

RESEARCH ARTICLE

A Quantitative Basis for Intersource Differences in Tracer Concentrations for Accurate Sediment Source Fingerprinting

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ABSTRACT

Sediment source fingerprinting provides support for targeted soil erosion control and land degradation mitigation. Tracer selection and source classification are the main considerations in this approach. However, the impact of tracer concentration divergences among different sediment sources on the accuracy of source apportionment remains uncertain and has not been quantitatively investigated. Here, artificial and virtual mixtures were created to explore the impact of low and high source differences on the accuracy of relative source apportionment. Additionally, global geochemical property data based on different source classification methods were collected to analyze the reliability of source classification. The results reveal that as the difference in tracer concentration between sources decreases (from 30% to 2%), the accuracy of source apportionment significantly decreased, with the mean absolute error (MAE) ranging from 5.9% to 60.1%. Furthermore, a relative deviation in tracer concentration > 10% between sources is suggested for reliable source classifications. Notably, approximately 40% of the existing papers exhibit potential issues due to low tracer concentration divergences between sources. The adoption of sediment source classification based on land use should be cautiously approached until tracer concentration divergences among major sources are examined. For catchments lacking long-term stable vegetation, source classification methods based on geological properties are recommended. Finally, we recommend the consideration of clustering methods or alternative tracers for specific study areas to improve the accuracy of sediment fingerprinting.

1 | Introduction

The worldwide occurrence of accelerated soil erosion poses significant challenges, including reduced land productivity, threats to water resources, and risks to human health (Amundson et al. 2015; Shukla et al. 2019). Sediment fingerprinting methods

have been widely used to identify potential sources of sediment caused by soil erosion (Haddadchi et al. 2013; Koiter et al. 2013b; Laceby et al. 2019). However, several uncertainties associated with source classification, sample collection, tracer type, unmixing model, particle sorting, and correction factors have been reported in recent publications (Collins et al. 2020;

Fathabadi and Jansen 2022; Owens et al. 2016; Pulley, Foster, and Antunes 2015). Differences in tracer concentrations between sediment sources are fundamental considerations to ensure accurate source apportionment (Collins et al. 2020; Evrard et al. 2022). However, quantitative studies on the key issue of source classification based on the differences in tracer concentrations are currently lacking, increasing the uncertainty in the source apportionment.

Selecting reliable tracers is crucial for effectively distinguishing between different sources. Several tracers, such as magnetic properties, geochemical elements, color characteristics, and biomarkers have demonstrated temporal stability and spatial variability in identifying sediment sources (Chen, Fang, and Shi 2016a; Latorre et al. 2021; Nascimento et al. 2022; Wallbrink, Roddy, and Olley 2002). However, the differences in tracer concentrations between sources are not always significant. In catchment areas characterized by geologic homogeneity, for instance, the geochemical elements from diverse sediment sources exhibit high similarities in their composition and concentration, resulting in weak discrimination (Evrard et al. 2022; Ramon et al. 2020). In addition, the large variability of the tracer concentrations within groups will reduce the source representativeness of the tracer (Pulley et al. 2017b). For example, regions characterized by frequent human activity, where fertilizer application, pesticide spraying, and human and livestock manures lead to high intra-group variability in tracer concentrations, affecting the accuracy of source apportionments (Foster and Charlesworth 1996; Zhou, Chang, and Zhang 2016).

Several source classification methods (e.g., land use, geology, soil type, subcatchment, soil depth, and topography) have been explored and applied in catchments of different sizes, with most being determined based on prior catchment investigations (Collins et al. 2010a; Koiter et al. 2013a; Upadhyay et al. 2020). Relevant reviews describe the majority of current studies on sediment fingerprinting, particularly in terms of land use for source classification (Collins et al. 2020; Haddadchi et al. 2013). Classifying sources by land use offers clear practical advantages; for example, predicting soil loss from cultivated land, pastures, or unpaved roads within a catchment area can guide the implementation of on-farm interventions (such as minimum tillage and no tillage) or serve as a reference for related management strategies (Collins et al. 2017). However, the effectiveness of this classification depends on the representation of different land uses by the chosen tracers. Small intergroup variability resulting from unreasonable tracer selection can potentially impede its utility (Horowitz and Stephens 2008; Pulley, Foster, and Collins 2017a). In contrast, other factors such as geology (Lacey and Olley 2015) or soil depth (Rode et al. 2018; Shu et al. 2024) may show sufficient variation across different spatial classifications. For example, geochemical tracers demonstrate significant concentration differences across various geological depths due to influences such as parent material, weathering processes, soil formation, and leaching (Grygar and Popelka 2016; Price and Velbel 2003). However, these significant differences in tracer concentrations between different geological/depth sources may be disrupted by specific changes within the catchment (Owens et al. 2016). For instance, wildfire-induced burnt soil in the catchment can result in the enrichment of nutrients and geochemical properties such as Pb, Zn, Ca, and Mg (Blake

et al. 2006; Muñoz-Arcos et al. 2021), increasing intrasource variability and reducing intersource differences. Therefore, source classification for specific research objectives is crucial to reduce uncertainty in sediment source fingerprinting.

The magnitude of differences in tracer concentrations between sources significantly affects the accuracy of sediment source apportionment (Pulley, Foster, and Collins 2017a; Vercruyssen and Grabowski 2018). When the tracer concentrations exhibit low differences between sources, source apportionment results are strongly influenced by the nonconservatism (or variability) of the tracer and sampling/measurement errors (Pulley, Foster, and Antunes 2015). For example, if there is only a 10% difference between two source groups, a small 5% variation may have a negative impact on the output result (Pulley, Foster, and Collins 2017a). Conversely, if the difference between two sources is greater than 100%, it is much more resistant to nonconservation (tracer variability) and error fluctuations. While statistical methods can always identify several “optimal fingerprint combinations”, small differences in tracer concentrations between intergroups may be ignored, potentially leading to low accuracy in predicting sediment sources. Therefore, quantitative investigations are necessary to guide researchers in assessing the reliability of source classification methods based on tracer concentration divergences between sources.

Artificial and virtual mixtures are considered reliable methods for accurately exploring sediment source fingerprinting (Derakhshan-Babaei et al. 2024; Gaspar et al. 2019; Gaspar et al. 2022; Gholami et al. 2019; Palazón et al. 2015). In addition to the advantage of low cost, these methods overcome the difficulty of obtaining comparable data (temporally and spatially) on sediment source contributions in the field environment (Batista, Lacey, and Evrard 2022; Gaspar et al. 2019). However, most current studies focus on validating unmixing models, tracers, and particle sorting effects through artificial and virtual mixtures (Gholami et al. 2019; Haddadchi, Olley, and Lacey 2014; Niu et al. 2022), often lacking a quantitative analysis of the impact of tracer concentration differences between sources on source apportionment accuracy. In this study, a determined range of tracer concentration divergences between sources corresponding to robust sediment source apportionment is tested based on artificial and virtual mixture experiments. Then, geochemical concentrations of different sources from existing studies were collected to evaluate the reasonability of sediment source classifications (Figure 1). This classification serves as a crucial factor influencing the accuracy of source apportionment in sediment fingerprinting. Finally, we provide some suggestions for the source classification to promote the further improvement of the sediment fingerprinting methods.

