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Patrick Belmont

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Review

Sediment source fingerprinting as an aid to large-scale landscape conservation and restoration: A review for the Mississippi River Basin

Zhen Xu [a,*], Patrick Belmont [a], Janice Brahney [a], Allen C. Gellis [b]

[a] Department of Watershed Sciences, Utah State University, Logan, UT, 84322, USA
[b] U.S. Geological Survey, Maryland-Delaware-D.C. Water Science Center, Baltimore, MD, 21228, USA

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ABSTRACT

Reliable quantitative information on sediment sources to rivers is critical to mitigate contamination and target conservation and restoration actions. However, for large-scale river basins, determination of the relative importance of sediment sources is complicated by spatiotemporal variability in erosional processes and sediment sources, heterogeneity in sediment transport and deposition, and a paucity of sediment monitoring data. Sediment source fingerprinting is an increasingly adopted field-based technique that identifies the nature and relative source contribution of sediment transported in waterways. Notably, sediment source fingerprinting provides information that is independent of other field, modeling, or remotely sensed techniques. However, the diversity in sampling, analytical, and interpretive methods for sediment fingerprinting has been recognized as a problem in terms of developing standardized procedures for its application at the scale of large river basins. Accordingly, this review focuses on sediment source fingerprinting studies conducted within the Mississippi River Basin (MRB), summarizes unique information provided by sediment source fingerprinting that is distinct from traditional monitoring techniques, evaluates consistency and reliability of methodological approaches among MRB studies, and provides prospects for the use of sediment source fingerprinting as an aid to large-scale landscape conservation and restoration under current management frameworks. Most MRB studies reported credible fingerprinting results and found near-channel sources to be the dominant sediment sources in most cases, and yet a lack of standardization in procedural steps makes results difficult to compare. Findings from MRB studies demonstrated that sediment source fingerprinting is a highly valuable and reliable sediment source assessment approach to assist land and water resource management under current management frameworks, but efforts are needed to make this technique applicable in large-scale landscape conservation and restoration efforts. We summarize research needs and discuss sediment fingerprinting use for basin-scale management efforts with the aim of encouraging that this technique is robust and reliable as it moves forward.

1. Introduction

Excess sediment in fluvial systems has been linked to impairments for river ecosystems, such as alteration of water chemistry and temperature (Bilotta and Brazier, 2008), enhanced transfers of carbon, nutrients and contaminants (Prosser et al., 2001; Debnath et al., 2021; Stackpoole et al., 2021), changes in channel morphology (Donovan et al., 2016; Call et al., 2017), an accelerated infilling of water-supply reservoirs (Dargahi, 2012; Murphy et al., 2018), the smothering of biotic habitats (Richards and Bacon, 1994; Henley et al., 2000; Kemp et al., 2011), a reduction of primary production due to increased turbidity and restricted light penetration (Wood and Armitage, 1997; Schwartz et al., 2011; McKenzie et al., 2022), and restricted activity of organisms that use visual searching cues (Breitburg, 1988; Shoup and Wahl, 2009). To limit the off-site impacts of excessive sediment delivery, scale-appropriate knowledge of different sources supplying sediment to riverine, lacustrine, and coastal systems is required for efficient conservation and restoration activities. Apart from direct conservation planning, quantitative information on sediment sources is also an essential prerequisite for robust calibration and validation of process-based hydrological and erosion models at a watershed scale (Kumarasamy and Belmont, 2018; Hansen et al., 2021). However, there is often a knowledge gap in sediment contributions from different source types or erosion processes as they cannot be readily or effectively

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* Corresponding author.
E-mail address: zhenxuprc@hotmail.com (Z. Xu).

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measured at the watershed scale.

Traditional techniques measuring or inferring riverine suspended sediment are limited in their ability to recognize the spatial and temporal variation in distinguishable sediment sources. This is especially true for discerning the relative importance of upland and near-channel erosion which is essential to inform effective management and policy (Mukundan et al., 2012; Belmont et al., 2014; Gellis and Gorman Sanisaca, 2018). For the purposes of this paper the term "near-channel erosion" is used for processes that deliver sediments through fluvial erosion (e.g., streambank) and from channel/near-channel sediment storage (e.g., bed/floodplain sediment storage) as opposed to upland erosion that refers to removal of sediment from the vast and relatively flat upland terrestrial surface (e.g., cropland). Most traditional sediment sourcing techniques, such as field mapping and monitoring, provide a means of sediment source apportionment based on small-scale erosion rate estimates and complementary information on sediment routing and sediment yield (Collins and Walling, 2004), which suffer from inherent spatial and temporal sampling constraints, the amount of fieldwork involved, and uncertainties associated with extrapolation approaches. Further, results obtained by such approaches may be less reliable due to a lack of constraints on connectivity within the river network and uncertainties associated with sediment routing (Walling, 1983; de Vente et al., 2007; Fryirs, 2013), which remains a concern even though the resolution of temporal and spatial patterns of catchment erosion has been improved by recent technological advances in field surveying, remote sensing, and photogrammetry (Collins and Walling, 2004). The use of watershed hydro-erosion models is often another alternative, which have advanced rapidly in process representation and sophistication in the past few decades (Singh and Frevert, 2010). However, only a few of the models can simulate streambank erosion (e.g., Darby et al., 2002; Janes et al., 2018), and such models often include numerous parameters that are hard to calibrate and are vulnerable to problems of equifinality (Kamali et al., 2017; Kouchi et al., 2017; Abbaspour et al., 2018; Kumarasamy and Belmont, 2018). Thus, while traditional approaches may provide useful context and information to understand watershed sediment dynamics, multiple independent lines of information are required to develop an understanding that is sufficiently reliable to serve as the basis for large-scale conservation and restoration planning.

Sediment source fingerprinting provides information about the relative importance of sediment sources in a way that is independent of other field or remotely sensed techniques. By only dealing with the geochemical and physical properties of target samples (e.g., suspended sediment, channel bed material sediment, lakebed sediment, or floodplain sediment) that are delivered to a given point at a watershed, sediment fingerprinting circumvents the sediment delivery problem that has plagued traditional sediment source identification techniques. Although this technique often does not directly inform people of sediment yield or distinguish a specific source location, when combined with water and sediment gaging information it could provide a quantitative constraint on sediment loading from various sources throughout the watershed. Sediment fingerprinting was initially developed and applied in agricultural catchments; nowadays this approach has been demonstrated in a variety of landscapes to be a powerful tool to inform watershed restoration projects and ensure that resources are used in ways and locations where they will be most effective (Caitcheon et al., 2007; Belmont et al., 2011; Hartranft et al., 2011; Smith and Blake, 2014; Cashman et al., 2018). Up to now most sediment source fingerprint techniques have been conducted in an *ad hoc* manner with a lack of standardization, which undermines the credibility of the fingerprinting approach in the longer term (Laceby et al., 2017; Collins et al., 2020; Owens, 2022). Considerable advances have been made in recent years outlining key methodological steps of sediment source fingerprinting, the application of which typically includes sediment source classification, source and target sample collection, tracer selection and analysis, corrections for non-conservative behavior of tracers, and source apportionment modeling (Collins et al., 2017).

Sediment source fingerprinting investigations have expanded greatly in recent decades both in number and scope, but sampling, analytical, and interpretive methods have varied considerably (Owens, 2022). Specifically, a vast range of fingerprint properties (commonly referred to as tracers) have been used by different studies, such as radionuclides (Evrard et al., 2020), elemental geochemistry (Raigani et al., 2019), bulk isotopes like stable $\delta^{13}C$, $\delta^{15}N$ (Ford et al., 2020) and radiogenic $^{87}Sr/^{86}Sr$, $^{144}Nd/^{143}Nd$ (Munoz et al., 2019), compound-specific stable isotopes (Reiffarth et al., 2016; Alewell et al., 2016), mineralogy (Nukazawa et al., 2021), magnetic susceptibility (Hatfield and Maher, 2008), and spectrocolorimetrics (Barthod et al., 2015). Site- or study-specific differences in the tracers used for sediment fingerprinting may be considered a benefit in the sediment fingerprinting approach, demonstrating that the approach can be adapted to a wide variety of local settings and research questions. Alternatively, inconsistency in fingerprinting properties used in different locations and studies could be seen as a problem in terms of developing standardized procedures for large-scale application of sediment source fingerprinting.

Smaller scale sediment fingerprinting studies have demonstrated success in identifying sediment sources at the watershed scale in order to inform management actions (e.g., Walling and Collins, 2008). However, some big questions remain: what are the best practices for the use of sediment source fingerprinting at large-scale landscapes? How good is its performance in current management frameworks? Is this technique ready to be scaled up and used in a more prominent way in conservation, restoration, and management efforts? In the context of the above, we explored means to use sediment fingerprinting to inform conservation and land management at the scale of large river basins (10^4–10^5 km^2). Specifically, we searched and reviewed established sediment source fingerprinting studies conducted within the Mississippi River Basin (MRB) to summarize what we have learned from sediment source fingerprinting. Goals of this review are to: 1) discern what information sediment source fingerprinting can provide that is distinct from other sediment source analyses, 2) identify methodological differences among studies and assess the corresponding consistency and reliability of different methods, and 3) provide prospects for the use of sediment source fingerprinting techniques as an aid to large-scale landscape conservation and restoration.

As the world's fourth largest river basin, the Mississippi River drains land from part or all of 31 U.S. states (3.2×10^6 km^2, 41% of the continental United States) and two Canadian provinces. The basin is comprised of six tributary basins including Upper Mississippi, Missouri, Ohio, Tennessee, the Arkansas-White-Red and Lower Mississippi (Fig. 1). Land-use in the MRB is highly heterogeneous but is dominated by agriculture located in the Missouri, Upper Mississippi, and Ohio River basins (Hassan et al., 2017), which makes the basin a potential hotspot for the application of sediment source fingerprinting techniques. Besides, many catchments within the MRB suffer from excess sediment-derived environmental problems; for example, extensive land use change since the mid-1800s has introduced high suspended sediment loads to the Upper MRB and its major tributaries (David et al., 2010), which are listed as impaired for turbidity by the U.S. Environmental Protection Agency (EPA). The investment required to reduce sediment loading and other non-point-source water quality problems is enormous, and effective use of funds dedicated to reducing sediment pollution requires accurate identification of sources and mechanisms of sediment supply. This issue is controversial, especially in determining the role of upland agricultural practice versus near-channel natural erosion (Belmont et al., 2011; Belmont and Foufoula-Georgiou, 2017). Sediment source fingerprinting techniques may be beneficial in this regard. It is worth noting that although this review focuses on the fingerprinting studies conducted within the MRB, generalized methodological insights from studies conducted by the rest of the global research community are also included.

