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3 authors:

Kazem Nosrati
Shahid Beheshti University

A. L. Collins
Rothamsted Research

Mohammad Madankan
Heriot-Watt University

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Fingerprinting sub-basin spatial sediment sources using different multivariate statistical techniques and the Modified MixSIR model

Kazem Nosrati[a,*], Adrian L. Collins[b], Mohammad Madankan[c]

[a] Department of Physical Geography, Faculty of Earth Sciences, Shahid Beheshti University, 1983969411 Tehran, Iran
[b] Sustainable Agriculture Sciences Department, Rothamsted Research, North Wyke, Okehampton, EX20 2SB, UK
[c] Geomorphology Laboratory, Department of Physical Geography, Faculty of Earth Sciences, Shahid Beheshti University, 1983969411 Tehran, Iran

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ABSTRACT

Information on the relative contributions of sediment from different sources is needed to target sediment control strategies to prevent excess sediment delivery to receptors like dam reservoirs. The overarching scientific objective of this study was therefore to apportion sub-basin spatial source contributions to the supply of fine sediment in an erodible mountainous basin in north-eastern Iran to inform management. The technical objective was to satisfy the scientific objective using a source fingerprinting procedure based on composite signatures selected by different statistical tests. Nine potential geochemical tracers were measured on 21 sediment samples collected to characterise the three sub-basin spatial sediment sources and seven sediment samples collected at the outlet of the main basin. The statistical analysis employed to select three different composite fingerprints for discriminating the sub-basin sediment sources comprised: (1) the Kruskal–Wallis H test (KW-H), (2) a combination of KW-H and discriminant function analysis (DFA), and (3) a combination of KW-H and principal components & classification analysis (PCCA). A Bayesian un-mixing model was used to ascribe sub-basin source contributions using the three composite fingerprints. Using KW-H, the respective relative contributions from sub-basins 1, 2 and 3 were estimated as 45.6%, 3.8% and 50.6%, compared to 46.8%, 18.8% and 34.4% using KW-H and DFA, and 61%, 2.5% and 36.5% using KW-H and PCCA. Kolmogorov-Smirnov test pairwise comparisons of the distributions of predicted source proportions generated using different composite signatures confirmed statistically significant differences. The root mean square difference between the predicted source proportions based on different composite signatures was ~12%. This study therefore provides more evidence that source tracing studies should deploy a number of composite signatures selected using independent statistical tests to permit appraisal of the consistencies or otherwise in predicted source contributions based on the tracers used. The outputs of this preliminary study will be used to inform the spatial targeting of sediment mitigation.

1. Introduction

Although soil erosion is a naturally occurring process, it can cause both on-site and off-site detrimental impacts where it occurs at elevated rates. A wide variety of negative effects have been reported, ranging from the deterioration of soil quality (Foley et al., 2005) on-site to the accelerated sedimentation of dams (Verstraeten and Poesen, 2000) and increased water treatment costs (Lal and Stewart, 2013) off-site. Consequently, accelerated soil erosion poses a serious threat to land management sustainability and water resource utilization in many areas of the world (Cerdà et al., 2009; Mukundan et al., 2010; Zhou et al., 2016). To combat such issues, specification and delivery of appropriate management solutions requires a robust understanding of the sediment problem at catchment scale and a focus upon the key sources involved (Collins et al., 2010b). Studies of sediment sources facilitate a better understanding of how soil erosion controls subsequent sediment transport, deposition and delivery in river catchments (Zhao et al., 2017).

Since traditional techniques for sediment source monitoring such as erosion pins and surveys of erosion features are time consuming and costly (Collins and Walling, 2004; Foster et al., 2007; Loughran and Campbell, 1995), applications of sediment source fingerprinting techniques have increased over time (Owens et al., 2017; Walling, 2013; Walling and Foster, 2016). Sediment source fingerprinting is founded upon a comparison of the properties or fingerprints of fine-grained sediment with those of the potential sediment sources present in a catchment (Collins and Walling, 2004; Pulley et al., 2015). Numerous types of fingerprint properties can be used to discriminate between the

* Corresponding author.
E-mail address: k_nosrati@sbu.ac.ir (K. Nosrati).