2 | Materials and Methods

2.1 | Artificial Mixing Procedures and Virtual Mixture Creation

The source soil used in the artificial mixing experiment was collected from three areas with differences in geology or land use in a catchment of Mizhi, Shaanxi Province (110°09' E, 37°54' N), China. Significant differences exist between loess

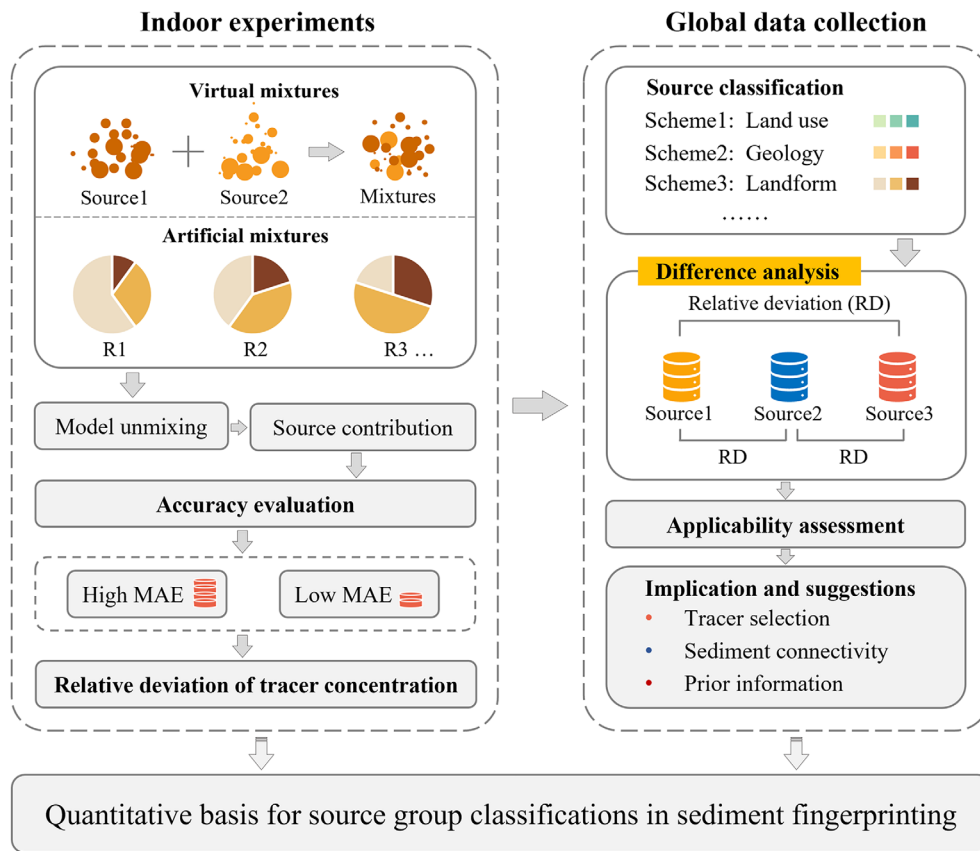


FIGURE 1 | The workflow for this study to explore rationality and applicability for different source classification schemes. The “R1, R2, R3...” represent different artificial mixing ratios. MAE represents the mean absolute error between the actual and predicted sediment source contribution. RD represents the relative deviation of tracer concentrations between any two sources. [Colour figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.com)]

TABLE 1 | Artificial and virtual mixtures creation for designed contributions of different sources.

Artificial mixtures				Virtual mixtures								
Mixture	Source contribution (%)			Mixing ratios (two sources)								
	Cropland	Gully	Paleosol									
R1	10	30	60	M1	2:8	M2	3:7	M3	4:6			
R2	20	40	40	M4	5:5	M5	6:4	M6	7:3			
R3	30	50	20	M7	8:2	—	—	—	—			
R4	33.33	33.33	33.33	Intersource difference of tracer concentration (%)								
R5	50	25	25									
				2	4	6	8	10	12	16	20	30

Note: The intersource difference refers to the relative deviation in tracer concentrations between any two sources, as shown in Equation (3).

(cropland and gully) and paleosol in structure and composition due to variations in depositional environments (Hou et al. 2020; Liu and Ding 1998). Paleosol was collected from a steep, vegetation-free gully. Both loess and paleosols are mainly formed by dust deposition (An et al. 1990). Paleosol is a soil layer formed during geological history due to changes in ancient climate (warm-humid climate) during the loess accumulation process, which was buried by later loess deposits (An et al. 1990). To exclude the impact of particle size on the apportionment accuracy, three particle size ranges < 125 (125–0), < 63 (63–0), and < 31 (31–0) μm were chosen, and the principle of selection is that most particles can be included in these ranges (Devereux et al. 2010; Laceby et al. 2017; Lizaga

et al. 2019; Sherriff et al. 2015). Considering difficulties in obtaining an adequate number of samples in the range of < 31 μm through dry sieving, all samples were obtained using a wet sieving method with a nylon sieve. Composite samples with known contributions were created by mixing samples from the three sources in different proportions. A total of five sets of composite samples (R1–R5) were prepared in the laboratory for each particle size fraction (Table 1), with three repeats in each set for laboratory determination. Ultimately, a total of 45 artificial mixtures were created. Artificial mixtures were created to compare the accuracy of source apportionments at both low and high intersource differences in tracer concentrations.

The apportionment accuracy of source contributions could be inconsistent for different tracer concentration divergences between sources when the measured mixture tracer concentrations exhibit a similar error caused by sampling or measurement. Virtual mixtures have proven to be reliable in assessing source apportionment accuracy due to their flexibility in generating combinations of different source proportions and low analytical cost (Gholami et al. 2019; Pulley and Collins 2018; Zhao et al. 2022). Therefore, we created virtual mixtures comprising two sources and two virtual tracers to systematically explore source apportionment accuracy under varying tracer concentration divergences between sources. Nine difference gradients (from low to high value) of tracer concentration between sources and seven mixing ratios (M1–M7) were set (Table 1). A total of 63 virtual mixtures were finally created. All of the mixture concentrations were added errors based on the ideal mixture tracer concentration calculated from known source ratios to simulate error fluctuation from field sampling and laboratory analysis, and the error remained consistent for each group of the mixture, around 1%.

