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Research article

Sediment source fingerprinting as an aid to catchment management: A review of the current state of knowledge and a methodological decision-tree for end-users

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ABSTRACT

The growing awareness of the environmental significance of fine-grained sediment fluxes through catchment systems continues to underscore the need for reliable information on the principal sources of this material. Source estimates are difficult to obtain using traditional monitoring techniques, but sediment source fingerprinting or tracing procedures, have emerged as a potentially valuable alternative. Despite the rapidly increasing numbers of studies reporting the use of sediment source fingerprinting, several key challenges and uncertainties continue to hamper consensus among the international scientific community on key components of the existing methodological procedures. Accordingly, this contribution reviews and presents recent developments for several key aspects of fingerprinting, namely: sediment source classification, catchment source and target sediment sampling, tracer selection, grain size issues, tracer conservatism, source apportionment modelling, and assessment of source predictions using artificial mixtures. Finally, a decision-tree representing the current state of knowledge is presented, to guide end-users in applying the fingerprinting approach.

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1. Introduction to sediment source fingerprinting

Reliable quantitative information on fine-grained sediment sources in river catchments is required to help target remedial actions for mitigating the impacts of excessive fine sediment loss on aquatic biology (Kemp et al., 2011; Jones et al., 2012). Such knowledge can also help reduce the contribution of high sediment loads to drinking water treatment costs (Lal and Stewart, 2013), the maintenance of water storage reservoirs (Verstraeten and Poesen, 2000), and navigation routes (Milliman and Meade, 1983). The role of fine sediment redistribution as a key vector for the transfer of nutrients and contaminants (Horowitz, 1985; Allan, 1986) across the land-to-water continuum has also been a key driver for the increased need for information on fine-grained sediment

provenance. The use of tracers to infer fine-grained (typically <63 μm) sediment provenance qualitatively dates back to the 1970s (Klages and Hsieh, 1975; Wall and Wilding, 1976; Walling et al., 1979). As the popularity of such approaches began to increase, statistical methods were introduced to improve the robustness of source discrimination (Yu and Oldfield, 1989; Walling and Woodward, 1995; Collins et al., 1996, 1997a). In addition, mathematical un-mixing modelling was introduced for the quantitative apportionment of sediment provenance (Walling et al., 1993; Walling and Woodward, 1995; Collins et al., 1996, 1997a) because it soon became apparent that no single tracer could discriminate robustly between multiple potential sediment sources. This realization also resulted in the growing application of composite signatures combining tracers with differing environmental controls (Walling et al., 1993; Collins et al., 1997a; Devereux et al., 2010). Accordingly, a wide range of tracer properties have been tested and applied in the growing body of studies using the fingerprinting approach (Collins and Walling, 2004; Walling, 2005, 2013;

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Haddadchi et al., 2013; Guzmán et al., 2013; Miller et al., 2015; Collins, 2015). The physical properties tested include colour (Grimshaw and Lewin, 1980; Krein et al., 2003; Croft and Pye, 2004; Martinez-Carreras et al., 2010; Barthod et al., 2015) and grain size (Kurashige and Fusejima, 1997; Weltje and Prins, 2003, 2007; Weltje, 2012). Chemical properties include clay mineralogy (Eberl, 2004; Gingele and De Deckker, 2005), mineral-magnetism (Yu and Oldfield, 1993; Caitcheon, 1998; Maher et al., 2009; Dearing, 2000; Zhang et al., 2008; Hatfield and Maher, 2009), geochemistry (Collins and Walling, 2002), fallout radionuclides (Wallbrink and Murray, 1993; Krause et al., 2003; Wilkinson et al., 2013; Belmont et al., 2014; Evrard et al., 2016), cosmogenic radionuclides (Perg et al., 2003), bulk stable isotopes and isotopic ratios (Yang et al., 2008; Fox and Papanicolaou, 2008), and biomarkers (Hancock and Revill, 2013; Alewell et al., 2016; Reiffarth et al., 2016). Biological properties include soil enzymes (Nosrati et al., 2011) and pollen (Brown, 1985). The underlying assumption in the application of these various tracer groups is that they provide a robust basis for discriminating potential sediment sources, although in many instances, source discrimination is tested by finding a statistical solution using either parametric (Collins et al., 2010a) or Bayesian (Stewart et al., 2014) approaches. Inherent in the sediment fingerprinting approach are the additional assumptions that the tracer properties are measurable, conservative (e.g. don't change from source to sink or evolve in a predictable manner), and representative. These assumptions have been and continue to be scrutinized (e.g. Foster and Lees, 2000; Koiter et al., 2013) and represent an area of much needed further research.

Despite the recent growing application of sediment source tracing (Walling, 2013; Guzmán et al., 2013; Haddadchi et al., 2013; Miller et al., 2015; Walling and Collins, 2016; Walling and Foster, 2016), there remains a strategic need to continue refining and, perhaps more importantly, standardizing the procedures therein. In response to the many questions being raised in this growing field, a technical workshop was organised by the International Commission on Continental Erosion (ICCE) at the 26th International Union of Geodesy and Geophysics meeting in 2015 in Prague to review methodological aspects of sediment fingerprinting, thus resulting in this special section. The following sections provide a brief overview of some of the issues discussed at that workshop and which are explored in the papers in this volume. The sections herein explore progress and remaining issues related to a number of fundamental steps required for the successful application of sediment source tracing including: source classification and sampling, target sediment collection, tracer selection, grain size considerations, tracer conservatism, source discrimination, and apportionment modelling and evaluation. The paper culminates in the presentation of a new decision-tree designed to guide end-users through a series of critical decisions needed to apply the fingerprinting approach to apportion fine-grained sediment sources in river catchments. This decision-tree builds on earlier versions of methodological flow charts including those presented in Lees (1999), Foster and Lees (2000), Walling and Collins (2000), Collins and Walling (2004), and Walling et al. (2003a, 2006), and critically, captures both historical and recent research experiences and lessons.

2. Sediment source classification

A key consideration in the application of sediment fingerprinting relates to the classification of potential catchment sources. The fundamental distinction (Collins and Walling, 2004) made here concerns individual source types (e.g. surface or land use-based versus subsurface i.e., stream banks) and spatial (e.g. geological units or tributary sub-catchments) sediment sources. Some recent

work has combined traditional sediment fingerprinting with particle tracking techniques to increase the resolution of land use-based source types (Collins et al., 2010a, 2013a). In some cases, the classification of sediment source types has been founded on the primary processes of sediment generation including mass wasting, and sheet, rill and gully erosion (Wallbrink and Murray, 1993; Gellis et al., 2009; Gellis and Walling, 2011; Miller et al., 2015); whereas others have combined sediment source types and spatial units (e.g. Collins et al., 1997b; Walling et al., 1999; Juracek and Ziegler, 2009; Wilkinson et al., 2009).

Classification of sediment source groups is most commonly performed *a priori* to align source apportionment estimates with land use patterns and corresponding management goals (e.g. Peart and Walling, 1986; Walling and Woodward, 1995; Collins et al., 1997a; Owens et al., 1999; Porto et al., 2005; Collins et al., 2010a,b,c,d; Smith and Blake, 2014; Lamba et al., 2015; Foucher et al., 2015). Classification by land use has clear practical advantages, as sediment loss assigned to cultivated or pasture land within a catchment, for example, can be targeted by relevant management strategies based on on-farm interventions such as minimum tillage or compaction management. Tracer concentrations in a land use-based source group are likely to be controlled by numerous factors including soil and colluvium parent material, pedogenic processes, anthropogenic inputs, or prevailing soil moisture conditions (e.g. gleying). These factors have the potential to increase within-source variability, with clear implications for the uncertainty ranges associated with predicted source apportionment. The fundamental requirement to reduce intra-group and increase inter-group tracer variability (Small et al., 2002; Collins and Walling, 2002; Pulley et al., 2015a) is likely to be complicated or even confounded if small differences exist in tracer concentrations between different land use or surface and subsurface sources. Small differences in tracer concentrations can be caused by, for example, pedogenic processes or anthropogenic tracer inputs (e.g. atmospheric fallout of particulate lead from combustion). Based on historic land use and/or the chemical properties of the sources, different land uses can be combined, such as in the case of combining pasture with cropland to produce a general source group called agriculture (Gellis et al., 2015; Collins, 2015). Additional complexities must be considered where the fingerprinting approach is used to reconstruct sediment sources through time using sedimentary deposits. Here, some tracers cannot be assumed to have remained constant (behaved conservatively) through time due, for example, to atmospheric pollution elevating concentrations in catchment topsoil sources, as is likely to be the case for heavy metals and nutrients in artificial fertilisers (Foster and Charlesworth, 1996; Foster and Lees, 2000) or as the result of post-depositional dissolution and remobilization.

Geology has commonly been used as a basis of spatial source classification, both alone (e.g. D'Haen et al., 2012; Laceby and Olley, 2015), or paired with land use based classification (e.g. Collins et al., 1998; Owens et al., 1999). Classification by geology may provide less useful information for management purposes as the areas of a catchment underlain by a specific geology may be scattered irregularly around a catchment or a catchment may have homogenous geology under which circumstances an alternative source classification scheme would be warranted. In many catchments, however, geology may provide a convenient basis for classifying different regions such as steep hillslopes and uplands used for grazing as opposed to valley bottoms utilised for intensive cultivation and habitation (e.g. Collins et al., 1998; van der Waal et al., 2015). In the latter situation, whilst the use of stratigraphic units aids source discrimination, there is a clear need to identify the major sediment generation processes within those source groups to ensure that management interventions are targeted.