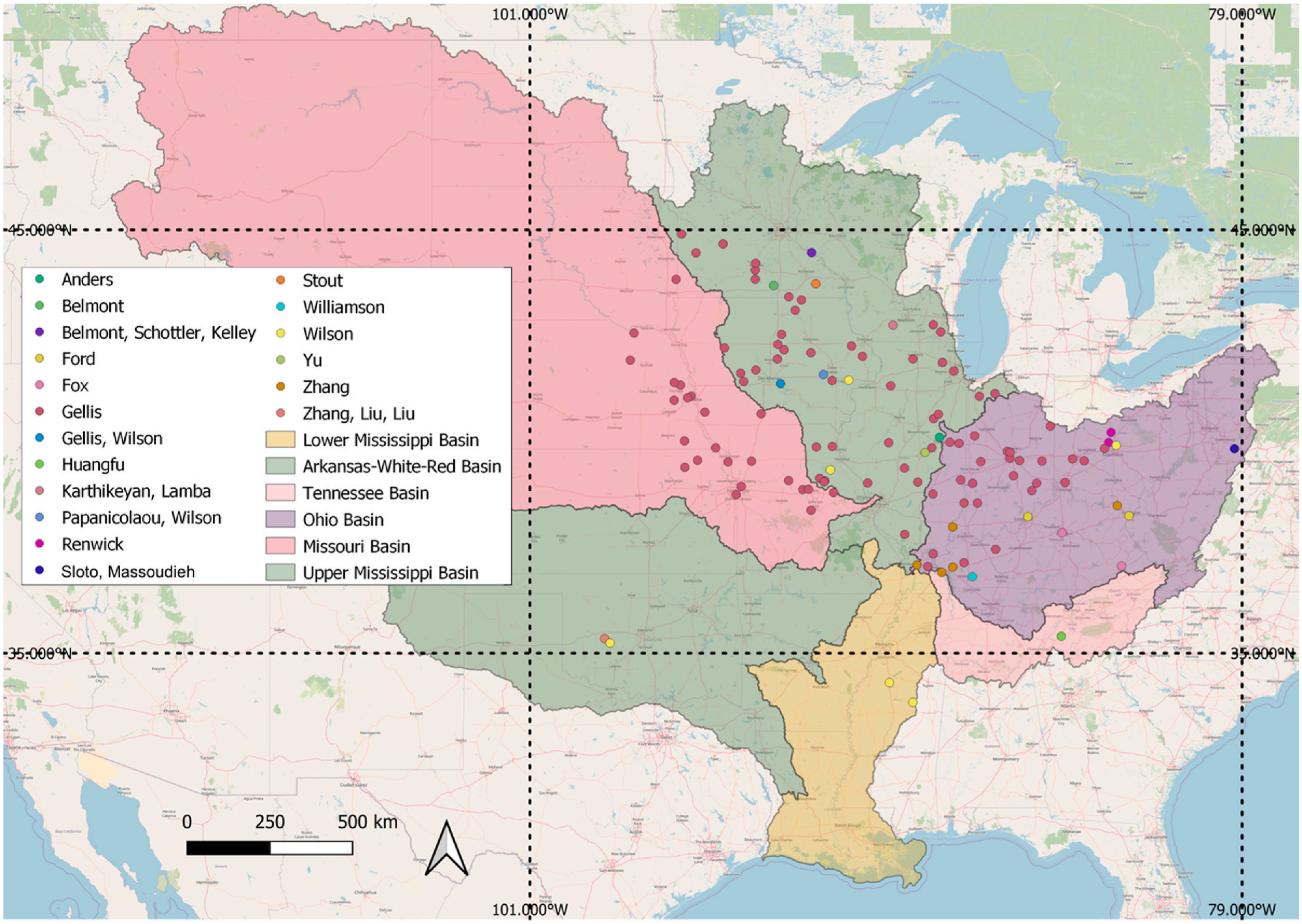

Fig. 1. Established sediment source fingerprinting studies conducted within the Mississippi River Basin (n = 46). Circles in different colors represent watersheds of interest in different publications. Last names of the corresponding authors are indicated next to circles. For a watershed investigated by multiple studies, last names of the corresponding authors from all studies are listed. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

2. Sediment source fingerprinting investigations in the MRB

Sediment source or provenance can be defined in different ways such as by land use or geologic provenance (Klages and Hsieh, 1975; Wall and Wilding, 1976; Walling, 2013). In this study we specifically investigate research attempting to discern sediment sources according to geographic regions (geographic sediment source fingerprinting) and source types (geomorphic sediment source fingerprinting). Geographic sediment source fingerprinting seeks to determine the relative importance of different geographic locations, as characterized by different rock types (e.g., by geology). The usefulness of this approach relies on the assumption that the landscape is naturally parsed into geochemically distinct units that align with relevant policy or management boundaries (e.g., by tributary, Zhang et al., 2012). Geomorphic sediment source fingerprinting mainly seeks to identify which landforms or land uses contribute sediment that passes a given location. Thus, geomorphic fingerprinting necessarily involves consideration of the processes responsible for sediment mobilization and deposition (e.g., Shi et al., 2021), which may be more useful for management, conservation, and restoration purposes. Combined information from both geographic and geomorphic sediment source fingerprinting could shed light on the fraction of sediment derived from each source group and from different geographic units (e.g., sub-basins) of a large watershed, which would be promising for the application of sediment source fingerprinting in a regulatory framework. Our literature search using Web of Science, Google Scholar and 32 search terms (Fig. S1) found 4 geographic and 42 geomorphic sediment source fingerprinting studies conducted within the MRB by research universities (81%), private organizations (4%), state (2%), and federal agencies (13%) (Fig. 1, Table S1). We excluded eight working papers that were later integrated into peer reviewed publications. A detailed method section for literature search as well as full lists of all search terms and reviewed publications are available in the supplementary material.

All reviewed studies were conducted since 2000, and the number of publications shows a rapid growth for the past two decades with most studies conducted during 2011–2015 (Fig. 2a). The near-exponential increasing trend is consistent with the upward trend for the global research community since the early 2000s (Walling, 2013; Collins et al., 2020). Most published studies targeted sites in Upper MRB (52.7%), followed by Ohio River Basin (26.3%), Arkansas-White-Red River Basins (10.5%) and Tennessee River Basin (2.6%). Several studies (7.9%) targeted sites across multiple tributaries (Fig. 2b, Table S1). Closer examination of reviewed publications reveals that most studies applied sediment source fingerprinting techniques to sites located in the Midwestern United States (Fig. 1), which is one of the most intensive and economically important agricultural regions of the country (Gellis et al., 2017). Specifically, if categorized using the U.S. Geological Survey (USGS) hydrologic unit code, most sites of interest are in the Upper MRB (44.3%), Ohio River Basin (32.8%) and Missouri River Basin (18.9%), with only 4.0% of studied catchments in Arkansas-White-Red River

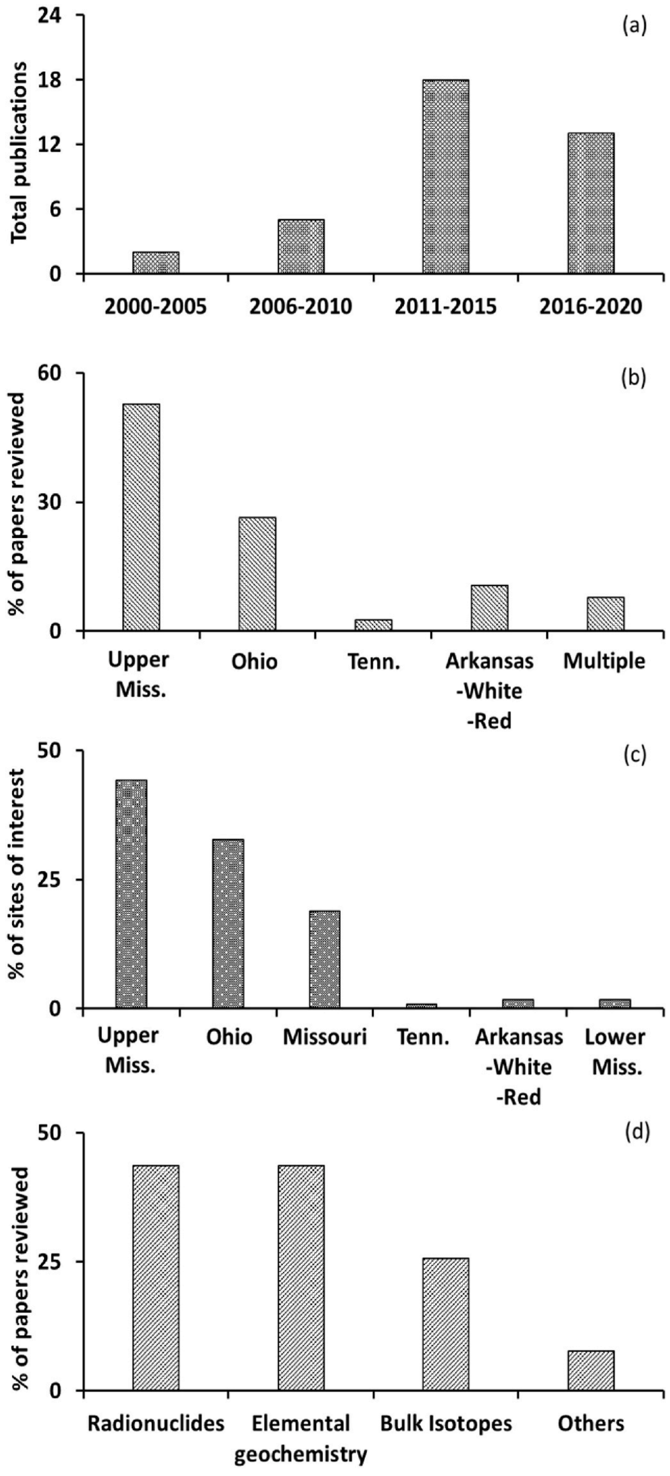

Fig. 2. (a) Number of published sediment source fingerprinting studies conducted within the Mississippi River Basin (MRB) during 2000–2020. (b) Percentage of studies targeting each tributary basin. (c) Percentage of individual site locations from reviewed studies in each tributary basin. (d) Percentage of composite signatures used in published MRB studies. "Miss." and "Tenn." are abbreviations for Mississippi and Tennessee, respectively. The sum of percentages indicated by the different bars in Fig. 2d would exceed 100% as one study could use more than one type of composite signature. A full list of reviewed studies is available in the supplementary material.

basins, Lower MRB and Tennessee River basin (Fig. 2c). The spatial pattern of sites of interest from established studies is correlated with the spatial sediment yield pattern estimated from hydrometric data (Hassan et al., 2017). Specifically, locations of most sites of interest overlap with regions of high sediment yield such as the valleys of lower Missouri, while fingerprinting studies were rarely conducted in the headwaters of the Missouri or Ohio Rivers, which constitute about half of both basins but have very low sediment yield (Fig. 1). The Lower MRB is an exception with a very high sediment yield but very limited sediment sourcing investigations. For the use of different composite signatures, the application of fallout radionuclides and elemental geochemical signatures dominate, followed by the use of bulk isotopes and other tracers such as C/N ratio (Fig. 2d, Table S1). The prevalence of utilized tracer signatures in the MRB is comparable to that used in the global research community during 2013–2019 (Collins et al., 2020).

3. Advantages of utilizing sediment source fingerprinting techniques

Based on studies conducted within MRB, one unique advantage of sediment source fingerprinting is its ability to determine the contribution of near-channel sediment sources to the total sediment load in a watershed. Specifically, 35 out of 38 reviewed publications are geomorphic fingerprinting studies, 33 of which specifically focus on distinguishing the relative importance of upland and near-channel sources for suspended sediment or fine sediment deposits (Table 1 and S1). Most of them (58%, n = 19, Table 1) found near-channel sources to be the primary sources for fine-sediment samples, whereas 42% of them suggested upland sources to be the dominant sources. Seventeen studies (Table S1) considered sediment sourcing dynamics over various timescales, which include a multi-season sampling strategy or involved sample collection across the entire hydrograph. Most of them (76%, n = 13, Table S1) reported significant temporal sediment source shifts driven by various factors. However, 74% of geomorphic fingerprinting studies (n = 26, Table 1) stopped at the stage in which the relative importance of source types is determined by the un-mixing model and did not combine fingerprinting results with other information either for load-weighted relative contributions from source types or for a sediment budget that shows a mass balance between inputs and outputs. Such information is critical to assist land managers especially for large river basins, as fingerprinting results alone do not say much about the magnitude of overall sediment yields (Lamba et al., 2015a; Wilson et al., 2014a) and cannot target specific locations of concern (Gellis and Walling, 2011).

A much-improved sediment yield or budget estimate is another advantage of utilizing sediment source fingerprinting techniques, which can be achieved by coupling fingerprinting results with other lines of evidence such as soil erosion and sediment yield measurement or modeling. One example is the work conducted in the Upper MRB by Belmont et al. (2011), which combined sediment source fingerprinting with sediment gaging data and remote sensing techniques to inform shifts of dominant sediment sources at scales of small watershed (10^2 km^2), large watershed (10^3 km^2) and large river basins (10^4 km^2). As for modeling, Fox and Martin (2015) showed the capability of using the sediment fingerprinting method for calibration of an erosion and sediment transport model. In their study investigating sediment yield from reclaimed surface mining sites located in the Appalachian region of Kentucky, the authors provided the proof-of-concept by using the results from sediment fingerprinting to calibrate the transport capacity coefficient, the sediment delivery ratio from reclaimed mining soils, and the eroding stream bank parameters (Fox and Martin, 2015). In addition, knowledge of sediment dynamics evidenced by sediment source fingerprinting can also be used to improve event-based watershed erosion models, which could be especially useful to quantify the short-term impacts of high-magnitude events (Wilson et al., 2014b). Adoption of watershed management approaches that combine sediment

Table 1

Summary of Mississippi River Basin (MRB) geomorphic fingerprinting studies that investigate the relative importance of upland and near-channel sources. Details of how upland and near-channel sources are defined and estimated for each study as well as additional notes for presented values can be found in the supplementary material.

