens et al., 2012; also see Fig. 2). Recent work has also used additional
444 radionuclides such as $^{239,240}\text{Pu}$ to discriminate hillslope and channel sources of fine sediment transported
445 by post-fire debris flows (Smith et al., 2012). In burnt forest environments, fallout radionuclides provide
446 the most useful tracer properties for watershed-scale source discrimination, and may be very effectively
447 coupled with process measurements and monitoring (Smith et al., 2013). For example, suspended
448 sediment flux measurements combined with tracer-based estimates of proportional source contributions
449 allow for calculation of source-specific sediment yields (Smith et al., 2011). Such information is important
450 for interpreting post-fire changes in proportional source contributions to sediment exports associated
451 with vegetation and soil recovery.

452 The discrimination of spatial sources according to burnt and unburnt areas or based on areas burnt at
453 different severities has received less attention. Previous studies have examined the potential for
454 geochemical (Blake et al., 2006a), mineral magnetic (Blake et al., 2006c) and organic compounds (Oros et
455 al., 2002) to discriminate burn-defined spatial sources, but did not attempt to estimate source

456 contributions to downstream sediment fluxes. To date, only Stone et al. (2014) have sought to
457 quantitatively apportion sediment contributions from spatial sources defined according to unburnt,
458 burnt only, and burnt and salvage logged sub-basins. However, this study did not identify any
459 environmental basis for why the selected geochemical tracer properties might discriminate these
460 sources. Difficulties may arise due to natural and burn-related variability affecting the basis for source
461 discrimination. Geochemical and fallout radionuclide tracer concentrations may increase in burnt soils
462 due to the loss of soil organic mass and with inputs of ash from burnt vegetation or decrease due to
463 vaporisation losses during soil heating (Johansen et al., 2003; Blake et al., 2006a, 2006c; Owens et al.,
464 2006, 2012; Perreault et al., 2012). Mineral magnetic properties can be enhanced in surface soils by
465 burning (Longworth et al., 1979; Clement et al., 2011), while heating and post-fire leaching and bio-
466 transformations contribute to changes in organic compounds in burnt surface soils (Oros et al., 2002).
467 Where the ash content of soil accounts for most of the difference in geochemical or fallout radionuclide
468 concentrations between soils burnt at different severities and unburnt areas, the requirement for
469 conservative behaviour of these tracer properties during transport is unlikely to hold. This reflects the
470 potential for density-related differences in the transport behaviour of ash and mineral soil, resulting in
471 the modification of surface soil tracer signatures during downstream transfer. Magnetic properties show
472 the greatest potential for burnt soil spatial source discrimination because burn-related changes are
473 associated with the mineral component of surface soils (Blake et al., 2006b).

474 The forested highlands of south-eastern Australia have been the focus of most source tracing and
475 fingerprinting work following wildfires. Studies were situated in the Blue Mountains in New South Wales
476 and the forested uplands in north-eastern Victoria, both regions subject to severe wildfires. In the Blue
477 Mountains, a multiple-catchment (17-629 km²) source tracing study was conducted over a 5 year period
478 following wildfire (Wilkinson et al., 2009). Fallout radionuclides were used to trace contributions from
479 hillslope surface and subsurface (river bank and gully) sources to in-channel sediment deposits. Post-fire

480 erosion resulted in a change in sediment sources from 80 % subsoil prior to the fire up to 86 % surface
481 soil for one of the study watersheds in the first year after burning (Wilkinson et al., 2009). Hillslope
482 surface contributions then declined after the fire. In north-eastern Victoria, hillslope surface erosion was
483 found to decline from approximately 100 % in the first year to 58% in the fourth year after wildfire in an
484 intensively monitored 1.36 km² research watershed (Smith et al., 2011). Coupling this information with
485 sediment load data showed that hillslope erosion accounted for 93 % of the total post-fire suspended
486 sediment yield over the study period, while the hillslope contribution in the first year after fire equated
487 to 75 % of the measured fine sediment output (Smith et al., 2011). Despite differences between the
488 studies in terms of watershed size, geology, topography, soil and forest type, hillslope surface sources
489 dominated in the first year after wildfire and showed similar declining trends with post-fire recovery. In
490 contrast, studies in North America have used sediment source fingerprinting to demonstrate the
491 dominance of channel bank sources following wildfire. For example, Owens et al. (2012) showed that
492 channel bank sources contributed ca. 91.5% to the sediment flux exported from the 135 km² Fishtrap
493 Creek watershed in British Columbia, Canada, for the period 2004-2010, following a wildfire in 2003. The
494 difference between findings for North America (see also Moody and Martin, 2009) compared to
495 European and Australian landscapes reflect differences in climatic drivers, and vegetation type and
496 recovery (Owens et al., 2013).

497

498 **3.3 Urban landscapes**

499 Compared to agricultural and forested landscapes, relatively few studies have quantitatively determined
500 the sources of sediment in urban watersheds with studies to date being concentrated in Australia (e.g.,
501 Ormerod, 1999; Charlesworth et al., 2000), Brazil (e.g., Poleto et al., 2009; Franz et al., 2014), China (e.g.,
502 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

527 (including the cities of Leeds and Bradford) the relative contribution from these source types decreased
528 (combined = 22-40 %) reflecting contributions from additional sources such as cultivated topsoils (20-45
529 %), solids from STWs (14-18 %) and road-deposited sediment (RDS, 19-22 %). The change in the
530 contribution of channel bank sources also reflects the artificial protection of many channel banks from
531 erosion and lateral migration in urban areas.

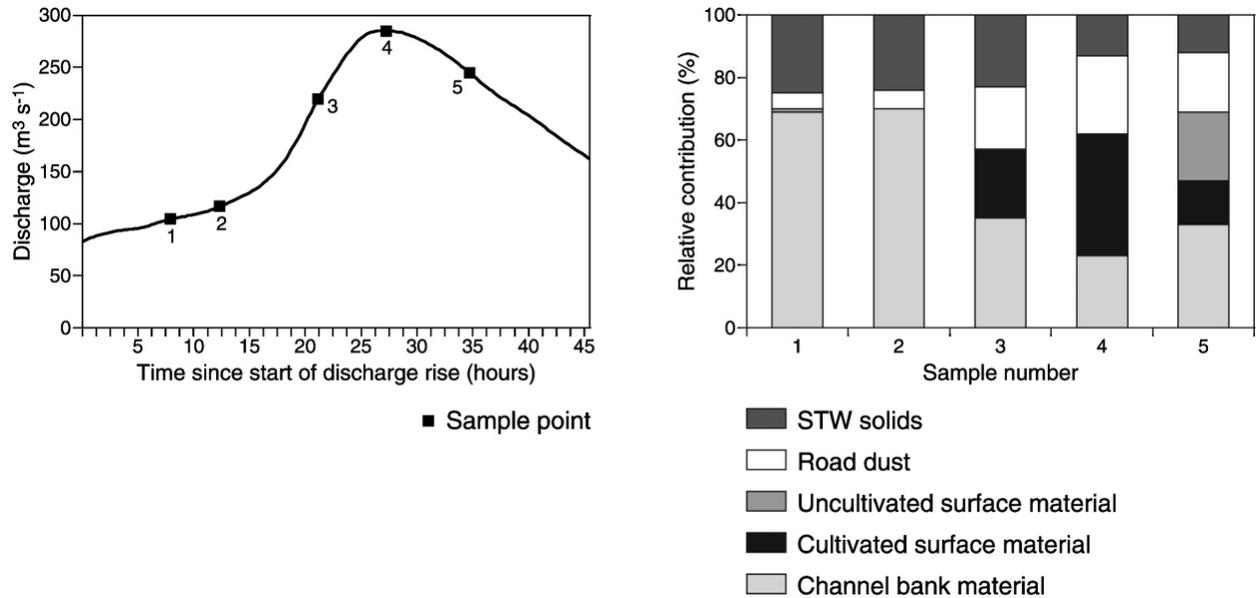
532 The high contribution of urban sources (ca. 40 % from STWs and RDS) illustrates the marked contrast of
533 sediments in urban watersheds to those in non-urbanized watersheds described in previous sections.
534 The results from the River Aire are consistent with those of Yin and Li (2008) who used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

