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Comparative study using spectroscopic and mineralogical fingerprinting for suspended sediment source apportionment in a river–reservoir system

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Abstract

The need to control soil erosion has received increasing attention, but quantitative data on the sources of suspended sediment in many river–reservoir systems is still lacking. The goal of this research was to compare the application of spectroscopic [mid-infrared (MIR)] and mineralogical [X-ray diffraction (XRD)] fingerprints for assessing relative sediment source contributions from different land use groups (agricultural lands, forests and human settlements) in the Konar–Damodar river–reservoir system in India. Source apportionment was estimated using partial least square (PLS) regression for spectroscopic tracers (MIR) and the Bayesian MixSIAR model for mineralogical tracers. Both methods identified differences between the pre- and post-monsoon sediment contributions of forests (overall contribution bounds of ∼35–43%). During monsoon seasons, both fingerprinting methods indicated agricultural land use as the primary source of suspended sediment. Although there were some temporal variations in the predicted contributions of the land use sources, the MIR-PLS and mineralogical–MixSIAR methods produced comparable ranges. The respective variations in contributions, using MIR-PLS and mineralogical–MixSIAR, were ∼31 to 66% compared with ∼36 to 61% for agricultural lands, ∼21 to 43% compared with ∼15 to 39% for forests and ∼16 to 37% compared with ∼19 to 32% for human settlements.

KEYWORDS
mineralogical fingerprints, MIR-PLS regression, MixSIAR, river–reservoir, sediment source tracing, spectroscopic fingerprints, uncertainty

1 | INTRODUCTION

Accelerated soil erosion and sediment delivery are considered one of the major concerns in all river basins globally due to adverse on-site and off-site consequences (Das et al., 2022). The on-site consequences include (i) decreased soil productivity due to removal of topsoil, (ii) reduction in the ability of soil to store water, (iii) exposure of subsoil with poor physical and chemical properties, (iv) loss of newly planted crops and (v) siltation in low lying areas (Schoorl & Veldkamp, 2001). Suspended sediment is among the most common pollutants in streams, rivers and lakes (Issaka & Ashraf, 2017). Sediment can indeed be both a pollutant and a natural component of the catchment system, serving essential ecological functions while also posing environmental challenges. The most adverse off-site effects of sediment in water bodies include (i) damage to aquatic biota, (ii) loss of reservoir storage capacity and (iii) degraded functioning of navigation routes and hydraulic structures. Therefore, reliable quantitative information on suspended sediment sources is required for implementing effective mitigation strategies (Collins et al., 2017).

The Chota Nagpur plateau is recognised as one of India's most vulnerable areas to water-driven soil erosion due to its predominantly water-eroded laterite soil. Soil degradation in this region is driven by geological formations of granite gneiss, underdeveloped soil profiles, undulating plateau topography with occasional hills and anthropogenic factors such as ongoing agricultural expansion, excessive groundwater extraction and irrigation (Mahala, 2017). Changes in land use, deforestation and inadequate soil and land management practices have further increased the region's susceptibility to soil erosion (Mahala, 2018). The study reported herein was conducted on a representative catchment of the Chota Nagpur plateau (i.e. the Konar River catchment).

Recently, significant attention has been directed towards using physical models to address sedimentation issues. For example, Das et al. (2019) discussed future directions for sediment distribution and transport modelling in river–reservoir systems. Comprehensive reviews in the literature highlight the growing application and scope of sediment source fingerprinting (Collins et al., 2017, 2020; Guan et al., 2017; Koiter et al., 2013; Owens et al., 2016; Smith et al., 2013). Although sediment distribution and transport models, such as USLE and its modified versions, EROSION-3D, WEPP and many others (Pandey et al., 20162016), are advancing, they often lack the ability to evaluate the proportional yield contributed by various sediment sources. Despite these technological advancements, the literature increasingly emphasises the need for direct methods (Collins et al., 2017). Few methods can directly identify, differentiate and quantify unique sediment sources in a catchment with minimal prior information, such as expert knowledge and semi-quantitative data (Davis & Fox, 2009). Sediment fingerprinting is a powerful technique used to identify and quantify the sources of sediment within a catchment. This method is advantageous in terms of using prior knowledge about catchment characteristics (i.e. land use, soil types, geology and topography) and erosion sources to guide sampling and the interpretation of results. By analysing the unique properties of sediment from various potential sources, source fingerprinting can be used to trace the origins of sediment found at the catchment outlet (Collins et al., 2020). Critically, sediment source fingerprinting links the sampled target sediment directly to the key sources without the need to explicitly quantify the intervening parts of the fine sediment delivery cascade and transportation to river channels (Collins et al., 2017).

The factors influencing soil erosion and sediment transport include inherent controls such as geology, geomorphology and pedology. In addition, sediment generation and transport are influenced by extreme weather, runoff patterns and human-induced land use change.

Discrimination of potential sediment sources has been accomplished using various combinations of properties or tracers. Techniques such as spectroscopy have been utilised to distinguish sediment sources based on their spectral characteristics. For instance, studies by Ni et al. (2019) and Tiecher et al. (2017) demonstrated how variations in spectral data can reveal differences in sediment origins. Geochemical analysis identifies unique chemical signatures of sediments from different sources. This method, used by Bahadori et al. (2019) and Nosrati and Collins (2019), leverages the distinct geochemical fingerprints that different environments impart on sediments. The mineral composition of sediments provides distinguishing features for source discrimination. Research by Sisinggih, Sunada and Oishi (2006) and Srivastava, Khare and Ingle (2011) highlighted how mineralogical profiles can effectively differentiate sediment sources. Radiometric tracers, such as isotopic signatures, have been used to trace sediment origins. Studies by Kim et al. (2013) and Navratil et al. (2012) utilised radiometric properties to provide insights into sediment provenance. Biological markers, including DNA and other biological residues, have the potential to differentiate sediment sources. Kraushaar et al. (2021) explored how biological properties can offer unique identifiers for sediments from various sources.

Similarly, different mixing model structures have been used for source apportionment studies for generating relative proportional quantification of sediment sources. Research by Collins et al. (2017, 2020) and Habibi et al. (2019) applied these models to accurately attribute sediment to its sources. Explicit estimation of uncertainties associated with the results is crucial for robustness and reliability. Gaspar et al. (2019) emphasised the importance of incorporating uncertainty estimation to enhance the credibility of source apportionment findings. By integrating these diverse properties and advanced modelling approaches, researchers can effectively identify and quantify sediment sources, providing valuable insights for catchment management and sediment control strategies.

Although most previous fingerprinting studies have used geochemical fingerprints to determine sediment sources (Wadman et al., 2017), the use of spectroscopic approaches has gained attention. Conversely, some initial studies showed that traditional fingerprints give more reliable results than spectral fingerprints (Tiecher et al., 2015), more successful spectroscopic fingerprinting studies have been conducted focusing on aspects including discrimination of land use classes, sub-catchments and lithological units in drainage basins (Legout et al., 2013; Poulenard et al., 2012; Tiecher et al., 2017).

The application of mineralogical fingerprints for tracing sediment sources is a traditional qualitative method for sediment source apportionment (Ramon et al., 2020; Rowntree, van der Waal, & Pulley, 2017). Mineralogical fingerprints are based on a unique set of mineralogical characteristics or profiles that can be used to identify the origin of sediment. These fingerprints are based on the specific minerals present in the sample, their relative abundances and their physical and chemical properties. Soil erosion from different land use types varies significantly due to their distinct physicochemical properties. Consequently, XRD analysis for mineralogical characteristics can be utilised to differentiate the relative contributions of various land use classes to sediment samples. Although the application of XRD spectroscopy to understand land use–related sediment dynamics in catchments is limited, this study aimed to investigate the influence of major land use classes (Das et al., 2023). There is a huge potential provided by the spatial and profile variability of soil mineralogy for conducting fingerprinting studies, as minerals constitute ~70% of river sediments by weight (Hillier, 2001). Mineralogical fingerprinting studies using commonly found minerals in soil such as quartz, mica, feldspar and calcite have been conducted and established satisfactory links between sources and target sediments (Eberl, 2004; Nath et al., 2007). The Damodar River basin is one of the mineral-rich areas of India, consisting of granites and granitic-gneisses of the Archeans, sandstones and shales of the Gondwanas and recent alluvial deposits (Singh et al., 2005). The Konar River catchment situated in the upstream portion of the basin consists of granites comprising quartz, mica and feldspar (Singh & Hasnain, 1999). The diversity of minerals in this study catchment therefore justifies the application of mineralogical fingerprinting to discriminate the sources of sediments therein.