2.2 | Data Collection

To explore the differences in geochemical concentrations across different sediment source classifications, global papers on sediment source fingerprinting using geochemical tracers were obtained. We have conducted a comprehensive search of all relevant research papers in English and Chinese. The databases Web of Science and China National Knowledge Infrastructure (CNKI) were searched, with the keywords (1) “sediment source”, “fingerprinting”, and “geochemical”, (2) “sediment”, “provenance”, and “geochemical”, and (3) “sediment”, “source apportionment”, and “geochemical” for the period 1995–2023. We collected a range of data from the identified papers, including the publication year, title, primary

author, study area name, location coordinates (latitude, longitude, and altitude), source classification, the optimal combination of geochemical elements, and corresponding mean and standard deviation of geochemical concentrations. To extract the geochemical data from digitized graphs, we utilized the GetData Graph Digitizer software (version 2.20). The extracted data are provided in the Supporting Information (Excel) in Data S1. Among the 217 retrieved papers, 78 contained information on geochemical properties (called valid data) from different classification sources (Figure 2). Land use, geology, subcatchments, soil depth (topsoil/subsoil), and landform types were included in source classifications.

In addition to the above geochemical data collected from 78 papers, we measured and analyzed different land use source samples from four catchments on the Chinese Loess Plateau. Specifically, the catchments are Shayangou (0.69 km²), Chabagou (205 km²), Dongjiaping (0.18 km²), and Hujiawan (27 km²). Notably, several sediment fingerprinting studies have been previously conducted in these catchments as part of our ongoing research (Chen et al. 2016b; Ni et al. 2019; Zeng et al. 2020). Each land use is confirmed through catchment surveys. Before collecting each sample, the surface debris and litter were removed and 10 surface samples of 0–5 cm were randomly collected and mixed evenly within a 5×5 m grid. More detailed catchment information and land use types are shown in Table S1.

2.3 | Sample Analysis and Unmixing

2.3.1 | Laboratory Analysis

In this study, all samples were dispersed to obtain particle size free of aggregates. Approximately 1 g of soil sample was placed in a container, and any fine roots or litter floating on

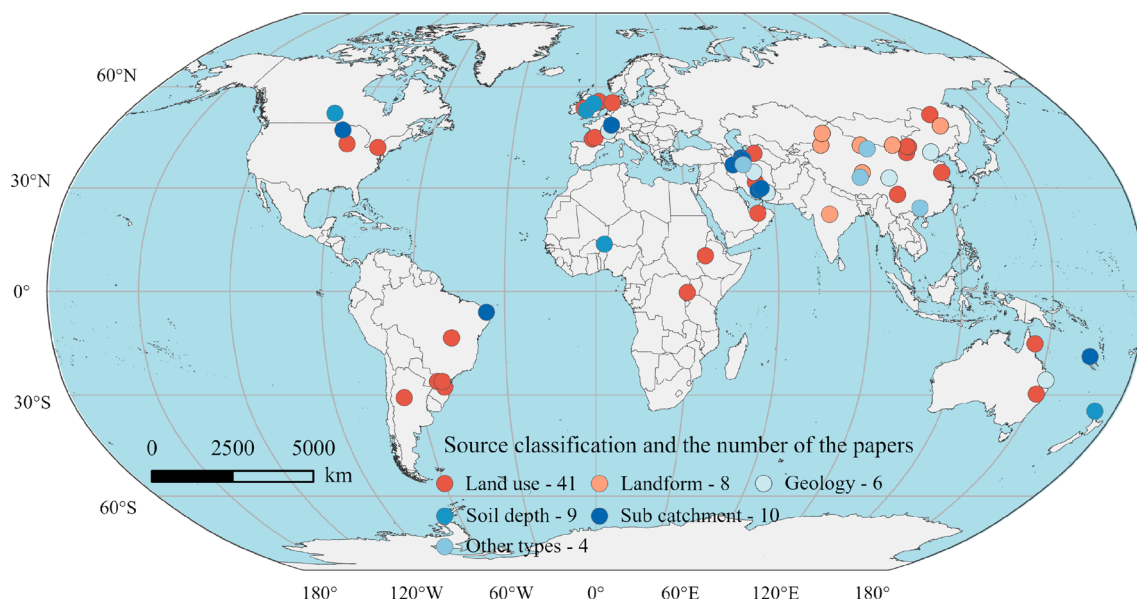


FIGURE 2 | Catchment location and source classification for sediment source fingerprinting based on geochemical properties in previous studies. The number after “-” represents the count of papers corresponding to different source classifications among the 78 papers, of which nine papers do not provide longitude and latitude information and are not marked on the map. Wiley acknowledges that the borders within the figure are subject to multiple territorial claims. [Colour figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com)]

the surface were removed after adding a small amount of water. The container was then heated (approximately 90°C) on an electric heating plate (Papuga et al. 2022), and 10% H₂O₂ was gradually added until all organic matter was removed. To eliminate soil carbonates, 10% hydrochloric acid was used, and the addition of the acid was halted when no air bubbles appeared in the container. The dispersed soil was then rinsed several times with distilled water until the solution was nearly neutral. Finally, 10 mL of 0.05 mol L⁻¹ sodium hexametaphosphate was added, and the dispersed soil particles were obtained by constant stirring. All processed samples were then transferred to the laboratory for particle size and specific surface area measurements using a Malvern Mastersizer 2000 laser particle size analyzer.

Samples with different particle size ranges were digested using the microwave digestion method, followed by the determination of geochemical concentrations in the laboratory using ICP-OES equipment. Detailed steps and methods can be found in the study of Dai et al. (2022). The geochemical properties include major elements (Al, Fe, K, Mg, Mn, P, and Ti) and trace elements (B, Ba, Bi, Ce, Cr, Dy, Er, Eu, Ga, Gd, Ho, La, Li, Lu, Mo, Nd, Ni, Pr, Pt, Rb, Sb, Sr, Sm, Tb, Tl, Tm, V, Y, Yb, and Zn). Throughout the experiments, several standard reference samples (GBW07408 (GSS-8), China) were added to verify the accuracy of the determination results (error 2.1%–4.0%).

2.3.2 | Optimal Tracer Combination

The selection of tracer groups exerts a significant impact on sediment source apportionment, whereby minimal intra-group variability and high intergroup differences can reduce the uncertainty of source apportionment (Collins et al. 2017). Generally, statistical analysis methods, including conservative test (range test), nonparametric Kruskal–Wallis H rank-sum (KW-H) test, and stepwise discrimination function analysis (DFA), can select the optimal combination of tracers (Collins, Walling, and Leeks 1997; Gellis and Noe 2013; Owens et al. 2016).

