1   **A review of source tracking techniques for fine sediment within a catchment**

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Abstract

Excessive transport of fine sediment, and its associated pollutants, can cause detrimental impacts in aquatic environments. It is therefore important to perform accurate sediment source apportionment to identify hotspots of soil erosion. Various tracers have been adopted, often in combination, to identify sediment source type and its spatial origin, these include: fallout radionuclides, geochemical tracers, mineral magnetic properties and bulk and compound-specific stable isotopes. In this review, the applicability of these techniques to particular settings and their advantages and limitations are reviewed. By synthesizing existing approaches, that make use of multiple tracers in combination with measured changes of channel geomorphological attributes, an integrated analysis of tracer profiles in deposited sediments in lakes and reservoirs can be made. Through a multi-scale approach for fine sediment tracking, temporal changes in soil erosion and sediment load can be reconstructed and the consequences of changing catchment practices evaluated. We recommend that long-term, as well as short-term, monitoring of riverine fine sediment and corresponding surface and subsurface sources at nested sites within a catchment are essential. Such monitoring will inform the development and validation of models for predicting dynamics of fine sediment transport as a function of hydro-climatic and geomorphological controls. We highlight, that the need for monitoring is particularly important for hilly catchments with complex and changing land-use. We recommend that research should be prioritized for sloping farmland-dominated catchments.

Introduction

Selective soil erosion and sediment transport, leading to downstream fining and an enrichment of organic matter content along sediment cascades have been well recognized at the catchment scale (Koiter et al. 2015). Field evidence, reported more than 30 years ago from the Jackmoor Brook catchment (9.8 km^2) in Devon, England, indicated that particle size controls selective erosion and transport; with the finest sediment fraction (diameter <1 μm) exhibiting a delivery ratio of 100% and coarser sediment (diameter 20–63 μm) exhibiting a delivery ratio of only 30% (Walling 1983). Fine sediment is a leading cause of impairment of waterways, particularly lowland rivers, in many countries around the world (Palmer et al. 2000; Grabowski and Gurnell 2016). Natural or anthropogenic disturbances (e.g., damming, agricultural practices, deforestation or timber harvests, urban, suburb or rural development, construction activities and wildfires) are the main causes of excessive fine sediment production and loading to rivers (Collins et al. 2010b; Jones and Schilling 2011).

Various adverse impacts are associated with excessive fine sediment (<63 μm or fine silt and clay) loads in the aquatic environment and these have led to an increasing concern for catchment management. Excessive amounts of fine sediment can increase turbidity, limit light penetration and potentially reduce photosynthesis and primary productivity of macrophytes. This damages aquatic habitats in the hyporheic zone (e.g., reduction in invertebrate biodiversity and changes in macrophyte communities) (Wood and Armitage 1997; Bo et al. 2007; Kemp et al. 2011) and important game fisheries (e.g., reduced feeding, impaired respiration, physiological stress and migration inhibition) (Collins and Walling, 2007). In great excess, sediment can smother the entire

74   stream bed and eliminate habitat complexity (Walser and Bart 2006). The chemically reactive nature

75   of suspended fine sediment makes it a potential vehicle for transporting contaminants (e.g.,

76   agrochemicals, antibiotics, persistent organic pollutants, trace metals and metalloids) rapidly over a

77   long distance with streamflow (Horowitz 1991; Zhang et al. 2015). Therefore, fine sediment, either

78   suspended, re-suspended or deposited, in the channel system represents a significant diffuse

79   pollution issue, and one that requires the attention of regulators to ensure safeguards are included

80   within catchment management regulations.

81   Conventional measures for dealing with excessive fine sediment accumulation in channels, such

82   as riverbed dredging and flushing, are largely impractical for large scale management due to high

83   labor and investment requirements. For different hydro-climatic conditions and catchment

84   characteristics, suitable measures should be developed and implemented along the main transport

85   pathways from source areas to a catchment's outlet (Mukundan et al. 2012; Vinten et al. 2014;

86   Lamba et al. 2015a). The precise design of effective fine sediment management and control policies

87   relies heavily on the quantitative identification of erosion sources, spatial soil redistribution on

88   slopes, in-channel sediment delivery flux and their temporal variation at a given location within the

89   catchment (Shackle et al. 1999; Walling et al. 2003). Knowledge of hydrological processes and the

90   spatial pattern of soil redistribution across the landscape (from slopes to catchment outlet), which

91   can change within a year and between years, is essential to apportioning fine sediment to surface

92   and subsurface sources. Generally, there are five main processes defined in a fine sediment tracking

93   framework, these are: (1) hillslope erosion, (2) gully erosion and deposition, (3) channel bank

94   erosion, (4) channel bed deposition/re-suspension and (5) fine sediment export at catchment outlet

95   (Bartley et al. 2007). The assessment of these processes is limited by our ability to resolve their

96   intrinsic spatial and temporal variability at the catchment scale (Grabowski and Gurnell 2016). With

97   the aid of remote sensing and GIS technology, spatial distribution of soil erosion and sediment

98   transport within a catchment can be estimated and erosion prone areas can be identified (Bhattarai

99   and Dutta, 2007; Ganasri and Ramesh 2016). In particular, specific remote sensing data, such as

100   color and standard reflectance spectra derived satellite images, can be used in combination with

101   other *in situ* physical, optical and acoustic measurements to reveal the dispersal, transport path and

102   fate of fine sediment across coastal margins as well as rivers (Warrick 2009; Loisel et al. 2014; Qu

103   2014). Remote sensing data are generally limited to the surface layer and are not as accurate as *in*

104   *situ* measurements. These limitations can be compensated, to some extent, by the spatial and

105   temporal coverage provided by remote sensing observations, particularly at large scales. However,

106   the available approaches that combine remote sensing data and GIS with soil erosion and sediment

107   transport models remain only moderately successful in reliably predicting (with high resolution)

108   spatial patterns of soil erosion and deposition and the dynamics of sediment transport in channels.

109   Most of the models such as USLE, and its revised forms, do not have the capability for routing

110   sediment through channels. Data on contributions from different sediment source types or erosion

111   processes to sediment are very limited because sediment production from many sediment sources

112   or erosion processes cannot be readily or directly measured in a catchment (Zhang and Liu 2016).

113   The use and development of various tracking techniques allow quantitative identification of the

114   sources of deposited and suspended sediment observed at a concerned location. The majority of

115   existing source tracking studies focus specifically on fine fraction of bulk sediment that is eroded

116   from the surface of diffuse upland areas and channel banks.

117   While recent reviews have considered the development of sediment source tracking techniques

118   and their application (e.g., Walling 2005, 2013; Davis and Fox 2009; Mukundan et al. 2012; Guzman

119   et al. 2013; Haddadchi et al. 2013; Smith et al. 2013) very recent developments, included, in

120   particularly, those relating to anthropogenic origin tracers have not been reviewed. Furthermore, the

121   principles and potential technical framework for combining existing approaches, at different

122   temporal and spatial scale, have not been fully established nor elaborated upon. Given the promise

123   such an integrated approach would have, for fine sediment source apportionment at multiple scales

124   in a catchment, this review seeks to highlight the possibilities. In this review, we revisit the topic

125   and evaluate the strengths and limitations of existing tracking techniques for fine sediment and

126   justify the need for future research directed towards sloping, farmland dominated, catchments.