Here, reliable variations in soil mineralogical composition due to different land uses have been found (Jeong, 2001), which further supports its application in sediment source fingerprinting.

Although other researchers have employed spectroscopic and mineralogical fingerprinting methods to establish relationships between target suspended sediment samples and their sources, a comparative study is still lacking in this context. Only a few studies have used more conventional geochemical methods to assess the results of spectroscopic analysis (Ni et al., 2019; Verheyen et al., 2014). There are comparative studies conducted on land use classes with both spectroscopic and conventional geochemical tracers, which have reported some disparities in the results (viz. Ramon et al., 2020; Verheyen et al., 2014), whereas studies including those by Tiecher et al. (2017, 2016) found the two approaches to be in good agreement. Here, explicit consideration of different mineralogical groups (viz. bulk minerals, clay minerals and heavy minerals) for such comparisons can also provide crucial information on sediment sources (D'Haen, Verstraeten, & Degryse, 2012).

Based on the above context, our research herein aimed to determine whether the sediment fingerprinting technique can effectively distinguish between different land uses that contribute to sedimentation. The major assumptions of our sediment fingerprinting study were as follows: (1) potential sediment sources (i.e. the land use classes) can be discriminated based on composite spectral and mineralogical signatures obtained from MIR and XRD spectroscopy; (2) these properties determined for both sources and target reservoir sediments provide a robust basis for apportioning the contributions of the land use classes to the sampled target sediment; and (3) the spectral and mineralogical tracers exhibit conservative behaviour because they retain their unique signatures from their source areas without undergoing significant alteration during transport and deposition. This tracer conservation is crucial for accurately identifying and differentiating sediment sources in sediment fingerprinting studies.

An important aspect of any sediment fingerprinting study is the selection of mixing models. The combination of partial least square (PLS) regression with spectroscopic methods is very well established (Legout et al., 2013; Poulenard et al., 2012, 2009) and the application of Bayesian models for quantitative unmixing of sediment sources using geochemical tracers has also received very wide attention in recent decades (Cooper et al., 2015; Huangfu et al., 2020). Hence, the specific objectives of this study were to explore (1) the applicability of spectroscopic and mineralogical tracers for discriminating land use classes based on sediment contributions and (2) the potential of PLS and Bayesian modelling techniques combined with spectroscopic and mineralogical tracers, respectively, to quantify land use source contributions in different seasons.

2 | MATERIALS AND METHODS

The key stages in the methodology adopted in this study are summarised into six main steps in Figure 1. Both modelling techniques (PLS regression for spectroscopic tracers and Bayesian mixing modelling—MixSIAR; Stock & Semmens, 2018) for mineralogical

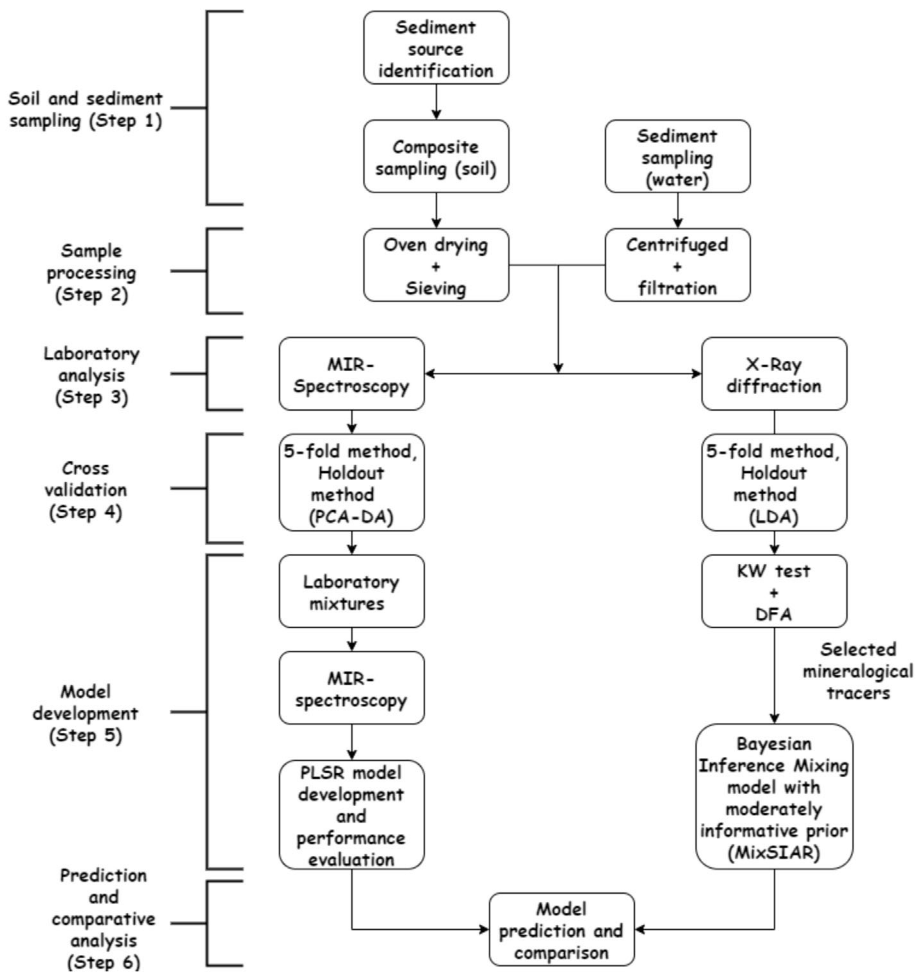

FIGURE 1 Overview of the methodological framework.

tracers were evaluated using laboratory mixtures and informative priors.

2.1 | Study area

This study was conducted in the Konar reservoir (990 km^2) catchment (23°51′23″N–24°8′26″N and 85°14′32″E–85°47′10″E) in Jharkhand, India, which is upstream of the Damodar River. The sedimentation rate in Konar has been estimated at 1.12 M m^3/year (Kumar, Raghuwanshi, & Mishra, 2015). The terrain of the region is very diverse in terms of topographical features, whereas the elevation and the land use are shown in Figure 2. Land use (Figure 2b) can be classified into three major classes (agricultural lands 37%, forests and shrubs 45% and human settlement areas 18%) and several minor classes (e.g. fallow lands and grasslands). The prevalence of agricultural lands, forests and human settlements alongside minor categories like fallow lands, channel banks and waste lands in this catchment complicates the identification of sediment sources. In addition to the minor land use classes like fallow lands and wastelands detailed in Das et al. (2022), channel banks can represent significant sediment sources, as supported by previous sediment fingerprinting studies (Smith & Blake, 2014). However, the presence of forests and grasslands covering most of the channel banks in the study catchment poses a challenge in distinguishing between sediment sources. To tackle this challenge, all minor land use classes were merged into major ones, streamlining the source discrimination process (Das et al., 2023). The catchment comprises deciduous and tropical forests and the farmers rely on rain-fed agriculture. Detailed descriptions of the land use classes are provided in Table 1. The forest areas mainly