Objective sediment source grouping, as opposed to *a priori* determination, especially in the context of minimising uncertainty associated with quantitative source apportionment estimates, has also been identified as an area of research worthy of more attention (see Pulley et al., 2016 this issue). Pulley et al., 2016 (this issue) expand on a method introduced by Walling et al. (1993) based on pre-selected tracers from cluster analysis to classify sediment source groups. Walling et al. (1993) found that land use was the primary controlling factor on the resulting four to six source groups. Walling and Woodward (1995) also used cluster analysis to classify source groups according to pre-selected tracers. In contrast to Walling et al. (1993), geology was identified as the major controlling factor affecting source group classification, presumably reflecting the larger and geologically more diverse drainage basin used in the latter study. Using a similar cluster analysis method, Pulley et al. (2016 this issue) used the tracer signatures of the source samples in a PCA and cluster analysis to select the source grouping which best fits the measured tracer signatures and this was combined with the modification of the cluster groupings to suit management goals. Wilkinson et al. (2015) modelled soil erosion and collected source fingerprinting samples with priority given to heavily eroding areas with the aim of reducing the potential impacts of within-source group variability and making the source samples more representative of heavily eroding sediment sources. Similarly, van der Waal et al. (2015) retrieved source samples from key erosional features, such as gullies, which were identified using aerial photography. These approaches do, however, require a clear understanding of the connectivity between eroding areas and the river channel, which continues to be a fundamental premise for targeting source sampling as eroded sediment will often not reach the river channel in short time periods and contributing areas will vary during runoff events of different magnitude and frequency (Fryirs, 2013). Few source tracing studies publish maps of actual source-to-river connectivity, but many (e.g. Gellis and Noe, 2013; Lacey et al., 2015) publish source sampling location maps (as opposed to just geology and land use maps) which can be assumed to be indicative of such connectivity given the underpinning assumption in applying fingerprinting procedures that active sources are sampled.

3. Catchment source sampling

Sampling protocols for catchment sources continue to be refined with recent developments including the combination of independent lines of evidence with more traditional strategies. Here, for example, some researchers have combined process-based model characterisation of catchments to help target source sampling (Wilkinson et al., 2015; Theuring et al., 2015). Geomorphic assessments (Wethered et al., 2015) and Google Earth (Boardman, 2016) can help ensure that source sampling strategies are better informed. Source material sampling is most commonly conducted during a single intensive campaign. For many tracers this is an adequate means of sampling; however, some organic tracers are strongly influenced by seasonality. For example, nutrients from decomposing post-reproductive salmon carcasses can be a significant seasonal contribution to aquatic organic matter in some rivers (Bilby et al., 1996; Ben-David et al., 1998). Sediment-associated organic matter in streams is primarily composed of bacteria, algae, soil-derived organic matter, macrophytes, leaf detritus, and human septic waste and these sources can have seasonal patterns of readily available material associated with, for example, vegetation dieback. In the absence of repeat source sampling campaigns over time, sediment-associated organic matter sources can be traced reliably, but for constrained time periods that do not include all seasons (Collins et al., 2013b; 2014). Given these issues, source

sample scheduling must either take into account seasonal issues associated with organic tracers, or alternatively, eliminate those tracers that are subject to substantial intra-annual variability. A similar issue is associated with agricultural crop rotations, that are common in lowland intensive agricultural landscapes, and which have been the feature of many sediment source tracing studies. Crop rotation in particular degrades the strong discrimination between arable and grassland surface soils as potential sediment sources provided by fallout radionuclides (e.g. Cs-137, unsupported Pb-210, Be-7) by generating more overlap between the measured concentrations to the depth of the plough layer. This complication is less evident for unsupported Pb-210 or Be-7 since fallout is ongoing, thereby providing a means of re-setting cultivation effects even in the context of down-profile transfers over time. However, re-setting of down-profile contrasts in fallout nuclide signatures, for example, will depend on nuclide half life; Be-7 (half life 53 days) will return to its pre-disturbance profile form much more quickly than Pb-210 (half life ~ 22 years). More research is required to fully understand the effects of land use change on down profile radionuclide, physical, chemical and magnetic signatures.

When tracing the sources of historically deposited sediment it also must be borne in mind that the concentrations of many tracers in source groups have the propensity to change over long time scales (decades and longer). Examples of tracers which may vary in concentration in source groups over long timescales include phosphorus, which is applied in artificial fertilisers, or lead and other heavy metals where atmospheric releases, through production, processing or consumption, have changed over time with increasing industrialisation and/or regulation (Foster and Charlesworth, 1996). Therefore, relevant timescales in the context of the temporal stability (conservatism) of tracers used to distinguish catchment source groupings over long time spans must be borne in mind (Foster and Lees, 2000).

Many sediment fingerprinting studies address sources by land use and are conducted in small <300 km² watersheds, referred to as management scale watersheds (Gellis and Walling, 2011). Sediment fingerprinting also has been conducted in larger watersheds (1000's km²) (Collins et al., 1997b; Douglas et al., 2003; Voli et al., 2013) but at this larger scale, source percentages by sub-basin and geologic provenance become more important. Here, a confluence-based approach can be the most efficient means of rationalising source sampling through the collection of sediment samples upstream and downstream of major tributaries (e.g. Vale et al., 2016). Gellis et al., 2016 (this issue) examined sediment sources for a large region of the United States, the Midwest cornbelt (648,239 km²), through sampling of fine-grained bed material in 98 Wadeable streams (ranging in area from 6.8 to 5893 km²). Building upon an approach using fallout radionuclides, ⁷Be and ²¹⁰Pb_{ex} (Matisoff et al., 2005). Gellis et al. (2016 this issue) estimated the percentage of surface versus channel derived sediment, and the age of this sediment to less than one year. Results indicate that the majority of sediment is channel derived with many samples being less than 100 days old.

A key outstanding issue is the optimisation of source sampling strategies informed by an understanding of the variability of tracers in the sources concerned (cf. McBratney and Webster, 1983; Oldfield et al., 1989; Sutherland, 1991). Here, one fundamental issue is the collection of sufficient sample numbers for statistical robustness. Probability sampling designs (cf. Collins et al., 2001a) have not been widely adopted, primarily because the implications are that many more samples will need to be collected than is currently normal practice and permitted by research budgets. In an attempt to deal with such issues, many studies collect multiple subsamples within the immediate vicinity of a specific point and bulk these into a composite representative of an individual sampling

location selected for any source category (e.g. Collins et al., 2010b). The statistical implications of such bulking approaches in the context of probability require further exploration. With respect to sampling target sediment, many studies use time-integrating devices for temporal coverage, but there remains a need to collect replicates to assess variation in sediment tracers at any individual channel location, especially since current un-mixing models can include such information in Monte Carlo routines, but also to consider carefully the need for sampling multiple channel locations to ensure appropriate attention is given to potential scale dependencies associated with process domains.

4. The collection of target sediment

A wide range of target sediment sample types have been used in source tracing investigations. Studies focussing on contemporary timescales, originally used either instantaneous suspended (e.g. Peart and Walling, 1986; Walling and Woodward, 1992; Collins et al., 1997a, 2001b; Russell et al., 2001; Carter et al., 2003) or channel bed sediment (e.g. Collins and Walling, 2007; Collins et al., 2012a,b) samples. The former were retrieved using either a submersible pump powered by a portable generator, a portable continuous-flow centrifuge, or an autosampler, whereas the latter were commonly collected using a re-suspension technique (Lambert and Walling, 1988; Duerdoth et al., 2015). But, over time, there has been a step change towards the collection of time-integrated suspended sediment samples (e.g. Walling et al., 2006, 2008; Collins et al., 2010b,d; Massoudieh et al., 2012) using simple traps developed by Phillips et al. (2000) and tested by Russell et al. (2000). Equally, time-integrated samples of interstitial sediment ingressing channel bed gravels have been collected using retrievable basket traps, sometimes inserted in artificial salmonid redds (Walling et al., 2003a; Collins et al., 2013b, 2014). Key advantages associated with using channel bed sediment samples is that they provide a surrogate for material transported continuously over multiple flood events and thereby permit the characterisation of sediment signatures using minimal sampling effort, whilst also avoiding the need to capture flood events and collect samples across hydrographs (Miller and Orbock Miller, 2007; Horowitz et al., 2012; Mukundan et al., 2012; Evrard et al., 2013; Miller et al., 2015). For recent timescales of a few years, some studies have collected surface scrapes from floodplains experiencing regular flood inundation (e.g. Bottrill et al., 2000), with such samples providing a basis for fingerprinting the sources of sediment conveyed and deposited during higher magnitude flood events resulting in overbank inundation.

Whereas many investigations have documented contemporary fine-grained sediment sources, others have sampled floodplain, reservoir, wetland, and lake deposits to attempt to reconstruct changes in sediment sources over a longer timeframe (Foster et al., 1998; Collins et al., 1997c; Owens et al., 1999; Walling et al., 2003b; Miller et al., 2005, 2013; Pittam et al., 2009; Foster et al., 2012; Pulley et al., 2015b; Walling and Foster, 2016). Here, it is assumed that the sampled depositional profiles provide a historical record of sediment material and its signatures. Accordingly, fingerprinting can be performed on sectioned profiles of core samples to reconstruct changes in sediment provenance for the depositional site, where tracer conservatism is shown not to be a problem although in many cases, such conservatism is atypical. The combination of the source apportionment estimates with a dated sediment profile, provides the basis for examining the impact of environmental change associated with, for example, documented land use change or extreme hydrological events on sediment source dynamics.