The fingerprint properties were first subjected to a range test to exclude those that may have been easily dissolved or redistributed (nonconservative) during sediment transport (Collins et al. 2020). Next, the KW-H test was used to select the fingerprint properties that showed significant differences among source categories, with *p* values less than or equal to 0.05. Fingerprint properties that did not meet this criterion were excluded. Finally, a stepwise DFA was used to evaluate the discriminant ability of the remaining fingerprint properties and determine an optimal tracer combination that maximizes the differentiation between source groups while using a minimal number of tracers.

2.3.3 | Estimating Source Contributions

Relative contribution of each source is quantified using an unmixing model based on mass balance equation. This equation minimizes the difference between the tracer concentration of sediment samples and the total concentration of sources with

varying proportions. The proportion of each source ranges from greater than 0% to less than 100%, with the total summing to 100% (Collins, Walling, and Leeks 1997). The unmixing model referenced here is the widely used model based on Collins, Walling, and Leeks (1996) and Walling (2005), specifically:

$$f = \sum_{i=1}^n \left(\left(C_i - \sum_{s=1}^m C_{si} P_s \right) / C_i \right)^2 \quad (1)$$

where C_i refers to the concentration of the *i*th tracer in the mixing sample, *n* refers to the total number of tracers selected, C_{si} is the tracer concentration in the *s*th source (*m* represents the number of sediment sources), and P_s represents the potential source contribution percentage estimated by the unmixing model.

The genetic algorithm (GA) in Matlab software was employed to obtain the optimum source contribution of sediment. GA simulates the evolutionary behavior of biological systems and excels at deriving the best-predicted source contributions through an adaptive iterative global search (Collins et al. 2010b). In contrast, methods like Excel Solver often get trapped in local search ranges, especially when the objective function has local extrema, leading to poor prediction results (Haddadchi et al. 2013). GA starts with a set of initial random source proportions and then iteratively advances through reproduction, crossover, and mutation (Collins et al. 2010b). To improve accuracy, we used 2500 iterations of Monte Carlo techniques based on Latin hypercube sampling instead of conventional random sampling (Collins et al. 2012; Haddadchi, Olley, and Laceby 2014).

2.4 | Accuracy Evaluation and Relative Deviation Calculation

The source apportionment accuracy of artificial and virtual mixtures here can be evaluated by comparing the model output contributions with actual contributions. The mean absolute error (MAE) can be used to quantify the deviation between the model and actual values, thereby assessing the apportionment accuracy (Haddadchi, Olley, and Laceby 2014).

$$MAE = \frac{1}{m} \left(\sum_{s=1}^m |X_s - Y_s| \right) \quad (2)$$

where X_s is the real sediment contribution from the *s*th source, Y_s refers to the predicted contribution from the *s*th source calculated by the unmixing model, and *m* represents the number of sources.

The relative deviation here reflects the difference in geochemical concentrations between any two sources, specifically expressed in Equation (3). When there are three or more target sources, the relative deviations of any two sources should be calculated separately to determine their differences.

$$R = \text{abs}(c_1 - c_2) / (c_1 + c_2) \times 100\% \quad (3)$$

R represents the relative deviation of geochemical concentrations between any two sources; c_1 and c_2 refer to the concentration of geochemical elements of the two sources, respectively.

3 | Results

3.1 | Quantitative Criterion for Tracer Concentration Divergences

Artificial mixtures were obtained from three sediment sources with different proportions (Table 1). To identify fingerprint properties with the highest discriminant ability, the range test, KW-H test, and DFA were conducted (Table S2–S5). The optimal fingerprint combination for the particle size range of $<125\mu\text{m}$ was determined to be Al, Ba, K, Ni, Sr., and Ti. For the $<63\mu\text{m}$ fraction, the optimal combination included Al and P, while for the $<31\mu\text{m}$ fraction, it comprised Al, Mn, P, V, and Y. Notably, the sources were correctly classified up to 100% for the three particle size ranges. The high MAE results indicated low source apportionment accuracy when classifying sources into three (Figure 3). Especially for R3 and R5 (particle size range $<63\mu\text{m}$), the MAE reaches the highest 24.3% and 38.5%, respectively (Figure 3). Further, low differences in geochemical concentrations were exhibited between cropland and gully, with relative deviations less than 10% (average 4.2%). In contrast, high differences were noted between paleosol and cropland (or gully) (Figure S1), with an average relative deviation of 24.4%. To explore the apportionment accuracy of geochemical properties in larger concentration divergences, cropland and gully with small concentration divergences were combined to form loess. The optimal fingerprint combination of the two sources was consistent with that of the three sources. Significantly low MAE can be observed for all five mixed percentages (R1–R5) and three particle size ranges when three sources are recombined to form two sources (Figure 3). The maximum MAE is 9.2%, and the minimum is 0.93%.

The accuracy of source apportionment was found to be significantly influenced by the difference in tracer concentrations between sources when the tracer concentrations of the virtual mixtures were set to a close error range (fluctuating around

1%) (Figure 4). Generally, as the differences between sources increase, the accuracy of source apportionment tends to improve. When the tracer concentration divergences are below 5% between sources, the MAE value exceeds 40% (43.9%–60.1%). Additionally, as the difference increases from 6% to 16%, there is a corresponding decrease in the MAE value from 29.2% to 12.0%. Eventually, when the difference reaches 30%, the MAE value further decreases to 5.9%. These results suggest that minor fluctuations (due to sampling and analysis errors) in the tracer concentration values of mixtures can significantly impact the source apportionment results, especially when differences in tracer concentrations between sources are small. This finding is also supported by previous studies (Pulley, Foster, and Collins 2017a). Additionally, previous artificial mixture experiments have shown that MAE values above 20% between predicted and true contributions are not acceptable for identifying reliable sediment sources (Du et al. 2019; Haddadchi, Olley, and Laceby 2014; Nosrati and Collins 2019; Shi et al. 2021; Vale et al. 2022). In our study, Figure 4 reveals a tracer concentration divergence of less than 10% between different sources, corresponding to an MAE greater than 20%. Therefore, it is considered that when the relative deviation of geochemical concentrations between two sources is close to or below 10%, lower apportionment accuracy is likely to occur.