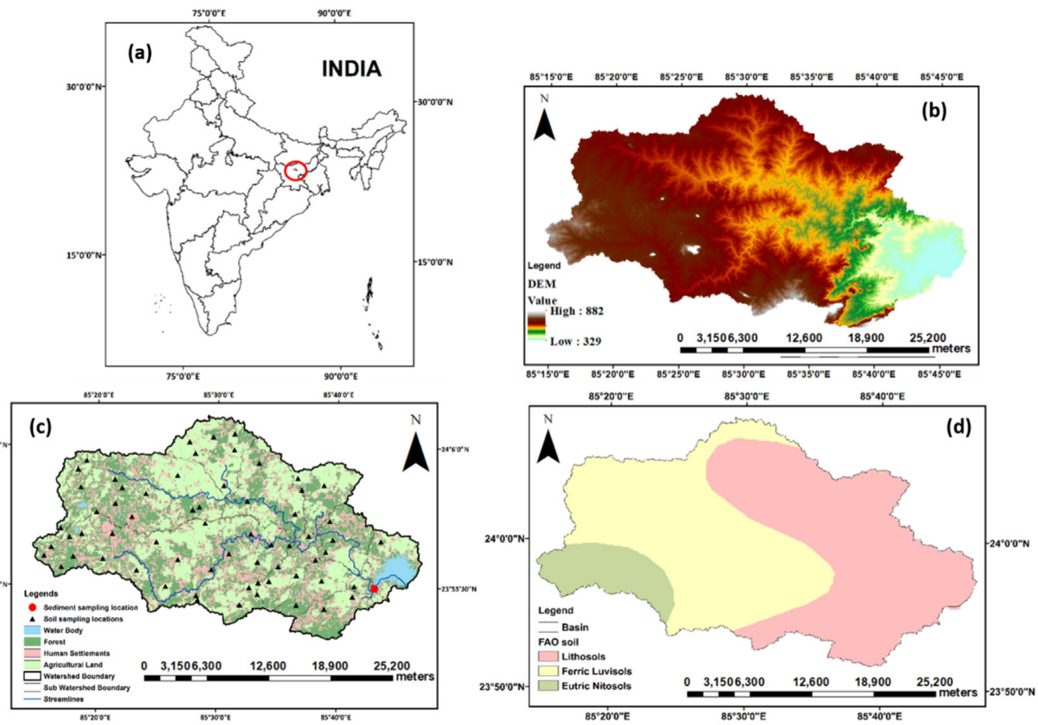

FIGURE 2 (a) Catchment location. (b) DEM of the study area. (c) Land use patterns in the study catchment with soil and target sediment sampling locations. (d) Soil map of the Konar catchment. [Color figure can be viewed at wileyonlinelibrary.com]

TABLE 1 Land use classes and their descriptions.

Land use class	Description	Area covered (%)	Sub-classes information	Soil class distribution
Agricultural lands	The land engaged in agriculture. It covers the rain-fed and irrigated cultivated lands along with fallow lands. Major crops grown are paddy maize, cereal and wheat.	37%	Around 50% of the agricultural lands are rain-fed and 30% are irrigated. The remainder of the lands in this class are fallow lands, cultivated in certain seasons and certain years.	Most of the agricultural areas (∼44%) are situated on lithosols and ferric luvisols (∼31%).
Forests	These lands mainly consist of dense and light forests along with grasslands. Forests are mostly tropical and deciduous.	45%	Around 40% of the forests consist of dense vegetation, and 30% of the land is covered by light and scattered vegetation. The remainder is covered by grasslands.	Forests in the catchment are mostly situated on ferric luvisols (44%), followed by lithosols (34%).
Human settlements	These lands consist of urban and rural areas dominated by houses, mines, paved roads, industrial areas and transportation services.	18%	Nearly 45% of the human settlements are rural, 30% of this class are urban and the remainder are mining and industrial areas.	Most of the human settlement areas are situated on eutric nitosols (51%) followed by lithosols (27%).

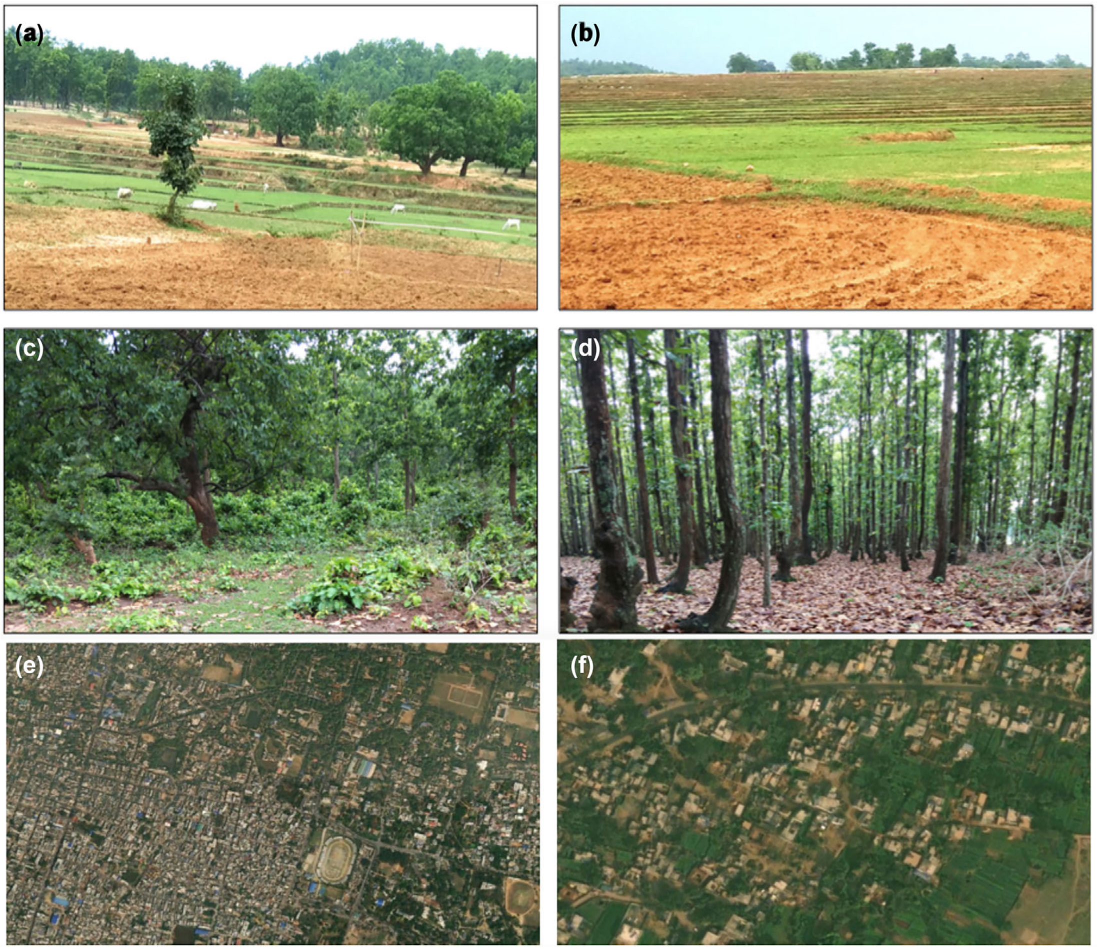

FIGURE 3 Images of agricultural and forest lands in the Konar study catchment. (a, b) Terrace farming practices in the region. (c, d) Dry and mixed deciduous forests in the region. (e, f) Human settlements in urban and rural areas of the region. [Color figure can be viewed at wileyonlinelibrary.com]

consist of dry tropical and mixed deciduous forests (Figure 3c,d). Recent land management practices in this region have resulted in a declining deforestation rate (i.e. ~10% in 2001–2010 to ~3% in 2010 to 2020) (Das et al., 2022). Major crops include rice, groundnuts and maize in the monsoon season and wheat, mustard and other vegetables in the non-monsoon period. Terrace farming is used in agricultural areas due to the uneven and steep terrain (Figure 3a,b). This region consists of human settlements with urban areas with dense settlements and rural areas with sparse settlements (Figure 3e,f). Although terrace farming practices reduce rain-induced soil erosion, compared to agricultural fields on bare slopes, they can still generate more sediment than other land use classes such as forests and human settlements. Three types of soils are present, that is, lithosols (46%), ferric luvisols (38%) and eutric nitosols (16%) (http://www.fao.org/soils-portal/soil-survey/soil-maps-and-databases/faounesco-soil-map-of-the-world) (Figure 2d). The presence of lithosols in the catchment soil implies that the climate is humid to arid (de Medeiros et al., 2019). Lithosols are not very suitable for agricultural practices due to their rocky texture. However, ferric luvisols and eutric nitosols have higher water-holding capacity and are more appropriate for agriculture. Ferric luvisols are rich in oxides of iron and alumina giving a distinct red colour, which provided a basis for the mineralogical fingerprinting in this study. Humidity ranges from 40% to 95%, with a temperature variation of 3°C to 44°C. The catchment receives an annual rainfall of 1250 mm and 80% occurs from July to September. This wet period delivers a considerable amount of sediment to the reservoir.