Reference	Site of interest	Drainage area (km^2)	Tracer properties	Apportionment results[a]	
				Upland	Near-channel
Abban et al. (2016)	South Amana Watershed	26	$\delta^{13}C$, $\delta^{15}N$	55.9%	44.1%
Abban et al. (2014)	South Amana Watershed	26	$\delta^{13}C$, $\delta^{15}N$	39.0%	61.0%
Belmont et al. (2014)	Maple River Watershed	2880.7	^{10}Be,$^{210}Pb_{ex}$,^{137}Cs	54.6%	45.4%
Belmont et al. (2011) [b]	Le Sueur River/Greater Lake Pepin Watershed	335–51500	^{10}Be,$^{210}Pb_{ex}$,^{137}Cs	26.1%	73.9%
Crain et al. (2017)	Little River Basin	150–1556	$\delta^{13}C$, C, Ca	33.3%	66.7%
Fox and Martin (2015)	Island Branch and Whitaker Branch watersheds	2.23-3.53	$\delta^{13}C$, $\delta^{15}N$, TOC, C/N ratio	35.5%	64.5%
Fox (2009)	Island Branch and Whitaker Branch watersheds	2.23-3.53	$\delta^{13}C$, $\delta^{15}N$	61.5%	38.5%
Gellis et al. (2019) [b]	Walnut Creek Watershed	52.6	TOC and 24 inorganic elements	63.0%	37.0%
Gellis et al. (2017)	91 MRB watersheds (out of 99 studied watersheds)	3–6348	^{7}Be,$^{210}Pb_{ex}$	38.0% for SS 29.0% for B	62.0% for SS 71.0% for B
Gillespie (2008)	O'Shaughnessy and Delaware Lake Reservoirs	1000–2536	^{137}Cs	49.0% for B	51.0% for B
Huangfu et al. (2020)	Oostanaula Creek Watershed	181.56	Co, Cr, P, Si, Ti	37.0%	63.0%
Huisman et al. (2013)	North Fork of Pheasant Branch Watershed	12.4	^{7}Be,$^{210}Pb_{ex}$,^{137}Cs	76.4%	23.6%
Huisman and Karthikeyan (2012)	North Fork of Pheasant Branch Watershed	12.4	^{7}Be,$^{210}Pb_{ex}$,^{137}Cs	79.5%	20.5%
Lamba et al. (2019)	Pleasant Valley Watershed	50	$^{210}Pb_{ex}$,^{137}Cs	23.1%	76.9%
Lamba et al. (2015a) [b]	Pleasant Valley Watershed	50	12 inorganic elements	68.0%	32.0%
Lamba et al. (2015c)	Pleasant Valley Watershed	50	^{7}Be,$^{210}Pb_{ex}$	76.7%	23.3%
Lamba et al. (2015b)	Pleasant Valley Watershed	50	16 inorganic elements	49.4% for B	50.6% for B
Liu et al. (2016)	Bull Creek Watershed	15.6	Br, Sr, As, Mn, Zr, Mg, Ca, Ba, Hf, C, Mo	57.0%	43.0%
Mahoney et al. (2019) [b]	South Elkhorn Watershed	61.8	$\delta^{13}C$, $\delta^{15}N$, TOC, C/N ratio	72.4%	27.6%
Neal and Anders (2015)	Wildcat Slough Watershed	61.4	$\delta^{13}C$, P, Mg, Mn, C	47.5%	52.5%
Schottler et al. (2010) [b]	Greater Lake Pepin Watershed	51500	^{10}Be,$^{210}Pb_{ex}$,^{137}Cs	30.5%	69.5%
Sloto et al. (2012)	Laurel Hill Creek Watershed	323.7	Bi, Nb, Y, P, Fe, Ti, Pb, Cu, Th, Cs, K	53.0%	47.0%
Stewart et al. (2015)	Laurel Hill Creek Watershed	323.7	$\delta^{13}C$, $\delta^{15}N$ and 25 inorganic elements	55.2%	44.8%
Stout et al. (2014)	Root River Watershed	4300	^{10}Be,$^{210}Pb_{ex}$,^{137}Cs	45.0%	55.0%
Williamson et al. (2014)	West Fork Beaver Creek Basin	253.9	In, P, C, Be, Tl, Th, Ti	15.6% for SS 50.7% for B	84.4% for SS 49.3% for B
Wilson et al. (2008) [b]	4 MRB watersheds (out of 5 CEAP watersheds)	51–780	^{7}Be,$^{210}Pb_{ex}$	32.4%	67.6%
Wilson et al. (2014a) [b]	7 MRB watersheds (out of 8 CEAP watersheds)	51–6417	^{7}Be,$^{210}Pb_{ex}$	35.6%	64.4%
Wilson et al. (2014b) [b]	South Amana Watershed	26	^{7}Be,$^{210}Pb_{ex}$,^{137}Cs	42.9%	57.1%
Wilson et al. (2012) [b]	South Amana Watershed	26	^{7}Be,$^{210}Pb_{ex}$	40.3%	59.7%
Yu and Rhoads (2018)	Upper Sangamon River	84.3	Ca, Sc, Be, S	17.0%	83.0%
Zhang and Liu (2016)	Bull Creek Watershed	15.6	19 inorganic elements	57.0%	43.0%
Zhang et al. (2016a)	Bull Creek Watershed	15.6	^{137}Cs and 19 inorganic elements	43.0%	57.0%
Zhang et al. (2016b)	Bull Creek Watershed	15.6	^{137}Cs	69.0%	31.0%

[a] Target sample is fluvial suspended sediment (SS) unless otherwise noted. Other target sample include bed sample (B).

[b] Denotes studies combine fingerprinting results with other lines of evidence for load-weighted relative contributions from source types or for a sediment budget that shows a mass balance between inputs and outputs.

source fingerprinting with budgeting or modeling efforts could lead to a better understanding of sediment source dynamics for developing and evaluating management strategies such as BMPs (best management practices), TMDLs (total maximum daily loads) and soil conservation strategies.

Sediment source fingerprinting techniques have also shown potential to improve our understanding of other sediment dynamics for effective land management. For example, Ford et al. (2020) used sediment fingerprinting results to provide evidence of sediment dynamics controlling deposition patterns in backwater confluences serving wetland and boat marina functions of the Ohio River. With the derived information of sediment transport efficiency and exchange processes of wetland and marina functions, the authors further quantified the drainage areas impacted by wetland and marina features for the Ohio River backwater confluences and discussed potential ecological and economic river management functions of both river features in the context of sediment transport and deposition (Ford et al., 2020). Mahoney et al. (2019) coupled sediment fingerprinting and watershed modeling methods to elucidate the role of the equilibrium sediment exchange process. Results from their study suggested that the equilibrium sediment exchange is a substantial but often neglected process, which should be considered when the sediment continuum is used to investigate the critical zone. Additionally, Fox (2009) used sediment fingerprinting in areas affected by coal mining in the southern Appalachian Mountains to determine that it takes longer for the sediment transport processes governing streambank erosion loads to reach geomorphologic equilibrium in watersheds after disturbance as compared to the surface erosion processes. Such information could be especially useful for managing watersheds with past disturbances such as reclaimed mines and retired agricultural land.

4. Consistency and reliability of methods

One outcome of the rapidly growing research efforts in sediment source fingerprinting is an expansion of methodological diversity driven by scientific curiosity and site-specific challenges due to natural and anthropogenic factors. Such inconsistency in methodological steps may raise doubts and uncertainties among land managers and policy makers in terms of applying sediment source fingerprinting techniques as strategic management aids. There have been trials to develop flow charts and decision trees summarizing critical steps needed to apply the fingerprinting approach (Collins and Walling, 2004; Collins et al., 2017). In this section we review methodological steps in MRB fingerprinting studies with reference to the latest decision tree presented by Collins et al. (2017) as well as recent advances in fingerprinting techniques made from the global research community. We also assess the consistency and reliability of methods and provide recommendations for the best fingerprinting practices for management purposes.

4.1. *Sediment source classification*

Proper classification of potential sediment sources within the studied area is the foundation for successful sediment sourcing, but it is probably the least thoroughly explored stage of the sediment fingerprinting approach (Pulley et al., 2017a). Previous studies have demonstrated that sediment source fingerprinting results can be strongly influenced by the specific source classification considered, driven by the effect of source discrimination and importance of source groups (Vercruysse and Grabowski, 2018). In the context of delivering useful results to land managers and policy makers, *a priori* sediment source group classification based on land use is by necessity more frequent as compared to approaches with catchment geology or soil types (Haddadchi et al., 2013). Specifically, sediment source classification is performed *a priori* for all fingerprinting studies within the MRB to align source apportionment estimates with land use or geographic unit. While being widely used, the *a priori* sediment source grouping approach may suffer from within-source group tracer variability as tracer concentrations under a given land use or overlying a specific geographic unit could vary spatially due to various factors including soil type and drainage (Blundell et al., 2009), anthropogenic pollutants (de Miguel et al., 2005), management practices (McDowell et al., 2016; Upadhayay et al., 2020a), and erosion intensity (Wilkinson et al., 2015), most of which cannot be easily corrected by statistical approaches or readily available datasets describing the geology (Laceby et al., 2015). Unawareness of such issues may lead to a scenario where classification into sediment source groups makes sense from a management standpoint but makes little sense in terms of tracer suitability. As a result, it would be beneficial to assess the suitability of potential source groups as an additional methodological step to determine if groups fit the tracers use, which is often termed as objective sediment source grouping in the literature (Pulley et al., 2017a, 2017b).

Objective sediment source groupings are under-investigated for studies within the MRB as well as the global research community (Collins et al., 2020) but have the potential to statistically improve discrimination among sediment sources and reduce uncertainty caused by within-source group tracer variability (Pulley et al., 2017b). The commonly used cluster-analysis-based method uses natural variability in tracer concentrations to define sediment source groupings (Walling et al., 1993; Walling and Woodward, 1995), and recent advances in this technique have been made to retain the naturally present cluster groups while accommodating the catchment management goal of discriminating between surface and subsurface sources (Pulley et al., 2017b). Alternatively, Pulley et al. (2017a) presented a simple tracing approach that compares the concentrations of all tracers measured for each source sample and a single target sediment sample. The simple approach provides qualitative results supplementing existing quantitative tracing methods and can act as a check to determine if the sediment source groups are likely to be representative of the actual catchment sources and as a way to refine sediment source area identification. These objective sediment source grouping approaches may not be used as a replacement for *a priori* sediment source group classification, as the conventional way is advantageous in defining management-oriented source groups. However, they could be applied as a supplement for optimal sediment source groupings. It is worth noting that the use of objective sediment source grouping techniques may lead to a greater spatial resolution of sediment provenance, which is not always beneficial to target remediation (Pulley et al., 2017b). Source groups heavily fragmented into small areas scattered around sites of interest would be difficult to interpret and likely be of little use for targeting management.

Agricultural land is the upland source that received most attention for geomorphic sediment source studies (77%, n = 27, Table S1) undertaken within the MRB, which makes sense as the land condition in areas of most sites of interest is either arable-dominant or mixed-arable (Hassan et al., 2017). The other commonly investigated upland source in MRB studies is pasture (29%, n = 10, Table S1). In contrast, some other human-derived sediment sources are under-explored. For instance, roads were not considered as an independent sediment source in most studies (86%, n = 30, Table S1), but they are worthy of attention especially for the case that a gravel road network exists within another studied upland source such as cropland. In a study investigating stream sediment sources in Midwest agricultural basins, Williamson et al. (2014) found the presence of a signature of sediment from roads in each of the suspended sediment samples (1–56%) using composite signatures of In, P, C, Be, Tl, Th, and Ti. If the un-mixing model was adjusted with road sources omitted, a significantly larger ($p < 0.001$) proportion of the sediment was identified as coming from a stream-bank source. The authors then suggested that roads should be included in any analyses of sediment sources especially in midwestern U.S. agricultural areas (Williamson et al., 2014). In contrast, Gellis et al. (2019) found unpaved road contributions in the agricultural Walnut Creek, Iowa watershed to be a small percent of the sediment load (<5%) compared to streambanks (27–36%) and cropland (48–62%). Additionally, in another investigation of 99 largely agricultural watersheds in the midwestern United States, Gellis et al. (2017) determined the age of fluvial sediment using the radionuclide $^{210}\mathrm{Pb}_{\mathrm{ex}}$ and $^{7}\mathrm{Be}$ and found that the surface-derived portion of sediment (top soil and street residue) had the youngest ages (<100 days), suggesting that management actions to reduce erosion in these areas may have short-term noticeable effects on sediment loadings.