127

128   **Selection of sediment source tracers**

129

130   In the past five to six decades, a number of tracers, or diagnostic properties, have been successfully

131   used to track sediment sources and quantify their relative contributions within a catchment. These

132   include artificial radionuclides (Brown et al. 1981a,b; Owen et al. 2012), naturally occurring

133   radionuclides (Wallbrink and Murray 1993), deliberately introduced radionuclides (Toth and

134   Alderfer 1960; Wooldridge 1965), geochemical elements (Olley and Caitcheon 2000; Haddadchi et

135   al. 2014), rare earth elements (Tian et al. 1994), bulk stable isotopes (Fox and Papanicolaou 2007),

136   compound-specific stable isotopes (Gibbs 2008), metals (Blake et al. 2012), fluorescent dye coated

137   particles (Young and Holt 1968), magnetic minerals (Yu and Oldfield 1989), color (Grimshaw and

138   Lewin 1980), plant pollen (Brown 1985) and diffuse reflectance infrared Fourier transform

139   spectroscopy (Poulenard et al. 2012). Although these tracers are useful, each has its limitations. In

140 this review, we have considered several factors of relevance to the use of a particular tracer, these

141 are: origin, applicable timeframe, applicable spatial scale, advantages and limitations. How a given

142 tracer performs in these regards has been summarized in Table 1. The readers may find it useful to

143 refer to Table 1 when they make use of this review. In practice, the selected tracers need to be easily

144 detectable by readily available analytical instruments.

145   The use of a composite fingerprint comprising several selected tracers or diagnostic properties

146 has proven to be reliable (Collins et al. 1997a,b; Carter et al. 2003; Krause et al. 2003). Multiple-

147 tracer approaches may provide information on sediment redistribution and offer a distinct advantage

148 over single tracer approaches. Multiple-tracer approaches are based on two assumptions (Walling et

149 al. 1999): (1) the fingerprints consist of selected diagnostic tracers, these are conservative during

150 sediment delivery and show distinctive differences between major source materials; and (2)

151 comparing the tracers or fingerprints of suspended sediment with those of potential sources leads to

152 quantitative estimation of the relative contribution of each individual source. At a given location, in

153 addition to the net deposition, the relative upslope source contributions to deposited sediment can

154 be quantified based on multiple-tracer measurements. Likewise, for areas of net soil erosion, offsite

155 fate of the mobilized sediment can be tracked; including, where the sediment is redeposited

156 downslope and how much is exported out of the catchment (Polyakov et al. 2004).

157   It is important to test for normality before proceeding with the selection of tracers. The statistical

158 representation of a tracer's spatial variation within each source area affects the results of sediment

159 source apportionment. By fitting normal distributions to source groups (Krause et al. 2003;

160 Wilkinson et al. 2009), using standard errors for estimated means (Hancock et al. 2014) or median

161 and a robust scaling estimator (Q_n) (Rousseeuw and Croux 1993), source soils may be represented

162 by the mean concentration derived from spatially random sampling locations in the catchment (Peart

163 and Walling 1986). Catchment-specific nature of tracer spatial variations can be defined using

164 sample data showing representative distributions (Caitcheon et al. 2012; Olley et al. 2013). The

165 statistical verification of a composite fingerprint, comprising multiple tracers, can be performed

166 using principal components analysis (PCA), genetic algorithm-driven discriminant function analysis

167 (GA-DFA) and the Kruskal-Wallis H-test (KW-H) or the Mann-Whitney U-test in combination.

168 Through the application of these statistical approaches the abilities of selected tracers for

169 discriminating spatially the sediment sources can be evaluated (Collins et al. 1997a,b,2012). If a

170 source type shows large differences among sub-catchments for each individual tracer, instead of

171 using a single set of tracers for the whole catchment, statistical analysis should be performed to

172 identify a subset of tracers that could discriminate the sources within each sub-catchment (Walling

173 2005; Smith et al. 2013). For example, a geochemical composite fingerprinting approach for

174 identifying statistically robust fingerprints was successfully used to apportion channel bed sediment

175 sources in the upper Kennet agricultural catchment (~214 km^2) in southern England (Collins et al.

176 2012). Optimum composite fingerprints were identified for each of the six bed sediment sampling

177 periods using a revised statistical procedure combining GA-DFA, KW-H and PCA and the numerical

178 mass balance mixing model described by Collins et al. (2010b).

179

180 **Mixing models, correction/weighting factors and uncertainty**

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182 When a set of composite fingerprints comprising n appropriate conservative tracer(s) has been

183 selected to discriminate $>(n+1)$ sources, multivariate mixing (or unmixing) models consisting of a

184   set of linear equations could be used to determine relative contributions from each source (e.g.,

185   Collins et al. 1997a,b; Walden et al. 1997). There are three main types of mixing models: mixing

186   model without uncertainty terms in the mixing equations (Eq. 1; Collins et al. 1997a,b,) or mixing

187   models with uncertainty terms in mixing equations that are accommodated by Bayesian (Eq. 2;

188   Franks and Rowan 2000) or Monte Carlo approaches (Eq. 3; Hughes et al. 2009).

189   The mixing model without uncertainty term can be written as:

190
$$\sum_{s=1}^{m} P_s S_{i,s} = C_i \quad with\ i = 1, n\ and\ \sum_{s=1}^{m} P_s = 1 \tag{1}$$

191   where C_i is the concentration of fingerprint tracer i in sediment sample, P_s is the optimized

192   percentage contribution from source s, $S_{i,s}$ is median concentration of tracer i in source s with or

193   without particle size and organic matter corrections, n is number of fingerprint tracers and m is the

194   number of sediment sources.

195   An uncertainty-inclusive Bayesian mixing model can be written as:

196
$$\sum_{s=1}^{m} P_s S_{i,s} = C_i + \varepsilon_i \quad with\ i = 1, n\ and\ \sum_{s=1}^{m} P_s = 1 \tag{2}$$

197   where ε_i is the error associated with the prediction of the target tracer value C_i.

198   The Hughes model (Hughes et al. 2009) applies a Monte Carlo approach and runs random

199   iterations of all individual source samples (not their mean) to minimize the errors. This is obviously

200   different from the Collins and FR2000 (Franks and Rowan 2000) methods, that both use mean

201   values for each tracer parameter pertaining to each source. The Hughes model can be written as:

202
$$\sum_{l=1}^{1000} \sum_{s=1}^{j} P_s S_{i,s,k,l} / 1000 = C_i \quad i = 1, n \tag{3}$$

203   where j is sample number index, each sample has k constituent concentrations and l is Monte Carlo

204   iteration number.

205   As given in Eqs. 1-3, for each tracer, a linear equation is used to relate tracer concentration in fine

sediment to the sum of each source's tracer concentration multiplied by unknown relative

contribution from each source. By minimizing differences between estimated tracer

properties/concentrations in fine sediment and those measured using an objective function (e.g., the

sum of relative errors or the squares of errors), each source's contribution can be obtained (e.g.,

Collins et al. 1997a,b).

The goodness-of-fit of the model can be assessed by comparing the actual tracer

properties/concentrations measured in fine sediment with the corresponding values estimated by the

optimized mixing model (Collins et al. 2010b). Many mathematical tools (e.g., MATLAB,

Mathematica and Excel) have built-in programs that can be used to solve these quadratic

programming problems. The Monte Carlo or Bayesian simulation could be employed to quantify

uncertainty associated with sampling and source estimates (e.g., Small et al. 2002; Caitcheon et al.

2012), either explicitly in mixing equations or separately. It is also essential to include particle size

correction factors, organic matter content correction factors, which are often site-specific, within-

the source variability and discriminatory power of each tracer, (Collins et al. 1997; Collins et al.

2010b). In particular, particle size effects are due to preferential adsorption of some tracers to fine

particles and selective transport that may cause a difference in particle size distribution between

source soil and fine sediment (Smith et al. 2013). Particle size corrections based on specific surface

area ratios, analysis of the fine fraction of suspended sediment, or thorium normalization could be

used to minimize particle size effects on tracer properties/concentrations (e.g., Collins et al. 1997a,b;

Wallbrink et al. 1999; Foucher et al. 2015). The hypothesis of positive linearity between particle

size and tracer concentration may not hold true for all tracers, or may differ between different tracers,

and precise relationship between particle size composition and tracer concentration for each tracer

11

228   must be routinely tested and developed for individual tracers if the linearity exists (Russell et al.