3.2 | Source Apportionment Accuracy for Different Source Classifications

The majority of studies (41 papers) selected land use as the primary method for source classification among the 78 extracted papers analyzed. When evaluating the relative deviations of geochemical concentrations between various sources, a significant portion of these papers (20 out of 41) showed low differences between at least two sources when classified based on land use. Notably, the relative deviation values observed were predominantly below or close to 10% (Figure 5). In addition, the four catchments on the Loess Plateau selected for this study showed generally low differences in geochemical concentrations among different sources when the sources were classified by land use. Especially for the Hujiawan catchment, all elements selected by DFA displayed close concentration values across the

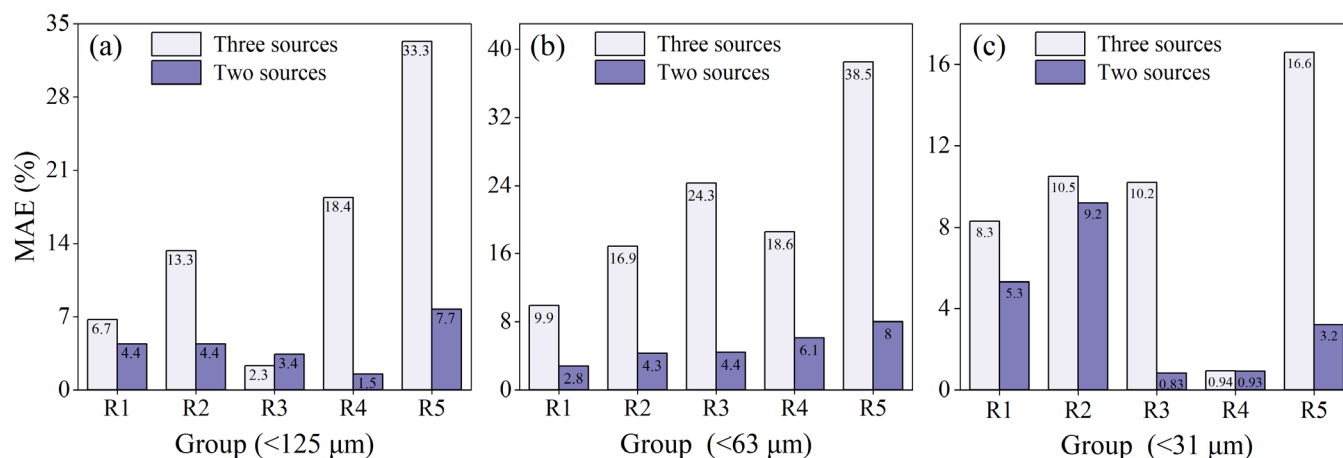


FIGURE 3 | The MAE values for three sources (cropland, gully, and paleosol) and two sources (paleosol and loess) of three particle size ranges (<125 , <63 , and $<31\mu\text{m}$). [Colour figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com)]

five land uses (Figure S2). Furthermore, the low relative deviation values among the different sources also reflect the small differences in the geochemical concentrations of different land uses in the selected catchments (Figure 6). Previous studies on

source classifications did not consistently exhibit high source differences (Supporting Information (Excel) in Data S1). For example, some geochemical concentrations exhibited low relative deviation values when classifying sources by geological

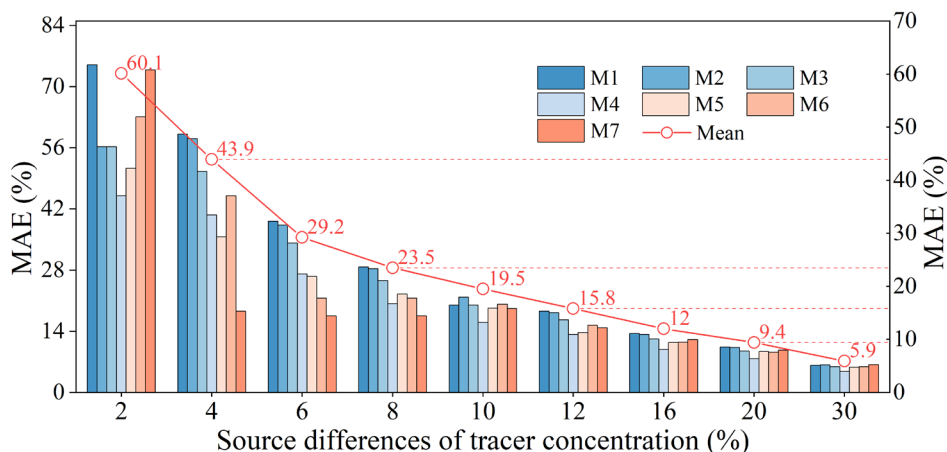


FIGURE 4 | The source apportionment MAE values corresponding to the nine source difference groups of tracer concentration, and a similar fluctuation range (error range) was set for all nine source difference groups. M1, M2, M3, M4, M5, M6, and M7 represent seven virtual mixing ratios, 20% and 80%, 30% and 70%, 40% and 60%, 50% and 50%, 60% and 40%, 70% and 30%, and 80% and 20%, respectively. [Colour figure can be viewed at wileyonlinelibrary.com]