As for near-channel sources, 89% of MRB geomorphic fingerprinting studies (n = 31, Table S1) investigated removal of sediment through processes of fluvial erosion including incision and undercutting of bluffs, banks, and ravines. However, far less attention was paid to other near-channel sediment sources such as variable amounts of storage between the source area and the suspended sediment sampling point of interest. Large watersheds typically contain opportunities for short- or long-term storage within the channel and floodplain. Channel-floodplain exchanges could alter geochemical properties of samples, cause erroneous interpretations for source apportionment and lead to incorrect judgements of conservation strategies (Belmont et al., 2014). An evaluation of sediment collected downstream of floodplain features needs to consider that some of the sediment is being derived from floodplains, the fingerprint properties of which may be different from those associated with the original sources depending on environmental conditions and storage times (Koiter et al., 2013b). However, only a few MRB studies (9%, n = 3, Table S1) isolated floodplain and bank sources, and most studies lack a clear consideration for channel-floodplain sediment exchange. In a study discerning how channel-floodplain processes may modify the sediment fingerprinting signature of floodplain and bank sources, Belmont et al. (2014) demonstrated the possibility for over-estimating channel source contributions when there is a moderate amount of sediment exchange between the channel and floodplain in large watersheds over sediment routing timescales. Such information is important for determination and implementation of landscape conservation strategies. Another example is in-channel sediment storage, the volume of which can be an important component of the total sediment budget, especially in streams with accumulations of large woody debris and beaver dams (Gellis et al., 2019). The in-channel fine sediment could create a "legacy" effect since sediment resuspension in stream channels plays an important role for large storm events, which leads to a "lag-time" between management practice implementation and achievement of desired water quality goals (Huisman et al., 2013; Lamba et al., 2015c). It was rarely sampled either as an independent source (14%, n = 5) or as a target sediment (11%, n = 4, Table S1) for MRB studies, probably due to concerns associated with particle sorting and organic matter enrichment. However, it is worth more attention in management cases with significant deposition of fine sediment on the stream bed (Lamba et al., 2015b).

4.2. Source and target sampling

Most MRB studies (61%, n = 23, Table S1) took a single intensive campaign for catchment source sampling with pre-knowledge from preliminary field assessment (e.g., Stout et al., 2014), which is often considered adequate as long as utilized tracers are conservative over the investigation timescale (Collins et al., 2017) or have predictable non-conservative behavior (Belmont et al., 2014). One concern for the single campaign sampling strategy is whether the one-time collected samples are representative for within-source group tracer variability, which is especially critical for samples from large areas of land. As mentioned above, this could be a problem as there are differences in tracer signatures related to different upland soil properties and management practices, a lack of consideration in which can greatly undermine the reliability of results. A randomized sampling approach, such as the randomized design which rasterizes each source area, assigns a unique value to each cell, and randomly selects grid cells within each source for sampling (Gellis et al., 2019), can avoid bias in site selection within a source and provide a random sample that is representative of each source. The random sampling design does not seem to be widely applied (13%, n = 5, Table S1) as most studies do not provide details for sample site selection. It can be used as an alternative to field-assessment-based sampling design for site selection. Furthermore, as the study scale increases from plot to river basin, it may be necessary to choose a sampling scheme stratified by major soil and land use types (e.g., Zhang and Liu, 2016), which could help constrain within-source group tracer variability at large-scale landscapes.

Another concern for source sample collection is whether a sufficient number of samples are collected for good representativeness and statistical robustness. There has been a lack of deep investigation for this matter in fingerprinting studies, and relevant techniques to address this issue, such as the probability-based sample number determination (e.g., Collins et al., 2001), was never considered in any MRB study and rarely adopted in the rest of the global fingerprinting community (Collins et al., 2017). The use of probability sampling designs would most likely lead to a condition that many more samples need to be collected than is currently normal practice. The collection of greater sample numbers per source can be seen as desirable, particularly with increasing catchment size and diversity of land use or geology, which are likely to enhance the heterogeneity of physical and chemical properties of source materials (Smith et al., 2015). Extensive sampling may also allow for a better fit of source group classification to tracer properties. Previous studies evaluating sediment fingerprinting by artificial sample mixtures have reported that the accuracy and precision of source apportionment results could be considerably improved with greater replication of sediment sampling (e.g., Smith et al., 2018). However, the probability sampling designs would assume a regular distribution (normal or Student's *t*) of tracer concentrations in the groups, which may not be the case in many real-world applications. Also, more samples being collected would require higher budgets for analysis, and greater justification is needed to determine whether it is worth it or not for management. In practice, other alternatives such as the bulking approach, which includes the collection of sub-samples around one sampling point and bulking them into a composite, are popular (e.g., Zhang and Liu, 2016), but the statistical implication for such practice is still under-explored (Collins et al., 2017).

As for target sampling, suspended sediment, either instantaneous or time-integrated (37% and 66%, respectively, Table S1) was the main target sample for most MRB studies. There is a one-time sampling approach to collect fine channel-bed material under low-flow conditions as a surrogate for the collection of suspended sediment to identify relative contributions from different sources within a watershed in the literature (e.g., Collins and Walling, 2007). However, according to the findings of MRB studies a disagreement exists over whether this approach could provide a similar result to the more complicated and costly sampling of suspended sediment. For example, when using both suspended and channel-bed sediment as target samples, Williamson et al. (2014) reported significant differences in the relative contribution of at least one of the land-cover sources (cropland, retired land, stream banks, and roads) for their sites at the Minnesota River Basin. The authors attributed the disparity to the fact that channel bed integrates sediment sources over unknown, longer time periods and can store sediment that can be remobilized during later events (Williamson et al., 2014). Gellis et al. (2017) found that suspended sediment was younger in age than bed sediment and suggested that bed sediment likely contains a higher proportion of older, previously deposited sediment than does suspended sediment. In contrast, Fox and Martin (2015) found no significant difference in tracer signatures between transported fine sediments and fine sediments collected from the streambed and attributed that to geomorphic characteristics of studied watersheds, which resulted in relatively small amounts of fluvial deposits (Fox and Martin, 2015). As a result, although the collection of channel bed deposits has the advantage of characterizing sediment signatures with minimal effort, its application requires pre-knowledge of geomorphology and hydrology of the studied watershed and should only be applied to proper systems (e.g., steep watersheds with relatively small amounts of fluvial deposits). Another related matter is that different sources may have distinct grain size distribution or density. If one source provides sediment that is considerably finer or lower density than others, it may be less likely to settle out in the bed. This issue could also be scale-dependent as bed deposits may mainly come from localized bank erosion in small streams but be more likely from deposition during the falling limb of a high flow event in large rivers. All these issues need to be taken into consideration if fine channel-bed sediment is used as target samples for sediment fingerprinting. It is also worth noting that recent overbank deposit in large rivers may be a useful target in some cases as it is easy to collect and represent samples from high-flow events when most sediment moves.

In addition to suspended sediment, lake sediment core samples were also used by a few studies (11%, Table S1) to reconstruct changes in sediment sources over a longer timeframe. Sediment fingerprinting investigations on historically deposited sediment (e.g., floodplain deposit, lake sediment core) could enhance our understanding of earth surface processes and establish baseline conditions for management strategy and policy development. For instance, to deal with the accelerated infilling of Lake Pepin, Kelley and Nater (2000) reconstructed sediment source changes over the past 700 years using sediment cores from the lake and reported that sediments from the Minnesota River had always been the dominant sediment source. Based on that, Belmont et al. (2011) further analyzed fallout radionuclide data of ^{210}Pb and ^{10}Be from Lake Pepin cores and documented major shifts in sources of sediment between upland and near-channel sources in the past five centuries. Combined results from both studies not only enhanced our understanding of past changes in landscape erosion but also highlighted the hotspot of contemporary sediment sourcing investigations for the greater Upper Mississippi-Lake Pepin watershed (Belmont and Foufoula-Georgiou, 2017). A major concern for the use of historically deposited sediment is the uncertainty associated with tracer conservativeness in a range of long-term depositional environments. Only limited studies have been conducted to explore that due to fact that an independent source of sediment provenance information is often lacking for result validation (Pulley et al., 2015). Pulley et al. (2015) assessed differences between sediment provenance predictions obtained using different tracer signatures (i.e., lithogenic radionuclide, geochemical and mineral magnetic signatures) on lake and floodplain sedimentary deposits and reported considerable differences between tracer group predictions (up to 100%). The authors attributed that to tracer non-conservatism and suggested that simple data corrections (e.g., particle size correction, weighting parameters) would not result in significantly greater agreement between the predictions of the different tracer groups. This work highlighted the importance of recognition of tracer non-conservatism for the use of historically deposited sediment and tracer selection, which merits further investigation.

About half of MRB studies (52%, n = 17, Table S1) investigating the relative importance of upland and near-channel sources conducted sampling campaigns with the consideration of temporal sediment sourcing variations. Considering temporal variability could improve the reliability of souring estimates as the contribution of various sediment sources may change significantly during runoff events or across seasons. For instance, in studies quantifying relative contributions of sediment sources in various watersheds in the MRB, Wilson et al. (2008, 2014a) observed significant shifts in sediment source contribution from the beginning of the rising limb to the end of the recession limb. The authors attributed that shift to bank failure during the recession limb of the hydrograph and source material exhaustion from the uplands (Wilson et al., 2008, 2014a), which addressed the role of hydrological control on fine sediment transport. These studies also highlight the fact that runoff events are highly unsteady. Approaches based on the steady state assumption, such as rating curves or models, could have significant errors especially for flush events. In contrast, the combination of field measurements and the load partitioning analysis using sediment source fingerprinting could lead to a comprehensive understanding on the relative importance of different sources to storm sediment loads. Additionally, targeting sediment source fingerprinting across the hydrograph could be especially important for low-relief landscapes as connectivity of sediment flux across the landscape is highly related to the changing magnitude of a runoff event (Neal and Anders, 2015; Yu and Rhoads, 2018). For seasonal variability, seven MRB studies (Table S1) took a multi-season sampling strategy that accounts for seasonal variations in land use and associated erosional processes as well as mobilization of stored sediment. Combined effects of the climatic forcing, land cover, and sediment availability were reported as the main controlling factors for the considerable seasonal sediment source dynamics (Abban et al., 2014, 2016; Lamba et al., 2015a; Neal and Anders, 2015). Specifically, Wilson et al. (2014b) emphasized that fingerprinting investigations over short time periods are only snapshots during a particular season of a year. They reported considerable variability in sediment contributions from near-channel sources (61–85%) at Goodwin Creek in different months of different years and called for the need long-term monitoring of sediment source contributions.

4.3. *Tracer selection for source discrimination*

4.3.1. *Fallout radionuclides*

Fallout radionuclides that originate from the atmosphere and are quickly and strongly bound to fine particles are the most commonly used sediment tracers by MRB studies (44%, Fig. 2d, Table S1). They are especially well-suited not only to apportion the sources of sediment but also serve as chronometers of sediment transfer in riverine systems (Matisoff et al., 2005; Belmont et al., 2011; Stout et al., 2014), which has clear management advantages. In particular, Belmont et al. (2014) proposed a process-based framework using radionuclides with different half-lives to apportion upland and near-channel sediment sources and estimate channel–floodplain exchange. The authors successfully applied the framework to a watershed in the Upper MRB using ^{10}Be, ^{210}Pb$_{ex}$ and ^{137}Cs measurements (Belmont et al., 2014). Other mathematical frameworks have also been proposed to accommodate radiogenic tracers within a sediment routing framework. For instance, Viparelli et al. (2013) presented a tracer routing model for the transport of radiogenic tracers associated with bed material and washload river sediment, which focuses on the average budget of sediment and tracers at reach scale. The model accounts for production and decay of radioisotopes in the floodplain and is applied to a generic river system (Viparelli et al., 2013). In another study, Lauer et al. (2016) developed a 1-D framework for simulating morphodynamic evolution of bed elevation and size distribution in a river that actively exchanges sediment with its floodplain. The program can track changes in radioisotopic concentration in particles in any size class and is applied to a catchment in France (Lauer et al., 2016). Such research advances reflect the strength and potential of the use of radiometric fingerprinting in management. Additionally, fallout radionuclides can be used independently to quantify the relative sediment contribution from ‘old’ and ‘new’ sources to further interpret fingerprinting results from other types of tracers. For instance, Gellis et al. (2019) apportioned the sources of sediment in an agricultural watershed of the Upper MRB into channel banks and surface-derived sediment using elemental tracers and determined the age of the surface-derived portion of sediment using fallout radionuclides with a three-box model.