229   2001; Smith and Blake 2014). A minimum number of potential sources and a maximum number of

230   composite fingerprints, each with a minimum number of non-contradictory tracers, have been

231   strongly recommended (Zhang and Liu 2016). However, source apportionment results may relate to

232   fine fraction and may not provide information on sources of bulk suspended sediment load.

233   The largest uncertainties of model prediction are often found in downstream sub-catchments,

234   which are usually the main contributors of fine sediment. Subsurface erosion of river bank occurs

235   mostly in downstream reaches (Theuring et al. 2015). Catchment-wide spatial variation of each fine

236   sediment source type has rarely been evaluated. Sampling location of sediment (e.g., headwaters

237   and downstream sections) affects the estimated contributions of sources, which are dependent on

238   the spatial and temporal variations in runoff generation.

239   Software tools have been developed for sediment source apportionment based on mixing models

240   and isotopic tracer measurements, such as, IsoSource (Phillips and Greg 2003), SIAR (Parnell et al.

241   2010) and CSSIAR (Sergio et al. 2017). IsoSource allows entering only one sediment mixture and

242   ten potential sources at the same time, and is not applicable for investigating large areas with

243   different sediment mixtures and sources. As an advantage over IsoSource, SIAR incorporates

244   uncertainties using a Bayesian approach. Therefore, SIAR has been more successful with wider

245   applications, using not only isotopic tracers but also other tracers or diagnostic properties such as

246   geochemical and radionuclide tracers and color (Parnell et al. 2010; Dutton et al. 2013; Koiter et al.

247   2013a; Barthod et al. 2015). Both IsoSource and SIAR require users to make some calculations (e.g.,

248   methanol correction for fatty acid methylesters, calculation of source soil proportions from isotopic

249   proportion) before and after the model runs. CSSIAR is a newly available open source tool based

250   on the Bayesian approach of SIAR for sediment source apportionment using $\delta^{13}C$ values of

251   compound specific stable isotopes (e.g., fatty acids and other tracer compounds), in which all

252   calculations are included and performed via a user-friendly interface (Sergio et al. 2017).

253

254   **Sampling of source soil and fine sediment**

255

256   Random or grid sampling of source soil focuses on potential source types (Wallbrink et al. 1998;

257   Fox 2005). Primary sources can be identified by investigating erosion-prone areas (i.e., sites

258   showing erosion scars and sparse vegetative cover and potentially connected to the channel network),

259   hydrologic processes and sediment delivery/redistribution paths in sub-catchments (Collins and

260   Walling 2007).

261   Taking into account geological, topological and hydrological variability, replicates should be

262   collected in each of the potential source areas including upland surface soils, river beds and banks

263   and other sources (e.g., construction sites and road verges) to ensure representativeness of tracer

264   datasets. In particular, repeated sampling of sources will be essential if ^{7}Be (a cosmogenic tracer

265   with a short half-life of 53.3 days) is to be used for tracking temporal changes in sediment sources

266   associated with runoff events or other human (e.g., land-use change and plowing) and natural

267   interferences (e.g., fire) (Smith et al. 2013).

268   Suspended sediment in channels at gaging stations can be monitored and sampled using an

269   automatic water sampler triggered by water-level variations. Samples of water and bulk surface

270   sediment at different locations on channel beds can be collected using a cylinder following the

271   procedure proposed by Lambert and Walling (1988). The depth of fine sediment ingress in channel

272 bed varies temporally and spatially, normally of the order of 1–10 cm (e.g., Frostick et al. 1984;

273 Collins and Walling 2007; Grabowski and Gurnell 2016). Therefore, a sampling depth of ca. 5 cm

274 into the channel bed is recommended. The water and sediment enclosed within sampling cylinders

275 are thoroughly mixed to re-suspend and collect fine sediment. Fine sediment recovered from bulk

276 surface sediment collected from a riparian area, a channel bed, a dam reservoir or a floodplain can

277 be used as surrogates for suspended fine sediment that is representative of a longer period of

278 deposition (Olley and Caitcheon 2000; Foster et al. 2007; Haddadchi et al. 2013). It should be noted

279 that the sources of suspended sediment may differ from that of recovered fine fraction of deposited

280 sediment (Nicholls 2001). Tracer data pertaining to the fine fraction of deposited sediment should

281 be applied carefully into source apportionment models, with accommodation made to incorporate

282 correction factors that account for differences in particle size selectivity between sediment delivery

283 and sediment deposition processes.

284   Fine sediment can be recovered from bulk surface water or re-suspended sediment by settling and

285 subsequent centrifugation, followed by freeze-drying and homogenization using a 63 μm sieve. A

286 composite sample is prepared by mixing all individual samples to obtain a sufficient amount of fine

287 sediment (e.g., 2–50 g) for analyses of radionuclides and other tracer properties or concentrations.

288   Other than water samplers used for rain event based monitoring, bed sediment samplers can be

289 used as an alternative means of collecting fine sediment undergoing continuous remobilization.

290 These are particularly useful for investigating seasonal patterns (Philips et al. 2000). Time-

291 integrated samples of suspended sediment have recently become more popular while point sampling

292 at specific times throughout the hydrograph of a runoff event (i.e., rising and falling limbs) may be

293 conducted in order to pinpoint erosion processes that vary temporally. It should be noted that, time-

294   integrated sampling approach requires correction or additional sediment concentration samples to

295   estimate total suspended sediment because time-integrated samplers usually show sediment trapping

296   efficiencies lower than 100% (Phillips et al. 2000). Point sampling approach is limited by the large

297   volume of sample required for tracer analysis (Davis and Fox 2009).

298   Localized spatial variations in fine sediment storage on a channel bed are dependent on the

299   variability of water depth, channel sinuosity and bed morphology (Collins and Walling 2007). Thus,

300   it is desirable for sampling campaigns that collect suspended and deposited sediment to be

301   complemented by surveys to define these attributes (Evrard et al. 2011).

302

303   **Strengths and limitations of tracers**

304

305   Radionuclides

306

307   Fallout radionuclides (e.g., ^{137}Cs, ^{7}Be and ^{210}Pb$_{ex}$) are generally abundant in surface soil

308   (particularly in association with fine particles) but are sparse in subsurface soil (Wallbrink and

309   Murray 1993; He and Walling 1996). Thus, they can be used to differentiate erosion of surface soil

310   from that of deeper sub-soil. ^{137}Cs and ^{210}Pb$_{ex}$ have been extensively used to trace surface and

311   subsurface sources of sediment suspended or deposited in river channels, reservoirs and lakes.