2.2 | Source and target sediment sampling

A total of 63 sampling sites (Figure 2c) were selected to characterise the potential sediment sources in different parts of the study catchment [25, 22 and 16 from agricultural land, forests and human settlement areas, respectively (Supplementary Table S1)]. Each sampling site was selected to represent an area of nearly 12–18 km^2. At each sampling site, composite sampling was undertaken to account for the variations in the erodible soil layer characteristics by mixing three to four sub-samples in a radius of 100–500 m, depending on the accessibility of the locations (Collins et al., 2017). The soil samples (0–5 cm depth) from potential sources were collected using a non-metallic trowel (Tiecher et al., 2017). The trowel was washed thoroughly after each sampling to avoid contamination. The source soil sampling was conducted in a single campaign, given that the lithological properties of the area are conservative. For each season, three target suspended sediment samples each of 2-L volume were collected from a depth of 0–10 cm from the reservoir inlet (Wang et al., 2019), and high-density polyethylene bottles were used to store the bulk sediment samples.

2.3 | Sample processing and laboratory analysis

The sediment samples were centrifuged and filtered to separate sediment for spectroscopic and mineralogical analysis. The soil samples were oven-dried for 12 h (Tiecher et al., 2017) and sieved through a 63-μm sieve to separate silt, clay and fine sand fractions (Bahadori et al., 2019). For assessing the accuracy of the MIR-PLS regression models, 30 laboratory mixtures were prepared with different ratios of source soil samples (Supplementary Figure S2). The MIR spectra of the samples were measured on KBr pellets (comprising ~1 mg of sample mixed with 200 mg of FTIR grade KBr) at wavelengths spanning 400–4000 cm^{-1} using a NICOLET 6700 (Thermo Fisher Scientific Instruments) FTIR spectrometer in reflection mode (2-cm^{-1} resolutions for all the sediment and soil samples with 32 scans per sample in triplicate). The spectra obtained from the pellets were corrected with respect to the pure KBr pellets and ambient air. Three spectral pre-treatments were used: Savitzky–Golay filter baseline correction, first derivative and multiplicative scatter correction (Ni et al., 2019).

All soil and sediment samples were also analysed using X-ray diffraction. Here, the spectra were measured in triplicate in the range of 10 to 80 degrees using a Bruker D2 phaser XRD system. The spectra obtained from XRD contain several peaks (~400 peaks) for different minerals, which were further analysed using the Xpert Highscore software to obtain the representative mineralogical composition by mass of the samples.

2.4 | Discrimination potential of spectroscopic and mineralogical tracers

The uncertainty due to the collection of a limited number of soil samples in representing the heterogeneity of the study catchment was examined using 'K-fold', 'Holdout' and leave-one-out (LOO) cross-validation methods. The K-fold cross-validation was executed using five folds of the sample data, and the 'Holdout' method was performed by partitioning 30% of the data for testing. The MIR spectral variables of the 63 soil samples from the three land use classes were analysed using PCA-DA to test the discrimination potential and obtain the misclassification rate of the source samples (Equation 1):

$$\begin{bmatrix} X_{1,1} & \cdots & X_{1,n} \\ \vdots & \ddots & \vdots \\ X_{63,1} & \cdots & X_{63,n} \end{bmatrix}, \begin{bmatrix} a_{1-26} \\ f_{1-22} \\ h_{1-15} \end{bmatrix} \tag{1}$$

(a = agricultural samples, f = forest samples, h = human settlement samples, n = 1869 spectral variables)

$X_{1,\,1}$ = variable 1 of spectral data of Sample 1.

For successful sediment source discrimination and apportionment, the input tracer data need to be conservative (i.e. no substantial change during transport between source and target sediment sampling location) and informative (i.e. how well it can differentiate between the individual sources) (Upadhayay et al., 2017). Mixing models consider that the mixing of sediment from the potential sources is homogeneous. Therefore, to differentiate between the individual sampled sources, the tracers should fall in the range of credibility. Considering the mineralogical composition of the source soil samples for the respective classes (Equation 2), the samples were cross-validated with all the tracers using linear discriminant analysis (LDA). The misclassification rate was determined to evaluate the source discrimination potential of the tracers:

$$\begin{bmatrix} Y_{1,1} & \cdots & Y_{1,p} \\ \vdots & \ddots & \vdots \\ Y_{63,1} & \cdots & Y_{63,p} \end{bmatrix}, \begin{bmatrix} a_{1-26} \\ f_{1-22} \\ h_{1-15} \end{bmatrix} \tag{2}$$

$Y_{1,\,1}$ = proportion of mineral 1 in Sample 1.
p = number of minerals used in the study.

2.5 | Source apportionment using mineralogy–MixSIAR mixing model

Unmixing of sediment samples was carried out to determine the proportional contribution of the individual sediment sources (i.e. land use classes):

$$X = \sum_{s=1}^{S} \alpha_s \cdot \beta_s \tag{3}$$

X = tracer composition of the mixture.
α_s = tracer composition of sources.
β_s = proportional contribution of tracer in source s.

For multiple tracers, the generalised equation is formulated as Equation 4:

$$X = \sum_{s=1}^{S} \alpha_{n,s} \cdot \beta_{n,s} \tag{4}$$

n = number of tracers.

The MixSIAR R package was used for source apportionment (Parnell et al., 2013) with the following values for our Markov chain Monte Carlo settings: number of chains = 3, chain length = 3,000,000, burn = 1,500,000, thin = 500. This open-source R software package, MixSIAR was developed by researchers taking into account some important considerations for source fingerprinting, including taking explicit account of (i) hierarchical structure, (ii) uncertainty in the means and variances of source data, (iii) covariance in tracer values and (iv) covariates in the mixing model (Stock & Semmens, 2018). The efficiency of Bayesian sediment fingerprinting models has been reported elsewhere (Cooper et al., 2015; Gateuille et al., 2019; Gholami et al., 2017).

To check the potential of the tracers for discriminating the sediment sources, a range test followed by a Kruskal–Wallis (KW) test ($p < 0.01$) was performed (Collins et al., 2017). As a result, a set of minerals was selected to execute the mixing model. To analyse the ability to discriminate between the source mineralogical signatures, a discriminant function analysis (DFA) was carried out using the statistical 13.4 software. DFA is mainly performed to determine the ability of the tracers to discriminate between two or more groups. Finally, the selected tracers were applied in the Bayesian mixing model for the apportionment of the sources (i.e. land use classes).

TABLE 2 Results indicating variation (mean and standard deviation) of mineralogical tracers in the three land use classes in the Konar catchment as obtained from XRD analysis.

	Agricultural lands (%)		Forests (%)		Human settlements (%)	
Minerals	Mean	SD	Mean	SD	Mean	SD
Quartz	34.88	7.66	29.88	4.85	34.36	8.16
Silica	29.63	7.12	31.11	9.85	24.85	12.8
Alumina	9.85	5.56	11.23	4.88	7.63	5.25
Halite	8.12	4.07	6.89	5.88	7.12	4.12
Calcite	7.82	5.21	5.63	5.01	5.03	4.23
Green cinnabar	6.85	3.36	5.67	2.67	6.1	4.8
Bornite	3.03	2.45	3.65	1.68	3.22	1.25
Fluorite	1.02	0.67	0.87	0.53	2.01	1.27
Silicon	0.92	0.4	0.45	0.48	0.81	0.23
Mica	0.69	0.32	0.51	0.37	0.46	0.11
Burnt ochre	0.09	0.13	0.49	0.1	0.03	0.06
Dolomite	0.07	0.019	0.48	0.03	0.025	0.06

TABLE 3 Variation of mineralogical tracers in the sediment samples collected from the reservoir.