541 One of the important aspects of these studies is that while the contribution from urban sediment
542 sources such as RDS and solids from STWs has been recognised (e.g., Foster and Charlesworth, 1996;
543 Charlesworth et al., 2000; Owens et al., 2011), fingerprinting has enabled such contributions to be
544 quantified, albeit in a relative sense. Furthermore, the work by Carter et al. (2003) also provided
545 important new information on the timing of such contributions, and how this changes during the course
546 of a rainfall event (Fig. 4). In the case of RDS, contributions from this source increased during the event
547 reflecting this increased connectivity of the road network to the channel system as the rainstorm
548 progressed (i.e., more runoff), while the contributions from STWs and other sources decreased during

549 the course of the event because of dilution effects. The complex hydrology and flashy nature of urban
550 catchments can pose significant challenges for sampling design and monitoring efforts.

551

552



553

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

558

559 Other source tracing and fingerprinting studies in urban environments have been concerned with
560 atmospheric sediment (i.e., dust and airborne fine particulate matter). Much of this interest has been
561 driven by the health concerns associated with the impacts of fine particulate material (e.g., PM_{2.5}) on the
562 human respiratory system. Thus numerous studies have attempted to fingerprint the sources of fine

563 airborne particulates in urban areas based on the geochemical properties of the sediment (e.g., Park and
564 Kim 2005), and these are reviewed in Section 6.

565

566 **3.4 Estuarine and coastal landscapes**

567 At the interface between watersheds and the marine environment, estuaries represent critical sink zones
568 for fine sediment and associated nutrients and contaminants. Estuarine morphology and fluvial and tidal
569 currents are key factors in determining the processes that control sediment accretion and, linked to this,
570 knowledge of sediment source is a key part of the management of sediment problems and remediation
571 strategies. Sediment source information is also critical to inform coastal management strategies
572 particularly in the context of management realignment schemes for flood defence and habitat
573 restoration (Rotman et al., 2008). Although limited in number compared to river basin studies (described
574 above), a range of sediment tracing and fingerprinting approaches have been used in estuarine and
575 coastal environments including recent advances in CSSIs (Gibbs, 2008), application of mineral magnetic
576 properties (e.g., Yu and Oldfield, 1989; Jenkins et al., 2002, 2005; Rotman et al., 2008), rare earth
577 elements (REEs) (Zhou et al., 2010) and radionuclide fingerprints (e.g., Hebinck et al., 2007; Yeager et al.,
578 2005, 2006) – with a focus on geogenic uranium and thorium series-based signatures. These tracers are
579 largely controlled by mineralogy and are therefore less likely to be affected by transformations
580 associated with transfers from freshwater to saline conditions. This is an important consideration for acid
581 soluble geochemical properties that have been extensively used in river basin systems (section 3.1).

582 Siltation problems are a key driver for knowledge of sediment sources and sediment fluxes to estuaries.
583 Gibbs (2008) presents a study where CSSI signatures were used to apportion sediment deposited in
584 estuarine mangroves back to potential upstream source areas. The agricultural watershed feeding into
585 the estuary in northern New Zealand presented a range of land uses from pasture (~70 %) to native

586 forest (~20 %) and plantation pine forest (~8 %) in the steeplands plus ~4 % urban coverage. In addition,
587 outer estuary sediments were sampled as a separate source. The study was driven by notable declines in
588 the abundance of a suspension-feeding bivalve which were hypothesised to result from enhanced
589 suspended solids linked to watershed disturbance with implications for oyster aquaculture sited on the
590 inter tidal flats. The isotopic signature highlighted the input of the pine plantation activities to sediment
591 stored in upper and mid estuary deposits. The lower estuary was dominated by reworked materials from
592 the coastal zone. In addition to identifying the potential of this relatively new sediment tracing and
593 fingerprinting tool (i.e., CSSIs; also see section 3.1.2), the study highlighted the wide variation in source
594 inputs in different zones of the estuary and the importance of comprehensive sampling strategies in
595 sediment sink zones.

596 As well as being perceived as an environmental problem in estuaries, sediment supply can also be a key
597 factor in the development of coastal zone habitats. Where management strategies are designed to
598 enhance these, or mitigate detrimental effects, knowledge of sediment source can be critical part of the
599 decision-making process. As such, sediment source fingerprinting has been used to identify sources of
600 sediment entering barrier reefs such as those in Australia (e.g. Douglas et al., 2010; Bainbridge et al.,
601 2016) and other sensitive coastal environments. In this context, Rotman et al. (2008) used mineral
602 magnetic signatures to develop knowledge of sediment sources contributing to the restoration of
603 intertidal flats and saltmarsh. A particular focus was to examine how the managed realignment sites
604 interacted with wider geomorphological processes in the estuary. The sampling programme focussed on
605 proximal sources of sediment to include established saltmarsh and intertidal deposits seaward of the
606 study site, plus terrestrial sources. Magnetic signatures were shown to discriminate the sampled sources
607 based largely on different magnetite concentration and magnetic grain size. Apportionment results
608 suggested that a large component of deposited material in the managed realignment site was derived
609 from existing eroding saltmarshes which did not align with the management goal of habitat creation.

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

613

614 **3.5 Oceanic environments and sedimentary basins**

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

630 Weltje and von Eynatten (2004) identified and described three main approaches to
631 characterizing the properties of sediment within provenance studies and include the analysis of: 1) bulk
632 sediment; 2) specific groups of minerals; and 3) single grains. The analysis of bulk sediment is one of the

633 more common approaches used in many of the other fingerprinting applications described in this review
634 (e.g., Section 3.1.1) and includes the determination of the concentration of major, trace, and rare-earth
635 elements (REE), isotopic ratios, and bulk mineralogy. It is interesting to note that REE concentration data
636 in these types of provenance studies, are often presented as normalized to either the average upper
637 continental crust (UCC) concentration (e.g., Padoan et al., 2011) or to chondrite (a stony meteorite)
638 concentration (e.g., Meinhold et al., 2007). This normalization makes it easier to display the values as
639 some elements are orders of magnitude different, and allows for the detection of anomalously high (> 1)
640 or low (<1) concentrations.