5. Tracer selection for source discrimination

Following early studies that tended to pre-select tracer shortlists (e.g. Peart and Walling, 1988), most applications of sediment fingerprinting have measured multiple tracers in source samples and then applied statistical tests to confirm source discrimination. Here, once again, recent work has underscored the need to consider carefully a number of critical factors pertaining to either pre-selection, or further screening, following analyses of source material samples. Firstly, confirmation of a sound physical basis for any tracer providing discrimination between potential sources is highly preferable (Foster and Lees, 2000). Accordingly, and by way of example, prior geochemical knowledge linked to geological variation can be used to guide initial tracer selection (Lacey et al., 2015). Equally, an understanding of tracer environmental behaviour, such as that responsible for the contrasting fallout radionuclide signatures of surface and subsurface sources, can be used as a basis for selecting these particular tracers (Walling et al., 2003a). Prior knowledge of the impact of weathering processes in enriching or depleting tracers in specific sources (e.g. surface soil) can also be used (Koiter et al., 2013). Secondly, in the context of the potential for tracer perturbation, composite signatures should not necessarily be based on reductionist optimisation, since larger composite signatures can reduce uncertainty and help counter problems associated with the perturbation of any individual tracer (Sheriff et al., 2015). Here, there is a need to consider expanding composite signatures in the context of goodness-of-fit metrics for un-mixing model performance. Thirdly, tracers with small differences between source groups should not be used since these generate larger uncertainties in estimated source proportions (Pulley et al., 2015a). Fourthly, tracers with greater between-group to within-group variability ratios should be pre-selected for inclusion in statistical tests applied for quantifying source discrimination (Pulley et al., 2015a). Previous work has shown that individual tracer property groups can provide robust discrimination (Collins and Walling, 2002), but where resources permit, the inclusion of properties responding to differing environmental controls is preferable. Although prior knowledge of tracer behaviour may not be for the precise physiographic setting in question, it is likely that sufficient general guidance on tracer pre-selection can be deduced from existing understanding of the typical environmental behaviour of most tracers.

6. Selection of grain size fractions for tracer analyses

The most common practice in published fingerprinting studies is to fingerprint the <63 μm fraction of sediment. The initial justification for this selection, above and beyond the dominant proportion of fluvial suspended sediment loads being represented by this size fraction, was to limit particle size effects given the knowledge that particle size exerts a strong influence on many of the tracers used for fingerprinting (e.g. Jonasson, 1977; Horowitz, 1991). But, because it has been shown that substantial variability in tracer concentrations can exist even within the <63 μm fraction (e.g. Horowitz and Elrick, 1987; Walling and Woodward, 1992; Motha et al., 2003; Hatfield and Maher, 2009; Pulley and Rowntree, 2016), an alternative approach is the use of narrower particle size fractions. Wallbrink (2004), for example, used only the <10 μm fraction, significantly reducing the capacity for particle size variability in the traced fraction. Hatfield and Maher (2009) found that the magnetic properties of catchment soils were significantly different between different particle size ranges of the same source material. As a result, they separated the sediment into 31–63, 8–31, 2–8 and <2 μm aliquots and the contribution of each fraction to the total magnetic properties of the sediment were quantified. Whilst

the methods of Hatfield and Maher (2009) have distinct advantages for identifying particle size effects and selecting the optimum particle size for tracing, they do require the measurement of tracers on multiple particle size fractions, significantly increasing the time and cost of analyses. Therefore, the selection of a narrow particle size range may be of benefit to many fingerprinting studies and, accordingly, some have pre-selected restricted ranges in their procedures (e.g. Wallbrink et al., 2003; Douglas et al., 2010; Wilkinson et al., 2013; Theuring et al., 2013, 2015; Lacey et al., 2015; Haddachi et al., 2015). Since finer fractions are more geochemically active, they are likely to provide more robust source discrimination; however this benefit can be counterbalanced because these finer grain size ranges also are more susceptible to transformation and non-conservative behaviour during transport. It also can be cost prohibitive to obtain sufficient sample masses of restricted size ranges to permit tracer quantification. Selection of any individual size fraction will only be appropriate if this is shown to be the size class that represents the majority of sediment in transport and indeed the fraction responsible for the environmental issue(s) (e.g. degradation of a coral reef or siltation of salmonid spawning gravels) in question. If sediment fingerprinting is to become a widely used management tool, the ability to source individual fractions and/or using very limited size ranges of fine-grained sediment may be cost prohibitive.

7. Tracer conservatism

Sediment source fingerprinting techniques are based on the fundamental assumption that selected tracer properties behave conservatively during mobilisation and delivery through the catchment system and that the properties of source material and sediment samples can therefore be directly compared. The significance of this assumption is increasingly recognised, but also challenged (Foster and Lees, 2000; Motha et al., 2002a,b, 2003, 2004; Koiter et al., 2013; Smith and Blake, 2014; Kraushaar et al., 2015). Early work highlighted the paucity of understanding on this critical assumption (Bubb and Lester, 1991; Zhang and Huang, 1993). Chemical transformations can occur in conjunction with a range of mechanisms throughout the sediment cycle, including, amongst others, scavenging by Fe/Mn oxides, chemical precipitation and incorporation into crystalline matrices (Forstner and Salomans, 1980; Foster and Lees, 2000). Despite these risks, published studies have included tracers prone to transformation, including phosphorus fractions (e.g. Owens et al., 2000). Whilst there are risks of non-conservative behaviour for actively transported fine-grained sediment, such risks are potentially elevated where sedimentary deposits are used to reconstruct catchment sediment sources through time. Post-depositional dissolution or diagenesis and the in-growth of bacterial magnetite can, for example, impact the conservatism of mineral-magnetic tracers (e.g. Foster et al., 2008; Pulley et al., 2015b). Short-lived radionuclides (e.g. ^{210}Pb , ^{137}Cs and ^7Be) are also unsuitable for long-term (more than 100–150 years old) tracing as their activities will be influenced by fallout histories and short half lives. Longer-lived gamma emitting radionuclides such as ^{40}K and ^{235}U , will be more suitable for long-term (centuries to millennia) source reconstructions, assuming they provide robust source discrimination, because of their much longer half-lives (Walling and Foster, 2016).

Of the sources of uncertainty highlighted in the published literature, the effects of changing sediment particle size and organic matter content on tracer signatures during the sediment cycle through catchment systems are often prominent. The effects of these factors on many of the geochemical properties commonly used as sediment source tracers was recognised early on (e.g. Goldberg, 1954; Rex and Goldberg, 1958; Goldberg and Arrhenius,

1958; Krauskopf, 1956; Kononova, 1966; Jones and Bowser, 1978; see Horowitz, 1991 for additional references), yet little of such work seems to have been integrated into current source tracing procedures. Associations of many elements with organic matter are often unpredictable, with some elements having a greater affinity than others (Swanson et al., 1966; Saxby, 1969; Rashid, 1974; Bunzl et al., 1976; Jonasson, 1977; Maulé and Dudas, 1988; see Horowitz, 1991 for additional references). The strength of these associations may differ between catchments (Gibbs, 1977) and organic matter can behave as both a diluent, (e.g. magnetic signatures (Walling and Foster, 2016)) or as a contributor (e.g., Horowitz, 1985; Horowitz and Elrick, 1987). Organic corrections are widely used in conjunction with the application of mineral-magnetic fingerprints. Efforts to mitigate the effects of particle size and organic matter in fingerprinting studies can therefore be seen as being in an early stage of development with many investigations neglecting to include any significant attempt to mitigate their effects other than to sieve to $<63\ \mu\text{m}$ and employ elementary corrections, as discussed elsewhere in this paper.

In the absence of comprehensive information and guidance on the conservatism of multiple tracers in different environments, the vast majority of studies continue to use a simple screening technique to evaluate the conservative behaviour of various tracers based on the so-called range or bracket test, using a variety of rules (e.g. Foster and Lees, 2000; Wilkinson et al., 2013; Collins et al., 2013b,c; Gellis and Noe, 2013). A principal danger with existing range tests is that whilst they confirm that non-conservative transformation is not significant in the context of the sampled source tracer ranges, they do not confirm the complete absence of any non-conservatism (Collins et al., 2013b,c). The use of the range test can be underpinned by tracer screening using literature reviews dealing with tracer geochemistry in conjunction with an understanding of the various effects of changing physicochemical conditions between the source area and the sink (Kraushaar et al., 2015) and this pragmatic approach merits further attention. Pulley et al. (2015c) produced bi-plots of magnetic properties for source samples comprising different particle size fractions and were able to identify if lake sediment samples exhibited similar linear relationships, suggesting the conservatism of these tracers in the deposited sediment. This approach represents a more robust form of the range test, although it does greatly increase the time and cost requirements for tracer analysis and is dependent on a relationship between at least two tracer variables.

Apportionment modelling in the procedures used by some researchers (e.g. Motha et al., 2004) has attempted to include the impact of non-conservative tracers explicitly. Here, for example, work by Collins et al. (2010b, 2012a,b, 2013b,c, 2014) has used probability density functions (pdfs) to construct deviate target sediment tracer values which are then sampled during un-mixing model Monte Carlo repeat iterations using a Latin Hypercube. This approach recognises explicitly that any individual sediment sample, or the sediment from any individual location in a catchment system, has the potential to be transformed due to selectivity and/or biogeochemical alteration (e.g. sorption, dissolution, precipitation, oxidation, reduction), but that collectively, those samples will provide a range of more and less altered tracer values which can be treated as a 'conservative' population (conservative in the context of using the simple range test). Sheriff et al. (2015) report the use of a tracer permutation algorithm developed by Franks and Rowan (2000) to determine the impact of non-conservative tracers on source apportionment predictions. The accuracy of predicted mean source contributions was reported to be significantly different between the maximum positive and negative levels of tracer corruption (-90 and $+155\%$), but uncertainty was not impacted by mimicking tracer transformation.