Sampling timescale	Mean/ SD	Quartz (%)	Silica (%)	Alumina (%)	Halite (%)	Calcite (%)	Green cinnabar (%)	Mica (%)	Bornite (%)	Dolomite (%)
July 2018	Mean	33.48	28.03	5.47	7.20	11.97	3.71	2.63	10.01	6.19
	SD	2.21	1.32	0.55	2.31	1.90	3.74	2.66	0.39	1.99
August 2018	Mean	35.67	29.33	3.67	8.67	15.00	2.33	1.65	3.67	7.46
	SD	4.51	1.53	1.53	2.08	3.00	2.52	1.79	3.06	1.79
October 2018	Mean	41.33	36.67	7.00	2.67	7.00	4.33	3.07	0.00	2.30
	SD	1.53	2.08	4.36	2.52	5.57	0.58	0.41	0.00	2.17
December 2018	Mean	35.00	27.00	13.17	3.00	8.33	5.67	4.03	4.67	2.58
	SD	6.24	3.46	0.76	1.00	2.52	2.08	1.48	1.53	0.86
March 2019	Mean	37.47	29.10	11.07	1.67	3.33	12.07	8.57	2.33	1.44
	SD	3.14	4.75	2.90	0.58	1.15	1.05	0.75	3.21	0.50
June 2019	Mean	37.23	36.17	1.36	6.13	11.69	1.79	1.27	4.29	5.27
	SD	0.75	0.76	0.21	0.06	0.50	0.06	0.04	0.18	0.05

Prior information about the study catchment was used with the Bayesian mixing model framework. Here, the Dirichlet distribution is most widely applied for informative priors (Stock & Semmens, 2018). The proportion of area covered by the individual land use groups was used as an informative prior vector ($\alpha = 45$, 37 and 18 for agricultural lands, forests and human settlements, respectively) in this analysis. To avoid skewing the model results, the priors were scaled using Equation 5:

$$\alpha_m = {kn_k}/{\sum n_k} \tag{5}$$

$m =$ respective source,

$k =$ rescaling factor (considered 5 for this study).

$n_k =$ (%) area covered by respective land use classes.

Based on Equation 5, the improved moderate prior vectors were estimated: 2.25 for agricultural lands (45%), 1.85 for forests (37%) and 0.9 (18%) for human settlements (Supplementary Figure S3).

2.6 | Source apportionment using spectroscopy–PLS regression mixing model

In spectroscopic studies, PLS regression is applied as a multiple regression approach for predicting a dependent variable from a set of predictor variables. In this method, the scores of variables are selected to maximise scores between the correlations within the variables. The main difference between PCA and PLS regression is that the scores of the variables are not selected based on preserving the maximum correlation between the variables (Long, 2013). In this study, the PLS regression model was used to predict the source contributions by decomposing both the predictor and dependent variables.

Three PLS regression models were entrenched to predict the contribution of the land use classes to the target sediment samples. The number of PLS variables selected for model calibration and validation was decided based on the lowest root mean square error of cross-validation (RMSECV). The PLS regression method was used to analyse the MIR spectra of the laboratory mixtures (x variate) and the contribution from the land use classes (*y* variate). The model performances were evaluated using statistical indicators: that is, RMSECV, root mean square error of calibration (RMSEC), root mean square error of prediction (RMSEP), coefficient of determination (R^2) and Nash–Sutcliffe efficiency (NSE) (Tiecher et al., 2017).

3 | RESULTS

3.1 | MixSIAR model based on mineralogical tracers

The main minerals detected in the source and target sediment samples were quartz, silica, alumina, halite, calcite, green cinnabar, bornite, fluorite, silicon, mica, dolomite and burnt ochre. The variation of the minerals in the soil samples is depicted in Table 2 and sediment samples in Table 3. Based on the range test (Figure S4), dolomite and mica were rejected as tracers for the analysis due to the lack of conservation.

TABLE 4 Results of the Kruskal–Wallis (KW) test to identify significant mineralogical tracers in the Konar study catchment.

Tracers	*H*-value (>28.13) *(test static)	*p*-value (<0.01)
Quartz	38.530	0.0001
Silica	31.980	0.0003
Alumina	32.950	0.0009
Halite	28.290	0.0028
Calcite	29.776	0.0056
Green cinnabar	30.757	0.0004
Bornite	34.029	0.0002
Fluorite	17.160	0.0098
Silicon	3.788	0.1532
Burnt ochre	9.060	0.1270

From the KW test results in Table 4, it can be seen that fluorite, silicon and burnt ochre failed to provide robust source discrimination. The results of DFA illustrated that minerals present in the source soil samples discriminated the land use classes more efficiently than spectroscopic variables (Figure 4). From the posterior distributions of sediment source contributions for July, August and March (Figure 5a,b,e), it can be observed that the major contributor of sampled sediment was agricultural lands. In contrast, the contribution of forests was dominant in June (Figure 5f). Moreover, we checked the applicability of the MixSIAR with the artificial mixtures (Figure S5).

3.2 | PLS regression models based on MIR spectra

The sediment samples collected from the reservoir were subjected to particle size analysis and based on the mean particle size distribution of the sediment samples (shown in Figure S1), 95% of the particles were found to be silt and clay (i.e. particle size range of <63 µm). The MIR reflectance spectrum of soil depicts the presence of minerals and organic compounds in different forms. The detailed information of different bonds corresponding to different peaks has been reported in several publications (Ni et al., 2019; Tiecher et al., 2017). Figure 6 shows pronounced variations in the spectral features of soil from the different land use classes, including in the characteristics peaks of C–H stretch (3500–3000, 3000–2800, 2250 and 2133 cm^{-1}), O–H stretch (3694, 3620, 1628 and 915 cm^{-1}), mixture of C–H, O–H and N–H stretch and Si–O stretch (1975, 1872, 1158, 1110, 810, 790 and 698 cm^{-1}) (Tiecher et al., 2017). The variations in carbonate peaks in the soil samples depict the presence of loess. The carbonate peaks are stronger in forest soil than the other two classes of land use in the Konar River reservoir system. The peaks in the range of the non-organic range are comparatively lower in the forest source samples than in the other two classes of land use. The discrimination potential of the MIR spectra for the soil samples collected from different land uses was determined based on the results of PCA-DA. The

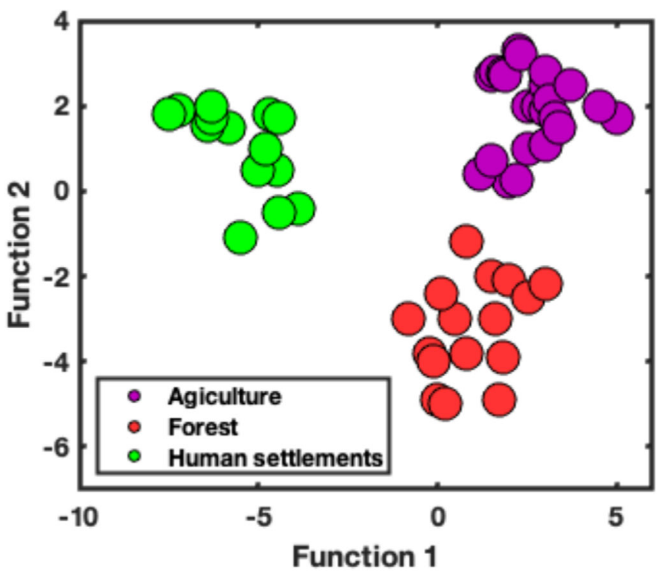

FIGURE 4 Results of DFA conducted on the mineralogical composition of the source samples collected to represent the land use classes in the Konar study catchment. [Color figure can be viewed at wileyonlinelibrary.com]