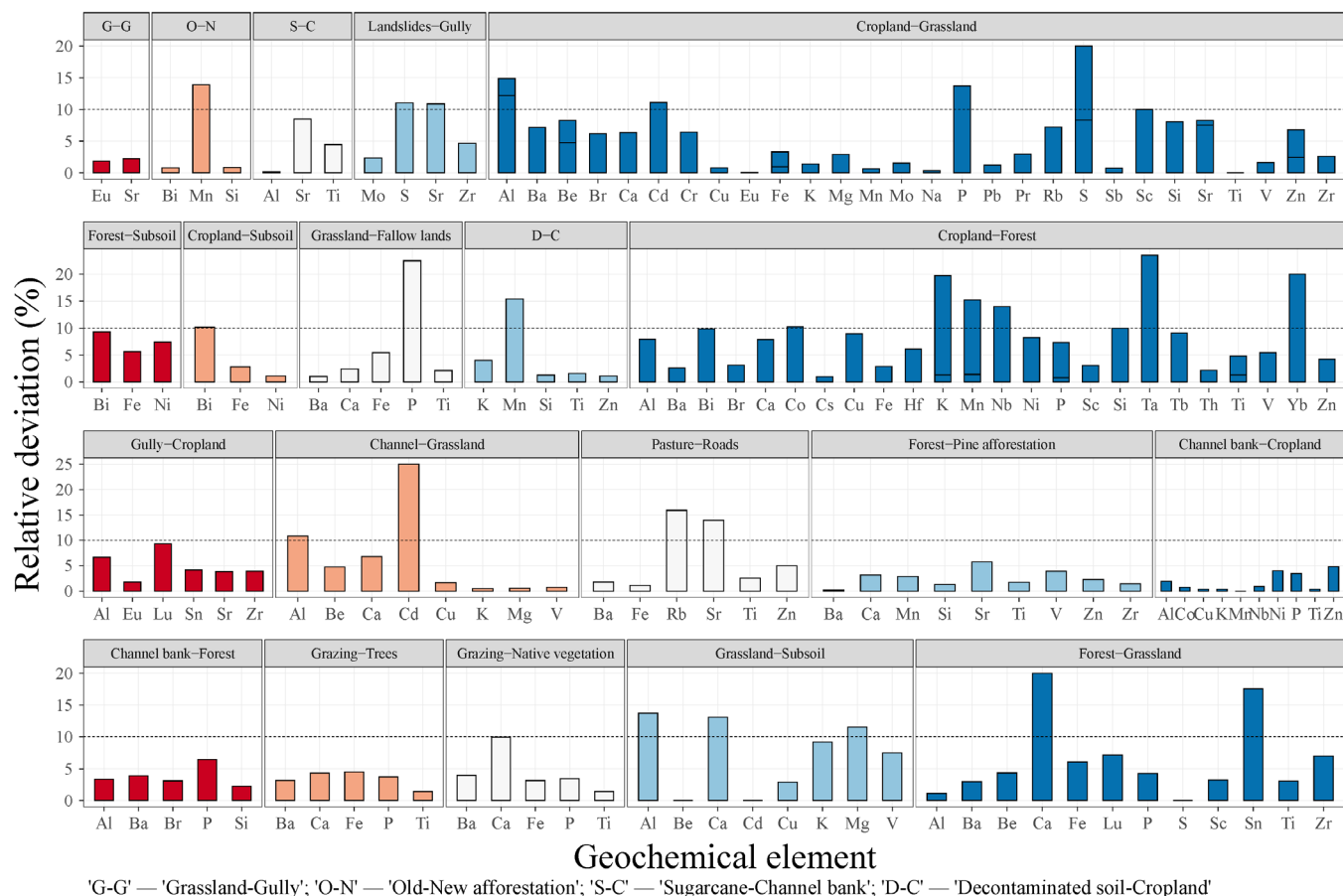


FIGURE 5 | The results of low relative deviation in geochemical concentrations between each of the two sources when classifying sources by land use (evaluation of published studies). The geochemical elements shown in the figure represent the optimal tracer combinations selected by statistical methods. The dotted lines parallel to the horizontal axis represent a relative deviation of 10%. [Colour figure can be viewed at wileyonlinelibrary.com]

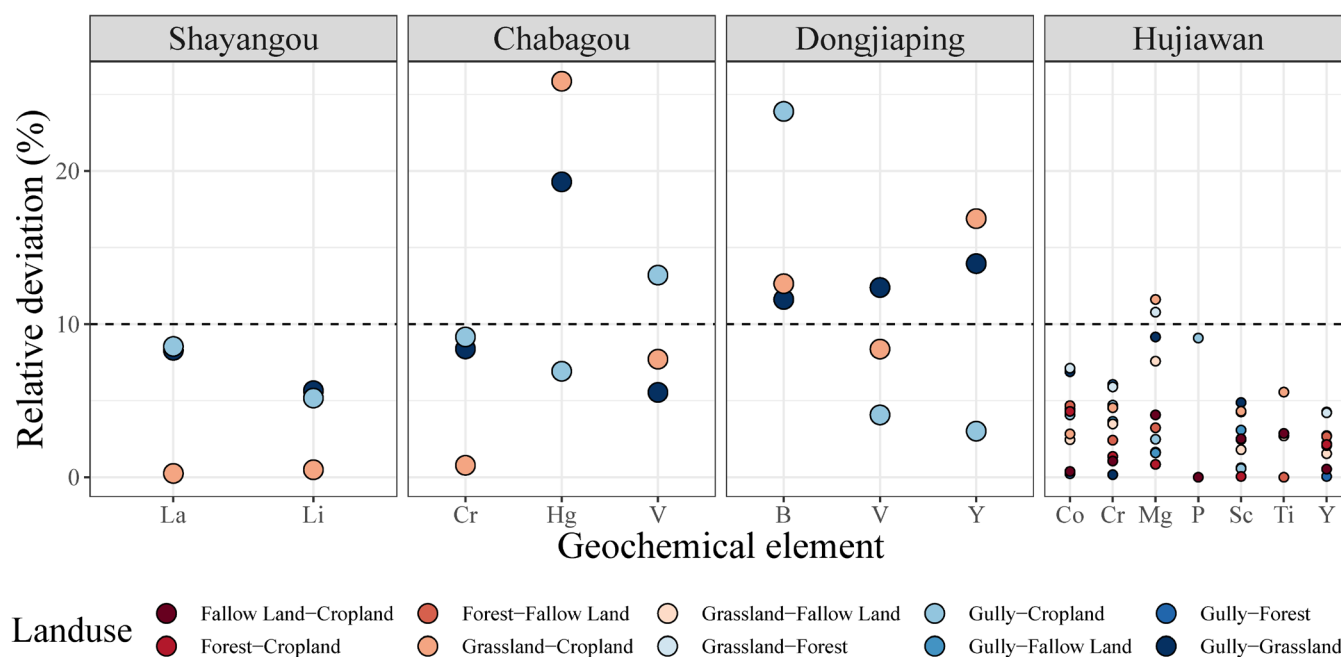


FIGURE 6 | The relative deviation (%) of geochemical concentrations between any two sources in the four catchments (Shayangou, Chabagou, Dongjiaping, and Hujiawan). The geochemical elements of each catchment represent the optimal combination selected by KW-H test and DFA. [Colour figure can be viewed at wileyonlinelibrary.com]

properties, including granite zone, limestone, and shale zone (Zhang et al. 2021) (Figure 7). In particular, limestone and shale sources have a low relative deviation in geochemical concentrations between them, with a minimum of 0.3% and an average of 8.5%. In addition, in regions (reported by other studies) with large areas of dune, sediment sources are classified by surrounding sandy land or mountains, not all showing great differences (Song et al. 2022; Wu et al. 2023). The results of the relative deviations of geochemical concentrations under all source classifications are presented in the Supporting Information (Excel) in Data S1. From the relative deviation results, we found that 31 out of 78 papers (approximately 40%) showed low intersource tracer concentration differences ($< 10\%$) for at least two sources. Alternative source classifications or tracers should be considered if sediment fingerprinting studies are conducted in these study regions in the future.