8. Source apportionment modelling

Use of mathematical techniques (e.g. Yu and Oldfield, 1989, 1993; Walling et al., 1993; Walling and Woodward, 1995; Collins et al., 1997a; Gellis and Landwehr, 2006; Hughes et al., 2009; Sheriff et al., 2015) to un-mix sediment samples represents a key methodological component of source fingerprinting procedures over the past two decades. Recent studies using un-mixing models have applied different composite signatures to assess variation in predictions dependent on the tracers used and to improve the use of multiple tracers provided by current analytical techniques including ICP-MS (Collins et al., 2012a, 2013c; Stone et al., 2014; Theuring et al., 2015) and NIRS (Collins et al., 2013b, 2014). The explicit assessment of uncertainty in conjunction with the growing application of un-mixing modelling was first introduced by Franks and Rowan (2000) and Rowan et al. (2000) in the form of Monte Carlo analysis. It is now standard to include an explicit assessment of uncertainties in conjunction with the use of source apportionment modelling.

The growing use of sediment un-mixing models has demonstrated that the range of uncertainty outputs from Monte Carlo routines is primarily driven by the within-source group variability in tracer concentrations and the corresponding differences in tracer concentrations between-source groups (Small et al., 2002). As a result, weightings have been applied to give a larger emphasis during un-mixing modelling to tracers with a lower within-source variability and greater discriminatory power (Martinez-Carreras et al., 2008; Collins et al., 2010c; Wilkinson et al., 2013; Gellis and Noe, 2013). The latter weighting has, in some cases, been used as a substitute for original weightings reflecting the analytical errors or precision associated with individual tracers (Mackay et al., 1987; He and Owens, 1995; Collins et al., 1997a). These weightings were developed in response to some papers identifying the need to explore their use (Walling et al., 1993). All such weightings should be carefully assessed in the context of evaluating modelled source proportions using goodness-of-fit metrics and artificial sediment mixtures (e.g. Laceby and Olley, 2015). The sensitivity of modelled source proportions to these types of weightings has been reported as limited based on some datasets (Pulley et al., 2015b) and where the impacts are greater, the weightings reflecting analytical precision or tracer discriminatory power are subtle compared to other weightings (Laceby and Olley, 2015).

More recently, variability ratios (of inter-/intra-source group variability) have been recommended by Pulley et al. (2015a) to capture the fundamental need to select tracers that maximize between- rather than within-group tracer variation. These variability ratios can be applied as an initial screen in the tracer selection procedure to remove tracers that are likely to result in elevated levels of uncertainty in both source discrimination (Collins and Walling, 2002) and un-mixing model outputs (Pulley et al., 2015a). Some work has also introduced distribution-based modelling, to ensure that multiple model iterations for uncertainty analyses maintain relationships between tracers during the iterative sampling of tracer distributions reducing the uncertainty ranges present in model outputs (Laceby and Olley, 2015; Laceby et al., 2015). In terms of the input tracer distributions, a critical decision is whether to represent source groups using the 25th–75th percentile range or the 5th–95th percentile range since this decision alone can influence the corresponding uncertainty ranges associated with modelled source proportions. Regardless of the scaling used, mixing model outputs are characterised by uncertainty ranges and a key decision is how to present this uncertainty to catchment stakeholders. Here, many existing studies have reported gross uncertainty ranges (e.g. 5th–95th percentiles, or the entire pdfs), the average mean or median source proportions with

associated uncertainty (95% confidence limits) and tested the convergence of the model runs (e.g. Collins et al., 2013c). Communicating the uncertainty ranges to stakeholders involved in decision-making for managing the sediment problem is important. To simplify the communication of uncertainty, whilst taking explicit account of this issue, some researchers have calculated relative frequency-weighted average mean or median source contributions (e.g. Collins et al., 2013b,c; 2014). The processing of sediment source tracing data for a single location within a study catchment will always be prone to bias introduced by the scale dependencies associated with spatial variation in the mixtures of potential sources and corresponding geomorphic processes driving sediment mobilisation and delivery.

The adoption of un-mixing models by many studies has been accompanied by the inclusion of particle size corrections. In the majority of studies, these continue to be based on the assumption of a simple linear relationship between particle size and tracer signatures (e.g. Collins et al., 1997a; Owens et al., 1999, 2000; Walling et al., 1999, 2003a, 2006, 2008; Smith et al., 2011; Smith and Blake, 2014). However, it has been recognised that relationships between particle size and many tracer signatures are non-linear, especially for specific surface areas $>1.0 \text{ m}^2 \text{ g}^{-1}$ (Horowitz and Elrick, 1987; He and Walling, 1996; Foster et al., 1998; Russell et al., 2001; Motha et al., 2003; Bihari and Dezs, 2008; Hatfield and Maher, 2009; Oldfield et al., 2009); thus introducing uncertainties in conjunction with simple linear corrections. Previous work has demonstrated that significant contrasts can exist in particle size composition between different source and sediment samples, even when all samples have been screened through a 63 μm sieve (e.g. Walling and Woodward, 1992; Russell et al., 2001). Such data imply that even post sieving to $<63 \mu\text{m}$, the tracers of source and sediment samples cannot be directly compared without further correction. Motha et al. (2003) measured tracer signatures associated with various particle size fractions and developed tracer signature-specific correction factors. Russell et al. (2001) also developed tracer-specific curvilinear corrections rather than assuming a generic linear relationship between concentration and grain size. Whilst such approaches help mitigate uncertainties associated with linear corrections they do, however, have disadvantages in terms of the time required for laboratory work. Due to these challenges, some studies have used enrichment factors based on the measured concentrations of tracers in sediment and source samples (e.g. Peart and Walling, 1986; He and Owens, 1995). Alternatively, other studies have adjusted source material tracer concentrations by using information on the grain size of target sediment and tracer concentrations associated with particle size fractions of source materials, to estimate property concentrations in source material with the same grain size composition as the target sediment (Walling and Woodward, 1992; Slattery et al., 1995; Motha et al., 2004). Recognizing that the relation of grain size and tracer property can be positive, negative, or have no relation, Gellis and Noe (2013) used regression analysis of the D_{50} of source samples against tracer concentration to produce a grain-size correction factor. This has the advantage that the fractionation of source samples and analysis of each fraction is not required and a linear relationship is not assumed. However, such methods may require extrapolation of a trend line beyond the range of values found in the source samples, thereby introducing uncertainty. An alternative to developing corrections for grain size effects, is to use narrower size fractions (e.g. $<10 \mu\text{m}$) to counter the potential influence of selectivity during the sediment delivery cascade (e.g. Theuring et al., 2015).

Elementary organic matter corrections have also been used (e.g. Peart and Walling, 1982, 1986; Collins et al., 1997a; Motha et al., 2003, 2004; Walling et al., 2003a, 2006, 2008; Gellis and Noe,

2013; Pulley et al., 2015b), driven primarily by correlations between tracer concentration and organic matter content and by the improvements in the goodness-of-fit outcomes for un-mixing. Again, these weightings were often developed in response to some researchers identifying the need for their inclusion (Walling et al., 1993), although such corrections are site-specific, meaning there is no universal correction factor. Although some research has highlighted the risk of such corrections biasing source predictions (Smith and Blake, 2014); alternatively, recent research has shown that they have limited impact on the source estimates (Pulley et al., 2015b). The importance of carefully assessing elementary corrections for grain size and organic matter on a dataset-specific basis and making informed decisions to avoid over-correction has long been underscored (e.g. Walling and Collins, 2000; Walling et al., 2003a). Noise associated with differing organic matter contents of source materials, or a high within-source group variability in tracer concentrations may mask relationships between D_{50} and tracers leaving them unaccounted for. Sediment-associated organic matter in the fluvial environment exists as loosely-bound particulate material (e.g. leaf litter), which in the case of many of the most commonly used tracers (apart from biomarkers) will act as a diluent, and as surface coatings for mineral particles, where it can act as a concentrator. Moving forward, this implies that the development of more informed correction factors for organic matter needs to take into account both grain-size and phase specific aspects of the problem. Such work has important resource implications.

Some research during recent years has been directed towards the comparison of variations in source apportionment depending on the applied un-mixing model. Haddadchi et al. (2014), for example, compared four different model structures using artificial mixtures with known proportions of sediment sources. There has also been a growing number of source tracing studies which, rather than using maximum likelihood/frequentist (see modelling papers cited above) methods, instead, use Bayesian (Fox and Papanicolaou, 2008; Rowan et al., 2011; Massoudieh et al., 2012; D'Haen et al., 2013; Cooper et al., 2014) modelling approaches. Uptake of the Bayesian models has benefitted from some of them (e.g. Barthod et al., 2015) being open source. The need to compare local and global solutions using the former types of models has been underscored by previous work (Collins et al., 2010d). Model structure and the robustness of the input data both have a strong bearing on the outputs, and end users must carefully assess model structures and approaches when applying fingerprinting procedures. Numerous uncertainties which are not fully accounted for in current fingerprinting procedures have been identified in recent publications, highlighting the need for further methodological refinements which, where appropriate and underpinned by replicated evidence based on multiple catchments and environmental settings, need to be incorporated into sediment un-mixing models (e.g. Motha et al., 2002a,b; D'Haen et al., 2012; Koiter et al., 2013; Smith and Blake, 2014; Pulley et al., 2015b; Lacey and Olley, 2015). An ongoing problem is that many papers assess specific issues for a single or limited set of study catchments/environments, and then propose generic guidance which simply may not be widely applicable. Importantly, however, these recent studies serve as useful reminders that source tracing datasets should be treated on a case by case basis.