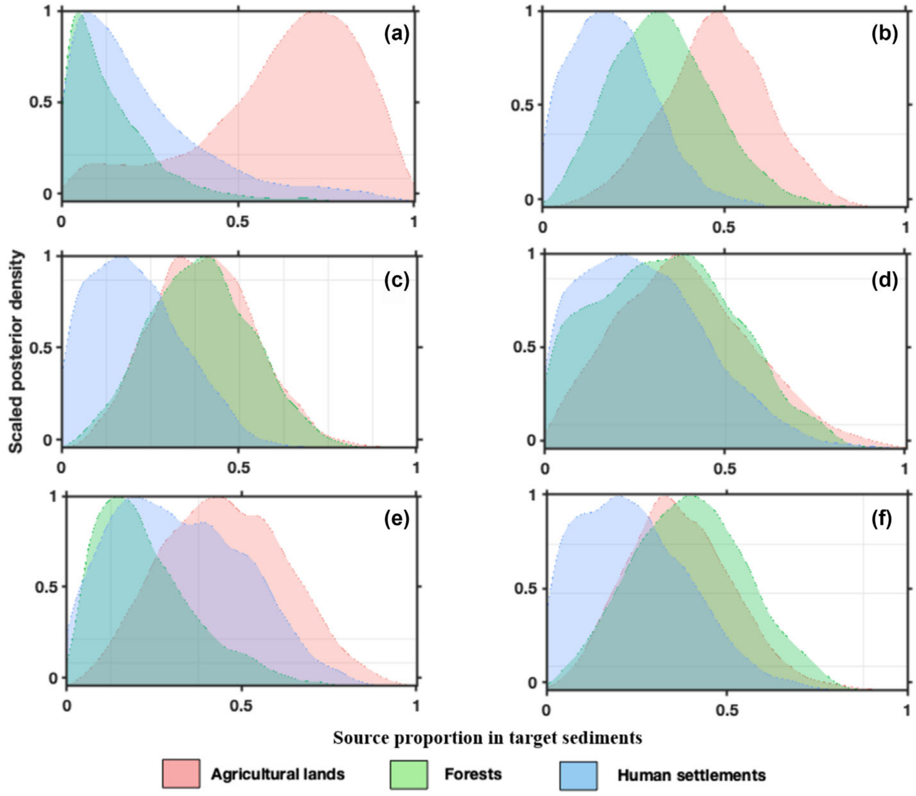

FIGURE 5 MixSIAR model density plots for the land use source contributions for different time periods: (a) July 2018, (b) August 2018, (c) October 2018, (d) December 2018, (e) March 2019 and (f) June 2019. [Color figure can be viewed at wileyonlinelibrary.com]

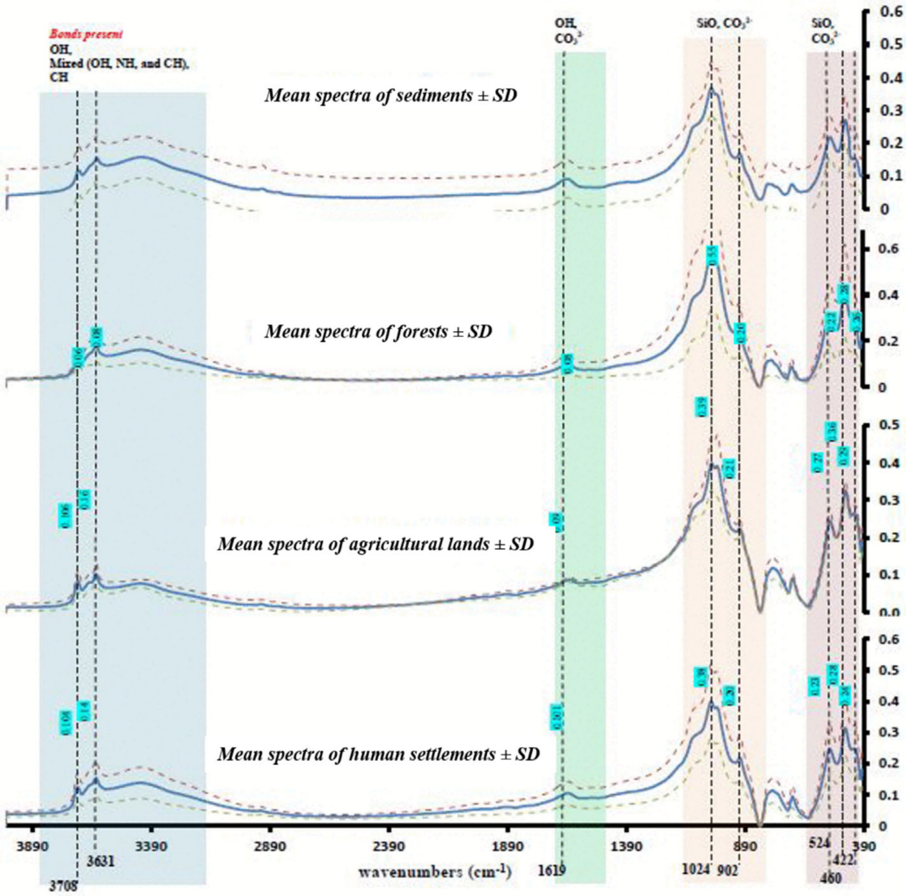

FIGURE 6 Mean MIR spectra of the soil and sediment samples collected from the Konar catchment. [Color figure can be viewed at wileyonlinelibrary.com]

PCA analysis indicates five principal components explained ~90% of the variance in the data (Figure S6). LDA was performed on these five principal components for the three land use classes (Figure 7). Although, from Figure 7, it can be seen clearly that there are some overlaps between the forest and agricultural soil spectra, discrimination between the three land uses is still evident. One possible reason for this overlapping of forest and agricultural source samples could be the presence of light vegetation and the merging of minor classes

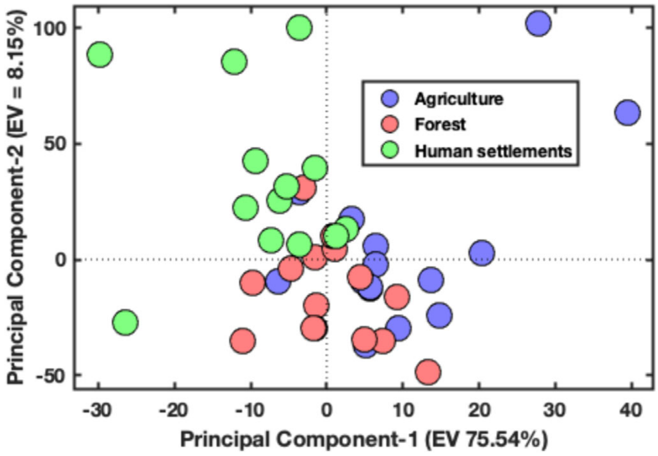

FIGURE 7 Results of PCA-DA analysis conducted on the MIR spectra of the land use classes using the first five principal components. [Color figure can be viewed at wileyonlinelibrary.com]

TABLE 5 Performance evaluation of the MIR-PLSR models based on spectroscopic variables.

Sources	PLS components	RMSECV (%)	RMSEC (%)	RMSEV (%)	R^2	NSE
Agricultural lands	5	4.21	2.86	2.72	0.89	0.76
Forests	5	3.89	2.14	2.93	0.87	0.74
Human settlements	5	4.26	3.71	3.79	0.82	0.71

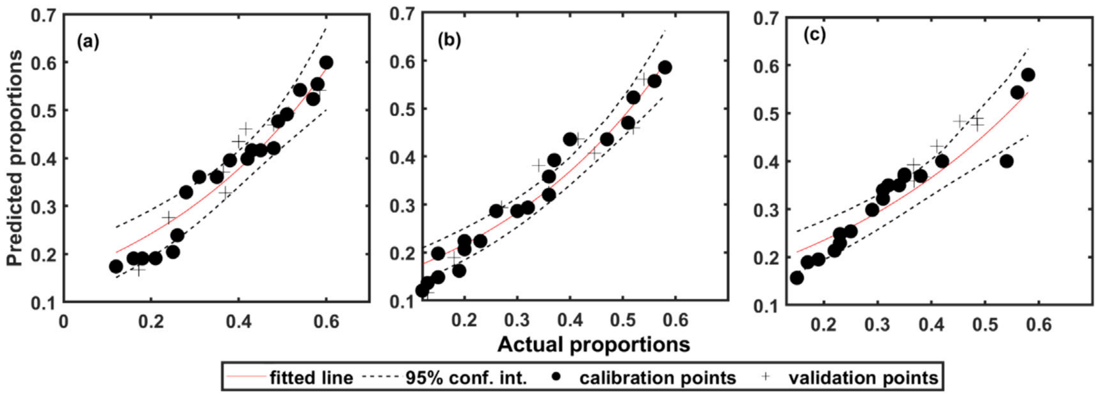

FIGURE 8 Results of the MIR-PLSR modelling showing the agreement between the actual (i.e. known using artificial mixtures) and predicted proportions of the land use sources sampled in the Konar study catchment: (a) agricultural lands, (b) forests and (c) human settlements. [Color figure can be viewed at wileyonlinelibrary.com]

including fallow lands or grasslands into the agricultural and forest classes, respectively.