4 | Discussion

4.1 | Error Analysis for Different Tracer Concentration Divergences

In artificial mixture experiments, the MAE of real and predicted contributions showed high values when there were three sources. This indicates a low accuracy in predicting the individual source contributions. In contrast, the combined two sources showed high accuracy for five mixing ratios and three particle sizes (Figure 3). The low tracer concentration divergence (relative deviation $< 10\%$) between source 1 and source 2 (Figure S1) resulted in low accuracy of source apportionment. Therefore, source discrimination analysis should be performed as part of the preparatory data analysis before source unmixing. In addition to assessing discrimination capability through DFA, it is

also essential to consider the relative deviation in tracer concentrations between sources. When sources are classified into three types, a high MAE value may be caused by a low concentration difference in the tracers between two of the sources. By implementing three source classification schemes, Pulley, Foster, and Collins (2017a) verified that low intergroup differences of tracer concentrations would significantly increase the uncertainty of the source apportionment. Qualitative descriptions are also mentioned in the review of Collins et al. (2017), noting that small tracer concentration differences between source groups can generate large uncertainties in source apportionment. Further, the virtual mixtures were created to quantitatively evaluate the impact of source differences on source apportionment accuracy (Figure 4). Although all tracers were conserved, small error fluctuations in the mixed tracer concentration resulted in large MAE values when the relative deviation between the sources was less than or close to 10%. This demonstrates that the accuracy of source apportionment is substantially impacted by fluctuations in mixed tracer concentrations, particularly for the case of low intersource tracer concentration differences.

4.2 | Rationality of Source Classification Methods

One of the key aspects influencing the accuracy of potential sediment source apportionment is the rationality of source classification. Generally, the source classification is based on study objectives and targeted soil erosion issues, while ensuring maximum tracer differentiation (Collins et al. 2017). Nevertheless, a few instances of unreasonable source classification can be observed after analyzing the results of the 78 extracted papers (Figure 5). The differences in soil geochemical concentrations between different land uses may be determined by the type of vegetation and human management of the land (Aleksiev

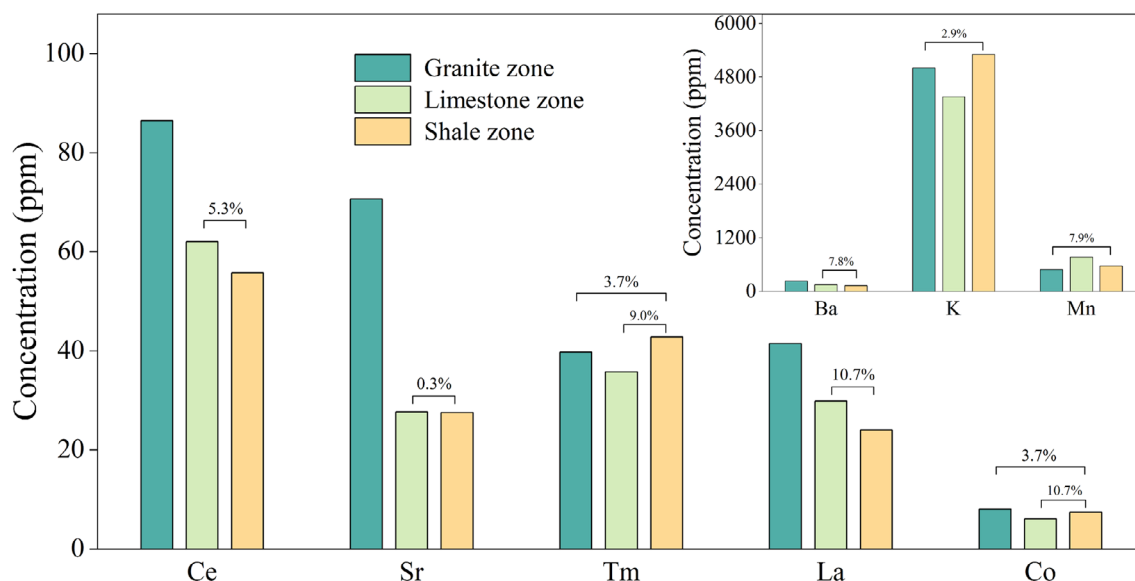


FIGURE 7 | The difference of geochemical concentration classified by geological properties for published study. The values on the bar chart represent the relative deviation between the two sources. [Colour figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com)]

et al. 2018; Santorufo et al. 2021; Tellen and Yerima 2018). For example, the Fe, Si, Al, and other mineral components of cropland, forest, and grassland show differences due to mineral transformations and the redistribution of mineral components (Alekseev et al. 2018). In addition, fertilization, irrigation, and pesticide application cause differences in the soil geochemical concentrations accordingly (Bai et al. 2010; Tilman et al. 2002). However, not all land uses showed significant differences in the geochemical concentrations to enable accurate source apportionments (Figure 5). A variety of factors, such as parent material of soil and colluvium, pedogenic processes, and human activities may influence the levels of tracer concentrations in a source group based on land use (Collins et al. 2017). For instance, catchments experience frequent changes in vegetation cover, where cultivated land is abandoned and reclaimed alternatively (Li et al. 2022; Madelene and Chen 2006; Molinillo, Lasanta, and Garcia-Ruiz 1997). As a result, it is difficult to maintain stable soil geochemical concentrations. Additionally, in areas like the Chinese Loess Plateau, where special pedogenesis (eolian soil) and uniform lithology prevail, the geochemical concentrations of slopes and channels also show slight differences. This is evident in the selection of four catchments on the Loess Plateau (Figure 6). In contrast, several catchments in UK, where land use is a reasonable way to classify sources as cropland, grassland, woodland, pasture, and channel bank, exhibit significant differences in geochemical concentrations (Blake et al. 2018; Blake et al. 2012; Collins et al. 2013; Cooper et al. 2015; Smith, Karam, and Lennard 2018). The main reason may be that land use in these regions is less dynamic, with the same type of vegetation coverage maintained for prolonged periods. Land use is a reasonable way to classify potential sediment sources, and it plays a crucial role in clarifying the different degrees of soil erosion caused by human activities. However, to classify sources based on land use, it is necessary to consider the age of vegetation cover and the spatial homogeneity of soil properties within the study area carefully. In general, classification based on land use has significant advantages, including the development of targeted prevention and control strategies (e.g., reduced tillage

or compaction management) according to the erosion characteristics specific to each land use (Collins et al. 2017). Furthermore, classifying sources by land use allows for efficient identification of agricultural pollutant sources, thereby mitigating the adverse effects of agricultural practices on the environment (Tiecher et al. 2017). Importantly, matching the appropriate tracer type with the source type is crucial for enhancing source discrimination. Tracers may perform worse if their inherent characteristics and environmental basis are not properly considered. For instance, compound-specific stable isotopes (CSSI) are less effective in areas with low soil organic matter content. In contrast, when the soil organic matter content is high, CSSI exhibits greater sensitivity and can more effectively discriminate among different sources (Vale et al. 2022).