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

652 Single grain analysis is typically limited to the coarse-grained sediment (> 63 µm) as the silt and
653 clay grains are too small given the resolution of the analytical equipment (Weltje and von Eynatten,
654 2004). However, it is important to consider that single grain analysis provides information on the source
655 of the mineral of interest which can result in biased results with respect to the source of the sediment as

656 a whole (Morton and Hallsworth, 1994). There are two main types of single grain analysis - geochemical
657 composition and radiometric dating - and von Eynatten and Dunkl (2012) provide a review of single grain
658 techniques. For geochemical analysis (also known as varietal heavy mineral analysis), the variability in
659 the concentration of major and trace elements of different minerals are used to differentiate between
660 sediment sources (e.g., Tsikouras et al., 2011). One of the more common single grain radiometric dating
661 analysis used to fingerprint sediment is zircon geochronology. The mineral zircon is highly resistant to
662 both chemical and physical weathering and is commonly found in many sedimentary deposits making it
663 ideal to fingerprint. The premise of using geochronology as a fingerprint is to link the sediment to the
664 source of sediment based on the age of the parent rock as the age of the mineral is interpreted to be the
665 crystallization age of a rock (Thomas, 2011).

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

679 Cook et al. (2013) presents an interesting case study using the isotopic ratios of $^{143}\text{Nd}/^{144}\text{Nd}$ and
680 $^{87}\text{Sr}/^{86}\text{Sr}$ as sediment fingerprints to identify the sources of sediment in a glaciated environment. In this
681 study, Cook et al. (2013) assessed the vulnerability of the east Antarctic ice sheet (predominately land-
682 based) to warmer temperatures as a result of a changing climate. This was undertaken by investigating
683 sediments that were deposited during an interval in the geological record when the temperature was
684 warmer than present. A core was drilled 310 km offshore (64°24'5" S 143°53'1" E; 3,465 m water depth)
685 and the section of the core that was studied was 75 – 125 m (below sea floor) and was deposited during
686 the Pliocene (5.3 - 3.3 million years ago) when the mean annual global temperature was 2 - 3 °C higher
687 than present. The core section was characterized by two alternating sedimentary layers: diatom-rich silty
688 clay sediment and diatom-poor clayey sediment layers representing warmer and cooler temperature
689 periods, respectively. Within the vicinity of the ice sheet, the isotopic ratios varied between the different
690 rock types and these variations were attributed to differences in both age and lithology and the isotopic
691 ratios provided good discrimination between the potential sources of sediment. The isotopic signatures
692 varied in a systematic manner that paralleled the alternating sedimentary layers suggesting a switch in
693 the dominate source of sediment between the warmer periods (ice retreat) and the cooler periods (ice
694 advance). By estimating the relative contributions from the different sediment sources the patterns of
695 erosion could be inferred and this information provided some direct evidence of the locations of ice
696 margin retreat. Overall, this research provided information that helps predict the future dynamics of the
697 Antarctic ice sheet under a warming climate.

698

699 **4. Archaeological and geoarchaeological applications**

700 There is a long tradition of geological “provenance” or source fingerprinting research in archaeology and
701 geoarchaeology. Two broad areas of research activity may be identified. The first is aimed at a better

702 understanding cultural processes of raw material exploitation and use, while the second involves a suite
703 of geological and anthropogenic processes associated with the creation of the archaeological record.
704 They may be summarised as studies investigating the procurement of sedimentary material or natural
705 depositional processes.

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

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

721

722 4.1 Fine-grained sediments as raw materials and sourcing clays for pottery

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

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

746 et al., 2015). Table 4 shows a number of sediment fingerprinting studies and a range of tracer properties
747 applied to the study of ceramic raw material sources.

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

761 On the Greek island of Aegina in the southern Aegean Sea, Christidis et al. (2014) developed an
762 integrated multi-method approach to establish the source of clays used by Bronze Age potters. They
763 made use of geographical, petrographic, mineralogical, mineral chemistry, and geochemical data to
764 create a substantial database to characterise the composition of the ceramics and to allow effective
765 discrimination between potential sources of clay-rich sediments on the island. Christidis et al. (2014)
766 were able to isolate a distinctive outcrop of Pliocene volcanic clay as the exclusive raw material source
767 for the distinctive Bronze Age ceramics on Aegina. They used the same approach to exclude older
768 Pliocene marls from the same location as a potential raw material source. In common with some of the

769 early quantitative sediment source fingerprinting work in river basins (e.g. Walling and Woodward,
770 1995), they stressed the importance of using a multi-tracer approach to avoid ambiguous outcomes.
771 Whether a project is seeking to establish the composition of a pot sherd or a sample of fluvial suspended
772 sediment or use this information to trace the source of the constituents in the wider landscape, the
773 guiding principles are essentially the same. It is also important to appreciate that suspended sediment
774 samples from river basins are complex environmental mixtures that typically include material from
775 multiple diffuse source types. In contrast, ceramics can be produced from raw materials from one or two
776 distinctive point sources. The principles of source identification are essentially the same in each case,
777 however, but this can sometimes make identification of the dominant raw material source both
778 theoretically and practically more straightforward in these archaeological contexts. There is clearly a
779 great deal of scope for collaboration and cross-fertilisation of ideas between these sediment source
780 fingerprinting communities – especially in relation to the statistical treatment of source data and the
781 unmixing of target samples whether these end products result from natural or anthropogenic mixing. In
782 fluvial geomorphology, watershed sediment budget investigations help us to better understand the
783 operation of watershed processes and the evolution of landscapes. In a similar way, linking the
784 composition of a pot to raw materials in the landscape helps us to better understand cultural processes
785 and decision making in the past and directly links archaeological sites and past societies to their wider
786 landscape (Michelaki et al., 2015; Ratto et al., 2015).