312   Estimates of sediment delivery and sedimentation using ^{137}Cs and ^{210}Pb$_{ex}$ have proven to be

313   consistent (Shi et al. 2012). The two radionuclides generate comparable results with data collected

314   using more traditional approaches (e.g., erosion pins, runoff plots and the monitoring of catchment

315   sediments yields) (e.g., Elliott et al. 1990; Mabit et al. 2002) and empirical erosion models (e.g., the

316 USLE and RUSLE models) (Mabit et al. 2008). As the traditional approaches cannot account for

317 uncertainties while upscaling the results, the use of the two radionuclides has the advantage of

318 determining spatial variations in soil erosion and sediment deposition across the landscape as well

319 as for specific landscape elements (Mabit et al. 2008). ^{137}Cs and ^{210}Pb$_{ex}$ can be used to distinguish

320 sediment eroded from uncultivated and cultivated lands (Wallbrink et al. 1998). Cosmogenic ^{7}Be is

321 usually concentrated in top 5 mm of soil. Thus, ^{7}Be can be used to discriminate very thin surface

322 soil layer from deeper layers and to confirm the relative importance of recently mobilized surface

323 soil due to its very short half-life of 53.3 days, as compared to ^{137}Cs (half-life 30.2 yr) and ^{210}Pb$_{ex}$

324 (half-life 22.3 yr) (Zapata 2003). ^{7}Be can be used to track sediment redistribution only in cases

325 where no significant erosive rainfall events have occurred for the past 5 months. This significantly

326 limits its application (Walling 2013a). ^{137}Cs and ^{210}Pb$_{ex}$ have been extensively included as a tracer

327 within the multivariate models to not only discriminate subsurface versus surface and cultivated

328 versus uncultivated sediment source in soil redistribution studies on slopes (Busacca et al. 1993;

329 Nagle et al. 2000), but also to reconstruct a chronological record of sediment deposition by using

330 cores extracted from floodplains, lakebeds and reservoir beds (Collins et al. 1997a; Hasholt et al.

331 2000; Zhang et al. 2007). Overall, fallout radionuclides can be the best available fine sediment

332 tracers for source discrimination if relevant correction/weighing factors are incorporated in model

333 estimation where necessary.

334

335 Geochemical tracers

336

337 A number of geochemical tracers including rare earth elements (REE; e.g., La, Ce, Pr, Nd, Sm, Eu,

338   Gd, Tb, Dy, Ho, Er, Tm, Yb and Lu), major elements (e.g., Fe, K, Na, Al, Ca and Mg) and trace

339   elements (e.g., As, Ba, Co, Cr, Cs, Hf, Sc, Ta, Ti, Th and Zr), in both source soil and sediment

340   samples, have been used in sediment fingerprinting studies (Horowitz et al. 2012). Eroded sediments

341   often maintain these distinct geochemical properties during transport processes (Hughes et al. 2009).

342   Major element geochemistry, particularly the relationship between Fe_2O_3 and Al_2O_3, can be used to

343   discriminate soils derived from different parent rocks (Dyer et al. 1996).

344   Active REE tracking method involves artificial mixing REE, normally as oxide powders, with

345   soils at different topographical positions, and sediment transport flux and loss of source materials

346   can be determined by analyzing REE concentrations in sediments (Tian et al. 1994; Zhu et al. 2010).

347   In very acidic soils, REEs may be partly leached and thus redistributed via desorption and

348   adsorption, limiting their applicability for tracking sediment movement (Land et al. 1999).

349   Furthermore, large errors may be introduced when REE is applied in light-textured soils due to their

350   poor aggregate-forming abilities (Zhang et al. 2001; Kimoto et al. 2006).

351   At undisturbed/uncultivated sites, physical properties such as water permeability and anti-erosion

352   ability of soil may be changed greatly due to artificial mixing of REE with soil. Therefore, REE-

353   derived data cannot truly reflect erosion process of originally undisturbed soil, and thus the

354   applicability of REE tracer method for uncultivated land is restricted (Polyakov et al. 2004). This

355   method may not be suitable for complex natural topographies because within-field deposition is

356   ignored (Zhu et al. 2010).

357   A non-intrusive method of spraying REE oxide powder suspension to soil surfaces has been

358   proposed to minimize soil surface disturbance (Deasy and Quinton 2010). It has been shown to be

359   effective on a temperate arable hillslope and may also be applicable to temperate grassland,

360   rangeland and semi-arid areas. Notably, in karst areas in southwest China, natural geogenic REE

361   signatures were found to be a potential proxy for identifying lacustrine sediment sources and soil

362   erosion rates (Wen et al. 2014). To date, the use of REE tracer methods has been limited, partly due

363   to its relatively high cost.

364   The use of geochemical tracers, particularly large numbers of major and trace elements, has

365   become increasingly common since the late 1990s (Rollinson 1993; Grimes et al. 2007; Franz et al.

366   2013). This can be partly attributed to recent advances in both quick analytical techniques for large

367   numbers of samples and numerical source apportioning models that allow a more detailed and

368   quantitative understanding of the uncertainty associated with estimated results. Fine sediment is

369   usually rich in clay minerals, Fe and Mn oxides and hydroxides and organic matter. There are three

370   approaches that can account for potential differences in sediment geochemistry relating to particle

371   size and mineralogy. One commonly used way of normalization is to use measured bulk

372   concentrations in the total sediment sample to account for differences in sediment geochemistry.

373   The diluting effects of non-reactive constitutes (e.g., coarse sediment, quartz and feldspar grains)

374   can be removed. The second way of normalization is to divide the concentration of the potential

375   tracer by the concentration of the conservative element (e.g., Al, Ti and Li). The third method is to

376   incorporate a correction factor into the mixing model (Collins et al. 1998). Moreover, the

377   geochemical tracer method can also be applied to contemporary and historic sedimentary deposits

378   for reconstructing temporal changes in sediment provenance at the catchment scale (Thevenon et al.

379   2013).

380

381   Properties of organic matter

382

Soil organic matter cycling that couples soil, plants and the atmosphere through biogeochemical

processes results in differences in total organic carbon (TOC), total organic nitrogen (TON) and

total organic phosphorus (TOP) between land-uses (e.g., forest and agriculture) (Collins et al.

1997b). TOC, TON, TOP and carbon to nitrogen ratio (C/N), either individually, collectively or in

combination with other tracers, have been used to represent plant cover, land-use and land

management at sediment sources (Collins et al. 1997b; Walling and Amos 1999; Carter et al. 2003;

Papanicolaou et al. 2003; Fox and Papanicolaou 2007). Fluorescence excitation-emission matrix

(EEM) spectra of sediment leachate can be resolved into fluorescent components using parallel

factor analysis (PARAFAC). A novel combined use of fluorescence spectroscopy and end-member

mixing analysis (EMMA) was proposed for source discrimination of fine sediment that is often rich

in organic matter (Larsen et al. 2015). Fluorescence EEM spectra could provide insightful

information on quality of organic matter that differ between sediment sources.

Stable isotopes of organic carbon (i.e., δ^{13}C) and nitrogen (i.e., δ^{15}N) show greater potential

detection sensitivity than total elemental composition and therefore can be useful tools to identify

sources and sinks of sediments (Papanicolaou et al. 2003; Alewell et al. 2008; Fox and Papanicolaou

2007; Turnbull et al. 2008). Soil depth profiles of stable isotopes (δ^{13}C vs % C and δ^{15}N vs % N)

may be used as qualitative indicators of soil erosion. The δ^{13}C measurements can reflect the

differences in contributions of different land-uses (e.g., forest, grassland and cropland) to sediments,

resulting from the differences in δ^{13}C value between C3 and C4 plants (Balesdent et al. 1988). Bulk

δ^{13}C signatures may be strongly affected by past crop rotations or other organic carbon sources such

as applications of manure and wastewater (Tang et al. 2014). The stable isotope ratio of nitrogen

404   (δ^{15}N) can be employed to discriminate subsurface sources (e.g., streambanks and construction sites)

405   of suspended sediment (Fox et al. 2010). High levels of δ^{15}N in cultivated soils and streambanks

406   complement well with the relatively low ^{137}Cs activities detected in these sources (Mukundan et al.

407   2010; 2012). Future work should be conducted to temporally and spatially identify the hydrologic

408   and biogeochemical controls on the isotopic signatures of surface and subsurface source soils,

409   suspended fine sediment and channel bed sediments where a significant portion of deposited fine

410   sediment is temporarily stored (Mukundan et al. 2012).