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potential sources of the sediment, such as physical characteristics (e.g., sediment size, shape, color), geochemical properties (e.g., trace metals), fallout radionuclides (e.g., ^{7}Be, ^{137}Cs, unsupported ^{210}Pb), mineral magnetic properties (e.g., magnetic susceptibility and isothermal remanence), and biological properties (e.g., compound-specific stable isotopes, microbial communities, pollen, and soil enzymes) (Collins et al., 2017; Haddadchi et al., 2013; Miller et al., 2015; Owens et al., 2017; Walling, 2005). Following source discrimination, quantitative analyses are used to determine the relative contribution of each potential source to the target sediment samples collected and these typically rely on either frequentist or Bayesian un-mixing models (Collins et al., 2017; Owens et al., 2017). Source fingerprinting techniques are being increasingly used in many different areas of the world (D'Haen et al., 2013; Haddadchi et al., 2014; Minella et al., 2008; Nosrati et al., 2014; Smith and Blake, 2014; Walling and Collins, 2008).

Robust discrimination between different potential sediment sources is a key requirement in fingerprinting. Here, multivariate statistical techniques including the Kruskal-Wallis H-test, discriminant function analysis (DFA), principal component analysis (PCA) and clustering techniques (Collins et al., 2012; Palazón et al., 2015; Palazón and Navas, 2017; Pulley et al., 2017; Tiecher et al., 2015) are widely applied to select optimum sets (i.e. composite signatures) of tracers for source discrimination.

A key consideration in the application of sediment fingerprinting relates to the classification of potential catchment sources (Collins et al., 2017). In some instances, researches have successfully investigated sediment source types such as surface or sub-surface (Peart and Walling, 1986), or different land use categories plus channel banks (Walling et al., 1999). In other circumstances, researchers have apportioned spatial sources represented by geological units (D'Haen et al., 2012) or individual tributary sub-catchments (Vale et al., 2016). Understanding the relative contribution of each or of major sub-basins to downstream sediment yield provides a basis for catchment managers to target sediment control strategies spatially. A sub-basin sediment source sampling strategy was therefore used in the work reported in this paper. More specifically, the overarching scientific objective of this study was therefore to apportion sub-basin spatial source contributions to the supply of fine sediment using a modified Bayesian un-mixing model in an erodible mountainous basin in north-eastern Iran to inform management. The technical objective was to satisfy the scientific objective using a source fingerprinting procedure based on composite signatures selected by different statistical tests. The work reported herein focussed on Iran since this country faces many challenges arising from erosion and sediment production in its mountainous catchments where the difficult terrain poses a serious challenge to installing and maintaining conventional sediment monitoring networks. As a result, improved evidence on the spatial sources of the sediment problem needs to be assembled using alternative research techniques such as source fingerprinting.

2. Materials and methods

2.1. Study area

The Mirabad drainage basin (228 km^2) is located near the town of Neyshabour, Khorasan province, in north-eastern Iran between 58°50′E to 59°00′E longitude and 36°17′N to 36°22′N latitude (Fig. 1). The topography of the Mirabad drainage basin is mountainous, with elevations ranging from 1213 to 3262 m, with a mean of 2100 m above sea level. The average slope gradient is 85%. The land use map of Iran provided by the Iran Forests, Range and Watershed Management Organization (IFRWMO) showed that land cover of the study area comprises rangelands and woodlands (159.6 km^2 area; 70%), cultivated land including orchards, dry land farming and cropped fields, (50.2 km^2; 22%) and residential areas (18.2 km^2; 8%).