Classification based on geological properties has proven to be effective in identifying distinct sediment sources (Lacey et al. 2015a). However, a low difference can be observed in optimal fingerprint (Ce, Sr, Tm Co, Ba, K, and Mn) concentrations when comparing limestone and shale sources in the published study (Figure 7). Additional attention should also be paid to source classification based on soil depth. For instance, Pulley, Foster, and Collins (2017a) classified sources based on soil depth (surface and subsurface), which showed low differences in concentrations of Li, Mo, Sr, and V (relative deviation was 1.6%–4.4%). Usually, the elemental content and distribution differ significantly between topsoil and subsoil due to natural processes like redox, leaching, mineral decomposition, and anthropogenic activities such as fertilization, pesticide spraying, and mining (Duan et al. 2015; Fan et al. 2020; Šajn et al. 2011). However, in areas with low human disturbance and homogeneous geology, the geochemical differences between topsoil and subsoil are weak, making it an unreasonable method for source classification.

In catchments spanning hundreds to thousands of square kilometers, conventional source classification methods may face limitations due to spatial complexities of geology, vegetation,

TABLE 2 | The relative deviation (%) of geochemical concentrations between different sources based on sources classification of subcatchments.

Relative deviation (%)								
Sources	Li	Al	Ba	P	Na	Fe	References	
Sub1-sub2 basin	0.23	7.89	8.83	9.59	3.58	4.70	Raigani, Nosrati, and Collins (2019)	
Sub1-sub3 basin	5.16	5.78	1.66	3.79	8.83	4.24		
Sub2-sub3 basin	5.39	2.12	10.47	13.33	5.27	8.92		
Sources	Cr		Th		Bi		Pr	References
Northern-central sub-basin	15.00		2.91		11.88		5.69	Habibi et al. (2019)
Northern-southern sub-basin	4.38		7.54		1.84		5.40	
Central-southern sub-basin	10.69		10.42		10.06		0.29	

and sediment transport deposition (Dabrin et al. 2021; Nosrati et al. 2021). As such, subcatchment classification provides another way of thinking (Fatahi et al. 2022; Kelley et al. 2006; Sellier et al. 2021). Source apportionments for subcatchments are manager-friendly (Collins et al. 2017; Collins et al. 2013) and can reduce intrasource variability, especially for large basins (> 1000 km²) with numerous and variable sources. Nevertheless, the geochemical concentrations among different sources across various subcatchments in several studies showed little difference (Habibi et al. 2019; Raigani, Nosrati, and Collins 2019), as indicated by the corresponding results of relative deviations in Table 2. This low source difference may be due to the close mean values of geochemical concentrations between subcatchments. In addition to conventional source classification methods, other methods classify sources based on geomorphic features, and several studies combine land use and geomorphic features, such as agricultural land and alluvial fans (Niu et al. 2020; Nosrati et al. 2022). While these classifications primarily rely on field surveys, it remains important to quantify whether the selected geochemical elements can accurately distinguish different sources.

4.3 | Implication and Suggestions

A careful evaluation of the geochemical elements is necessary to determine their applicability in the study area when performing source classification. First, rigorous investigation and meticulous recording of critical information, including land use, geology, and soil depth, are essential to enhance the feasibility and significance of the research. Then, a quantitative analysis of inter-source differences is necessary to reduce uncertainty in source apportionment. Clustering methods can be utilized to amalgamate similar sources, such as combining cropland and grassland into a single category referred to as “slopes” (Collins 2015; Collins et al. 2017). This posterior clustering method can obtain new source proportions and ensure the reliability of clustering (Stock et al. 2018). When geochemical properties alone cannot distinguish between different sources, alternative tracers should be considered. These may include soil biological information (n-alkanes, sterols, lignin, and DNA) strictly dependent on land use, as well as carbon and nitrogen stable isotopes and CSSI (Bianchi, Mitra, and McKee 2002; Chen, Fang, and Shi 2016a; Frankl et al. 2022; Hirave et al. 2021; Laceby et al. 2015b; Reiffarth et al. 2016).

In some cases, the inclusion of prior information is necessary for sediment source classification. For example, assigning a higher weight value to areas experiencing severe soil erosion can reduce uncertainty caused by within-source group variability and improve apportionment results (Wilkinson et al. 2015). However, determining prior information requires a rigorous investigation of the catchment, including an assessment of erosion patterns under varying rainfall conditions and the upper boundary for source contributions (Fryirs 2013; Owens et al. 2016; Tiecher et al. 2017; Upadhayay et al. 2020).

In this study, we focus on the impact of differences in tracer concentrations between sources on the accuracy of sediment source apportionments. However, within-source variability of tracers is also an important factor contributing to source apportionment uncertainty (Collins et al. 2020; Pulley, Foster, and Collins 2017a; Wilkinson et al. 2015). Specifically, low within-source variability reflects high sample representativeness, whereas high within-source variability reduces the reliability of source apportionment. Future research should further explore the potential impact of within-source variability on the accuracy of source apportionments. Additionally, in complex field catchments where more sources may be included in source groups, it is important to quantitatively assess the impact of tracer concentration differences among sources on the accuracy of source apportionments. Considering the difference in tracer concentration between any two sources among multiple sources will greatly ensure the accuracy of source apportionment.

5 | Conclusions

This study highlights that source classifications based on differences in tracer concentrations ensure the reliability and robustness of the source apportionment results in sediment fingerprinting. Low differences in tracer concentrations between sources result in significant source apportionment errors, while higher differences (exceeding 10%) enhance the accuracy of source apportionment and improve resistance to uncertainty caused by sampling and analysis errors. Unfortunately, a significant proportion (at least 40%) of existing studies have not adequately addressed these errors. Land use classification proves valuable for targeted catchment management as it provides information on potential soil erosion linked to human activities. However, classifying sources based on land use alone can

sometimes lead to erroneous apportionments due to low differences in geochemical concentrations. Rigorous catchment surveys are necessary to ensure that each land use remains stable over time in the study area. Alternative methods for classifying sources based on catchment characteristics, including geology, landform, soil depth, and subcatchment, can also be effective in predicting sediment source contributions.

In areas with uniform geological properties or human disturbances, the differences in geochemical concentrations between sources may be insignificant. Therefore, it is crucial to explore alternative tracers across different temporal and space scales to ensure the reliability of sediment fingerprinting. These tracers may involve soil biological information (n-alkanes, sterols, lignin, and DNA) that is strictly dependent on land use, as well as carbon and nitrogen stable isotopes and CSSI. Furthermore, the combination of multiple tracers is worth considering to improve the robustness of source discrimination.

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Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Supporting Information

Additional supporting information can be found online in the Supporting Information section.