411   Although geochemical properties, radionuclides, anthropogenic pollutants and bulk stable

412   isotopes have been increasingly used to inform the need and effective strategies to control fine

413   sediment erosion (e.g., Collins et al. 2010b), the application of sediment source tracking techniques

414   has its limitations. For example, the use of these approaches cannot give quantitative information

415   on sediment source with specific crop, which is crucial in agriculture-dominated catchments. Gibbs

416   (2008) demonstrated that the use of compound-specific stable isotopes (CSSI) (e.g., δ^{13}C of fatty

417   acids) allowed the identification and apportionment of sediment to source soils under different land-

418   uses or cropping systems in New Zealand. The plant-produced fatty acids (FA), particularly those

419   having an even number of carbon atoms (C14:0 to C24:0, range of lipid numbers in the form C:D

420   where C is the number of carbon atoms and D is the number of double bonds), can be readily leached

421   from leaves and roots and bound strongly to soil particles which may be subsequently transported

422   to channels by water erosion (Williams et al. 2006). Isotopic fractionation of carbon leads to a range

423   of isotopic values for each FA. Acid group of FAs in sediment and source soil samples should be

424   replaced by a methyl group to form a non-polar fatty acid methyl ester (FAME) prior to performing

425   analysis with GC-combustion-IRMS. Different plants or crops produce the same FAs which show

426   different CSSI signatures (Chikaraishi and Naraoka, 2003). For example, the C16 FAs show rather

427   different δ^{13}C values in soil under maize (−25.7‰) and pasture (−30.1‰) (Blake et al. 2012).

428   Selected FAs are chemically conservative and the δ^{13}C value of FAs does not change over time

429   during sediment transport (Blessing et al. 2007). The δ^{13}C signatures of fatty acids associated with

430   particles can therefore be used to track sediment in rivers back to uplands under specific vegetation

431   or crop cover. This method could provide unique insights into the development processes of

432   sediment source fingerprinting (Blake et al. 2012). Palmitic (C16:0) and Oleic (C18:1) acids exhibit

433   larger differences across different land-uses and have been reported to be the most useful in source

434   fingerprinting (Gibbs 2014). Myristic acid (C14:0) may also be a good tracer but it is sometime

435   missing in old sediments due to its volatilization. It has been reported that shorter chain FAs can be

436   degraded preferentially by soil microorganisms (Matsumoto et al. 2007). Thus, the dynamics of

437   CSSI fingerprint development and microbial activities in the soil and sediment are potentially

438   influencing factors that may affect the results of fine sediment source tracing. Given these dynamic

439   factors the use of CSSI fingerprinting needs careful consideration if it is to be applied meaningfully

440   at different temporal and spatial scales. Furthermore, the CSSI approach is often not a 'standalone'

441   quantitative approach, and complementary tracer data or information are highly desirable (e.g.,

442   measured sediment loads in channels, modelled sediment loads within geographical information

443   system, climate/meteorological data and estimates of erosion and sedimentation rates e.g. based on

444   fallout radionuclide measurements). CSSI tracer approaches have been mostly used in contemporary

445   (not annual mean) source apportionment of surface sediment deposition to channel beds or

446   floodplains (Gibbs 2008).

447

448   Anthropogenic pollutants

449

450   Increased concentrations of various anthropogenic contaminants (e.g., persistent organic pollutants,

451   heavy metals and pesticides) discharged into aquatic environments have been reported during the

452   last 200 years (Takeda et al. 2004; Bravo-Espinoza et al. 2009; Tang et al. 2014). Provided that,

453   dates of benchmark events (e.g., first introduction, marked increased or decreased inputs to the

454   environment and/or ban on production and use of the chemicals) are known it is possible to use

455   contaminant profiles of deposited sediment to reconstruct the temporal changes of soil erosion and

456   sediment transport at the catchment scale (Hom et al. 1974; Latimer and Quinn 1996; Middelkoop

457   2002; Hartmann et al. 2005).

458   Heavy metals (e.g., Pb, Hg, Zn and Cu) are the best recorded anthropogenic contaminants due to

459   high affinity to fine sediment (Chillrud et al. 2003; Franz et al. 2013). In addition, persistent organic

460   pollutants (POPs) (e.g., polychlorinated biphenyls, polycyclic aromatic hydrocarbons,

461   dichlorodiphenyltrichloroethane, benzotriazoles and alkylbenzenes) are ubiquitous in aquatic

462   sediment (Helm et al. 2011). Pesticides may be useful in separating agricultural soils from other

463   types of land-use/land-cover. Sources of fine sediment may be apportioned into individual

464   agricultural fields according to pesticide fingerprints and particle size distribution (Tang et al. 2014).

465   More reliable and detailed information about temporal changes in sedimentation rate and

466   contributing sources over periods of decades can be obtained by inter-comparison and combined

467   use of different dating markers such as radionuclides, heavy metals, persistent organic pollutants,

468   stable isotopes and optically stimulated luminescence (Fox et al. 1999; Boonyatumanond et al. 2007;

469   Hobo et al. 2010). However, it should be noted that most successful studies have been limited to

470   lake and estuary sediments, where sedimentation conditions are more ideal than river sediments

471   where stronger physical mixing, resuspension and bioturbation have been reported to disturb the top

472   few centimeters of sediment. At stream gaging stations, it is often not practical to take a large volume

473   of water using an automatic water sampler and this limitation constrains the mass of anthropogenic

474   micropollutants (associated with suspended fine sediment) ultimately collected. This then restricts

475   the opportunity to reliably quantify the presence and concentrations of micropollutants in the sample.

476   Therefore, some of these micropollutants (e.g., organic micropollutants) may not be suitable to be

477   used for tracking sources of fine suspended particles transported during individual rain events.

478   Alternatively, contaminant profiles of deposited sediment at slower moving section of channels and

479   other surface water bodies can be used to apportion sources and reveal temporal variations, despite

480   the inherent limitations relating to selective transport of sediment that is dependent on hydro-climate

481   condition and channel geomorphological characteristics.

482   Recent studies have shown that stable isotopes of metal contaminants (e.g., Cd, Cu, Hg and Zn),

483   in terms of isotopic ratios such as $^{114}Cd/^{110}Cd$, $^{65}Cu/^{63}Cu$, $^{202}Hg/^{198}Hg$, and $^{66}Zn/^{64}Zn$, may hold

484   particular promise to track sources of contaminated sediments (Gao et al. 2008; Petit et al. 2008;

485   Weiss et al. 2008; Mil-Homens et al. 2013). However, the use of these 'non-traditional' isotopes is

486   complicated by multiple physical and biological fractionation processes that occur during

487   contaminant transport and following sediment deposition (Wombacher et al. 2004; Rehkämper et al.

488   2011). At present, these non-traditional metal isotopes may play only a supportive role to

489   complement data obtained using other well established sediment source tracking techniques.

490

491   Mineral magnetics

492

493   Mineral magnetic properties have been used to discriminate sources of fine sediment entering lakes,

494   rivers and estuaries (Walden et al. 1997; Walling 2013a, b; van der Waal et al. 2015). In particular,

495   mineral magnetics have been successfully used to trace sediment sourced from burned landscapes.

496   In these landscapes fine-grained ferrimagnetic minerals are accumulated in surface soil (Longworth

497   et al. 1979; Smith et al. 2013). Changes in oxidation/reduction conditions in surface soil during

498   burning can convert less magnetic iron oxyhydroxides into more magnetic minerals (Clement et al.