The catchment lithology is primarily Triassic sedimentary deposits including black shale and sandstone and slightly metamorphosed shale and limestone (C-TR ssh), shale, quartz, red sandstone and conglomerate (Cf), slightly metamorphosed conglomerate and sandstone (Jb), Miocene red conglomerate (Msc), Devonian fossiliferous limestone (Db), Quaternary older terraces and recent alluvium (Fig. 1). The soils within the study catchment are mainly sandy clay loams. Four soil samples collected from the study basin comprised 30.8% clay, 25.45% silt and 43.75% sand. The soil map of Iran provided by IFRWMO showed that the soil orders within the catchment are mainly Entisols and Inceptisols. Based on the data provided by the Iran Meteorological Organization, the long-term (30 years) mean annual precipitation in the Neyshabour station next to the study area is ca. 247 mm. In the upper parts of the region, precipitation is mostly snow. Mean annual discharge based on the 41 years (1973–2013) of record from the hydrological station at the outlet of the drainage basin provided by the Iran Water Resources Research Organization is estimated at 1.11 m^3 s^{-1}, with most discharge occurring in April and May: 3.61 m^3 s^{-1} and 2.87 m^3 s^{-1}, respectively. The high average slope gradient of the study catchment, the variety of potentially erodible lithological formations, overgrazing problems, and medical plant harvesting are important factors increasing soil erosion and sediment yield in this area. On the basis of 12 field surveys and associated observations over a 12 month period, it was established that three sub-basins comprising sub-basin 1 (44.7 km^2), sub-basin 2 (115 km^2) and sub-basin 3 (18.6 km^2) (Fig. 1) in the study area dominate tributary sediment inputs to the main stem. These three tributary sub-basins were monitored for discharge over the 12 month observational period. Stream discharges were manually measured at the end of each month during 2015 by means of the velocity-area method (Gordon et al., 2004). This method requires measurement of the area of a stream cross-section and the average stream velocity. Discharge is then calculated as $Q = V \times S$: where Q is discharge (m^3 s^{-1}), V is average velocity (m s^{-1}) and S is cross-sectional area of the water (m^2). Area was calculated from cross-section measurements. Flow velocity was measured with an OTT current meter (Z30 counter). The relative contributions to mean annual discharge (based on the 12 months of record at the three sub-basin outlets) delivered by sub-basin 1, sub-basin 2 and sub-basin 3 were estimated as 65% (0.73 m^3 s^{-1}), 9% (0.1 m^3 s^{-1}) and 25% (0.28 m^3 s^{-1}), respectively.

Runoff and erosion has important off-site impacts in this drainage basin with, for example, muddy floods affecting roads and property. The flooding in August 2016 affected roads (Fig. 2a) and the fine-grained sediment also covered fruit orchards (Fig. 2b). The hillslopes are linked directly to the river channel by steep gradients in sub-basins 1 and 3. In sub-basin 2, however, the hillslopes have longer lengths. The average slope gradients of sub-basins 1, 2 and 3 are 91%, 81% and 108%, respectively. The lithology of sub-basin 1 comprises C-TR-ssh (88%), Db (9%) and Quaternary older terraces (3%). The lithology of sub-basin 2 is 65% C-TR-ssh, 23% Db, 7% Cf and 5% Quaternary older terraces. In sub-basin 3, the geological formations comprise 85% C-TR-ssh, 7.5% Db and 7.5% Jb. Collectively, these three sub-basins were representative of the main combinations of exposed highly erodible lithologies in the study area (Fig. 1).

2.2. Field sampling and laboratory measurements of sediment source tracers

Samples of sediment deposited on the river bed were collected at the overall outlet of the main drainage basin and at the three sub-basin outlets (Fig. 1). Samples collected at the former were used as the target sediment for apportioning the relative contributions from the upstream sub-basins (see schematic in Fig. 3). River bed fine-grained sediment samples that appeared to have been recently deposited were collected at each of the channel sampling sites (Figs. 2c-f). These samples have been referred to as 'drape' sediment deposits in previous work (Collins and Walling, 2007; Olley et al., 2013; Walling et al., 1998). This approach has previously been used in Iran to fingerprint sediment in a