The set of 30 artificial mixtures created in the laboratory was used to examine the performance of the MIR-PLS regression models (Poulenard et al., 2012; Tiecher et al., 2017). Based on the lowest RMSECV values, five PLS components were selected for the agricultural, forest and human settlement models (Table 5). Figure 8 shows the agreement between the actual and the predicted contributions of the land use classes based on the set of 30 artificial laboratory mixtures. The variations between observed and predicted source contributions in the calibration and validation datasets, based on RMSEP and RMSEV, were consistently less than ~4%, with an R^2 exceeding ~0.8 and NSE values ranging between 0.71 to 0.76. The correlations (Table 5) between actual and predicted proportions were higher for the agriculture and forest sources (R^2 of 0.89 and 0.87, respectively) but weaker for the human settlement source (R^2 of 0.82).

3.3 | Comparative inspection of the modelled source proportions

The predicted contributions of the land use classes from both models are compared in Figure 9a. The results indicate that agricultural land use is predicted to be the largest contributor of sediment (~41% to ~60%) during monsoon months (July–August) by both models. However, there are disparities in the estimated contributions of forests and human settlement areas. During the post-monsoon months (October–December), the results reveal a sudden increase in the

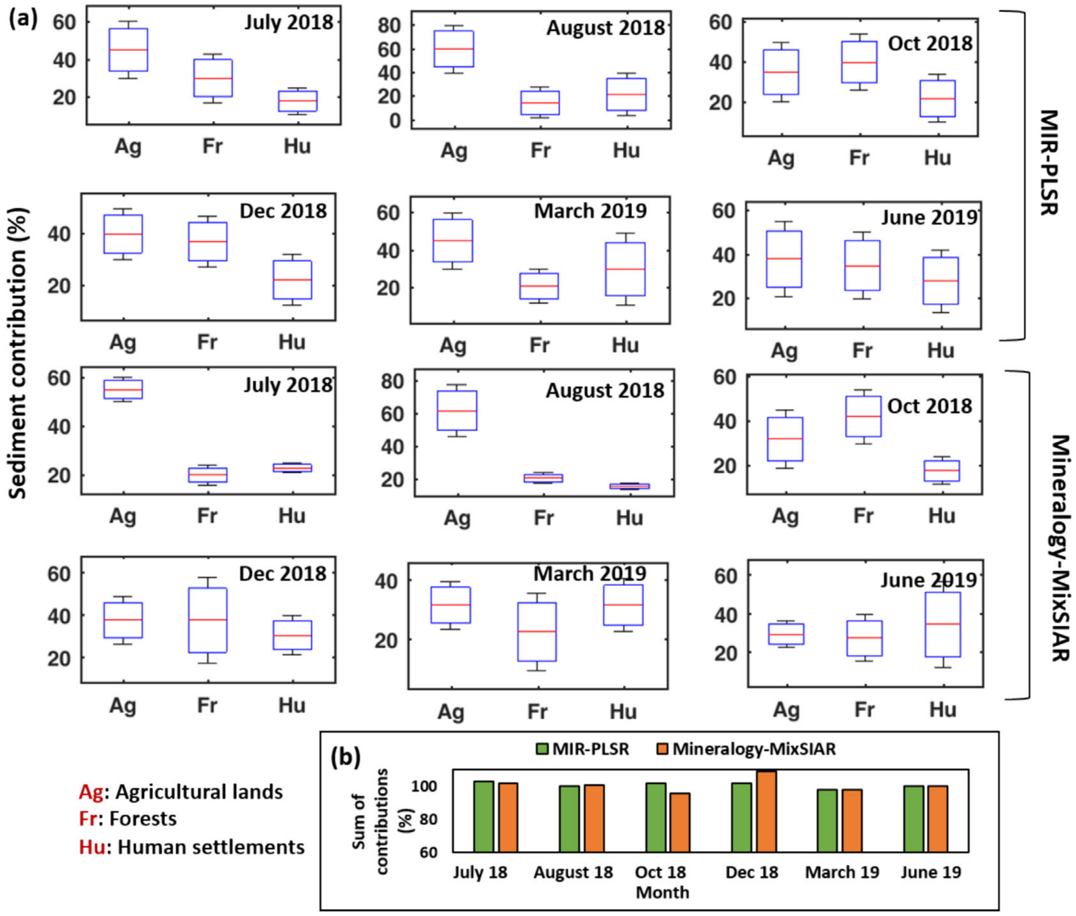

FIGURE 9 (a) Box plots, with 95% confidence intervals, showing the comparison of spectroscopic and mineralogical tracing results in the Konar study catchment using MIR-PLSR and mineralogy–MixSIAR modelling. (b) Sum of predicted contributions for the MIR-PLSR and mineralogy–MixSIAR modelling. [Color figure can be viewed at wileyonlinelibrary.com]

TABLE 6 Results of cross-validation for the spectroscopic and mineralogical tracers.

Type of variables	No. of variables	Model used	Misclassification rate: K-fold cross-validation ($K = 5$)	Holdout cross-validation ($p = 0.3$)	Leave out cross-validation ($n = 1$)
Spectral (MIR)	1869	PCA-DA	0.23	0.22	0.20
Mineralogical	10	LDA	0.19	0.16	0.17

sediment contribution from forest areas (~35% to ~43%) and a decrease in agricultural lands. The results of both modelling approaches, in general, showed good agreement. The sum of predictions of the models varies between 94% and 102% for MIR-PLSR and 96% and 111% for mineralogy–MixSIAR model (Figure 9b). In most seasons, the sum of predictions from both models is close to 100%. However, in the post-monsoon month of December, the MixSIAR model sum of prediction is 111% but exhibits better accuracy for the remaining seasons.

3.4 | Source discrimination

The cross-validation tests executed on the MIR spectra and mineralogical composition of the source samples revealed the uncertainty in the discrimination potential of the variables (Table 6). The misclassification rates of the source samples for the MIR spectral variables using K-fold, holdout and LOO methods were 0.23, 0.22 and 0.20, respectively. The corresponding misclassification rates for the mineralogical variables were 0.19, 0.16 and 0.17, respectively.

3.5 | Geological insights on the source discrimination results

In view of the high erodibility of ferric luvisols and the fact that agricultural lands were found to be the primary source of sampled sediment in this study, it can be deduced that more than half of the agricultural lands are located on the ferric luvisol, which is most critical in terms of soil erosion and sediment yield (Figure 10). Our results are consistent with the findings of Kuhn et al. (2009). Considering that roughly half of the forest is located on lithosols with a low propensity

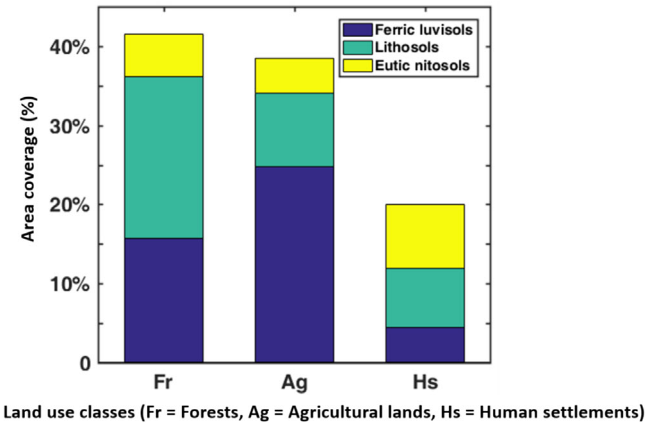

FIGURE 10 Proportion of soil classes in the land use classes of the Konar catchment. [Color figure can be viewed at wileyonlinelibrary.com]

for erosion, this section of forest can be expected to contribute the least sediment in the study region (Elliot, Page-Dumroese, & Robichaud, 2018). Studies that analysed soil erosion using regional geology and soil classifications including lithosols, nitosols and luvisols also reported similar conclusions (Bekele & Gemi, 2021).