499   2011). Mineral magnetic properties have shown potential for discriminating soil burned at different

500   severities with respect to unburned areas. For example, recent application in the South African

501   Karoo found good discrimination between the sedimentary sources (soils and subsurface material)

502   and dolerite soils due to the much higher magnetism of dolerite soils. Moreover, it was

503   recommended that further fractionation of the <32 μm fraction or a narrower particle size range (e.g.,

504   <10 μm) should be performed to separate fine sediment (<63 μm) and larger particle size fractions

505   of soils and sediments, so that more reliable correction factors can be obtained (Pulley and Rowntree

506   2016a).

507

508   Physical properties

509

510   Color signature and particle size distribution were among the earliest tracers developed (Davis et al.

511   2009). Sediment color, which represents the spectral response of its components (i.e., iron oxides,

512   organic matter, water molecules and clay minerals) to visible light, is an inexpensive tracer despite

513   the reported high uncertainty (Krein et al. 2003). Color is an easy-to-establish trait that can be simply

determined using an ordinary color scanner. The uncertainties of color measurement resulting from

organic matter and particle size can be minimized by sample pre-treatment with H_2O_2 and separating

the sediments into two size fractions (Pulley and Rowntree 2016b). Moreover, variations in

sediment moisture content and chemical reactions of particles with natural elements can alter the

color of the sediment during transport and storage. Therefore, color signature has been less

frequently used in recent years. Notably, color parameters measured by Diffuse Reflectance Infrared

Fourier Transform Spectroscopy (DRIFTS) can be used to characterize potential sources of

suspended sediment in rivers (Martínez-Carreras et al. 2010; Poulenard et al. 2012). For example,

in the Galabre (20 km^2) catchment (French Southern Alps), DRIFTS and a chemometric technique

(i.e., partial least square analysis) were used in parallel to estimate the contribution of potential

sources (i.e., molasse, manly limestones, black marls and gypsum) to the sediment flux during the

floods (Poulenard et al. 2012). In addition, the use of DRIFT allows direct quantification of the

gypsum proportion in soils and sediments (Böke et al. 2004; Poulenard et al. 2012). This method

requires only a small amount of sediment contained on filter paper. Both near-infrared and mid-

infrared spectroscopy, which are quantitatively linked to geochemical properties, could be employed

to rapidly characterize fine sediment sources. Particle size distribution is applicable to sediments of

heavily contrasting textural origins but may be subject to aggregation or disaggregation during

transport. Particle size distribution has been applied for correction purposes rather than for tracing

(Collins et al. 1997b).

Other tracers

The multi-scale, complex and stochastic nature of sediment production, delivery and transport

processes complicates the diagnosis of fine sediment sources, pathways and impacts (Grabowski

and Gurnell 2016). Geochemical tracers, fallout radionuclides (FRNs) and mineral magnetic

properties are the most extensively used for tracking sources of fine sediment (D'Haen et al. 2012).

Most of tracers reviewed above may not be applicable for evaluating the effects of recent changes

of land use on the relative contributions of sources to fine sediment. Alternatively, active approaches

that make use of tracers injected into sources, such as fluorescent fine sediment surrogates (e.g.,

Harvey et al. 2012) and radioisotope-tagged particles (e.g., ^{56}Fe, ^{134}Cs and ^{60}Co; Wooldridge 1965;

Newbold et al. 2005; Greenwood 2012). These have been proposed for evaluating the reach-scale

(i.e. ~100 m scale) effect caused by such short-term changes. Using artificial radionuclides for

tracking the transport of fine sediment requires radioisotope permits and adequate facilities to store

and dispose of radioactive waste. Therefore, the application of artificially introduced radiotracers in

the field has been limited.

In addition to these tracers, there are some temporal markers (i.e., benchmark events with known

dates), such as forest fire, flood and construction activity, that can be identified with related

properties and used to separate the erosion history into several periods showing likely different

sedimentation rates (Hobo et al. 2010; Zhang et al. 2012; van der Waal et al. 2015). For example, in

the sediment profile at the Jiulongdian Reservoir, southwest China, a thin charcoal layer (2-3 mm

thick, observed at 21 cm depth) marked a forest fire in 1998 and has been used as an additional

temporal marker (Zhang et al. 2012). Combined use of these temporal markers and suitable

fingerprint tracers could provide information on temporal changes in relative contributions of

sources to deposited sediment.

26

558

Aspects of channel morphology, discharge and sedimentology

560

561   Fine sediment deposition usually occurs in slower moving sections where slopes are less steep, in

562   wetlands, riparian zones and along channel margins. Channel cross sections and longitudinal

563   surveys can be conducted using an automatic level to determine channel morphology and

564   morphometrics (i.e., channel cross-sectional area, bankfull width and depth, width-to-depth ratio

565   and bed slope) (Wethered et al. 2015). Sedimentological units such as benches, bars and channel

566   beds at regular intervals can be sampled for determining the amount of unconsolidated sediment

567   stored within the channel (Collins and Walling 2007). The average thickness of soft fine sediment

568   for each channel transect can be calculated, which is then averaged for the selected transects in a

569   reach. The volume of fine sediment deposition in each reach can then be estimated by multiplying

570   the average thickness of fine sediment by the average channel width and the reach length. Flow

571   velocity, bankfull discharge and unit stream power can be estimated from the morphometric data

572   using standard equations. Geomorphic units, bank sediment profiles, riparian vegetation and

573   Manning's *n* (roughness) coefficient should also be recorded for each reach. Usually, the sediment

574   sample is sieved through 63 μm and the >63 μm fraction is further dry sieved at specific size

575   intervals to determine the size distribution. This in-channel information can be used to quantitatively

576   evaluate seasonal or inter-annual variation in sediment delivery and deposition along the channel

577   pathways with respect to geomorphological and hydro-climate controls. In a survey of 230 streams

578   in agricultural catchments across England and Wales, total fine sediment in channel beds showed a

579   highly significant relationship with stream power (calculated for bankfull flow and used to index

580   the capacity of the stream to transport sediment; $p < 0.001$) as well as flow velocity ($p < 0.001$)

581   (Naden et al. 2016). Interplay between fine sediment supply, stream flows and aquatic and riparian

582   vegetation is the key to understanding how the channel system has evolved over time and will

583   respond to future land management and climate change (Grabowski and Gurnell 2016).

584

585   **Research needs in sloping farmland-dominated catchments**

586

587   A hierarchical framework, as given in Fig. 1, can be employed to investigate fine sediment sources

588   across multiple scales; four elements are highlighted: (1) identifying catchment-scale spatial

589   distribution of fine sediment sources and temporal changes in sediment production, (2) measuring

590   features (e.g., riparian vegetation, wetlands, dams and hydrologic connections) along the channel

591   system that could intercept fine sediment; (3) determining fine sediment delivery capacity of the

592   river at the segment scale; and (4) quantifying spatial and temporal (seasonal and inter-annual)

593   changes of channel dimensions and fine sediment characteristics at the reach scale. Future work

594   could consider the following aspects.

595

596   Combined use of multiple composite fingerprints

597

598   It is well accepted to use an optimum composite fingerprint comprising a minimum number of

599   tracers in sediment source apportioning (Zhang and Liu 2016). Nevertheless, Sherriff et al. (2015)

600   reported that uncertainty in predictions of source contributions became smaller if more tracers were

601   used. This inconsistency needs further investigation through more case studies. It was recently noted

602   that the link between a tracer's ability to discriminate among sources and its reliability in estimating

603   source contributions have received less scrutiny and may be weak (Zhang and Liu 2016). Multiple

604   composite fingerprints (i.e., different sets of tracers) can be used in parallel to improve the accuracy

605   and reliability of the contribution estimates. Mean contributions averaged over multiple composite

606   fingerprints may be more reliable and closer to the population means than estimates derived using

607   a single composite fingerprint alone and may provide greater certainty of contribution estimates.