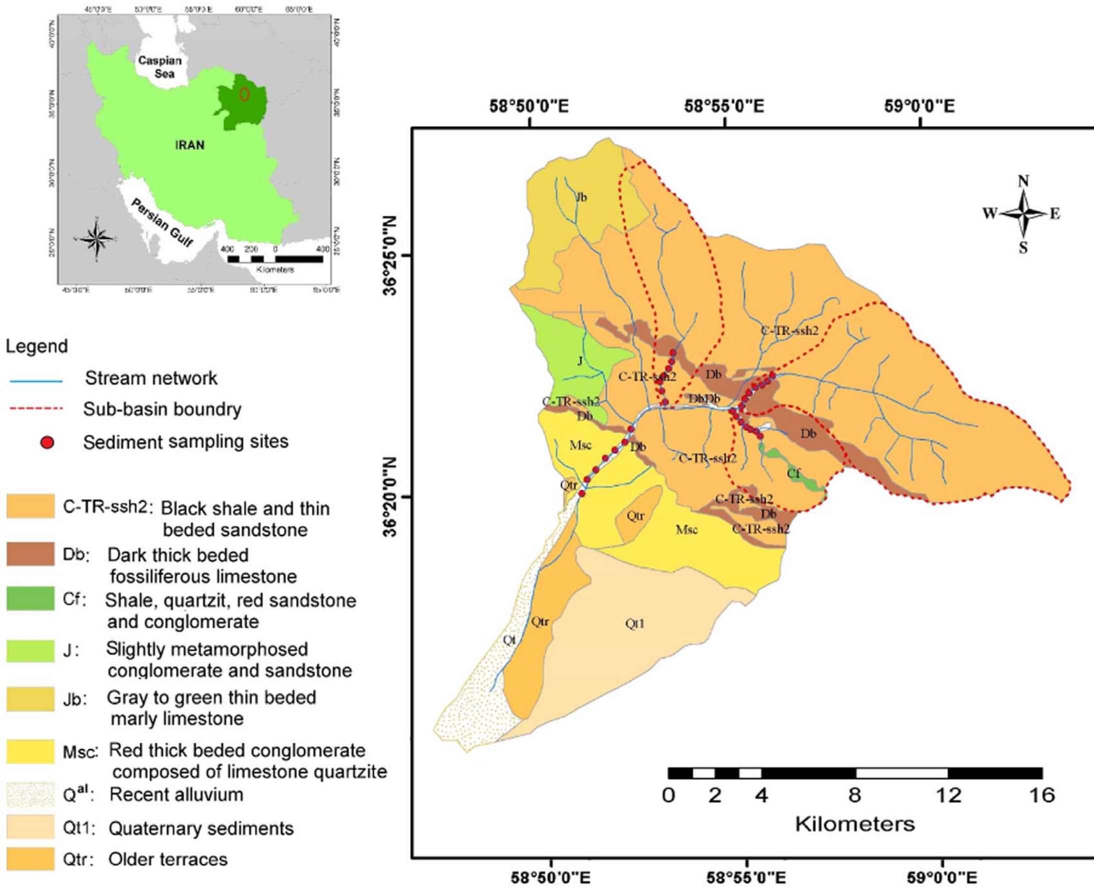

Fig. 1. Map of the Mirabad River Basin, showing the location of the catchment in north-western Iran, in Khorasan Province, geology and sediment sampling locations in the three representative sub-basins and downstream on the main stem.

region in which it was difficult to sample suspended sediment during high flow conditions due to poor access and remoteness (Nosrati, 2017). Fine-grained sediment was sampled after a high magnitude flood event with a 50-yr peak discharge of 10.5 $m^3 s^{-1}$ (23 August 2016) from the main channel and three major tributaries in the study area (Fig. 1). This major flood was assumed to have transported a major portion of the annual suspended sediment load. In order to ensure the sediment samples were as representative as labour and financial resources permitted, 10 sub-samples were collected in a ~20 m long reach (interval 2 m) at each sampling site and combined into individual composite samples. Seven composite samples were retrieved from each sub-basin outlet and a further seven composite samples from the overall outlet. Composite sample masses were ~500 g. A composite sampling procedure based on replicated sub-samples is needed to take account of potential sediment property variability resulting from water mixing and stream reach characteristics.