There are certain concerns regarding the application of fallout radionuclides, and not all of them were addressed in MRB studies. For instance, Wilson et al. (2008) found temporal and spatial variations of atmospheric influxes of radionuclides (^{7}Be and ^{210}Pb$_{ex}$) within and across their studied watersheds (0.04–7.70 mBq cm^{-2} day^{-1} for ^{7}Be and 0.01–0.46 mBq cm^{-2} day^{-1} for ^{210}Pb$_{ex}$), which calls for the need for atmospheric deposition corrections. However, 35% of MRB studies (n = 6, Table S1) utilizing fallout radionuclides did not mention that in their work. The local scale heterogeneity by potential fallout deposition patterns could be another issue in areas with local to regional fallout from nuclear accidents (e.g., Chernobyl and Fukushima), which may make it impractical to use radionuclides such ^{137}Cs as correcting for spatial variability could be very costly. In addition, sediments from non-channel sources with lower radionuclide activities could be entrained in the suspended sediments and misinterpreted as channel contributions. For example, extensive gully erosion could erode deeper during a single runoff event and introduce sediment with low radionuclide concentrations, whereas re-suspended bed sediments could have low radionuclide activities due to radioactive decay (Wilson et al., 2008, 2014a). It may not be realistic to expect a full elimination of such sources in practice, and in some cases, gullies may only form when the study is ongoing. So, radionuclide signatures of the fine suspended sediment should be exercised primarily in cases such as in watersheds without deep incision of the landscape, in low-to-moderate intensity runoff events with gullies likely being eroded gradually to shallow depths, and in sand/gravel-bed streams with little fine material resuspension (Wilson et al., 2012, 2014a). A correction factor may be helpful to constrain such uncertainties, but there has been a lack of systematic research in this topic for questions like how deep gully erosion needs to be to bias results or how spatially extensive the gullying needs to be to affect fingerprinting results at various scales. Another issue that needs attention is that as fallout radionuclides decay with time, the availability of full datasets with reference dates is of particular importance for radionuclide activity decay-corrections at a later date or data comparison between studies (Evrard et al., 2020). However, this information is lacking in 65% of MRB studies (n = 11, Table S1).

Even with the concerns mentioned above, in terms of management radionuclides are overall the most robust tracers to discriminate between upland and various subsurface sediment sources including channel banks, gullies, landslides, unpaved roads and construction sites (Pulley et al., 2017; Evrard et al., 2020). Motha et al. (2002) found that the sediment generation process, after accounting for differences in particle size and organic matter content, had an effect on some geochemical properties for some of the sources (i.e., non-conservative behavior) but no effect on radiochemical properties. The commonly used radionuclides in the global research community are ^{137}Cs, ^{210}Pb$_{ex}$ and ^{7}Be (Evrard et al., 2020), and research in past decades has added other radionuclides as alternatives for researchers and land managers to choose from, including ^{10}Be (Belmont et al., 2007, 2014), ^{226}Ra (Schuller et al., 2013), ^{232}Th (Zebracki et al., 2015), ^{4} K (Sellier et al., 2020) and plutonium isotopes (Hobgen et al., 2014). Based on their needs, they can either use just a single radionuclide (e.g., ^{137}Cs) for a robust discrimination between surface and subsoil sediment sources, or a combination of several radionuclide measurements for additional information on different erosion processes such as channel-floodplain exchange (Belmont et al., 2014) and vertical and horizontal subsoil erosion (Hancock et al., 2014). Although analysis of radionuclides is relatively expensive (McKinley et al., 2013) and only available at a few laboratories, radiometric fingerprinting should still be considered as one of the most powerful aids for sediment sourcing, especially for large scale land management that could cost millions to billions of dollars.

4.3.2. *Elemental tracers*

Elemental concentrations are also commonly used as tracers for geochemical characterization for MRB studies (44%, Table S1). One major advantage of using elemental tracers in sediment source fingerprinting is that they are commonly measured by samples collected by various routine water quality monitoring programs, which permits investigations of sediment sources without dedicated sampling and analytical costs being incurred (Zhang et al., 2012). Also, compared to radionuclides, elemental analysis can provide results for more than 40 elements by only a small mass of samples (often $<$0.5 g) at significantly less cost. Additionally, elemental tracers are the only type of tracer used in both geographic (100% studies) and geomorphic (40% studies, Table S1) sediment source fingerprinting for MRB studies, making it a cost-effective choice for different types of fingerprinting investigations.

One concern with the use of elemental tracers is that the degree of conservatism differs among tracers, which depends on the tracer itself and how it is bound to the sediment. For example, matrix-bound elements (e.g., Si and Al), the rare earth elements (e.g., Y and Sm) as well as certain chemically stable minerals (e.g., TiO_2) and sulfides (e.g., Pyrite) are likely to be most conservative. In contrast, minerals that can be affected by changes within the normal pH range (e.g., carbonate) and elements associated with sediment surfaces via sorption onto Fe oxide coatings or organic carbon films are likely to be less conservative (Collins et al., 2017). In practice, the conservatism of elemental tracers correlates with their solubility, so that the choice of digestion approaches would be significant for the reliability of analysis and comparability of results between studies. In this context, different digestion procedures were used for MRB studies utilizing elemental tracers. Specifically, 53% of studies applied total digestion for elemental analysis, and 47% used partial digestion ($n = 9$ and 8, respectively, Table S1). Different analytical protocols would likely lead to different fingerprinting results. In a study conducted in the Tennessee River basin, Huangfu et al. (2020) tested the sediment source fingerprinting technique with samples prepared by both total dissolution and nitric acid extraction and found that source apportionment results were very sensitive to the digestion procedure. Although total dissolution has the advantage of a complete decomposition of the samples and the unambiguous measure of total metal levels (McGrath, 1998), this method is risky due to the dangerous reagent hydrofluoric acid. It may not be realistic to expect an extensive application of total dissolution by management agencies for large-scale landscape conservation efforts. In addition, it is not fully clear whether a complete quantitation of tracer concentration from sediment samples would always be superior in terms of providing tracer values that are most useful to differentiate between potential sources (Collins et al., 2017). There is a lack of systematic comparisons between the use of total and partial digestions, and up to now, selection for digestion procedures may still have to be a trial-and-error exercise for each case of study. This makes it challenging to use elemental tracer data from some easily accessible sources such as the USGS National Geochemical Database, as they contain tracer results obtained by both total and partial digestions. Impacts of digestion procedures on fingerprinting merit further investigations, the results of which could help in developing standardized fingerprinting analytical protocols. It is also worth mentioning that all MRB studies used ICP-OES/MS (inductively coupled plasma-optical emission/mass spectrometry) techniques for analysis instead of non-destructive methods such as XRF (X-ray fluorescence), and it is not sure whether a certain analytical method is more reliable than others for the use of elemental tracers.

4.3.3. *Biogeochemical tracers*

Information generated by geochemical tracers to differentiate between broad categories of sediment sources (e.g., forest, cropland and pastureland) may sometimes not be of sufficient detail for river basin managers to make effective and well-informed decisions. In contrast, biogeochemical tracers, such as stable isotopes of carbon and nitrogen (δ^{13}C and δ^{15}N, respectively), have the potential to discriminate between a greater number and variety of sediment sources (e.g., differentiate between different crop types or tree species) and provide greater detail on sediment sources. δ^{13}C and δ^{15}N are the most used biogeochemical tracers for MRB studies (26%, Table S1), which can differentiate sediment from land use origin as sediment can retain the isotopic ratios of its parent soil (Riddle et al., 2022). Specifically, δ^{13}C of plants can be integrated to soil organic matter as plants undergo degradation (δ^{13}C enrichment is often relatively small during degradation), whereas δ^{15}N of soil is dependent on nitrogen inputs to the soil, the nitrogen outputs, and land management practices. These MRB studies showed that, in addition to discriminating the upland and instream sediment sources (Mahoney et al., 2019), δ^{13}C and δ^{15}N could also provide useful information on the relative contributions of allochthonous and autochthonous organic matter sources for a better organic matter correction (Abban et al., 2016) and differentiate soil organic matter and geogenic organic matter for the management of disturbed watersheds (Fox, 2009). δ^{13}C may also discriminate between sediment derived from soils with C3 vegetation (majority of tree or temperate grass species) compared to those covered with C4 vegetation (grass and cropping species). One MRB study (Wilson et al., 2012) suggested a coupled use of radionuclides (^{7}Be and ^{210}Pb$_{ex}$) and biogeochemical tracers (δ^{13}C and δ^{15}N) for better spatial resolution of source areas, with radionuclides used for temporal heterogeneity of the sediment movement and stable isotopes as an indicator of the vegetation cover.

However, concerns remain regarding the usefulness of δ^{13}C and δ^{15}N due to uncertainties associated with their conservativeness. Both isotopes may be subject to transformations within channel-floodplain environments that may be difficult to detect by standard statistical approaches (Riddle et al., 2022). δ^{13}C and δ^{15}N of source soils are typically treated as constant in fingerprinting studies, but significant differences of their tracer signatures have been reported within the time frame of a few years for upland sources (Fox and Martin, 2015). Both isotopes can also be modified during transport or storage, notably in channel primary production and respiration. A number of studies removed one or both bulk isotopes from fingerprinting practices due to a lack of tracer conservativeness or other unknown errors in using δ^{13}C and δ^{15}N (Rose et al., 2018). In a recent publication investigating the use of carbon and nitrogen isotopic ratios for sediment fingerprinting, Riddle et al. (2022) conducted virtual sample mixture tests to verify whether δ^{13}C and δ^{15}N of sediment are conservative in various processes that could potentially impact their usefulness in sediment fingerprinting and concluded that algae accrual, concentration dependency mixing, physical loss of organic matter during transport, and seasonality of the in-stream sediment source could significantly impact the use of δ^{13}C and δ^{15}N in the stream environment. As a result, δ^{13}C and δ^{15}N should be used with caution in certain systems such as low gradient agricultural systems with substantial algal load (Ford et al., 2017). Riddle et al. (2022) also suggested that both $\delta^{13}C$ and $\delta^{15}N$ could be considered as potential tracers as they performed similarly in the tests, and the mean isotopic ratios of sediment sources should be separated by at least 1‰ to lessen tracer conservativeness concerns.

4.4. Particle size selection and tracer non-conservatism assessment

All MRB studies addressed particle size impacts on fingerprinting studies by utilizing at least one of the main approaches widely used in the global research community (Laceby et al., 2017), either fractionation (89% MRB studies), concentration correction (37%) or both (34%, Table S1). For fractionation, it is considered the best practice to measure the particle size distribution of the sources and target sediments and to check whether significant differences exist between the two (Collins et al., 2020). 11% of MRB studies ($n = 4$, Table S1) did that, and the rest simply chose the <2 (6%), <53 (29%), <63 (53%), 2–63 (6%), <64 (3%) or <125 (3%) μm fraction. A unique fractionation selection is simple and straightforward, but it may not be a robust way to avoid the sorting effect of particle sizes when applied at large watersheds. The use of fractionation means that the corresponding source apportionment results would only relate to the fraction being used. Failing to use the primary fraction that poses the major problem to the area of interest could lead to unreliable fingerprinting interpretations. For instance, the use of narrower size ranges (e.g., <2 μm) could minimize the variability in tracer concentrations, but in some cases its fingerprinting results may not be representative for the primary fraction of sediment being transported. The use of broad particle size ranges such as <2 mm could suit certain management needs (e.g., to target the source of sand for issues of channel adjustment and smothering) but may be scientifically unsound as properties of sands and silts/clays are unlikely to be comparable. Previous studies have reported that the choice of different particle size fractions for samples from the same watershed can lead to divergent apportionment results (e.g., Haddadchi et al., 2016), highlighting the concern of using one particle size range for fingerprinting investigations. In addition, numerous sub-catchments within a large watershed may have different grain size distributions of the source materials, and there can be significant spatial variations in the surface particle size distribution across areas as small as 0.35 km^2 (Mzuku et al., 2005). The key to using fractionation is to ensure that such differences are addressed for samples being collected and analyzed. A further consideration is to understand the relationship between particle size distribution and event magnitude, as larger runoff events with stronger driving force likely result in less particle size selectivity. In brief, for large watersheds particle size distributions may vary within and among watersheds, and geomorphic processes further fractionate particle sizes and densities over time and space. Thus, all aspects of the sediment fingerprinting sample design, from source classification, tracer selection, sample collection, analysis, and interpretation, must all consider grain size dynamics in a way that is commensurate with the research or management questions being addressed.