787

788 4.2 Sediment sources and archaeological site formation

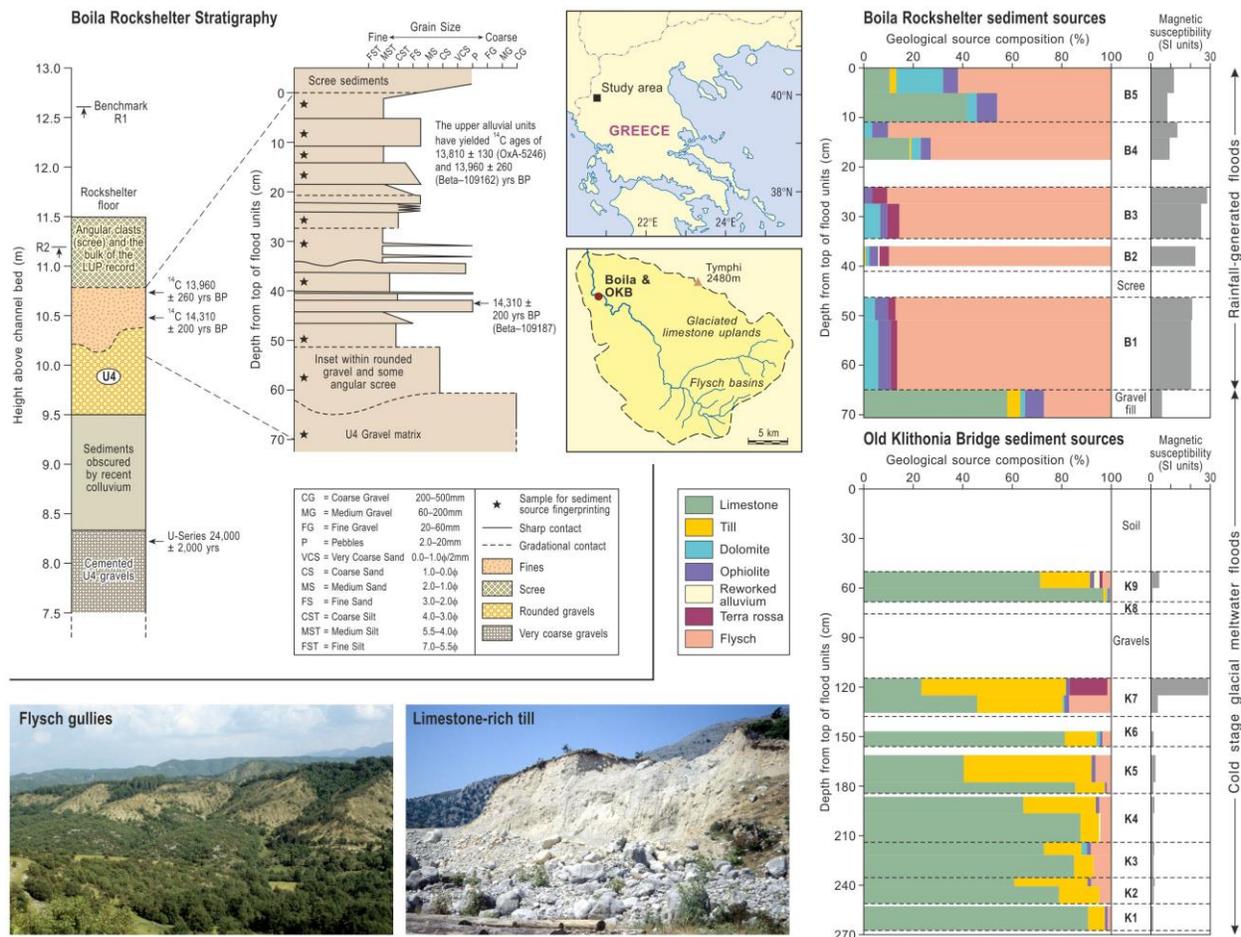
789 An understanding of sediment sources can also provide valuable information on the formation of
790 depositional records in many kinds of archaeological site (see Goldberg and Macphail, 2006). In the case
791 of rockshelters and caves formed in hard limestone bedrock, the fine-grained components within

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

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

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

825



826

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

836 2006). A key feature of these Late Pleistocene sediment fingerprinting data is the abrupt end of the
837 meltwater-dominated system during the last deglaciation.

838 **5. Forensic applications**

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

848 Soil forensics draws many parallels with the other applications of sediment source fingerprinting outlined
849 in this review. Similarly, locating and appropriately characterizing source materials (e.g., spatial
850 variability; see Section 7.1) is important and other evidence can be used to narrow the sampling area
851 (e.g., near footprints) (Pye et al., 2006; Dawson and Hillier, 2010). In some cases, the potential sources of
852 the soil or sediment may not be immediately known and the potential source needs to be deduced from
853 the evidence collected (e.g., Lombardi, 1999; see below for a description of the case). The selection of
854 independent measurements (i.e., some soil properties are correlated; see Section 7.2) is important as
855 this provides the strongest case for determining the origins because the use of non-independent
856 properties as supportive evidence may be problematic in a court of law (Morgan and Bull, 2007a). The
857 contamination or mixing of soil/sediment pre-, syn- and post-forensic investigation are important
858 considerations as this may lead to erroneous conclusions (Morgan and Bull, 2007a). This is where the

859 development of protocols in documenting, collecting, preserving, preparing, analyzing and interpreting
860 the data is important (also see Section 7.3). In a similar way to the issues surrounding particle size
861 correction factors (also see Section 7.4), it is important to have an understanding of the mechanism of
862 soil/sediment transfer as the process can be particle-size selective (e.g., clay tends to stick to footwear
863 whereas sand does not) (Dawson and Hillier, 2010).

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

874 Very few cases are reported fully in the literature due to the sensitive nature of criminal investigations.
875 Lomardi (1999) presents an interesting case report in which soil forensics contributed to a murder
876 investigation. On March 16, 1978, the Italian Prime Minister, Aldo Moro, was kidnapped and on May 8,
877 1978 he was found dead, from gunshot wounds, in the trunk of a parked car near the centre of Rome.
878 Small amounts of sand and soil were collected from the victim's trouser cuffs, shoes and coat pockets as
879 well as from the floor, fenders and tires of the vehicle in which the victim was found. Along with the soil
880 and sand there were also traces of plant material, asphalt, fibres and an assortment of building materials
881 (e.g., brick chips). These samples were used to help identify the origin of the vehicle and the last

882 whereabouts of the victim. A range of analytical techniques was used to characterize both the sand and
883 soil samples including: particle size analysis (sieving), particle morphology (microscopy), mineralogy (x-
884 ray diffraction, scanning electron microscopy and polarized microscopy), soil colour (microscopy), pollen
885 analysis (microscopy) and the identification of micro-fossils (microscopy).

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

895 The soil samples contained a mixture of halloysite-rich clay, glassy scoriae and an assemblage of volcanic
896 minerals with an overall reddish-brown colour. The soil samples were identified as volcanic in origin;
897 however, this type of soil is very prevalent in the area covering more than 6000 km². The vast areal
898 extent of this soil type prevented any meaningful insight to the origin of the soil. Pollen extracted from
899 the soil was identified as coming from Cypress and Hazel trees, which is produced in the winter. This
900 information established that the soil adhered to the vehicle prior to the abduction of the victim. The
901 fingerprinting of the soil and sand evidence found on the victim and car was one line of enquiry that
902 helped to reconstruct a timeline of events leading up to the murder and to corroborate the testimony
903 from potential suspects.

904

905 **6 Human health applications: fingerprinting airborne particles**

906 Soils and sediments, and the contaminants associated with them, can be a risk to human health
907 (Abrahams, 2002). The main pathways by which soils and sediments and associated contaminants enter
908 humans are: direct ingestion; inhalation through nose and mouth; and adsorption through the skin.
909 Consequently, there has been an interest in determining the origin of soils and sediments known to be a
910 risk to human health. One such application is the identification of the sources of very fine airborne
911 particles, especially in urban areas. In this context, particulate material with a diameter less than 10 μm
912 (i.e. PM_{10}) or 2.5 μm ($\text{PM}_{2.5}$) is of greatest concern because when they are inhaled they are able to
913 penetrate deep into the alveoli where they can be deposited and absorbed. The effects include chronic
914 lung disease, lung cancer, influenza, asthma, and increased mortality. In addition to the small size of such
915 particles, they are also detrimental to human health because of the contaminants associated with them,
916 such as metals and polycyclic aromatic hydrocarbons (PAHs). Numerous studies have collected such
917 particles and used geochemical fingerprints to identify sources. The approach is broadly similar to that
918 described above for other applications, and involves establishing diagnostic chemical (e.g., trace
919 elements, organic elements, PAHs) and physical (e.g. particle morphology) fingerprints, and the use of
920 statistical and modelling approaches (e.g. Receptor Modelling, Positive Matrix Factorization, Principal
921 Components Analysis) to quantitatively determine source types or spatial areas (Breed et al., 2002;
922 Dogan et al., 2008; Callén et al., 2014; Huang et al., 2014; Suman et al., 2014). Typically, airborne
923 particles are collected using specialised samplers with filters. Some studies in the urban environment
924 have determined sources of road-deposited sediment (RDS) which is itself a major source of PM_{10} and
925 $\text{PM}_{2.5}$ and an important contributor to the fine-grained sediment load of urban rivers (Owens et al.,
926 2011; Karanasiou et al., 2014).