Grain-size effects resulting from selective sediment delivery between catchment sources and river channels (Stone and Walling, 1997; Viparelli et al., 2013; Walling et al., 2000) can bias fingerprint property data (Laceby et al., 2017; Pulley et al., 2017). Such bias can confound the direct comparison of source and target sediment samples. To date, the most common approach for addressing this issue has been particle size fractionation, using either the < 63 μm (Walling et al., 1993) or < 10 μm (Douglas et al., 2003) fractions. Dry sieving revealed that the < 63 μm fraction was more representative (by mass) of the surface drape sediment samples collected in this study. Consequently, only the < 63 μm fraction of the sub-basin outlet and downstream main stem sediment samples were used for the analysis and comparison of fingerprint properties. Fractionation was undertaken using dry sieving.

Sieving to a broad size fraction such as < 63 μm may not address all uncertainties associated with particle size effects in situations where the samples being compared are comprised of contrasting grain sizes within that overarching fraction. Consequently, in addition to undertaking sample fractionation, many fingerprinting studies have also applied additional particle size corrections to take account of contrasts between the < 63 μm fractions of source and sediment samples (Collins et al., 1997; Owens et al., 2000; Walling et al., 2006; Walling et al., 2008). Given the uncertainties associated with comparing soil and sediment samples and the corresponding corrections applied for grain size effects resulting from slope to channel sediment delivery, some previous studies have also adopted a tributary confluence sampling design similar to the one adopted here (Caitcheon, 1993; Collins et al., 1996; Vale et al., 2016; Walling et al., 1999). The assumption here is that comparing sub-basin outlet sediment samples used to represent potential catchment sources, with downstream overall outlet sediment samples, removes much of the uncertainty associated with slope to channel routing and delivery. Using sub-basin outlet sediment samples as spatially-integrated samples of the material mobilised from potential sources also avoids the resource issues and uncertainties associated with attempting to be representative of spatially-distributed slope-based source categories such as different land cover types. This is especially relevant to larger drainage basins.

In order to measure the concentrations of geochemical tracers, one gram of the sediment samples (< 63 μm) was digested in aqua regia (HCl–HNO3; 3:1) using a Velp Thermo-reactor at 95 °C for 2 h. After filtering the extracts through S&S ME24 (0.2 μm) filter papers, the solutions were analysed by a Varian SpectrAA-20 Plus calibrated using an element standard solution (Merck KGaA, Frankfurt, Germany) for Ca, Cu, Fe, K, Mg, Mn, Na, and Ni in the Geochemical Laboratory at the Faculty of Earth Sciences, Shahid Beheshti University, Tehran. The