608   Therefore, a minimum number of potential sediment sources and a maximum number of composite

609   fingerprints, each with a minimum number of tracers, are recommended (Zhang and Liu, 2016). In

610   addition to the widely used statistical approach, prior knowledge of biogeochemical, hydrological,

611   soil erosion and environmental processes and related parameters should be considered in tracer

612   selection.

613

614   Temporal and spatial variation of fine sediment sources and delivery

615

616   Sediment sources are commonly defined either spatially (e.g., reach sub-catchments and geological

617   sub-areas) or typologically (e.g., land-use types, surface *vs* subsurface sources) (Collins and Walling

618   2002). Land-use time series maps and agricultural census records are useful to identify temporal

619   changes in intensive crop cultivation and livestock farming in source uplands. Rainfall regime

620   governs temporal variability of soil erosion and sediment production. Agriculture could be the

621   dominant contributor to suspended sediment during cropping season while river banks could be an

622   important source of suspended sediment during winter (particularly snowmelt) months (Lamba et

623   al. 2015b). In channels, the bed downcuts slowly during moderate- and low-flow hydrologic events.

624 Deposition and replenishment of sediment to the bed is pronounced as a result of very high-flow

625 hydrologic events (Fox et al. 2010). The channel bed in a lowland catchment functions as a

626 temporary storage zone of fine sediment. The effect of temporary fine sediment storage (known as

627 'memory effect') may alter the representativeness of fine sediment collected downstream for source

628 apportionment, particularly in large braided river catchments (Navratil et al. 2010; Evrard et al.

629 2011). Transport of suspended fine sediment during storm events and resuspension of bed fine

630 sediment are different from baseflow periods. Improved understanding of seasonal and inter-annual

631 variability of fine sediment production and delivery is essential for identification of the

632 geomorphological impacts of excess fine sediment within river systems. Based on identification of

633 fine sediment sources by multiple-tracer approaches, mapping of delivery pathways and

634 measurement of fine sediment fluxes, a complete, spatially distributed sediment balance from source

635 uplands to catchment outlet should be made at the catchment scale, and temporal dynamics of

636 sediment delivery, to and within, the channel system quantified as a function of landscape position

637 or hydrologic connection (Koiter et al. 2013b). Reliable identification of critical fine sediment

638 source types and erosion hotspots is essential for optimizing fine sediment management and

639 informing best soil conservation practices at the catchment scale. Combined use of soil erosion and

640 sediment transport models, such as the Revised Universal Soil Loss Equation 2 (RUSLE 2) (USDA-

641 ARS 2006) and Water Erosion Prediction Project (WEPP) model (Flanagan and Livingston 1995),

642 with sediment source tracking techniques with selected tracers (Lamba et al. 2015b) and temporal

643 markers could be an effective approach toward this end.

644

645 Development of mixing models

646

647   Multivariate mixing models provide relative contribution estimates of fine sediment sources

648   (Collins et al. 1997a,b; Franks and Rowan 2000). In addition to particle size, organic matter

649   correction factors and other weighing factors, a temporal dimension should be incorporated to

650   represent the changes in fine sediment source over time (Walling, 2013a,b). Additional uncertainties

651   associated with non-linear additivity need also to be considered in modeling (Lees 1997; Small et

652   al 2004). Few attempts have yet been made to develop precise guidelines for selecting appropriate

653   tracer properties to be incorporated into the mixing model (Davis and Fox 2009).

654

655   Effect of construction activities and road verges

656

657   Fine sediment deliveries per unit area via runoff from construction sites and damaged road verges

658   are usually much greater than those from agricultural lands (Owens et al. 2000; Collins et al. 2010a).

659   Fine sediment from construction sites can be a significant contributor of sediment flux to channel

660   systems if erosion controls are not implemented. The magnitude of fine sediment transport from the

661   site is governed by soil type, site slope, erosion control practices and rainfall characteristics. In

662   particular, rainfall intensity plays a much more important role in fine sediment transport than the

663   "first flush" generated by the first rainfall (Owens et al. 2000). Additionally, attention should be

664   given to the potential for fine sediment transport resulting from intensive human disturbances in

665   hilly areas.

666

667   Development of an environmental forensics tool

668

669 Sediment fingerprinting offers a potentially valuable tool for accurate assessment of fine sediment

670 sources, in support of the development efficient mitigation strategies for excess fine sediment

671 loading and resultant environmental problems in a catchment (Davis and Fox 2009). Nevertheless,

672 it has rarely been practiced as a catchment management tool for fine sediment within a regulatory

673 framework. Regarding the multidisciplinary nature, forensic-style investigations of sediment

674 sources should use robust quantitative schemes incorporating various data and modeling

675 uncertainties and be practically resourced to achieve adequate accuracy and precision (Rowan et al.

676 2012; Small et al. 2004). When compared with the current source assessment tools in developing

677 total maximum daily load standards (TMDLs) for sediment (USEPA, 1999), sediment fingerprinting

678 offers significant improvement regarding source type and spatial location. A conceptual framework

679 for applying sediment source fingerprinting along with sediment budgeting and modeling in a

680 regulatory framework was proposed for developing justifiable sediment TMDLs, with special

681 reference to the U.S. TMDL program (Mukundan et al. 2012).

682

683 Practices for fine sediment control

684

685 Lower surface hydrological connectivity in well-drained catchments as a result of a dominant role

686 of subsurface flows can cause decrease in sediment transport (Sherriff et al. 2016). Management of

687 hydrological connectivity becomes an effective approach to preventing long distance transport of

688 fine sediment and associated pollutants. Temporary measures such as sediment fences in fields

689 subject to high soil erosion risk may be taken prior to the occurrence of extreme rainfall events

690 (Vinten et al. 2014). To minimize river bank erosion, limiting livestock (cattle, ducks etc) access to

691 rivers and river bank stabilization are recommended (Lamba et al. 2015a). Other engineering

692 practices for sediment control include stormwater detention facilities (e.g., small dams and pools),

693 riparian buffers, wetlands and targeted in-channel management (e.g., installation of various

694 sediment traps) (Mukundan et al. 2012; Lamba et al. 2015a). Tiessen et al. (2011) reported that small

695 dams in an agricultural river basin could reduce the annual sediment load by 70%. Riparian buffer

696 zones and wetlands could slow the stream flow, resulting in the deposition of fine sediment, and the

697 trapping efficiency was largely dependent on the vegetational and geomorphical conditions of these

698 features. Constructed and restored wetlands receiving agricultural runoff could achieve sediment

699 removal efficiencies greater than 50% (O'Geen et al. 2010). However, relatively few data of particle

700 size distribution and organic matter content have been reported for suspended sediment leaving

701 dams or wetlands although fine sediment would be expected to be preferentially exported from these

702 features. Cost-effective catchment-wide practices for reducing sediment control are increasingly

703 needed with respect to diffuse fine sediment sources.

704 Sediment source tracking at multiple scales within a catchment can support the formulation and

705 continued optimization of fine sediment control and management strategies. A general procedure

706 for this purpose, which is modified from the procedure described by Madej (2007), may include the

707 following seven steps: 1) define the critical problems relating to soil erosion and fine sediment

708 transport; 2) acquire regional background information by reviewing various available sources of

709 literature; 3) subdivide the concerned catchment into sub-catchments or terrain units with respect to

710 land uses, vegetation and spatial patterns of hydrological processes, soil erosion and sediment

711 transport; 4) identify the locations and timing of hillslope sediment sources, channel bank erosion

712 and channel incision using suitable erosion models and/or monitoring data; 5) assess fine sediment

713 delivery and apportioning its sources using optimum composite fingerprint(s); 6) prioritize,

714 prescribe and implement measures for fine sediment control; 7) and monitor the effectiveness of the

715 fine sediment control practice implemented using source tracking techniques in order to prescribe

716 and take more effective management actions.