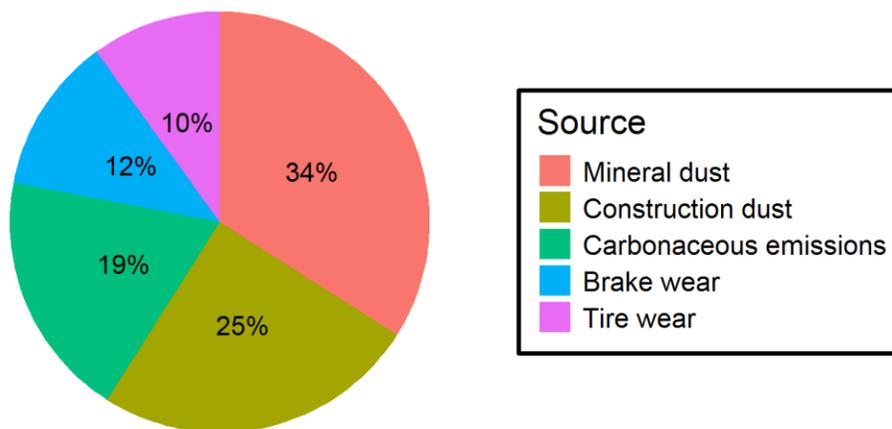
927 For example, Karanasiou et al. (2014) collected samples of RDS from Madrid, Spain, in 2009. They used
928 chemical analysis (major and trace elements, carbon) of the $<10 \mu\text{m}$ fraction to establish fingerprints and

929 applied Positive Matrix Fractionization to determine its source. Their results (Fig. 6) illustrate the
930 importance of both natural and anthropogenic sources of fine-grained RDS and thus PM₁₀, including
931 sources from the construction industry. Such information was used to provide guidance on street
932 sweeping approaches in Madrid to minimise detrimental impacts on human health. Other studies (e.g.
933 Merefield et al., 1999, 2000) have used similar approaches (including SEM/EDAX and XRD analysis) to
934 determine sources of airborne particles associated with construction, mining and other resource
935 extraction industries.

936

937

938



939

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

943

944 **7. Methodological considerations and recommendations**

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

949

950 **7.1 Collection of soil and sediment samples**

951 As with most areas of earth and environmental sciences, field sampling of the materials under
952 investigation (i.e., soils and sediments) represents a key consideration. The design of the sampling
953 programme needs to be flexible to suit the specific environment and research objective (e.g., the various
954 applications described in Sections 3 to 6), since there cannot be a single approach to the sampling of
955 soils and sediments. However, it is important to optimise the sampling of soils and sediments in order to
956 characterize both the spatial and temporal variability of fingerprint properties. For example, in the case
957 of surface soil material, samples of the upper 0-2 cm layer are usually collected and this is assumed to
958 represent the material mobilised by erosion processes and delivered to stream channels. Even with
959 adjustments for particle size effects (see section 7.4) such material is unlikely to be truly representative
960 of the material that is delivered to channels, coastal areas or oceans. Thus, it may be more appropriate
961 to sample eroded soil and sediment that is being actively transported. This could be achieved by
962 sampling during runoff events or installing suitable samplers (i.e., Gerlach troughs). Furthermore, some
963 studies (e.g. Walling et al., 1999; Laceby et al., 2015b) have used suspended sediment samples collected
964 from tributary streams to characterize the sources (type and spatial) of suspended or deposited
965 sediment in downstream waterbodies, thereby reducing the need for correction factors. In addition,
966 there are concerns surrounding the nature of the most appropriate sampling design and how effectively

967 it characterizes a particular source (e.g., random, transect or strategic sampling) or tracer property
968 (Wilkinson et al., 2015). Understanding the geomorphological context in terms of the erosional history of
969 the landscape (at least in a general way) and the dominant water and sediment transport pathways can
970 be helpful in the design of an appropriate sampling framework.

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

984 It is also necessary when developing a source material and sediment sampling protocol to consider the
985 requirements of the statistical and numerical approaches that are used to convert fingerprinting
986 property data into quantitative estimates of sediment sources. For example, in the case of source
987 materials, there is a need consider the number of samples to be collected so as to provide a meaningful
988 representation of the fingerprinting properties of that source. The number of samples to be collected
989 should be guided by the spatial variability of the fingerprint properties in that source, which is likely to

990 vary for different properties (e.g. fallout radionuclides, geochemical properties, colour parameters, CSSIs)
991 and different sources (e.g. cultivated fields, forest, channel banks). In addition, the spatial variability of
992 source materials is likely to increase with larger study areas. Furthermore, while the concentrations of
993 some properties may be essentially random within sources, others (e.g. fallout radionuclides) may
994 exhibit some spatial structure due to erosion and deposition processes or environmental gradients (e.g.
995 Wilkinson et al., 2015) and may require a stratified sampling protocol (e.g. Wilkinson et al., 2015) and/or
996 the collection of samples along transects or environmental gradients (e.g. Koiter et al., 2013b). Thus,
997 while some studies have provided estimates of the number of samples to be collected (typically of the
998 order of 30 composite samples per source type), in reality each study area is different and requires site-
999 specific considerations.

1000

1001 **7.2 Selection of fingerprint properties**

1002 While it may be possible to measure soil and sediment samples for hundreds of different physical,
1003 chemical and biological properties, of paramount importance is the selection of properties that make
1004 sense in terms of how fingerprints have developed (Collins and Walling, 2002). This is required for three
1005 reasons: (i) so that the source and sediment samples are collected in the correct way (e.g., soils and
1006 sediment to the correct depth, as in the case of fallout products like ^7Be or artificial amendments like
1007 fertilisers); (ii) so that laboratory analysis is cost-effective (i.e., avoid unnecessary analysis); and (iii) so
1008 that the user can understand and interpret the data. Again, this will depend on the study site and the
1009 purpose of the investigation. In most cases the reason for the study can help to inform the selection of
1010 suitable fingerprints. Thus, if the aim is to investigate the relative contributions of topsoil compared to
1011 channel bank sources, then properties that either label surface materials (e.g., some organic properties,
1012 fallout radionuclides and other fallout products, and agricultural amendments) or are enriched in subsoil

1013 materials (i.e., geochemical properties that reflect bedrock or surficial materials) are likely to be worth
1014 considering.