9. Use of artificial sediment mixtures to assess source apportionment modelling

The use of artificial mixtures of known quantities of sediment sources (cf. Stott, 1986) has gained increasing popularity in recent years and represents an important component for the development

of robust, widely applicable source tracing procedures. A limitation of fingerprinting research is that it is difficult to validate estimated source proportions using independent evidence as the monitoring and measurement techniques required face their own limitations in terms of the practicalities and costs of deployment both spatially and temporally (Collins and Walling, 2004). Validation of fingerprinting estimates against independently measured data assembled using alternative techniques therefore continues to be rare, although some examples exist (e.g. Peart and Walling, 1988; Collins et al., 1998; Stone et al., 2014). Mixtures of known proportions of sediment sources thereby provide a pragmatic opportunity to assess the accuracy of a fingerprinting procedure on the basis of its estimated source proportions. Early studies using artificial mixtures include the work by Lees (1997) who identified non-linear additivity associated with the use of the mineral magnetic properties of sediment. Franks and Rowan (2000) used five artificial mixtures consisting of five source types based on major chemical groups to assess a source tracing procedure. Small et al. (2004) used a Bayesian modelling approach and artificial mixtures to explore source sampling related uncertainties and the number of source samples required to limit uncertainty in modelling results. Additional studies using artificial mixtures to assess un-mixing model outputs include those by Hughes et al. (2009), Poulenard et al. (2012), Legout et al. (2013), Brosinsky et al. (2014), Haddadchi et al. (2014) and Lacey and Olley (2015). Given the laboratory work associated with generating and analysing the tracer content of artificial source mixtures, some recent studies have introduced synthetic mixtures based on Monte Carlo routines (Palazón et al., 2015; Sheriff et al., 2015) as an alternative. Whilst virtual sample mixtures can be deliberately corrupted to mimic uncertainty (Sheriff et al., 2015) they do, however, have limitations including, for example, different source groups having contrasting particle size distributions (Palazón et al., 2015).

10. A decision tree for guiding application of sediment source tracing

Progress continues to be made in the refinement of sediment source fingerprinting procedures but much scientific debate is ongoing. Following four decades involving preliminary applications, acceptance of the need for composite signatures and the introduction of statistical and numerical modelling approaches, including uncertainty assessment, recent work has re-visited critical assumptions and challenged some recent proposed methodological modifications. In the context of ongoing studies, and the diverging opinions on some aspects of fingerprinting procedures, it is timely to propose a revised decision-tree for supporting critical choices that have to be made by end-users applying the technique. This decision-tree (Fig. 1) attempts to capture the current state-of-the-art, and hopefully serves as one means of synthesizing the lessons gleaned from the past 40 years of research.

Currently many steps of the methodology presented in the decision-tree are in the early stages of research and firm instruction cannot be given due to many factors or processes being site-specific. However, the decision-tree aims to provide an overarching comprehensive methodology which includes important steps for evaluation, validation and uncertainty analysis. It is intended that the decision-tree will provide a framework from which researchers and reviewers can structure their methods and interpretation(s) of sediment fingerprinting results. The goals and resource availability of different studies will likely mean that not all stages of this decision-tree can be strictly followed but in such situations, end-users must identify limitations and shortcomings in the procedures actually applied when reporting their results.



Fig. 1. A methodological decision-tree for guiding application of sediment source fingerprinting (Rousseeuw and Croux, 1993).

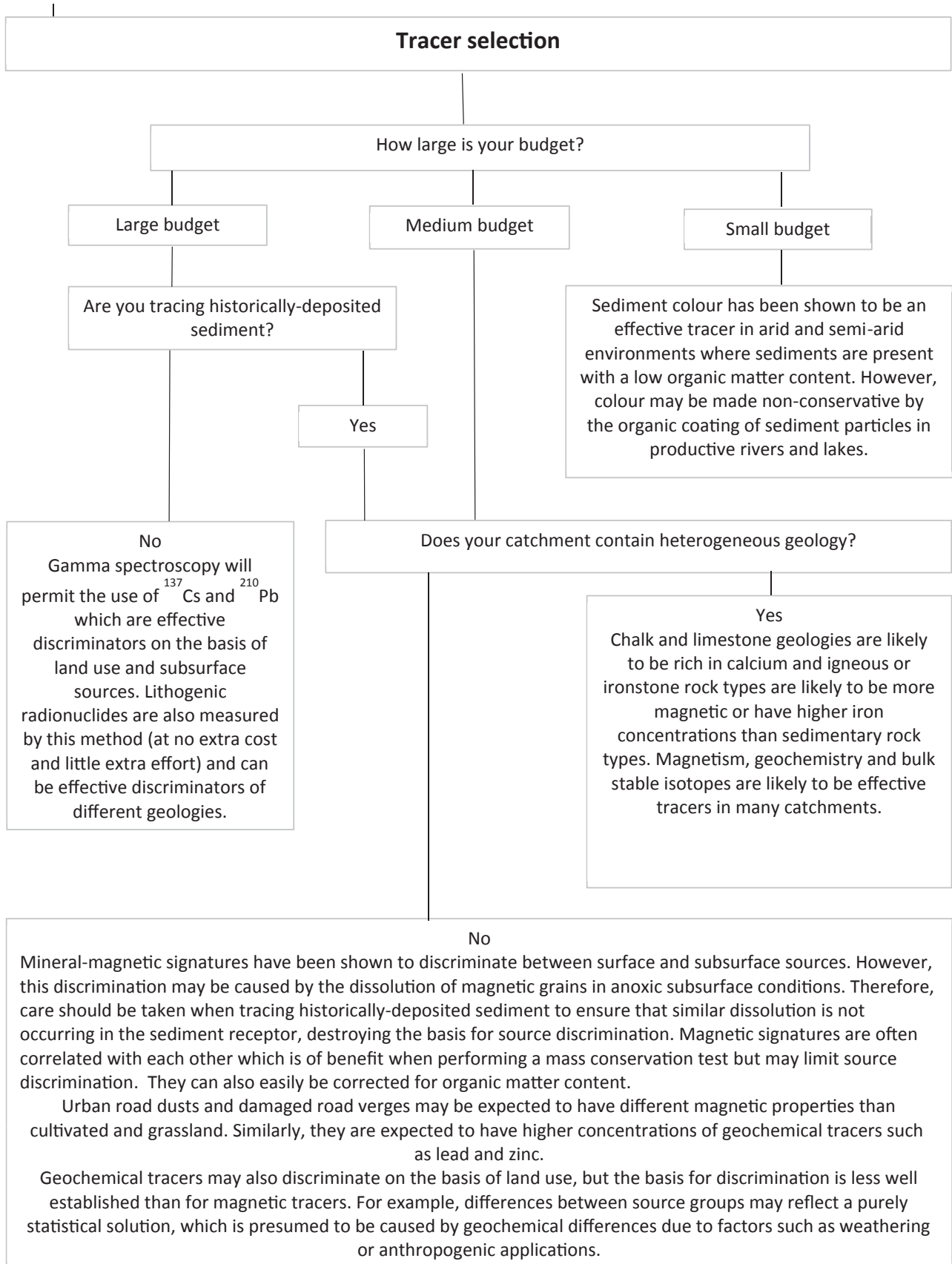


Fig. 1. (continued).

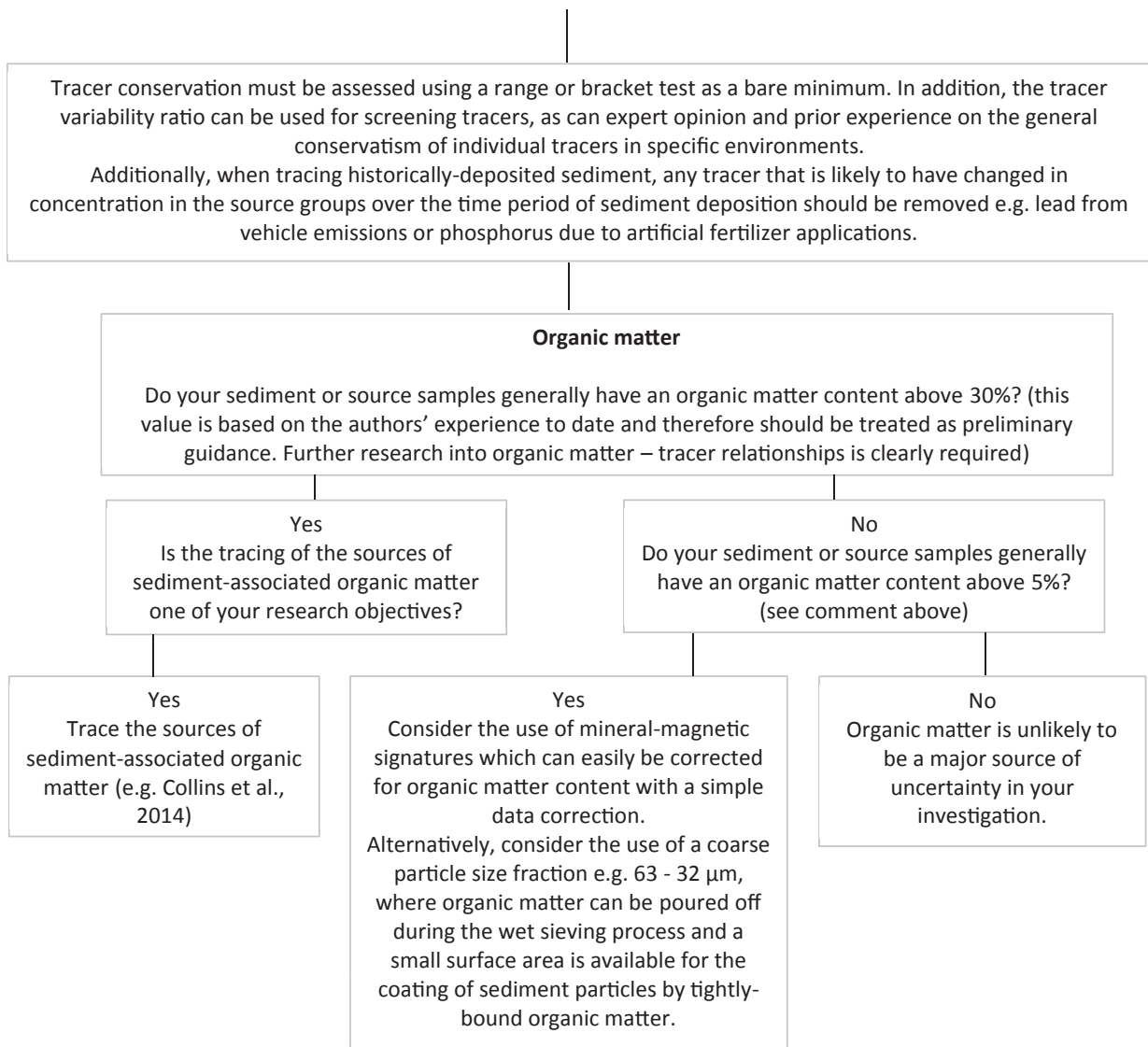


Fig. 1. (continued).