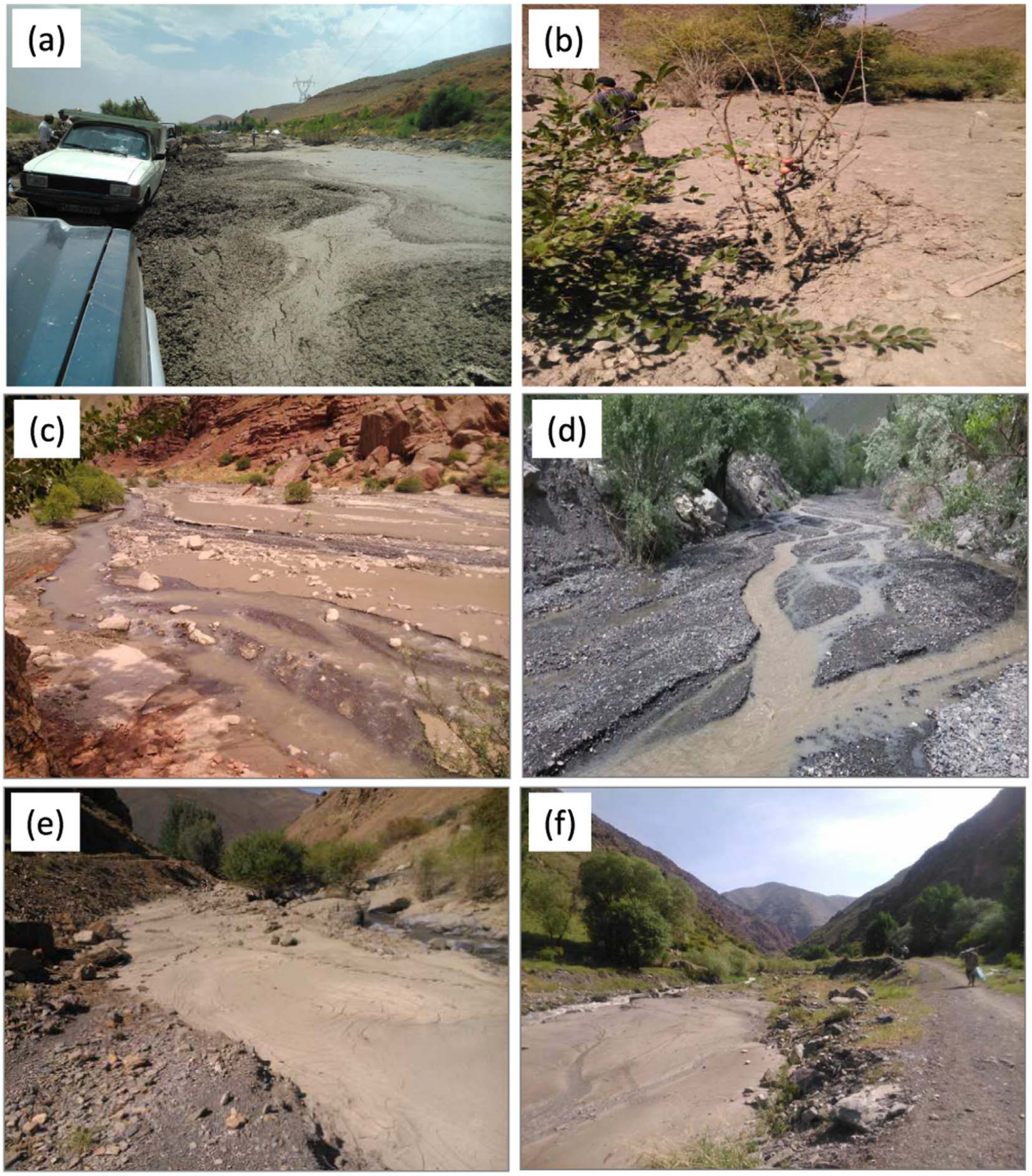

Fig. 2. Photos showing the off-site impacts of sediment mobilisation and delivery on (a) roads and (b) property. Photos taken at the outlets of the (c) main basin (target sediment sampling location for apportioning sub-basin sources) and three upstream sub-basins (representing potential spatial sediment sources): (d) sub-basin 1, (e) sub-basin 2, and (f) sub-basin 3. Refer to Fig. 1 for the locations of these main stem and tributary sub-basin outlet sediment sampling sites.

results showed that analytical error was < 5% for all elements. Here, it is useful to note that although the extraction for geochemical elements provided pseudototal concentrations in the absence of complete dissolution using a more powerful acid matrix, source fingerprinting compares samples and thus the consistency in tracer extraction across samples is, in the main, more important than the absolute magnitude of the tracer concentrations. Use of aqua regia reagents to extract geochemical fingerprints has been widely reported in the literature (e.g. Owens et al., 2000; Walling et al., 2006; Walling et al., 2008). Total organic carbon content was measured by the Walkley-Black method (Skjemstad and Baldock, 2008) in the Geomorphology Laboratory at the Faculty of Earth Sciences, Shahid Beheshti University, Tehran.