1015 Once soil and sediment properties have been selected and measured, an addition step prior to further
1016 statistical and numerical analysis is to plot the data (i.e. bi-plots; Figure 2) and consider if the data make
1017 sense in the context of any prior knowledge concerning the environmental distribution of a given
1018 fingerprint property. For example, if fallout radionuclides are elevated in subsoil or channel bank
1019 material compared to topsoil, then either the property is not appropriate as a fingerprint and/or may
1020 have been compromised (e.g., soil disturbance) or there are additional processes that are occurring
1021 which warrant further investigation. For example, Owens et al. (2012) measured some subsoil samples
1022 with high ^{137}Cs and unsupported ^{210}Pb values in a wildfire-affected watershed in British Columbia,
1023 Canada (see Fig. 2). In this case, the complete combustion of individual trees had exposed subsoil
1024 materials to new fallout of unsupported ^{210}Pb and some redistribution of old ^{137}Cs -enriched surface soil.
1025 For the fingerprinting technique to be robust and repeatable it is crucial that the processes controlling
1026 their behaviour within the particular landscape or river basin are well understood and can be predicted.
1027 Furthermore, fingerprints that fall below the detection limit in one or more samples are typically
1028 discarded without consideration as to whether the fingerprint has the potential to provide good
1029 discrimination. There are statistical procedures available to deal with left-censored data but their uses in
1030 sediment source fingerprinting studies have not been fully explored.

1031

1032 **7.3. Laboratory analysis and data reporting**

1033 While there has been a reasonable amount of debate on appropriate field sampling and statistical
1034 procedures (e.g. Walling, 2005, 2013; Koiter et al., 2013a; Smith et al., 2015), there has been much less
1035 discussion of laboratory analysis. In this respect, appropriate sample storage and preparation prior to

1036 analysis represent fundamental requirements, particularly as the requirements are likely to vary for
1037 different analyses. This is likely to be especially so for organic tracers, and to some extent for colour
1038 analyses due to issues associated with geochemical stabilization. As sample analysis can be destructive
1039 for some determinants (e.g., particle size, organic matter via loss-on-ignition, some geochemistry) there
1040 is also a need to consider the order of analysis if a sample is being analysed for a range of determinants
1041 (i.e., particle size, colour, geochemical, radionuclide, CSSI properties). This is especially relevant for low-
1042 mass samples.

1043 Once samples have been analysed, then there should be a detailed and complete reporting on analysis,
1044 including the reporting of any deviations from standard protocols. This should include the reporting of
1045 fingerprint data in publications. At a minimum, this should take the form of summary table(s) containing
1046 source and sediment fingerprint data without any corrections or modifications. It could also extend to
1047 the inclusion of complete datasets tabulated in supplementary material. Such a step would then allow
1048 readers to run the same procedures described in the paper to confirm the outcomes of the data
1049 processing and source unmixing. This enhanced level of transparency would be likewise supported by
1050 reporting of specific statistical programmes or R packages used in the analysis, along with inclusion of
1051 any bespoke code in the supplementary material (see section 7.6). Such steps could make significant
1052 progress towards reducing ambiguities and ensuring reproducibility in data treatment and analysis
1053 between sediment source fingerprinting studies.

1054

1055 **7.4. Particle size and organic matter correction factors**

1056 One of the most controversial aspects of the sediment source fingerprinting technique is the use of
1057 correction factors to account for differences in some properties between sources and sediments. The
1058 two main correction factors relate to the particle size distribution and organic matter content, as many

1059 fingerprint properties are often related to these two parameters (Horowitz, 1991). In many studies (e.g.,
1060 Walling and Woodward, 1995; Collins et al., 1997a; Walling et al., 1999; Owens et al. 2000) relatively
1061 simple correction factors are determined based on the ratio of measures of the particle size distribution
1062 (i.e., median particle size or specific surface area) and/or organic matter content (i.e., OM%) of the
1063 source and sediment samples. It is typically assumed that the relation between tracer property
1064 concentration and particle size or organic matter content is linear. However, some studies (e.g., Foster et
1065 al., 1998; Russell et al., 2001; Smith and Blake, 2014; Taylor et al., 2014) have demonstrated that such
1066 relations can be more complex, and may be tracer- and site-specific. Other studies (e.g., Stone and
1067 Walling, 1997) have shown that the particle size composition and organic matter content of sediment
1068 can change as it moves from source to sink. For example, Koiter et al. (2015) used a recirculating flume to
1069 simulate river channel conditions, and documented that both particle size and organic matter content
1070 changed over time (i.e. distance travelled) and that the degree of change was influenced by the depth
1071 and porosity of the channel bed sediment. Indeed, some studies (e.g., Martinez-Carreras 2010; Dutton et
1072 al., 2013; Koiter et al., 2013a; Smith and Blake, 2014; Palazón et al., 2015a) have stated that particle size
1073 and organic matter correction factors are inappropriate. Smith and Blake (2014) have demonstrated that
1074 such manipulation of the raw data can significantly change source apportionment results. Thus it is
1075 important to consider if such corrections factors are appropriate, and clearly further research is required
1076 to address this concern.

1077 Other studies have attempted to overcome problems of particle size and/or organic matter influences by
1078 focussing on a specific size fraction. While most studies restrict analysis of source materials and sediment
1079 samples to the <63 μm fraction, other studies have fractionated source and sediment samples prior to
1080 analysis. For example, Haddadchi et al (2015) separated samples into fine sand (63-212 μm), silt (10-63
1081 μm) and fine silt and clay (<10 μm). The selection of finer size fractions can remove some of the
1082 confounding influences of changing particle size composition between source and sink, but it may also

1083 limit the representativeness of the results, as most suspended sediment in rivers is $>2\ \mu\text{m}$ (Walling et al.,
1084 2000).

1085 The choice of particle size fraction for analysis can influence the source apportionment results
1086 (Haddadchi et al., 2015). Hence, it is important that studies explain the choice of size fraction for analysis
1087 in relation to the research context and objective. Previously the choice of the $<63\ \mu\text{m}$ fraction was
1088 justified because it was considered representative of sediments largely transported in suspension that
1089 were of most concern in terms of water quality impacts and the transport of particle-bound
1090 contaminants (e.g. Walling et al., 1999). However, such a choice may be unsuitable in environments
1091 dominated by sandy materials or where the research objective is unrelated to water quality.
1092 Alternatively, a focus on a finer fraction such as $<10\ \mu\text{m}$ may be justified by concerns related to the
1093 specific impacts of these finer-grained sediments on particular ecosystems, such as coral reefs
1094 (Bainbridge et al., 2016) and oceanic environments. It may also be justified from a process perspective,
1095 for example, in the study of sediment source contributions to post-fire debris flows, where fine sediment
1096 supply is an important factor in debris flow initiation from burnt hillslopes (Smith et al., 2012).

1097

1098 **7.5 Conservative behaviour of soil and sediment properties**

1099 One of the fundamental assumptions underpinning the source tracing and fingerprinting technique is
1100 that the properties of the collected sediment samples are directly comparable to those in the potential
1101 source materials. In some cases, allowance is made for differences in the particle size and organic matter
1102 content between the two types of materials (see section 7.4). However, Koiter et al. (2013a) have
1103 recently reviewed the broader environmental literature on the range of soil/sediment properties used as
1104 fingerprints and demonstrated that for several properties there exists the potential for considerable
1105 modification in concentrations as sediments move through river basins; i.e., from hillslopes through

1106 riparian zones, into and through river channels, and into deposition in floodplain, lake/reservoir,
1107 estuarine and marine environments. Several properties, such as phosphorus, are known to exhibit non-
1108 conservative behaviour during fluvial transport and short-term storage in river channels (e.g., Withers
1109 and Jarvie, 2008), while other properties are known to undergo transformations in medium- to long-
1110 term storage elements such as floodplains, lakes, wetlands and oceans due to changes in redox, pH,
1111 salinity and other environmental conditions (Hudson-Edwards et al., 1998; Owens et al., 1999; Foster et
1112 al., 2006; Pulley et al., 2015).