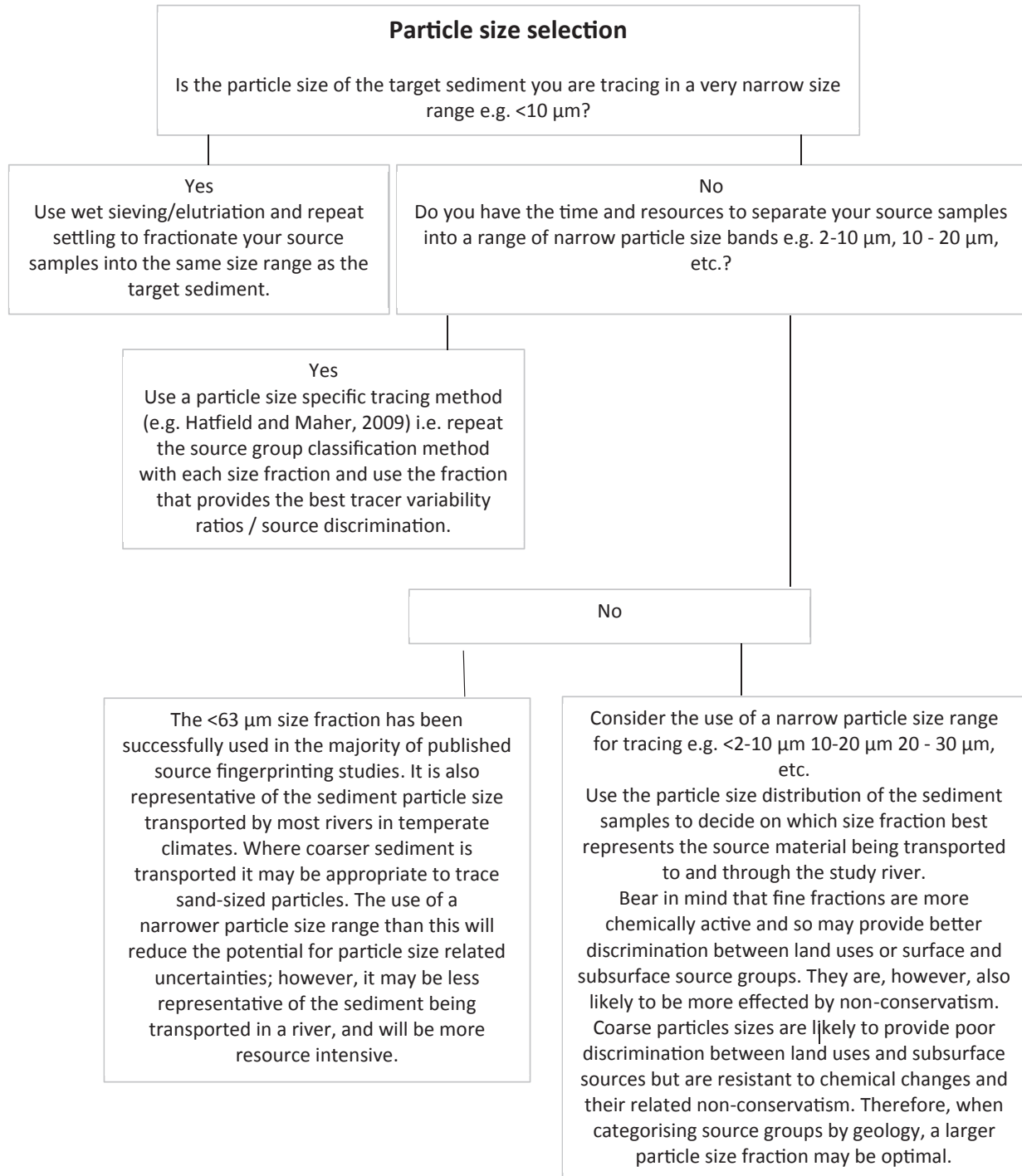


Fig. 1. (continued).

Although the steps in this new decision-tree may increase the resource requirements compared with those used by past published studies, it is our view that adequate recognition must be given to the current state-of-the-art for sediment source

fingerprinting by providing appropriate caveats and/or reporting levels of uncertainty, especially where the results are intended to inform catchment management and sediment mitigation planning.

Notes

It is likely that sediment source discrimination will be in some way particle size dependant in many catchments. For example, sedimentary limestones, chalks and ironstones are likely to have the majority of their calcium and iron concentrated in very fine grain sizes, meaning that discrimination could be weaker in the coarse silt and sand fractions than in fine silts. Additionally, weathering and soil formation processes are likely to result in the preferential precipitation of weathering products on the large surface areas of fine silt and clay particles; as might the adsorption of fallout radionuclides take place. Therefore, prior knowledge of the controls on source group tracer concentrations combined with objective particle size selection may be useful for achieving optimal source discrimination.

The potential for particle size related uncertainties in a tracing methodology is likely to be proportional to the range of particle size selected for analysis. For example, there is far less scope for particle size change in the <10 μm fraction than in the <63 μm fraction. Therefore, when using wide particle size ranges, it is important to demonstrate that particle size effects have been properly investigated and, where necessary, accounted for. The result validation section provides guidance on the use of artificial mixtures to demonstrate the range of uncertainty potentially caused by particle size effects. It is also good practice to compare the particle size distribution of the prepared source samples to the target sediment samples collected from the lake, floodplain or river to show if there are significant differences between the two.

Fig. 1. (continued).

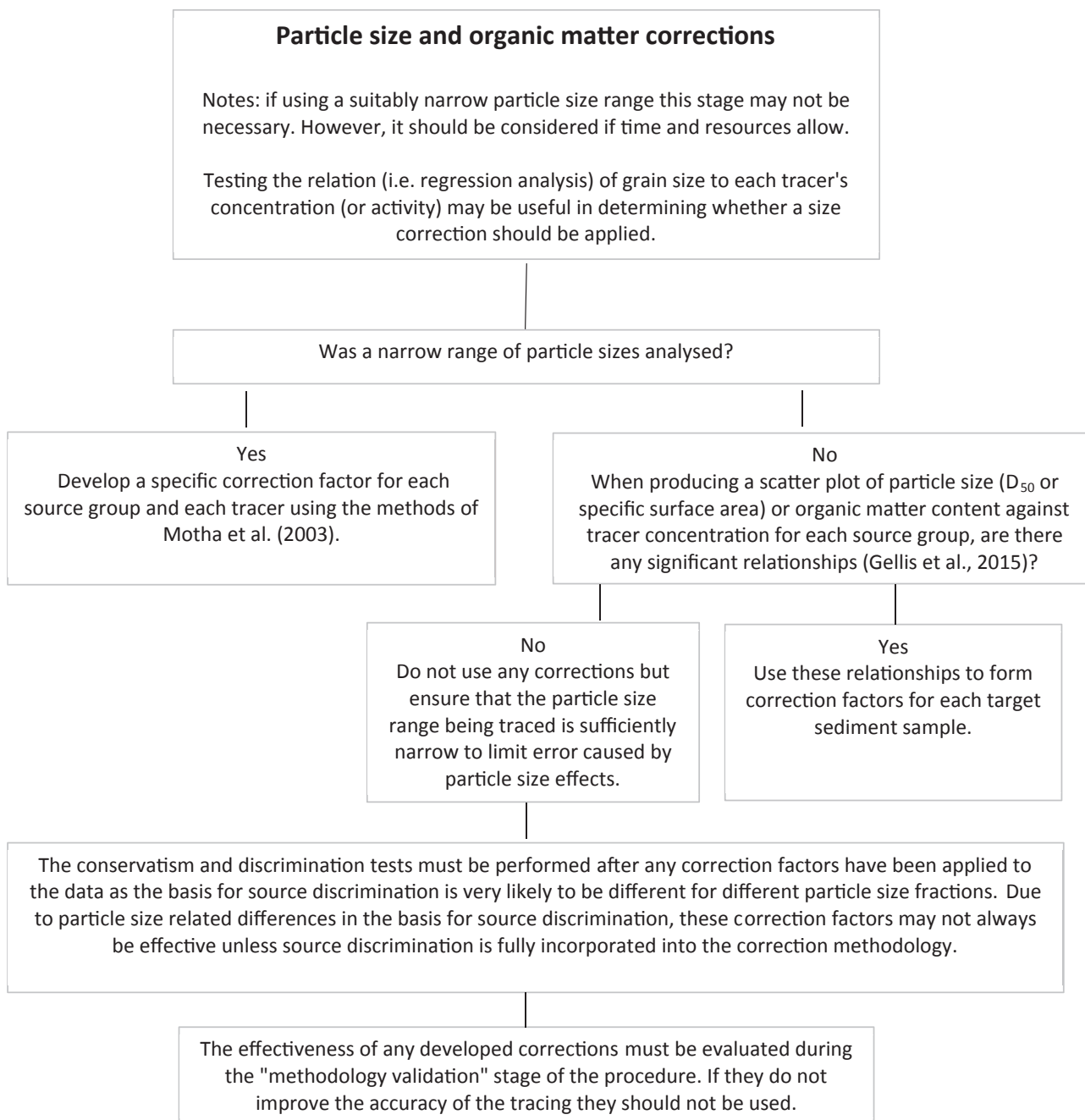


Fig. 1. (continued).