2.3. Statistical discrimination of tributary sub-basin sediment sources

A standard bracket or range test (Foster and Lees, 2000) was used to identify significantly non-conservative tracers, whereby the tracer concentrations in the target sediment samples collected from the main stem outlet were compared with the corresponding ranges associated with the sub-basin samples (Zhang and Liu, 2016). This test has been, and continues to be, applied worldwide as part of sediment source tracing procedures (e.g. Collins et al., 2017; Gellis and Walling, 2011; Gellis and Noe, 2013; Laceby et al., 2015; Mukundan et al., 2010; Owens et al., 2017; Walling, 2013; Wilkinson et al., 2013). Tracers failing the bracket test (i.e. tracer concentrations measured for the downstream target sediment samples fell outside the corresponding ranges of the upstream sub-basin sediment sample tracer concentrations) were removed from further analysis. The range test does not confirm the complete absence of tracer property transformation but, instead, provides a rudimentary screening for removing tracers undergoing significant change during transport between upstream source and downstream sediment sampling sites.

The statistical analysis employed to identify different composite fingerprints for discriminating between the potential sub-basin sources used three approaches: (1) the Kruskal–Wallis H-test (KW-H), (2) a combination of the KW-H as step one and discriminant function analysis

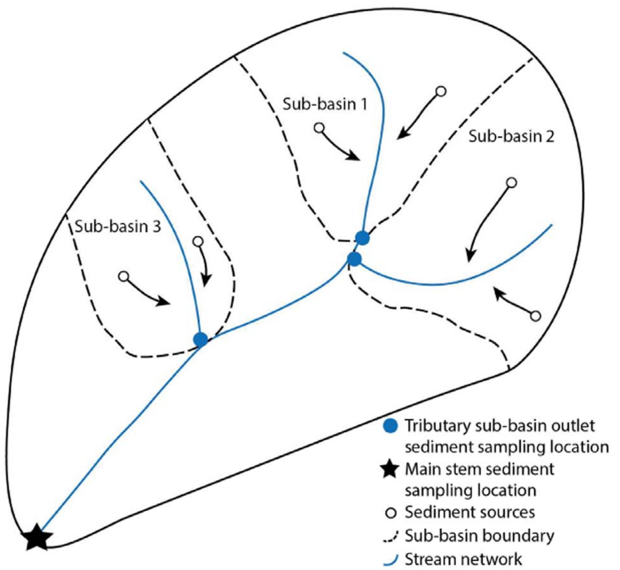

Fig. 3. Schematic of a tributary sub-basin sampling design for sediment source fingerprinting.

(DFA) as step two, and (3) a combination of the KW-H as step one and principal component & classification analysis (PCCA) as the second step. Three final composite signatures were selected on this basis. All statistical analyses were performed using STATISTICA V.8.0 (StatSoft, 2008). The scientific basis for using different statistical methods to identify alternative composite signatures is now well-established in the international literature (e.g. Collins et al., 2012; Palazón et al., 2015) and reflects the desire to take explicit account of the impact of different composite signatures on estimated source apportionment. Since independent tests are based on different principles and rules, their application ensures a multi- rather than single-dimension analysis of the available tracer data. This is considered more informative than running the same test multiple times to identify different composite signatures (i.e. with increasing numbers of tracers). Previous work has already investigated the impact of differing numbers of properties in composite signatures on the goodness-of-fit between source-weighted and measured tracer concentrations in sediment mixtures (e.g. Sherriff et al., 2015). Although three combinations of statistical tests were applied in this study, more combinations could have been used. It is important, nevertheless, to rationalise data processing and the three approaches used provided a range of tests founded on different principles.

2.3.1. Kruskal–Wallis H-test

The KW-H is a non-parametric equivalent of one-way ANOVA to compare more than two groups, and tests the null hypothesis that the different groups in the comparison were drawn from the same distribution or from distributions with the same median. However, unlike one-way ANOVA, it does not make assumptions about homogeneity of variance or normal distributions. Thus, the interpretation of the KW-H is basically similar to that of parametric one-way ANOVA, except that it is based on ranks rather than means (Dytham, 2011).