717

718 **Conclusions**

719

720 A reliable identification of contributing sources and delivery pathways of fine sediment into riverine

721 systems is of pivotal importance for sustainable catchment management. Accurate provenance of

722 fine sediment is still limited by: problems associated with representativeness of sediment and source

723 soil samples; the complexity and stochasticity of transport processes; uncertainties associated with

724 source tracking approaches, and; lack of temporally and spatially distributed monitoring data for

725 fine sediment production and delivery. This review provides an overview of conservative tracking

726 and source apportionment modeling techniques to pinpoint fine sediment to individual sloping fields

727 and non-field areas according to their fingerprints. These fingerprints may comprise: selected fallout

728 radionuclides; geochemical properties; mineral magnetics; elemental composition; fluorescence of

729 organic matter; anthropogenic pollutants; stable isotopes; physical properties, or; other tracers.

730 Despite varying forms of erosion processes and particle size selectivity, mobilized fine sediment

731 can maintain the same fingerprint as the original sources or secondary sources (e.g., those re-

732 suspended during rain events). The multiple tracer approach along with numerical mixing models

733 could provide a valuable forensic technique to resolve a legal dispute between land users and

734 catchment managers regarding environmental pollution issues relating to fine sediment transport,

735 and could be useful in developing total maximum daily load for fine sediment. The quantitative

736 relationships between tracers and particle size composition and organic matter content are likely to

737 vary between tracers and the use of a single correction factor for all tracers may not be appropriate.

738 Through combined use of multiple-tracer approach and seasonal and rain event based monitoring

739 of suspended and deposited fine sediment and channel geomorphological properties at nested sites

740 within a catchment, spatial distribution of fine sediment sources and temporal changes of their

741 contributions at a specific location can be identified. Such information will be essential for

742 prioritizing the implementation of erosion control measures to reduce the loading of fine sediment

743 and associated contaminants to surface water bodies, particularly for agriculture-dominated

744 catchments, catchments known to have high agrochemical or contaminants loadings and catchments

745 with complex and changing land-use. However, this integrated scheme is demanding in terms of

746 resources (i.e., time, funding and personnel), and this represents an important constraint on wider

747 application.

748

755

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1245　**Figure caption**:

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1247　**Fig. 1**　A conceptual framework of integrated, spatial distributed source tracking approach for fine

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1251 **Table 1**   A review of tracers for fine sediment tracking

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Category	Tracer	Origin	Applicable timeframe	Applicable spatial scale	Advantage	Limitation	Reference
Radionuclides	^{137}Cs	Man-made	Since 1954	Plot to catchment	Capable of distinguishing uncultivated and cultivated lands and reconstructing retrospective sedimentation histories	The chronology of sedimentation is crude, which is separated by a turning year of 1963 (main peak)	Wallbrink and Murray (1993); Wallbrink et al. (1998); Zhang et al. (2007)
	^{7}Be	Natural cosmogenic	Runoff event; <5 months	Plot to short reach	Capable of estimating runoff events based and seasonal erosion	Not for serious erosion, gully erosion, long term erosion estimation or distinguishing cultivated and uncultivated surface soils	Walling (2013a)
	^{210}Pb$_{ex}$	Natural geogenic	<~100–150 years	Plot to catchment	Capable of distinguishing uncultivated and cultivated lands and reconstructing retrospective sedimentation histories	Requires skilled staff; measurement with low energy gamma spectrometry is not easy	Wallbrink and Murray (1993); Wallbrink et al. (1998); Mabit et al. (2008)
Geochemical elements	Rare earth elements	Artificially added or natural geogenic	Runoff event (artificial) or since 1772 (natural)	Laboratory and pilot scale (artificial); catchment (natural)	Capable of estimating soil redistribution at small temporal and spatial scales (artificial); for reconstructing the histories of soil erosion from deposited sediment	Not for undisturbed soils and acidic soils	Tian et al. (1994); Land et al. (1999); Wen et al. (2014)

					(natural)		
	Major elements	Natural geogenic	Runoff event based; <~150 years	Catchment	Applicable in catchments showing significant spatial variations in geochemistry	Not for catchments with spatially homogeneous geochemistry	Rollinson (1993); Franz et al. (2013)
	Trace elements	Natural lithogenic	Runoff event based; <~150 years	Catchment	Applicable in catchments showing significant spatial variations in lithology	Not for catchments with spatially homogeneous lithology	Collins et al. (1997a); Grimes et al. (2007)
Properties of organic matter	TOC, TON, TOP	Plants	Runoff event based; decades	Catchment	Capable of distinguishing different land-uses	Relatively low discriminatory ability	Walling and Amos (1999); Carter et al. (2003)
	3D Fluorescence spectroscopy	Plants	Runoff event based; seasonal	Catchment	Capable of distinguishing different types or quality of organic matter sources	Relatively high demand for statistics	Larsen et al. (2015)
	^{13}C, ^{15}N	Plants	Runoff event based; several hundred years	Catchment	Capable of distinguishing different land-uses and land covers	Not standalone	Fox and Papanicolaou (2007)
	CSSI (^{13}C of fatty acids)	Plants (including crops)	Runoff event-based; seasonal; decades to thousands of years	Catchment	Capable of distinguishing between land-uses and cropping systems	Not standalone; not for situations where the soil is very low in organic matter	Gibbs (2008)
Anthropogenic pollutants	Heavy metals	Industry and	Rain event based;	Catchment	Capable of reconstructing sedimentation rates in	May be limited by mixed or unknown sources of	Chillrud et al. (2003); Franz

Category	Property	Source	Timescale	Spatial scale	Advantages	Limitations	References
		agriculture	seasonal; decades		floodplains & catchments having historic river pollution	each metal	et al. (2013)
	Persistent organic pollutants	Industry and agriculture	Seasonal; decades	Catchment	Capable of identifying agricultural sources	Not for individual runoff events	Helm et al. (2011)
	Pesticides	Agriculture	Seasonal; decades	Catchment	Capable of tracking crop-specific agricultural sources	Not for individual runoff events	Tang et al. (2014)
	^{114}Cd, ^{65}Cu, ^{202}Hg, ^{66}Zn	Industry	Seasonal; decades	Catchment	Capable of tracking industry-affected sources	Not for individual runoff events	Gao et al. (2008); Weiss et al. (2008); Petit et al. (2008); Mil-Homens et al. (2013)
Mineral magnetics	Magnetic mineralogy, magnetic properties (χ_{lf}, χ_{fd}, χ_{ARM}, SIRM, Soft IRM, HIRM)	Soil formation	Decades; runoff event based	Plot to catchment	Capable of reconstructing history of sediment sources and discriminating soil burned at different severities	May be changed in oxidation/reduction conditions	Yu and Oldfield (1989); Walden et al. (1997); van der Waal et al. (2015)
Physical properties	Color signature	Natural	Decades; runoff event based	Catchment	Quickly, easily measurable	May be subject to moisture content and chemical reactions	Krein et al. (2003); Pulley and Rowntree (2016)

	Particle size distribution	Natural	Decades; runoff event based	Catchment	Easily measurable	Relatively low discriminatory ability	Collins et al. (1997b)
Other tracers	Fluorescent fine sediment surrogate	Artificial	Runoff event based	Slope	Results are precise with high spatial resolution	Relatively costly and man-power demanding	Harvey et al. (2012)
	Radioisotope-tagged particles	Artificial	Runoff event based	Slope	Results are precise with high spatial resolution	Relatively costly; needs radioisotope permits and related facilities.	Wooldridge (1965); Newbold et al. (2005); Greenwood (2012)

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