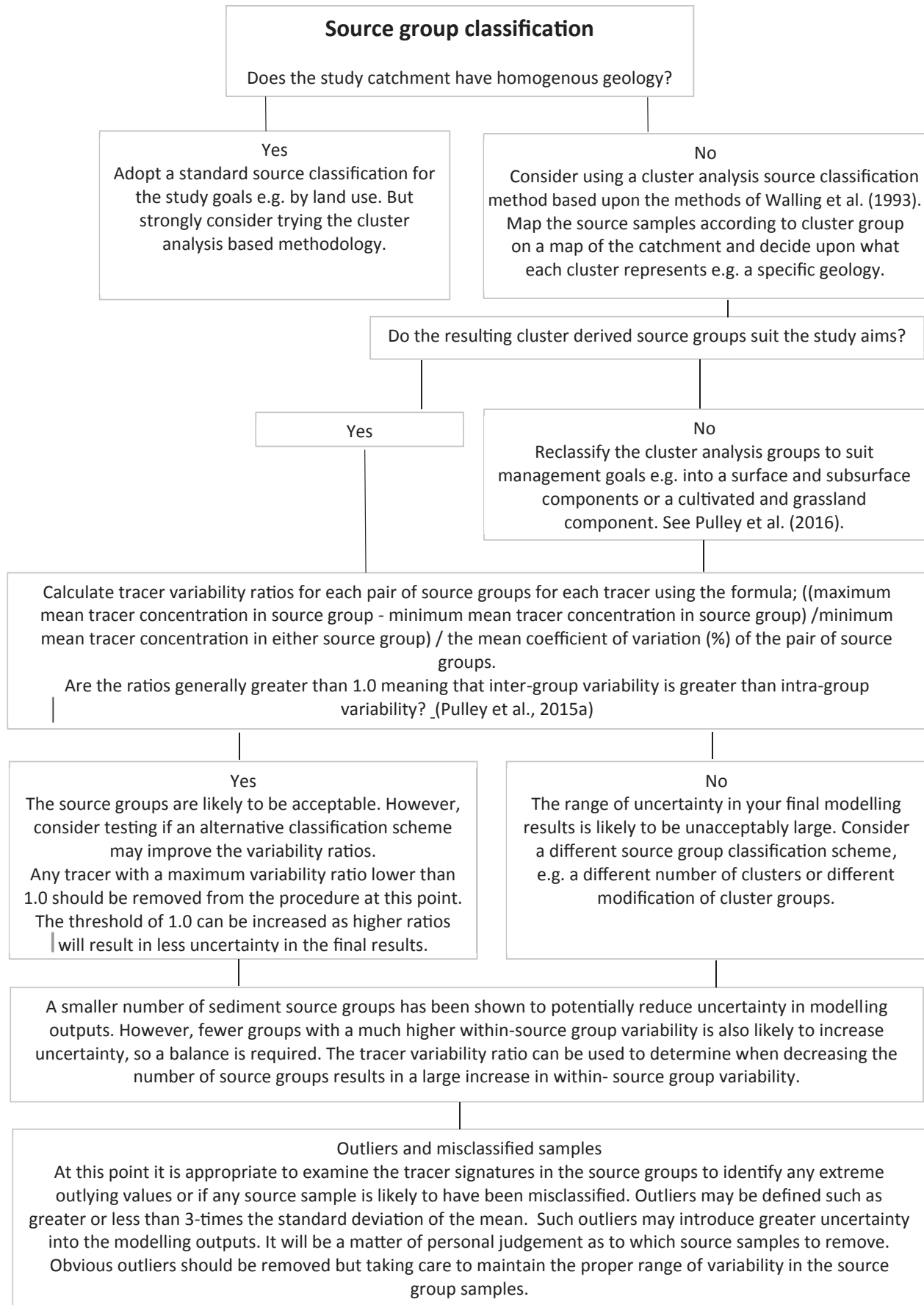


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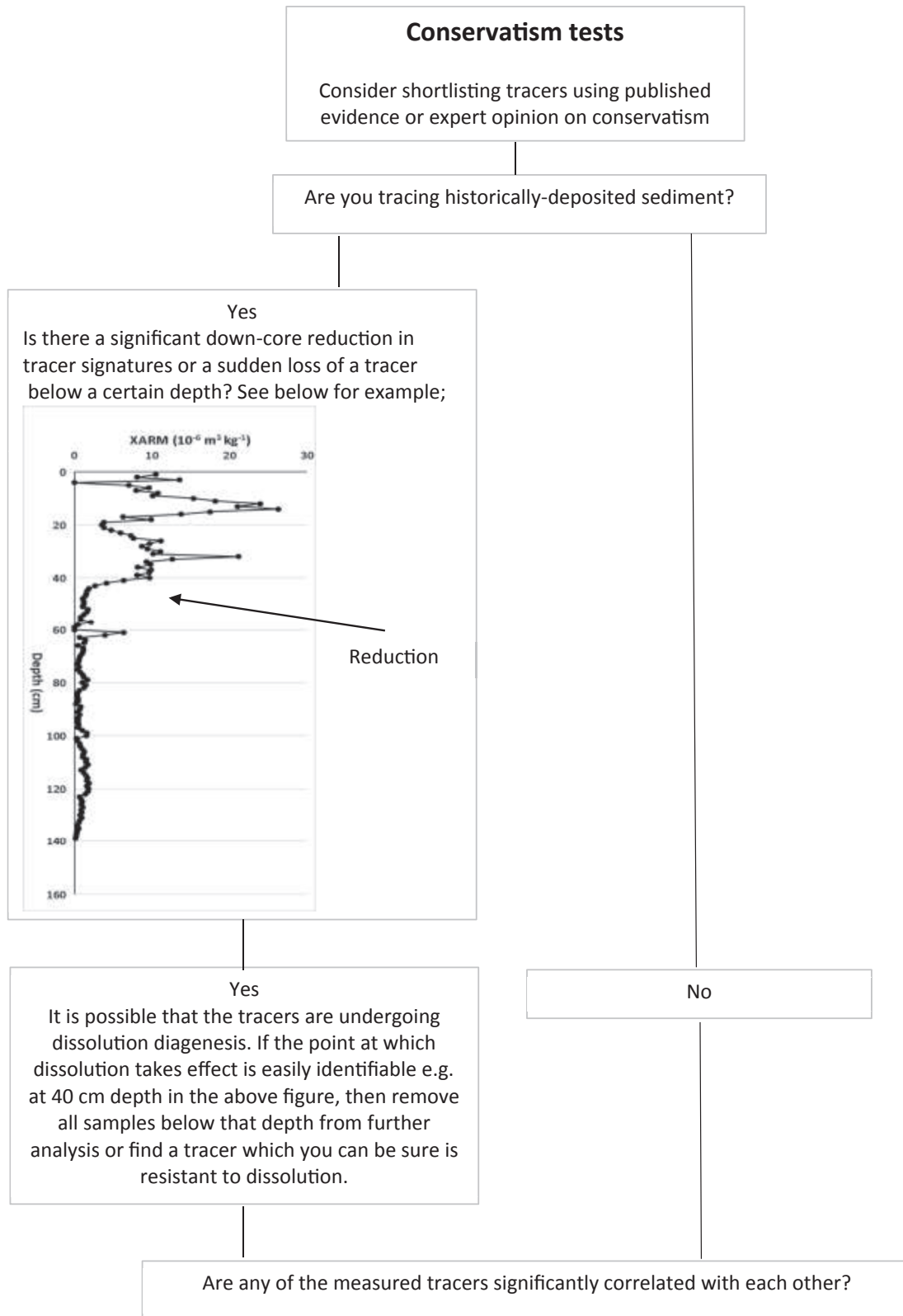


Fig. 1. (continued).