Concentration corrections for particle size were sometimes conducted to mitigate differences in the particle size distributions of source and target samples (Table S1), with specific-surface-area (SSA) being the most common metric for normalization. Although not mentioned in many MRB studies, there are uncertainties relating to the fundamental assumption of particle size corrections, such as whether assumed positive linearity between SSA and tracer concentration applies to all tracer properties being used and whether selected tracer properties have the significant linear correlations across all sources and target sediments (e.g., Belmont et al., 2011; Smith and Blake, 2014; Collins et al., 2017). A simple SSA ratio-based model weighting may not be a rigorous approach for concentration correction. In a study identifying sediment sources and sinks in a 4,300 km^2 basin located within the Upper Mississippi River basin, Stout et al. (2014) did not use a single reference surface area for the entire watershed but corrected upland and alluvial samples by suspended samples from each corresponding sub-watershed. Such approaches appear to be much more robust for concentration correction in a large watershed. Alternatively, if accurate SSA ratio-based corrections cannot be achieved due to limits of personnel and funding, particle size corrections may just not be applied as suggested by some researchers (Martinez-Carreras et al., 2010; Koiter et al., 2013a). Instead, the physical reasoning of the sediment transport process and sample fractionation can be used as the main control for the particle size sorting effect.

The conservativeness of tracer properties during sediment transport has been considered a fundamental assumption for sediment source fingerprinting, while only 34% of MRB studies ($n = 13$, Table S1) tested tracer conservatism by using a widely used range or bracket test. Nearly all recent published source fingerprinting studies from the global research community assessed tracer conservatism through such tests (92% of 2018–2019 publications, Collins et al., 2020). So, it could be beneficial to include at least a simple screening test, such as a range test which determines if tracer concentrations of target sediments fall within the ranges of source samples, as part of a fingerprinting routine, especially in cases when sediment fingerprinting is used to inform land management decisions. It is worth mentioning that these commonly used techniques are mostly black box testing lacking comprehensive information on the conservatism of multiple tracers in different environments, and there is very limited literature explicitly investigating tracer conservatism (e.g., Motha et al., 2002) and no formally agreed approach to detect tracer transformation during sediment generation, transport and deposition. Few studies have also considered that tracers could be polluted or non-conservative after they pass a range test. In this regard, Lizaga et al. (2020a) proposed a quantitative approach to identify tracers with values that are inside the source range but that have non-conservative behavior, which was referred as "dissenting tracers" (Lizaga et al., 2020a; Latorre et al., 2021). Such approaches merit further investigation as tests from artificial sample mixtures show that neither frequentists nor Bayesian apportionment models could effectively handle the "dissenting tracers" (Latorre et al., 2021). Additionally, one study taken in the MRB showed that tracers with non-conservative behavior that is predictable and verifiable can be immensely useful for sediment fingerprinting, and that the combined use of tracers that exhibit conservative (i.e., ^{10}Be) and non-conservative behavior (i.e., $^{210}Pb_{ex}$ and ^{137}Cs) can provide spatially integrated, yet temporally discrete insights to constrain sediment sources at a river network scale (Belmont et al., 2014). Utilization of similar approaches is still under-investigated and requires knowledge on source-river connectivity of sediments and sediment routing through fluvial systems.

4.5. Sediment source apportionment

Statistical tests are sometimes conducted for pre-selected tracers to find the minimum number of tracers with the best discriminating power for sediment sources. This is especially true for the use of elemental tracers. Specifically, 76% of MRB studies ($n = 13$, Table S1) utilizing elemental tracers included the use of statistical analysis to identify a subset of tracer properties to best discriminate sources, 69% of which ($n = 9$, Table S1) took a two-step process of Kruskal-Wallis H-test and discriminant function analysis (KW-DFA). The two-step process is also the most widely used approach in the rest of the global research community. In a study comparing three statistically selected optimum composite fingerprints (i.e., by KW-DFA, DFA and principal components analysis), the statistical discrimination based on the widely used KW-DFA tracer selection was found to be the most effective option yielding the most reliable results (Palazon and Navas, 2017). There have been trials to find better alternatives for the commonly used tracer reduction exercises. The theoretical base is that uncertainty in source predictions would be better reduced by increasing, rather than decreasing the number of tracers after excluding non-conservative tracers, especially for cases with no particle size or organic matter corrections (Sherriff et al., 2015). Some studies further proposed selection procedures that removed tracers on the basis of non-conservative behavior after a bracket or range test, which was shown to lead to more accurate source apportionment results as compared to the KW-DFA that prefers tracers with strong discriminating power (e.g., Smith et al., 2018; Lizaga et al., 2020a). Most of such procedures received less attention from the research community but have potential to benefit tracer selection routines. Nevertheless, maximizing tracer data can be considerably resource demanding, and whether the benefits would outweigh costs could be determined on a case-by-case basis. Most of these alternatives were tested by artificial sample mixtures but have not been widely applied in the field. Further research could verify their performance in a natural environment. In addition, it is commonly considered that proportional contributions of $n+1$ different sources can be uniquely determined by using n different tracers. However, in complex systems the number of potential sources could exceed $n+1$ with n available tracer signatures. Some methods were proposed to determine ranges of source contributions in these cases, such as the development of the IsoSource program for the use of isotope tracers (e.g., Phillips and Gregg, 2003), but relevant investigations are, in general, limited. Further research may be needed for definitive solutions.

Traditional statistical analysis for tracer selection will always result in one optimum composite fingerprint, which is often considered adequate for source apportionment modeling. However, Zhang et al. (2016) argued that multiple composite fingerprints that have similar discrimination abilities could produce quite different estimates of source proportions (Zhang and Liu, 2016; Zhang et al., 2016a). This lack of correlation between ability of the tracer to discriminate and its rigor in estimating source contributions challenges the common use of one optimum fingerprint. In contrast, the authors suggested a new approach to use the maximum number of multiple composite fingerprints in lieu of a single optimum fingerprint. In their studies conducted at sites located in the Arkansas-Red River basins, Zhang and others reported that estimated source contributions varied greatly among different composite fingerprints due to differences in the measurement errors and degrees of the conservativeness between tracers and demonstrated that their proposed approach substantially increased the accuracy of source proportion estimates while significantly reducing the associated uncertainties (Zhang and Liu, 2016; Zhang et al., 2016a). Although not being widely applied, this approach provides greater assurance that tracer non-conservatism or poor discrimination will not lead to great uncertainties in fingerprinting results. Elemental tracers seem to be the best candidates for the use of the multiple composite fingerprints approach, as elemental analysis usually gives results of multiple inorganic elements at a time. However, it is worth noting that use of multiple elemental composite fingerprints is still subject to the same limitations of one optimum fingerprint in other aspects, such as the incapability to discriminate floodplain sediments from upland and channel sources.

The use of an un-mixing model for sediment source apportionment is a critical step in sediment source fingerprinting. Approaches to modeling have significantly advanced over the last 20–30 years, shifting from deterministic optimization procedures to stochastic frameworks. The latter relies on Bayesian and/or Monte Carlo methods and has the advantage of producing distributions of source apportionments and assigning prediction intervals (Batista et al., 2022). In terms of Bayesian and frequentist methodologies, both frequentist (87%, $n = 33$) and Bayesian (16%, $n = 6$, Table S1) un-mixing models were used for MRB studies. In recent years, the Bayesian modeling approach has been increasingly used by the global research community (Collins et al., 2017). While some literature suggests that it produces comparable apportionment results to frequentist un-mixing models (e.g., Nosrati et al., 2018), others suggest their model outputs can be very different based on the same datasets (e.g., results from Song et al. (2022) prefer frequentist modeling). There is one study taken within the MRB that utilized both a frequentist and a Bayesian model for sediment source apportionment for an agricultural stream in the Iowa River basin (Wilson et al., 2014b), the results of which suggest that the frequentist model results compare favorably with those of the Bayesian model.

There are limited studies comparing different apportionment models (e.g., Haddadchi et al., 2014), and it is hard to say whether there is a certain model that is more robust than the rest. The frequentist model presented by Collins et al. (1997) and its derivatives were extensively used in MRB studies. However, one study comparing accuracy of apportionment model outputs reported that the performance of Collins model was less accurate than the other three widely used models (Hughes et al., 2009; Devereux et al., 2010; Laceby and Olley, 2015) due to overuse of weighting parameters (Haddadchi et al., 2014). While this study does not necessarily mean that one model is superior to another, it demonstrates the dependence of source attribution on model selection. As for MRB studies, findings from Fox and others also showed that different frequentist un-mixing models could result in different apportionment results using the same datasets (Fox, 2009; Fox and Martin, 2015). Another related issue is for models using local and generic algorithms (e.g., Collins et al., 2010b). For instance, Haddadchi et al. (2013) compared mixing models applying local and global optimization methods to datasets from two different catchments and showed that model outputs could change remarkably depending on which mixing model was used. The inconsistency in model performance highlights the need to validate models before or after use since sediment source fingerprinting often involves extensive and time-consuming procedures of field sampling, sample preparation and laboratory analysis. It is important to ensure that data collected from these costly programs can accurately ascribe the source contributions, especially in cases that fingerprinting results are further used as management aids. In addition, there is a lack of research discerning whether different types of un-mixing models could better suit different types of datasets or tracers, which merits further investigation.

The use of artificial sample mixtures appears to be the most robust way to validate model outputs. These can be created in the laboratory by physically combining known masses of the source material (e.g., Haddadchi et al., 2014; Uber et al., 2019; Gaspar et al., 2019). Specifically, artificial mixtures have been successfully applied to provide information on the quality of the source discrimination afforded by the tracer suite, investigate the impact of within-source tracer variability, assess the influence of corrupt or non-conservative tracers on model outputs and discern the importance of different tracer selection procedures (Pulley et al., 2017; Pulley et al., 2020; Shi et al., 2021; Latorre et al., 2021). Although artificial mixtures can provide a powerful tool for evaluating fingerprinting models, their use in sediment source fingerprinting studies is relatively rare (Batista et al., 2022). This may be attributed to additional costs for sample analysis. Also, biases might be introduced due to particle size effects and sample mixing (Collins et al., 2020). If limited by budget and staff, the use of virtual sample mixtures is another option (e.g., Pulley et al., 2020; Gholami et al., 2020a; Nosrati et al., 2021). For example, the values of all the selected tracers can be multiplied by known source proportions for the individual sources, and the results can be input to the frequentist or Bayesian apportionment models. Subsequently, the predicted and known source proportions should be compared by statistical indicators such as root mean square error and mean absolute error (Gholami et al., 2020b; Li et al., 2020). A limitless number of virtual mixtures can be created without cost, and a recent study comparing the use of artificial and virtual mixtures has reported that virtual mixtures can be as useful as artificial mixtures for model testing when analytical errors are negligible (Batista et al., 2022). Therefore, the use of virtual sample mixtures for model output validation could be included as a routine for sediment source fingerprinting.

Traditionally, model goodness-of-fit (GOF) is also used to ensure reliability of model outputs (26% of MRB studies calculated GOF, Table S1), but there are arguments regarding its robustness. For example, through experiments simulating natural processes to validate un-mixing model outputs, Gaspar et al. (2019) demonstrated that high GOF values do not necessarily correspond to accurate predictions of source contributions, and care should be taken when using only this parameter to assess the performance of un-mixing models. In another study where both the frequentist and Bayesian models were applied for sediment source apportionment, Song et al. (2022) found that while both types of models had very high GOF values, the virtual sample mixture test indicated that the frequentist model performed better in terms of model accuracy. As a result, the use of GOF is considered inferior to the use of artificial or virtual sample mixtures for model output validation.

Despite model accuracy (low error), decision-makers and stakeholders often require sediment source information to be precise (low uncertainty) as well. Accordingly, a Monte Carlo uncertainty test is often considered part of the sediment source fingerprinting routine. For MRB studies, 45% conducted uncertainty analysis ($n = 17$, Table S1) with 88% of them ($n = 15$, Table S1) using the widely used Monte Carlo routine. Zhang et al. (2006b) reported that Monte Carlo simulation tends to underestimate relative errors and standard errors (SE) based on their study at Fort Cobb Reservoir in Oklahoma and suggested to report interquartile range or standard deviation of the Monte Carlo simulation results. If objective sediment source grouping is not used at the beginning, a trial to re-group sediment sources after the model run may reduce uncertainty and can be achieved as part of the uncertainty analysis (e.g., the source verification test, Gellis et al., 2019).