1113 While it may be possible to account for such behaviour, most sediment source fingerprinting studies do
1114 not do this in a rigorous or standardized way. The conservative behaviour of tracers can be assessed
1115 through experimentation. For example, Poulenard et al. (2012) assessed the conservative behaviour of
1116 Diffuse Reflectance Infrared Fourier Transform Spectrometry (DRIFTS;) including colour) properties by
1117 placing samples in micro-porous bags and submerging them in the river for up to two weeks. They found
1118 that the DRIFTS signature was sufficiently conservative to be used as a potential fingerprint property. In
1119 addition, Legout et al. (2013) assessed the conservative behaviour of the colourimetric properties of
1120 source materials in a similar experiment for up to 63 days, and found comparable results.

1121 A common procedure to help identify if fingerprint properties are behaving conservatively is to compare
1122 the property values (mean, median or range) of the sources to those of the collected target sediment,
1123 sometimes called the “range test”, with the idea that if the sediment values for a particular property fall
1124 outside of the values for the potential sources that either the property is exhibiting non-conservative
1125 behaviour or that not all sources have been identified or fully characterised. Properties that fail this test
1126 are often discounted as potential fingerprints and removed from further analysis. This approach
1127 represents a useful test of property behaviour, although it represents only part of the solution. The range
1128 test cannot definitively identify all tracers that are behaving non-conservatively. Rather it could also

1129 reveal the existence of an un-sampled source or a tracer property may have been altered by non-
1130 conservative behaviour during mobilization and deposition but remains within the source range. This
1131 could affect the source apportionment but would not be captured by the application of the range test
1132 alone. Others (e.g. Koiter et al., 2013b; Kraushaar et al., 2015; Laceby et al., 2015b) advocate that tracer
1133 selection should not be based purely on statistical procedures and that knowledge of the hydrological,
1134 geomorphological and geochemical processes controlling tracer behaviour should also be used to help
1135 select appropriate soil and sediment properties for sediment fingerprinting. Koiter et al. (2013a) make
1136 some more general recommendations to help identify, and reduce, problems associated with non-
1137 conservative behaviour of fingerprinting properties, but further research is needed to understand such
1138 behaviour and incorporate such understanding into the sediment source fingerprinting technique.

1139

1140 **7.6 Statistical and unmixing model approaches and incorporation of uncertainties**

1141 Statistical tests and unmixing models are crucial for enabling raw data on property concentrations to be
1142 converted to meaningful values of source contributions. There are several approaches available for both
1143 the statistical procedures (e.g., Walling et al., 1993; Yu and Oldfield 1993; Walling and Woodward, 1995;
1144 Collins et al., 1997a) and unmixing models used. Others (e.g., Collins et al., 2010, 2012a, b) have
1145 suggested some recent developments to the statistical framework. In the case of unmixing models,
1146 earlier frequentist-based approaches used optimization to minimise residuals between sources and
1147 sediment mixtures to estimate the unknown proportional source contributions (e.g., Collins et al., 1997a;
1148 Walling et al., 1999). These approaches are now increasingly being superseded by Bayesian models
1149 drawn from studies of diet within ecology (i.e., IsoSource, SIAR, MixSIAR; Parnell et al., 2013) which are
1150 showing much promise, and consequently are recommended by organisations such as IAEA and are
1151 increasingly used in recent studies, especially in earth sciences (e.g., Dutton et al., 2013; Koiter et al.,

1152 2013b; Barthod et al., 2015; Cooper et al., 2015a, b). Bayesian unmixing models have several advantages
1153 over frequentist-based models, including: (i) better incorporation of prior information; and (ii)
1154 incorporation and reporting of uncertainties. Indeed, unlike many previous unmixing models, Bayesian
1155 models can explicitly capture many of the sources of uncertainty presently associated with the sediment
1156 source fingerprinting approach including: spatial variability in fingerprint properties across the study
1157 area, uncertainties associated with instrumental precision, covariance between fingerprint properties,
1158 and residual model errors (Cooper et al., 2015b). Furthermore, several of the Bayesian models are open-
1159 source with standard operating procedures (SOPs), often with graphical user interfaces (GUIs) and
1160 operated using R (e.g. MixSIAR), which has increased their usage in a variety of settings. Such an
1161 approach provides the opportunity for some level of standardisation and should enable researchers to
1162 reproduce and compare results.

1163 While studies in earth sciences have tended to use unmixing models (either frequentist- or Bayesian-
1164 based), other fields have used different approaches, such as Positive Matrix Factorization models,
1165 although often the underlying concepts are similar. In several cases the sources are inferred using
1166 principal components analysis or similar statistical procedures (e.g. studies of airborne particulates
1167 within human health: section 6), or through the use of elemental ratios (e.g. provenance studies in
1168 ocean sciences: section 3.5).

1169 The limitations of unmixing models and the fingerprinting approach as a whole, including estimation of
1170 uncertainties, are not well quantified and can be difficult to assess. Haddadchi et al. (2014b) attempted
1171 to assess the accuracy of several different unmixing models, and found that some perform better than
1172 others. Furthermore, weightings factors have been widely used in the frequentist optimization-based
1173 models to account for the differing ability of tracers to discriminate between sources or to enable tracers
1174 with lower within-source variability to exert a greater influence on source apportionment results (Collins

1175 et al., 2010). However, work by Laceby and Olley (2015a) and Haddadchi et al. (2014b) showed that the
1176 inclusion of such weightings actually reduced the accuracy of source contribution predictions compared
1177 to known artificial mixtures. It follows that the inclusion of any such parameters requires explicit
1178 justification and support based on the evaluation of model outputs compared to experimental or
1179 synthetic datasets. Indeed, the use of synthetic or virtual sediment mixture data is an efficient way of
1180 evaluating the performance of different unmixing model structures or data treatments (Palazón et al,
1181 2015b), and it is recommended that this practice be more widely adopted by sediment fingerprinting
1182 studies alongside measured datasets.

1183

1184 **7.7 Linking source fingerprinting to sediment budgets**

1185 Most sediment source fingerprinting studies have determined the *relative* contributions from different
1186 sources and not absolute mass contributions. In most cases, the former information is adequate.
1187 However, when determining sediment sources over time (e.g., events, seasons, years, millenia) it is more
1188 important to either determine mass contributions from the sources and/or consider sediment flux and
1189 storage information. For example, it may be possible for the relative contribution from two sources to
1190 remain constant from year to year but the absolute mass flux to change. Thus some studies (e.g., Smith
1191 et al., 2011) have linked sediment source data to sediment flux data to determine if sediment mass
1192 contributions from different sources have changed.