4 | DISCUSSION

4.1 | Sediment sourcing using spectroscopic fingerprints

As spectral data are highly correlated and noisy with the presence of more variables than samples, the PLS regression model is the most effective tool for processing such data (Karaman et al., 2013; Vercruysse & Grabowski, 2018). Variation in reflectance of the spectra signifies the influence of particle size, organic compounds and minerals in the soils of the study area and similar factors have been discussed in other studies (Brosinsky et al., 2014; Ni et al., 2019). The results of PCA-DA performed on the spectroscopic variables revealed an overlap between forest and agricultural soils, whereas the human settlement samples were very well distinguished. This probably reflects the presence of organic matter and finer particles in both forest and agricultural soils. As the Konar catchment is situated in one of the mineral-rich valleys (Damodar valley) of India, soil organic matter (SOM) and mineral-associated (high-density) SOM-based tracing could provide some extra important information on soil erosion management as organo-mineral associations are very prominent in different land uses, especially forests (Ludwig et al., 2015). Nevertheless, huge challenges remain for this SOM research theme because of (i) the unstable nature of and other uncertainties associated with biomarkers and (ii) high degrees of environmental and analytical uncertainty (Li et al., 2020).

A study by Ni et al. (2019) performed PCA-DA analysis for samples collected from topsoil and channel sediments and found promising results. However, in our case, the overall reflectance of forest soil spectra showed greater variation, which most likely reflects the higher contents of finer particles compared with the other land use classes, which are more prone to water erosion (Figure S7).

Previous work has reported successful application of spectral fingerprints using PLS regression models (e.g. Tiecher et al., 2015; Uber et al., 20192019). One limitation associated with the use of spectral fingerprints includes sediment-bound organic matter, which is non-conservative, making spectral fingerprints unsuitable for suspended sediment source tracing (Collins et al., 2014). Stevens et al. (2008) highlighted one limitation of PLS regression associated with its site-specific nature.

4.2 | Sediment sourcing using mineralogical fingerprints

The results of DFA performed on the mineralogical variables in this study showed that the discrimination of the land use classes is feasible without any overlapping of the samples. Previous sediment fingerprinting studies have employed mineralogy for the (i) development of effective beneficial best management practices (BMPs) and understanding of the connectivity of sediment delivery and land uses (Koiter et al., 2013) and (ii) determination of the provenance of floodplain deposits (D'Haen et al., 2013) and many more applications. In our case, although, the mineralogical and spectroscopic tracers generated similar results, the 95% confidence intervals of the mineralogical method were more precise (Figure 9a). Laceby et al. (2015) highlighted the importance of including meaningful mineralogical elements and their geological basis for use as tracers and discussed the uncertainty of merely relying on statistical techniques alone. Batista et al. (2019) argued that statistical methods using geochemical element fingerprints can yield very similar results to the use of any knowledge-based (i.e. of pedogenetic processes) tracers in the case of finer particle size fractions but that greater disparities can occur in the case of coarser sediment particles.

4.3 | Comparison between mineralogical and spectroscopic source apportionment

There are very few studies comparing spectroscopic and geochemical analysis (Tiecher et al., 2016; Verheyen et al., 2014). The existing studies suggest that both methods can deliver source discrimination but that some disparities can exist between the results because of the nature of the specific variables selected or mixing models applied. Evrard et al. (2013), for example, used diffuse reflectance infrared Fourier transform spectroscopy and showed that its results were consistent with the conventional geochemical approach but emphasised the need to consider the organic carbon content of soils as the results were influenced by the presence of organic matter. Our study herein also advocates the possibility of combining both mineralogical composition and spectroscopic information for robust estimation of sediment sources. In the post-monsoon season, the mineralogy–MixSIAR model predicts the contribution of forests to be closer to that of the agricultural land use class, whereas the results are different in the case of the MIR-PLS regression model. Artificial mixtures of known source contributions provided an efficient opportunity to assess the accuracy of the sediment fingerprinting methods (Cooper et al., 2014; D'Haen et al., 2013).

4.4 | Catchment land use properties

Our study catchment includes forests as a major land use class and a large proportion of the forest areas are located very near to the reservoir with reasonable sediment connectivity as reported by Das et al. (2022). This is most likely justification for the estimated contribution of forests in both the monsoon and non-monsoon seasons. The appearance of sediment from forests in post-monsoon seasons could be connected to the time lag and residence time of suspended sediment and the nature of monsoon-driven erosion patterns in the study catchment. Another possible reason for the increase in sediment inputs from forests could be the fragmented forest cover and mixed forests resulting in a reduction in infiltration and concomitant increase in runoff as reported by Rajbanshi and Bhattacharya (2020). However, most of the rainfall in this region (~80%) takes place in the monsoon season. From this irregular pattern of sediment generation, it can be inferred that the high predicted contribution of sediment (~35% and 43% by the PLSR and MixSIAR models, respectively) from forests in the post-monsoon season is not illogical. Moreover, the explanation for these seasonal differences could also be associated with seasonal cropping patterns and agricultural management practices, as well as the influence of pre-monsoon showers. Seasonal patterns in the density of crop cover generate a seasonal control on sediment delivery in our study area (Kretz et al., 2021; Loaiza, 2008).

4.5 | Limitations and uncertainties

The source apportionment reported in this study inevitably has some limitations. A common limitation of the sediment fingerprinting method concerns difficulty in validating the source proportions in the absence of independent monitoring data (Collins et al., 2017). The total number of samples collected from the potential sources inevitably depends on some practical constraints such as research budget, accessibility of the locations and the requirements of any tests used for statistical source discrimination. The findings of this study are specific to the studied catchment and might not be directly applicable to larger or different river basins with varying geomorphological and land use characteristics. In some catchments, channel banks may serve as significant source groups, contributing to a more comprehensive understanding of sediment dynamics. This aspect was not considered in our study. However, recent studies by Pulley and Collins (2024) and Pulley and Foster (2017) have emphasised the importance of channel banks as potentially substantial sources of sediment. To enhance the robustness and accuracy of our results, future research should include sampling from channel banks. The merging of minor land use classes into major categories could oversimplify the complex interactions between different land uses and sediment production. The precision and accuracy of the XRD and MIR analytical techniques used for tracer identification might introduce uncertainties in the results. Moreover, the non-linearities in modelled outcomes compared to laboratory mixtures arises probably due to complex sediment transport processes, spatial and temporal variations in sources, mixing and transformation of sediments, non-conservative behaviour and simplified modelling assumptions. Addressing these challenges highlights the need for advanced modelling techniques. Other potential uncertainties could be associated with the spatial heterogeneity of rainfall, slope and lithological characteristics of the catchment. To explore temporal uncertainty, sediment sampling was performed for a water year (Nosrati, 2017). Our study was conducted during 2018–2019, which can be considered as a typical, rather than atypical, water year (Bahadur et al., 2020).

We performed limited interpretation of the effect of geomorphologic events [(e.g. landslides as mentioned in Pickup and Marks (2000) and Fathabadi and Jansen (2022)]. Future research could investigate sediment provenance by focusing on processes like rock deterioration (Pola et al., 2014), other environmental stresses (Collins et al., 2020) and geomorphological events such as landslides (Pickup & Marks, 2000). As mentioned in Jacq et al. (2019), sediment fingerprinting has much more potential for deriving new knowledge if there are proper collaborations between palaeo-climatologists, geomorphologists and hydrologists. A better understanding of sediment sources within gully systems and the quantification of gully sediment transport at the catchment scale are essential for effective management and control policies (Lin et al., 2015). However, this aspect is not considered in our study. Thus, future sediment sourcing work in the Konar drainage basin should acknowledge that sediment transport processes are rather complex in nature and can be triggered by various factors including channel morphology or channel properties and landslides (Xiong et al., 2022) as well as by conventional fluvial and hydrological dynamics. Although we recognise the importance of economic considerations and the transferability of methods, as discussed in Pulley and Collins (2021), this study has not considered the cost–benefit evaluations of multiple tracer and data processing analyses. For future research, we recommend incorporating cost–benefit evaluations and exploring the broader applicability and economic feasibility of various methods, such as sampling strategies of MIR spectroscopy and XRD with MixSIAR modelling, to enhance the transferability of our findings.

5 | CONCLUSIONS

Our work illustrated a reasonable agreement between both source tracing techniques in the Konar study catchment for the monsoon and post-monsoon seasons. The findings suggested that agricultural land use was the major sediment source, especially during the monsoon season, whereas forests deliver a greater proportion of the target sediment in the post-monsoon months. These results are critical for prioritizing the implementation of land use and erosion control measures. Our study illustrates how a detailed evaluation of sediment sources with different tracer sets, fingerprinting techniques and mixing models, is advisable to help confirm management targets more robustly.

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