Other metrics to evaluate sediment fingerprinting models have also been proposed. For instance, if multiple composite fingerprints are used, reporting of standard error or 95% confidence interval (CI) along with the estimated mean source proportions could improve accuracy of result interpretations. It is based on the recognition that predicted proportions by the un-mixing model are not very meaningful if the 95% CI of the predicted proportions are quite large for the studied watersheds, which could indicate insufficient sample collection and uncertainty due to spatial variation of sources and temporal variation of sediment mixtures (Zhang et al., 2016b). For models using stochastic frameworks, Batista et al. (2022) suggested that modeled source apportionments should be tested as distributions, instead of point-based estimates. Their study further suggested use of the continuous ranked probability score (CRPS) to compare modeling approaches or tracer selection methods, the modified Nash-Sutcliffe model efficiency (NSE) coefficient for defining limits of acceptability of model error, and the contingency metrics to assess if the fingerprinting approach can, at the very least, identify the major source in a catchment (Batista et al., 2022). There are also trials to apportion sediment sources without the use of un-mixing models, such as using discriminant function analysis as an intuitive method for characterizing sediment source contributions (Liu et al., 2016). Most of these trials are easier to use and may provide quick access to preliminary results. However, they are often less reliable than the modeling approach especially for complex conditions with multiple source groups.

5. Prospects for sediment source fingerprinting as a management aid to large watersheds

It is important to emphasize that large-scale decisions about policy and management may waste funds if due diligence is not done to identify the sediment sources and understand key processes eroding and transporting sediment through the system. In this regard, the use of sediment source fingerprinting techniques at the scale of large river basins could have tremendous societal and economic benefits. In addition to standardizing fingerprinting steps to advance this approach, efforts to scale it up for its use on large-scale landscapes and make it available to non-scientist users could be beneficial. In this section we discuss applications of sediment source fingerprinting technique in large-scale river basins, review and discuss its use by non-scientist users, and talk about its usage under current management frameworks. The management frameworks being discussed here are mainly those implemented in the United States as this review has a focus on the MRB. However, insights of the relevant discussion should be applicable to management strategies implemented in the rest of the world.

5.1. Sampling approach and effective scales for application of sediment source fingerprinting at large-scale river basins

Sediment source fingerprinting studies taken within the MRB have shown the capacity of this approach to provide critical insights at the scale of large river basins. Specifically, 16% of MRB studies informed the sources of sediment in large-scale river basins (10^4 km^2 or larger, Table S1), all of which provided credible results and informative interpretations. However, they were achieved by different sampling approaches. For example, Gellis et al. (2017) examined sediment source types of the Corn Belt region (648,239 km^2) in the midwestern United States through intensive sampling of fine-grained material in 99 wadeable streams (median drainage area: 167 km^2 ranging from 3 to 6,348 km^2), while Zhang et al. (2012) only used data from samples collected at four major tributary outlets of the Ohio River Basin (median drainage area: 90,084.5 km^2 ranging from 46,392–160,579 km^2) for a total investigation area of 526,024 km^2. The former approach could minimize potential uncertainties and biases associated with non-conservative behavior or sediment deposition/erosion processes but may be financially unacceptable in many management cases. The latter approach is financially and logistically feasible, but interpretations of its results have clear limitations in terms of informing management actions. A hybrid approach that samples at outlets of large basins and intensively at certain watershed(s) of concern may have the strengths of both approaches mentioned above and be more cost-effective for management purposes. As mentioned earlier, the sediment sourcing investigations undertaken in the Upper Mississippi-Lake Pepin (UMLP) basin can serve as a good example of the hybrid approach. Previous fingerprinting studies (e.g., Kelley and Nater, 2000) show that almost all of the recent sediment deposited in the UMLP basin (51,500 km^2) originated in the Minnesota River catchment, whereas the Le Sueur River watershed (2,820 km^2) is the primary contributor of sediment to the Minnesota River. Accordingly, Belmont and other (2011) applied geochemical fingerprinting to sediment cores collected from the basin outlet (Lake Pepin) to interpret shifts in the proportion of sediment derived from upland vs near-channel sources for the UMLP basin. At the same time, the authors developed a sediment budget using sediment fingerprinting and other lines of information for the Le Sueur River watershed to identify sediment source locations and mechanisms. Combined information from both Lake Pepin and Le Sueur River provides strong evidence that the dominant source of large sediment loads in the UMLP basin has shifted from upland soil erosion to the channel network that only comprises a very small percentage of the landscape (Belmont et al., 2011).

Another alternative could be the confluence-based (or tributary-based) approach that continuously models sediment as a source and a sink (Caitcheon, 1993; Walling et al., 1999; Olley and Caitcheon, 2000; Hatfield and Maher, 2008; Laceby et al., 2015; Vale et al., 2016). Its fundamental concept is that collected sediment can be used as target samples to infer sediment sources from upstream tributaries and simultaneously as source samples for sediments sampled further downstream (Fig. 3), which helps remove the concern for new and emergent properties as the scale of observation increases from plot to river basin (Laceby et al., 2017) and incorporates knowledge of the hydro-geomorphological connectivity (Koiter et al., 2013a). The size of the base unit in this sampling approach could be at the scale of a large watershed (10^3 km^2), which is the investigating scale for 13% of MRB studies (Table S1). Previous research has suggested a smaller watershed size of <250 km^2 as the management scale (Collins and Walling, 2004; Walling, 2005; Gellis and Walling, 2011). Although a smaller scale may help constrain uncertainties, it would cost too much to sample every 250 km^2 at the appropriate timescale to discern basin-scale sediment source information. Considering the fact that numerous fingerprinting studies from the MRB and global research community were successfully conducted at the scale of 10^3 km^2, this scale may be the most appropriate

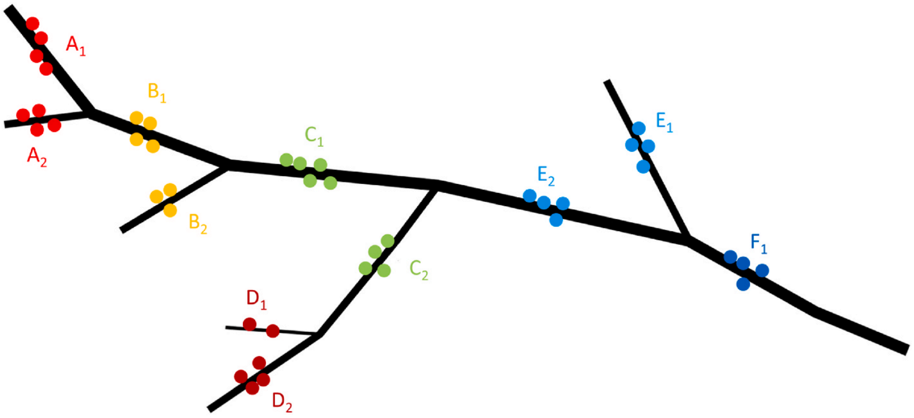

Fig. 3. An example of a confluence-based sampling design for a theoretical catchment with multiple tributaries. This sampling approach continuously models sediment as a source and a sink, which is indicated by colors, letters and subscripts. For example, sediment collected at site B_1 is used as target sample comparing to sediment collected at sites A_1 and A_2. At the same time, it is also used as part of source sample (together with B_2) comparing to sediment sampled further downstream at site C_1 (target sample). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

one to ensure local source fingerprints match local suspended sediment fluxes while balancing costs and benefits. In practice, the size of a base unit can be larger depending on heterogeneity in landscape characteristics and land use types. The more homogeneous the studied catchment is, the larger the base unit can be.

In addition, developing tracer signature libraries using existing data is another way to support basin-scale sediment source fingerprinting applications. Taking full advantage of large, existing datasets can be a necessity for wide applications of sediment source fingerprinting techniques in each and every sub-catchment of a large river basin. For instance, the above-mentioned basin-scale study by Gellis et al. (2017) used published data on $^{210}Pb_{ex}$ in agricultural soils from the literature for the midwestern United States as source sample values. Specifically, these tracer signature libraries could include all published tracer data for different sediment types (e.g., upland soils, streambank samples, floodplain deposits, suspended sediment, riverbed deposits) and for specific ecologically and geographically defined regions of interest (e.g., ecoregion, river basin by hydrologic unit code). For the purposes of fingerprinting, these libraries could also include informative data such as particle size range (or particle size distribution), dates of collection, GPS coordinates of sampling sites, and additional information for certain tracer groups including date of analysis for fallout radionuclides and digestion procedures for elemental tracers. A developed tracer signature library at the scale of a large river basin would be useful for management purposes and could be even more powerful if the library had data output portals for modeling and spatial analysis toolboxes. Data sources for such libraries could include, but not be limited to academic research publications, reports, and publicly accessible databases from governmental soil conservation and environmental monitoring projects, and consulting investigations from private companies. To the best of our knowledge, up to now there is only one standalone database specifically developed for MRB which includes all published sediment source fingerprinting data across the entire basin (Belmont et al., 2022). Further efforts are still needed to transform this standalone database to a server-based database with multiple user access and to develop useful extensible interfaces for research and management needs of broader audiences. Similar attempts to develop tracer signature libraries could be beneficial for other large river basins. It should be noted that fingerprinting data from a library should always be used with caution, as tracer signatures may change with management practices, anthropogenic pollution, or natural erosion.

5.2. Development and use of open-source software

With the increasing complexity of data analysis in procedural steps of sediment source fingerprinting to constrain uncertainties associated with tracer selection and sediment source apportionment, streamlining sediment source fingerprinting data processing could make it more accessible for use by non-scientist end-users such as landowners, catchment officers, policymakers, as well as academics with low programming and statistical skills. The development and use of open-source software could support a wider uptake of the fingerprinting approach, which would be critical for its application at large-scale watersheds. Several toolboxes were developed based on the statistical software R (R Core Team, 2019) in the past years to guide end-users through all critical steps in processing fingerprinting data, which include the Sediment Source Assessment Tool (Sed_SAT), the Sediment Fingerprinting Tool (SIFT) software and the FingerPro package.

Developed by USGS (Gorman Sanisaca et al., 2017), Sed_SAT uses a push-button interface based on R Shiny (https://github.com/tim7en/sedsatv2_shinyapp). Sed_SAT can identify outliers, perform grain size and organic content corrections to the source data, evaluate the conservative behavior of tracers by a bracket test, find tracers with the highest discriminatory power using stepwise DFA, and conduct sediment source apportionments using a frequentist model (modified from Collins et al., 2010a) with Monte-Carlo simulation, source verification test and tracer-by-tracer plot used for error evaluation. This toolbox has been successfully applied to a watershed at the Upper Mississippi River Basin (Gellis et al., 2019) as well as several sub-basins of the Chesapeake Bay watershed (e.g., Cashman et al., 2018; Jiang et al., 2020).

The Rothamsted SIFT has a Shiny user interface as well (Pulley and Collins, 2018a; Pulley and Collins, 2018b). SIFT can run additional bi-plot-based tracer conservation assessments, has GOF functions for model output evaluation, but does not provide options for particle size and organic content corrections. SIFT also includes features including the reclassification of *a-priori* source groups by a combined use of linear discriminant analysis and cluster analysis to ensure adequate discrimination and testing of multiple model configurations using virtual sample mixtures. This software has been successfully tested in multiple catchments in the UK (e.g., Pulley et al., 2019; Pulley and Collins, 2021a; Pulley and Collins, 2021b).

The FingerPro package (Lizaga et al., 2020b) utilizes a step-by-step procedure divided into three main sections: 1) a range test for tracer conservatism and KW-DFA test for tracer selection, 2) a standard linear multivariate mixing model for sediment source apportionment and a GOF function for output validation, and 3) Monte Carlo simulations for uncertainty tests. Specifically, FingerPro allows users to reject tracer selection suggestions by KW-DFA after viewing the results and manually select tracers based on “expert judgment”. It can also exclude discriminant tracers that are shown to have non-conservative behavior after passing a range test (Latorre et al., 2021). The FingerPro package has been tested with artificial samples (Gaspar et al., 2019) and applied in several case studies in Europe and Asia (e.g., Navas et al., 2020; Lizaga et al., 2020a; Song et al., 2022). It is worth mentioning that FingerPro has also been applied to trace the source of wind-borne sediments (Song et al., 2022).

Some other packages were also created to only assist in the sediment source apportionment modeling step. One example is the MixSIAR package in R, a framework that allows users to create Bayesian un-mixing models based on their data structure and research questions (Guerrero and Rogers, 2020). MixSIAR has been used in many sediment source fingerprinting studies by providing a Bayesian approach for un-mixing modeling (e.g., Upadhayay et al., 2020; Song et al., 2022) and can serve as a good addition to existing step-through fingerprinting software.