1193 In a similar way, changes in upstream storage could mask changes in sediment sources (e.g., Trimble,
1194 1983). For example, an increase in hillslope erosion rates in headwater areas could be offset by
1195 concomitant increased sediment storage on floodplains, such that no net change in relative sources is
1196 determined at a downstream sediment collection site if all other sources remain constant. Incorporating
1197 sediment source information within wider sediment budget investigations should help to ensure that an

1198 overall picture of sediment dynamics within a river basin or marine environment is obtained. Thus,
1199 sediment source fingerprinting represents one tool and is most effective when it is utilised with other
1200 tools (e.g. sediment budgets, monitoring, remote sensing).

1201

1202 **8. Conclusion and perspective**

1203 Sediment source fingerprinting has emerged within the last few decades as an important tool that can be
1204 used for a range of applications, including those in earth and ocean sciences, (geo)archaeology, forensics
1205 and human health. While applications in forensic and health sciences may have longer histories, there
1206 has been a dramatic increase within earth sciences, particularly since the 1990s, reflecting the fact that
1207 the approach is able to provide useful information on landscape and watershed processes and,
1208 especially, as it can be used to inform management decisions. To date, within the earth sciences it has
1209 mainly been used within agricultural landscapes but its use within other settings (e.g., forested, urban,
1210 estuarine, marine) is increasing. This partly reflects the development of new tracers, such as CSSIs, REEs
1211 and clay mineralogy, which can help to tackle new research questions and be used in new environments.

1212 It is apparent from this review that there is considerable commonality between the approaches of the
1213 various groups using fingerprinting to identify the sources of airborne and aquatic sediments. These
1214 include: (i) the need to collect representative samples of source materials and airborne or aquatic
1215 sediments; (ii) the selection of soil/sediment/dust properties that can effectively distinguish between
1216 potential sources; and (iii) the broad use of statistical and numerical approaches that are able to
1217 quantitatively apportion sediments to sources. Despite this commonality, there are often differences
1218 between the approach used which include: (i) the use of different tracer properties and combinations of
1219 properties; (ii) differences in the approaches used to account for issues of particle size and
1220 conservativeness; and (iii) differences in statistical and numerical approaches, and how to deal with

1221 uncertainty. As such, each discipline can learn from the others, and there exists the potential for cross-
1222 fertilisation. Indeed, the recent up-take by fluvial geomorphologists and other earth scientists of organic
1223 tracers such as stable isotopes, CSSIs and DNA – many of which were developed by biologists and
1224 ecologists for other applications (e.g., food webs) – shows that the process is underway. There are also
1225 some useful approaches to the selection of appropriate fingerprinting properties and statistical
1226 procedures used within other disciplines that could benefit earth science applications. These include
1227 procedures to select less mobile elements as fingerprint properties and the use of procedures to
1228 normalise data so as to remove some of the confounding influences associated with changes in particle
1229 size composition and organic matter content, as used in sediment provenance studies in marine
1230 environments (i.e., section 3.5).

1231 One recommendation is that there is a need for comparison between the various tracer groups; for
1232 example, comparison between geochemical elements, fallout radionuclides, colour properties and CSSIs
1233 (e.g. Blake et al., 2012; Verhayen et al., 2014; Barthod et al., 2015). While each tracer type may give
1234 different results, reflecting the sources that they are able to distinguish – topsoil vs subsoil in the case of
1235 radionuclides, different crop types in the case of CSSIs – there should be internal consistency in the
1236 results. For example, if CSSIs identify that most of the organic component of the sediment is coming from
1237 the surface of cropland, then if other tracer groups identify that subsurface soils dominate (e.g., due to
1238 gully erosion) then there needs to be further investigation to assess if the difference is real (i.e., organic
1239 and mineral component derive from different sources) or if one tracer group is exhibiting non-
1240 conservative behaviour.

1241 Similarly, there is a need to compare source fingerprinting results to other, independent lines of
1242 evidence. Such an approach is typical in geoarchaeological (section 4) and forensic (section 5)
1243 applications, where it is important to both place source fingerprinting or provenance findings within a

1244 broader context and to determine the validity of the findings; as such, earth science applications could
1245 learn from these disciplines.. Thus, if fingerprinting results suggest that channel bank erosion is the main
1246 sediment source yet ground-truthing (e.g., monitoring) and aerial photographs show no evidence for
1247 such erosion then there is a need for further investigation to either determine which approach is correct,
1248 or to identify the cause of the difference (e.g., intermediate storage between source and sediment
1249 collection). In both of these cases (i.e., internal comparisons and comparisons with independent
1250 approaches) there is a considerable amount to be learnt about both the techniques themselves and how
1251 landscapes function. Sediment source fingerprinting should be viewed as only one component of
1252 investigations into sediment (airborne, terrestrial and aquatic) processes and dynamics, and it should be
1253 part of a more holistic assessment of landscape and watershed behaviour and functioning.

1254 The sediment source fingerprinting approach offers many advantages over more conventional
1255 techniques, such as landscape and watershed monitoring, given that information can be assembled
1256 relatively quickly and cheaply; although this in part depends on the actual approach adopted. In reality,
1257 the fingerprinting approach should be viewed as a complementary approach to these more conventional
1258 approaches. Realisation of the potential of the fingerprinting approach has seen its use expand into new
1259 applications as well as a growing interest and recognition of similar approaches in other disciplines. This
1260 review has highlighted some of these new applications, reviewed developments in cognate disciplines,
1261 and illustrated some of the areas requiring further work if the potential of the technique is to be fully
1262 realised.

1263

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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 Basin)	(River Basin)	Basin area (km ²)	Contribution from agricultural land	Other main sources	Sediment tracers	Study
USA (Pleasant Creek, WI)	Valley	50	45-97% (34%)	Channel banks 3-47%	Geochemical elements	Lamba et al. (2015a)
USA (Laurel Hill Creek, PA)		324	50-95% (27%)	Stream banks 5-50%	Geochemical elements, $\delta^{13}\text{C}$, $\delta^{15}\text{N}$	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	17 183 446 629	31 99 57 69	FRN (¹³⁷ Cs, ²¹⁰ Pb _{ex})	5 years	10 86 45 71	n/a 55-68 29-51 21-63

^aBurned (B); Unburned (UB)

^bNot available (n/a)

^cFallout 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

^dChannel sources constitute the remaining percentage contribution for each of the studies.

^eThis 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 geochronology (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	$^{143}\text{Nd}/^{144}\text{Nd}$ $^{87}\text{Sr}/^{86}\text{Sr}$	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 ($^{40}\text{Ar}/^{39}\text{Ar}$)	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: $^{208}\text{Pb}/^{204}\text{Pb}$ $^{207}\text{Pb}/^{204}\text{Pb}$, $^{206}\text{Pb}/^{204}\text{Pb}$	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 2 nd and first half of the 1 st century BC.	a shipwreck between the islands of Ponza and Palmarola (<i>n</i> = 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 (<i>n</i> = 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 (<i>n</i> = 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 Murder	crime Trace element mapping – particle-induced x-ray and γ -ray emission	Shovel Vehicle	Bailey et al. (2012)
Wildlife Hit and Murder	crime run 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	NA	Cox et al. (2000)
Experimental	Soil colour – Munsell color chart	NA	Guedes et al. (2009)
Experimental	Soil colour – spectrophotometer	NA	Khodakova et al. (2014)
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)