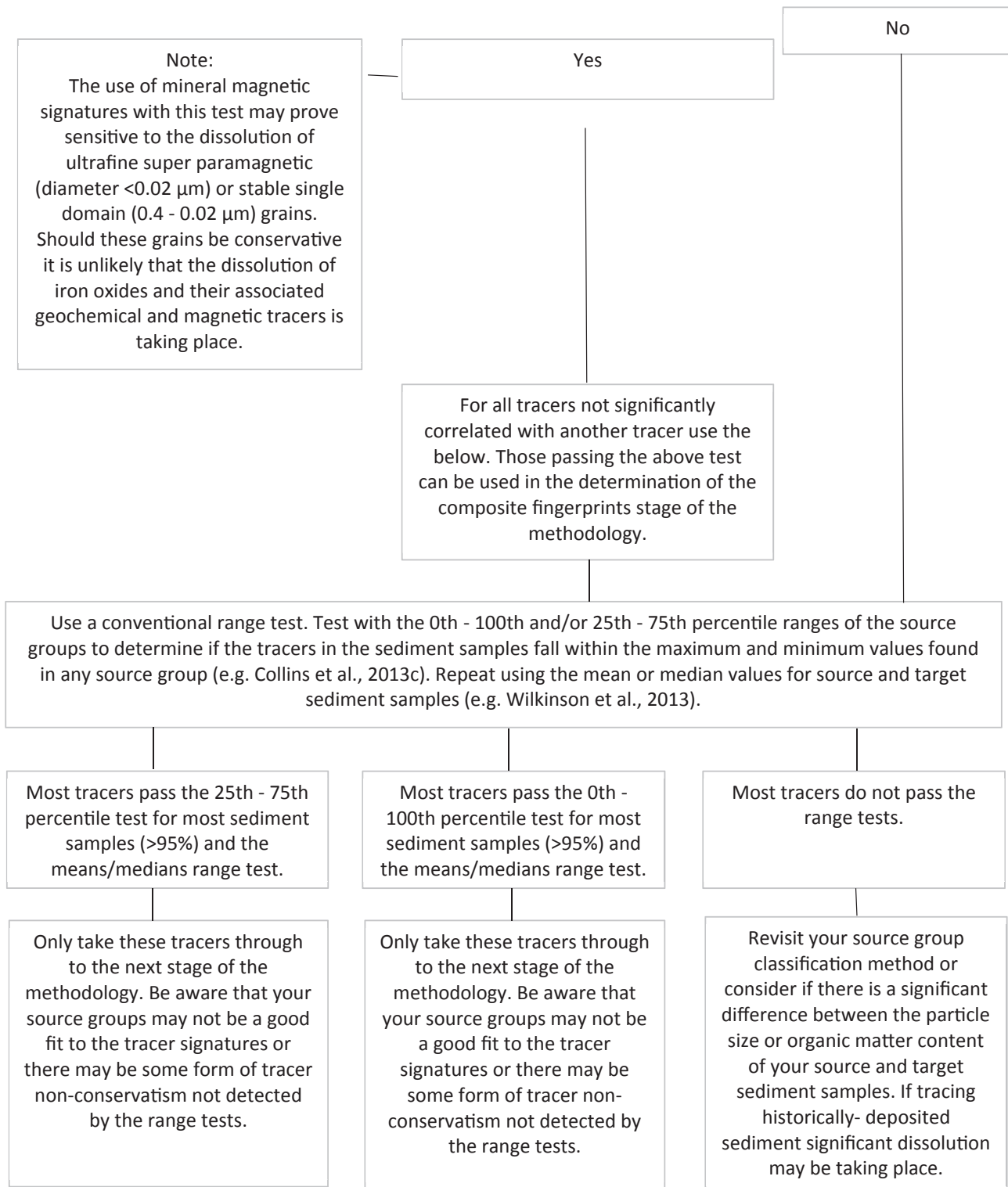


Fig. 1. (continued).

Source discrimination

Consider the use of a number of independent statistical tests to identify multiple composite signatures for discriminating the study catchment sediment sources. Consider the use of the Kruskal-Wallis H test, Principal Component Analysis, cluster analysis and linear discriminant analysis, amongst others.

Consider the use of a two-step process with step one testing the ability of each individual tracer to provide some degree of discrimination between your source sources using the independent tests such as a Kruskal Wallis H-test. Consider ranking the results from each test to select the most powerful individual tracers (e.g. Collins et al., 2012a).

Step two takes the ranked results from each independent test and passes them through Discriminant Function Analysis driven by a stepwise algorithm to finalise the optimum composite signature. Consider driving the discriminant function analysis using a genetic algorithm.

Different optimum composite fingerprints

As we have few ways to validate the outcomes of a fingerprinting study, the replication of the modelling using multiple composite fingerprints comprising different sets of tracers is an important part of the procedure.

Larger composite fingerprints of tracers have been shown to reduce uncertainty in modelling outputs tested by the use of artificial sediment mixtures. However, this must be balanced with the need to minimise mixing model errors represented by the difference between source-weighted and measured sediment tracer values since larger fingerprints will return greater errors using a goodness-of-fit based on absolute error.

Fig. 1. (continued).

Source apportionment

The choice of mixing model structure will influence the outputs generated and the appropriateness of different structures should be explored. Consider the combined use of frequentist and Bayesian approaches.

Models must include some form of Monte Carlo based uncertainty analysis to capture uncertainty in characterising the source and target sediment tracer values. Models must include the distributions of tracer signatures in both source groups and target sediment.

If sufficient composite samples (at least 20; Small et al., 2002) are collected per source group and/or for target sediment, use the distributions of the measured tracer values to construct pdfs for the apportionment modelling. If fewer composite samples are collected, Normality tests should be used to establish the most appropriate location (mean / median) and scale (standard deviation, median absolute deviation, Qn, Sn; Rousseeu and Croux, 1993) estimators for constructing the source and sediment tracer pdfs.

Using a model which maintains correlations between tracers in each source group can reduce the range of uncertainty in its outputs.

Run the un-mixing model, using an error threshold (e.g. disregarding iterations with an error >15%; Walling and Collins, 2000) to predict pdfs of source proportions. Use these to establish full uncertainty ranges. Test the reproducibility (convergence) of the model solutions by repeating the Monte Carlo analysis. Consider expressing uncertainty using relative frequency-weighted average mean or median source proportions (Collins et al., 2012a).

Estimate 95% confidence limits for these average means or medians.

Assess the goodness-of-fit between source-weighted and measured sediment tracers using a combination of absolute mean relative error (AMRE; Collins et al., 1997a) and mean relative error squared (MRES; Motha et al., 2003). Assess the relationship between these two estimators of goodness-of-fit for measured tracer values.

Divergence between the two estimators is possible, especially with larger composite signatures. Acceptable results using these goodness-of-fit tests still need to be interpreted alongside those under 'apportionment validation' using artificial mixtures.

Check the consistency of your source apportionment predictions using your different optimum composite signatures. Are the predictions based on each signature consistent?

To test the robustness of your model, put the source samples in as target samples and see how accurately they are ascribed.

Consider generating final source apportionment estimates by weighting the model results generated using each independent composite signature on the basis of a weighting combining the discriminatory efficiency of the signature and the associated AMRE.

Do the final source apportionment estimates make environmental sense for your study catchment?

Weightings may be included based upon within-source group variability or discriminatory power (Martinez-Carreras et al., 2008; Collins et al., 2010c) or tracer variability ratios (Pulley et al., 2015a). However, these may have a detrimental effect on model accuracy and should be tested using artificial mixtures of source groups before inclusion in the final methodology.

Apportionment validation

Prior to running the un-mixing model, methodological validation should be performed using the artificial mixing of known quantities of the sediment source groups. The mixtures should be used to validate the following:

Fig. 1. (continued).

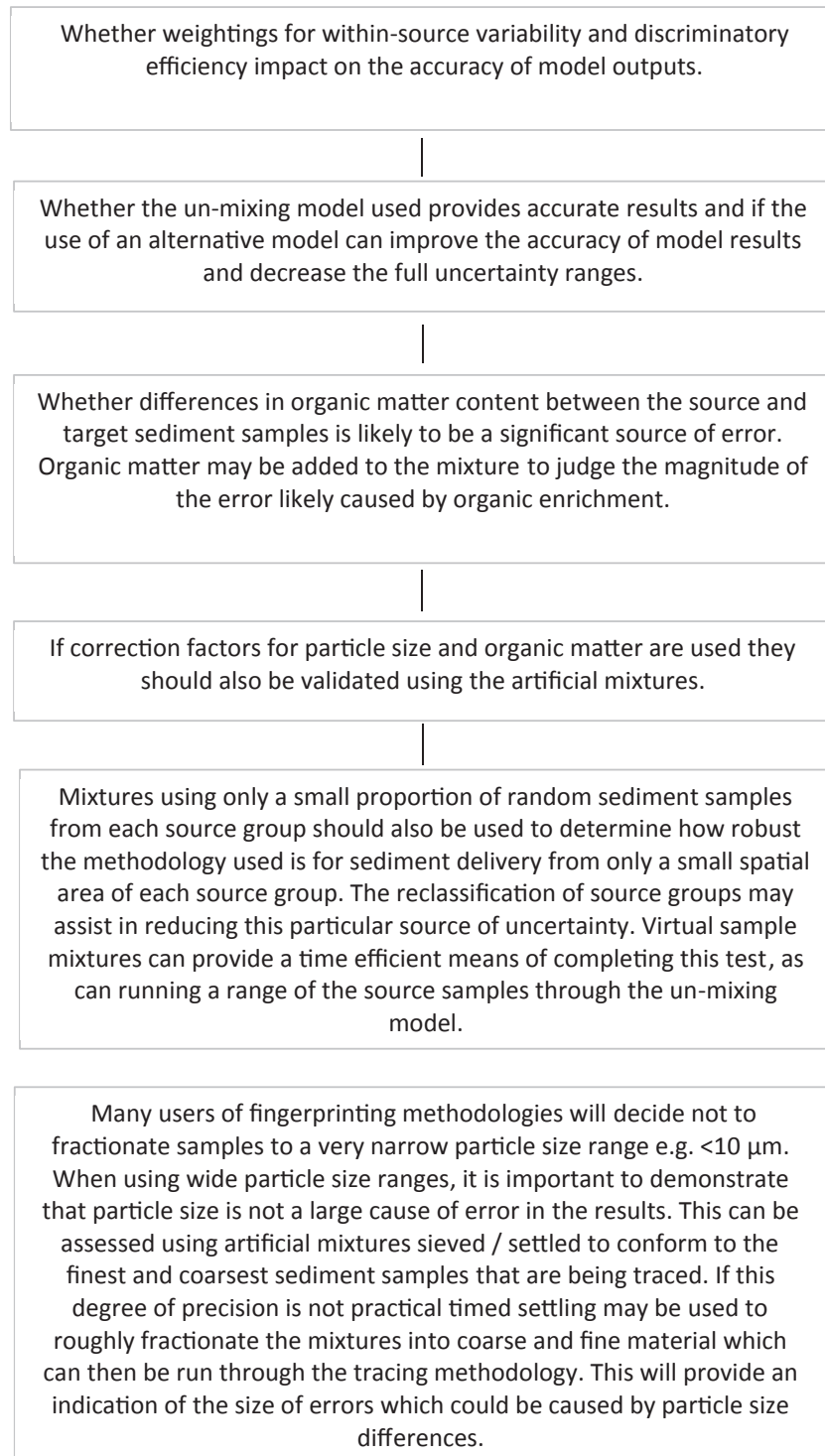


Fig. 1. (continued).

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