Despite the recent advances in software development, current fingerprinting data processing tools are still not designed for some end-uses to aid large-scale landscape management. First, existing fingerprinting packages do not incorporate functions to support robust sampling designs, which may be a critical research need. One software improvement could include support functions of extracting data from external databases such as the USDA Soil Survey Geographic database (https://data.nal.usda.gov/dataset/soil-survey-geographic-database-ssurgo) and USGS National Land Cover Dataset (https://www.mrlc.gov/) to make it easier for end-users to develop sampling schemes stratified by major soil and land use types (Zhang and Liu, 2016). Another improvement could be portals to GIS toolboxes so that a randomized sampling design can be implemented with the use of a GIS and random number generator (Gellis et al., 2019). Additional features, such as probability-based sample number determination, could also be integrated in this step to provide support beyond tracer data processing. Although existing software includes key steps of fingerprinting data processing, most only offer limited customization options. As end users may have background knowledge in geomorphology, hydrology, or geology for targeted watersheds, customizing options such as whether to perform particle size or organic matter corrections and whether to accept statistical test results for tracer selection could better fit the needs of end-users. Other software improvements might include more statistical approaches for tracer selection, different types of un-mixing models for source apportionments, and various metrics to constrain uncertainties and validate model outputs; also, software could include upgrade capacity for ongoing development of novel techniques. For example, the previously mentioned approach that uses conservative and predictably non-conservative fallout radionuclides for floodplain sediment source apportionment is not available in all existing sediment source fingerprinting toolboxes. If the software includes these features as individual packages, it could catch up with developments in sediment source fingerprinting techniques by the global research community. Lastly, a push-button interface benefits non-scientist end-users but has significantly less flexibility than the coding interface; ideally both interfaces would be integrated into the software for users with different programming skills.

5.3. The use of sediment source fingerprinting under current management frameworks

The total maximum daily loads (TMDLs) project is the most widely applied national regulatory program in the United States for water quality impairment and sediment reduction, which typically includes long-term and costly efforts to remedy point source and non-point source pollutants. Given the level of investment needed to remedy watershed-scale water quality impairments caused by excess sediment or turbidity, it is essential that TMDLs are informed by robust sediment source analysis approaches, such as sediment source fingerprinting. Sediment source fingerprinting could provide robust source assessment results, especially at a large watershed scale, as compared to the scaled-up estimates based on local-scale applications of the Universal Soil Loss Equation (USLE) and derivatives thereof as implemented in simulation models such as the Soil and Water Assessment Tool (SWAT) and the Water Erosion Prediction Project (WEPP). Furthermore, sediment fingerprinting is often more cost-effective and diagnostic than full-scale field erosion measurements. The ability of sediment fingerprinting to estimate the relative contribution of near channel sources to the sediment load in a watershed further enables it to separate pollutant load allocations for individual non-point source categories such as bank erosion and floodplain sediment (e.g., Belmont et al., 2014). This is a major advantage over other techniques, which provides valuable feedback for developing appropriate monitoring and remediation strategies to target non-point source pollutant loads. One previous study (Mukundan et al., 2012) reviewed sediment source fingerprinting techniques with an attempt to transfer it from a research tool to a management tool for state and regional TMDL programs implemented in the United States. The authors proposed an additional component of source assessment to the existing TMDL framework that combines sediment source fingerprinting with sediment budgeting and modeling, which could support all steps in development of a robust and effective TMDL. They emphasized the role of sediment source fingerprinting in linking targets and sources, load allocation and follow-up monitoring in the sediment TMDL procedure (Fig. 4), the point of which remains valid today. In practice, Belmont et al. (2011) demonstrated how sediment source fingerprinting can be coupled with sediment budgeting to allocate loads among different sources for the Le Sueur River in the Upper MRB. Similar approaches have also been applied to watersheds in other regions of the United States such as the Piedmont plateau (e.g., Mukundan et al., 2010; McCarney-Castle et al., 2017), Chesapeake Bay drainage area (e.g., Cashman et al., 2018; Gellis and Gorman Sanisaca, 2018; NOE et al., 2020) as well as other countries around the world (e.g., Gellis and Walling, 2011; Sherriff et al., 2019). Nearly all of the studies considered sediment source fingerprinting a reliable and cost-effective method to identify and target sediment sources in mixed-use watersheds.

Based on our literature review, Minnesota appears to be the only state within the MRB that incorporates sediment source fingerprinting in state TMDL projects. The state TMDL is the maximum amount of a pollutant a body of water can receive without violating water quality standards, and the TMDL process identifies all sources of a pollutant and determines how much each source must reduce its contribution in order to meet the standard. In terms of sediment, the state has reported that the majority of the suspended sediment load in the south metro Mississippi River, which lies between the mouth of the Minnesota River and Lake Pepin, comes from the Minnesota River and has successfully identified near-channel sources including ravines, bluffs and streambanks as the greatest contributors to increased sediment in the Minnesota River based on their previous investigations using radiometric fingerprinting techniques (Wilcock et al., 2010; Gran et al., 2011; Belmont and Foufoula-Georgiou, 2017). Specifically, Belmont and Foufoula-Georgiou (2017) reported that upland sediment from agricultural fields, which constitutes 78% land of the whole watershed, dominated in the mid-twentieth century. While sedimentation rates in Lake Pepin have remained high since 1950, the source of sediment has shifted to predominantly near-channel bluffs and stream banks after that. That fact indicated that sediment reduction efforts by improving tillage practices and taking agricultural land out of production would be ineffective, as enhanced artificial drainage and increased precipitation that amplified erosion of near-channel sediment sources were the key factors controlling sediment dynamics. Instead, careful targeting of the installation of water detention features in as little as 5% of the landscape at upper portions of the watershed could reduce sediment loading by as much as half (Belmont and Foufoula-Georgiou, 2017). With this credible and robust information, the Minnesota state agencies changed their initial perspective on the role of agricultural field erosion in causing excessive sediment loads, developed a sediment reduction strategy to meet the Minnesota River sediment TMDL by 2040 and considered the development of sediment source fingerprinting technique as one of the priorities for the success of the sediment reduction strategy for the Minnesota River Basin and the south metro Mississippi River (Gunderson et al., 2015). It is worth mentioning that the successful application of radiometric fingerprinting in the state of Minnesota does not suggest

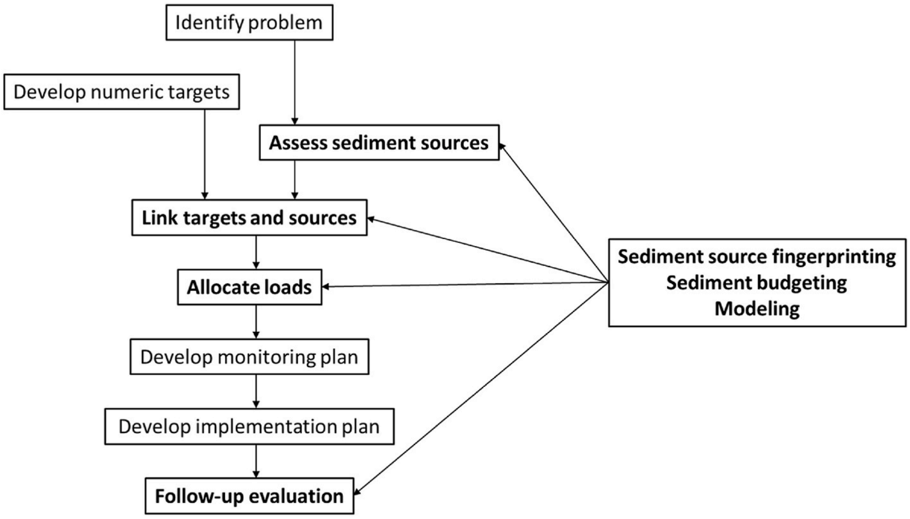

Fig. 4. A framework of general procedures of total maximum daily loads with sediment source fingerprinting techniques incorporated.

that the use of radionuclides is superior to other types of tracers in TMDL projects. Optimal cost-saving regional tracer suites will be based on land managers' needs and budgets for a streamlined application of sediment source fingerprinting (e.g., McKinley et al., 2013).

Appropriate implementation of BMPs are important means to achieve TMDL-specific targets. BMPs describe specific ways to manage agricultural, urban and forest lands and activities to mitigate non-point source pollution of surface and groundwater, in which sediment source fingerprinting can not only help target implementation locations but also provide a reliable way to evaluate BMP efficiencies. Sediment is often the main target, especially for forest and agriculture BMPs (Cristan et al., 2016; Liu et al., 2017). For example, surface soil erosion has been cited as the most important water quality concern related to forest practices in the United States (US-EPA, 2005), which could have profound effects on stream headwater environments as well as substantial effects on areas far downstream due to the capability of suspended sediment to travel exceptionally long distances. Accurate source assessments by sediment fingerprinting can greatly help land managers and policy makers to find the small areas of the landscape that would most benefit from implementation of conservation practices. More importantly, sediment fingerprinting offers a better way to evaluate the efficiency of implemented BMPs. Traditionally, BMPs efficiency for soil erosion reduction is evaluated based on monitoring at the outlet of a catchment (e.g., Xu and Xu, 2018), which provides an estimate of total sediment loading but includes no specific information regarding whether the change is due to sediment reductions from targeted sources. The use of sediment source fingerprinting in BMP evaluation could lead to a clearer sediment budget to better indicate relative changes in individual source contribution. Furthermore, only limited studies have explored BMP performance at a large watershed scale (Cristan et al., 2016; Liu et al., 2017; Xu and Xu, 2018), which could be attributed to the fact that large watersheds tend to have numerous sinks that may delay or attenuate the sediment signal from the land. If the area of storage is not permanent, substantial amounts of sediment could be flushed from the watershed and delivered downstream in extreme precipitation and runoff events, leading to BMP efficiencies being incorrectly evaluated. The use of radiometric fingerprinting (e.g., Belmont et al., 2014) or a combined application of sediment source fingerprinting and age dating techniques (e.g., Gellis et al., 2019) could quantify sediment storage time in channels and determine the lag time between BMP implementation and achievement of desired water quality goals, which may resolve this concern.

6. Implications of sediment source fingerprinting implementation in the MRB

Past sediment source fingerprinting research conducted within the MRB and described in this review shows that this technique is progressing from a research tool to a management tool, but with challenges to be overcome and knowledge gaps to be filled. Most studies included all necessary methodological steps for successful applications of sediment source fingerprinting, but the comparability of their results is limited by a lack of standardization in procedural steps. Reliable sediment source apportionment results were calculated by most studies with near-channel sources being reported as the dominant sediment sources in most cases, but only a few combined fingerprinting results with other lines of information to investigate sediment mass balance between inputs and outputs or the location of these sources. A wider application of the fingerprinting approach to the MRB for robust sourcing information is expected as this technique continues to advance. However, this approach is not currently an easily accessible management tool. Due to the small spatial scale of most papers (71%, Table S1) and the lack of investigations of the headwaters of most major tributaries as well as the Lower Mississippi River, it is currently hard to develop a full understanding of how sediment sourcing varies between different land use patterns or geographic units for the MRB. Although the findings of MRB studies have indicated that this technique is a reliable sediment source assessment approach to assist land and water resource management under current frameworks, in general the approach is still in a development phase with many research gaps needing to be addressed, such as the number of sediment samples needed for good representativeness, the impacts of digestion procedures on fingerprinting results, the use of appropriate correction approaches when using radionuclides in landscapes with deep incision, the applicability of conservatism-based tracer selection procedures in a natural environment, the relationship between particle size distributions and event magnitude, the suitability of different un-mixing models for different types of dataset or tracers, the functions of study design support in fingerprinting software, and definitive solutions for situations where the number of potential sources exceeds $n+1$ with the use of n different tracers. It is also worth mentioning that future method developments to resolve the above-mentioned issues could lead to a further divergence of fingerprinting procedural steps. This does not mean to discourage research advances but just to remind the fingerprinting community that it may result in a departure from the goal of applying this approach in a changing world to inform our understanding of sediment dynamics and provide reliable information for management needs (Kelly et al., 2017; Owens, 2022).

Lastly, it would be beneficial for study comparisons if every fingerprinting study could include a standardized reporting table summarizing key methodological steps (e.g., sediment source classification, source and target sample collection, tracer selection and analysis, assessment of tracer non-conservatism, tracer data correction, and source apportionment modeling) and fingerprinting results (e.g., upland vs near-channel sources). In summary, making advances in sediment fingerprinting research, standardizing the protocol for best practices, developing easy-to-use software for non-scientist end-users, and ensuring that fingerprinting is robust and reliable as it advances will help in using this technique for large-scale landscape conservation and restoration.

Author statement

Zhen Xu: Data curation, Investigation, Methodology, Visualization, Writing – original draft and editing. Patrick Belmont: Conceptualization, Funding acquisition, Resources, Supervision, Writing – review & editing. Janice Brahney: Conceptualization, Funding acquisition, Resources, Supervision, Writing – review & editing. Allen C. Gellis: Resources, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

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