2.3.2. Discriminant function analysis (DFA)

Those tracers exhibiting statistically significant differences between the tributary sub- basin sediment sources, using KW-H, were included in the DFA. The basis of DFA is to provide a set of weightings that allow the source groups to be distinguished. The weightings can then be used on individuals that are not assigned to a group to provide a probability of them belonging to each of the possible groups. Different tests including eigenvalue, canonical correlation, Wilks' lambda, and squared Mahalanobis were used to determine whether the discriminant functions were statistically significant. Membership of the spatial sediment source groups was the dependent variable, whereas the measured tracers constituted the independent variables.

2.3.3. Principal component & classification analysis (PCCA)

PCCA can be used as a classification technique in addition to reducing the dimensions of the original variable space so that the relations among variables and cases can be highlighted. To do this, the variables and the cases are plotted in the space generated by the factor axes. This technique works in very much the same way as Principal Component Analysis (PCA) but with one crucial difference; the individuals must be assigned to groups before the analysis. The test then calculates the variable weightings that will maximize the differences between groups rather than individuals as is the case with PCA. The PCCA produces weightings that will allow you to identify those variables that are the most different between groups and discard those that are the same.

Only those tracers with significant differences between the spatial sediment sources, using KW-H, were included in the PCCA. Principal components with eigenvalues > 1 were retained and subjected to a varimax rotation to minimize the number of tracers that have high loadings on each principal component (PC). Under a particular PC, each tracer is given a weight or factor loading that represents the contribution of that tracer to the composition of the PC. Only the highly-weighted tracers were retained from each PC. Highly weighted tracer loadings were defined as having absolute values within 10% of the highest tracer loading. When more than one tracer was retained under a single PC, multivariate correlation coefficients were employed to determine if the tracers could be considered redundant and, therefore, eliminated from the final set of tracers (i.e. composite fingerprint). If the highly- weighted tracers were not correlated (assumed to be a correlation coefficient < 0.60) then each was considered important, and thus, retained in the final composite signature. Among well correlated tracers, the tracer with the highest PC loading (absolute value) was chosen for the composite fingerprint. Once the composite signature was chosen, a final check was undertaken to identify significant differences among the spatial sediment sources based on the PC scores of each sample using one-way ANOVA (F-test) and Tukey HSD post hoc tests ($p < 0.05$).

2.4. Source apportionment using the Modified MixSIR Bayesian un-mixing model

Some recent sediment source tracing studies applying un-mixing models have used the Modified MixSIR Bayesian model (Nosrati et al., 2014). This model was used here to compare the sub-basin spatial sediment source contributions predicted by the three final composite signatures. Modelling source contributions using more than one composite signature permits an assessment of the potential uncertainty resulting from different fingerprint property sets (Collins et al., 2012).

The Modified MixSIR Bayesian statistical approach, proposed by Nosrati et al. (2014), quantifies the relative contributions of sediment from different sources by calculating probability distributions for the proportional contribution (f_i) of each source i to the downstream target sediment samples in three stages: 1) determination of the prior probability distributions for model parameters, 2) construction of a likelihood function for the statistical model, and 3) derivation of the posterior probability distributions for the parameters using the Bayes rule to adjust the prior distribution based on the observed data. The Bayes rule states that the posterior probability distribution for all f_i is proportional to the prior probability distributions multiplied by the likelihood, and then dividing by their sum.

$$P\left(f_q \;\middle|\; data\right) = \frac{L(data\ |f_q) \times p(f_q)}{\sum L(data\ |f_q) \times p(f_q)} \tag{1}$$

where L(data|fq) is the likelihood of the data given f_q, $p(f_q)$

representing the prior probability being true, based on prior information, and f_q is the proportional source contributions of q proposed vectors.

The relative contributions of sediment are factored into the model by defining mean and variance parameters for each sediment source i and the final sets of tracers (composite fingerprints) j.

The proposed tracer distributions for the target sediment mixture collected from the overall main stem outlet are determined by solving for the proposed means $\hat{\mu}_j$ and standard deviations $\hat{\sigma}_j$ of the sediment mixture based on the randomly drawn f_i values comprising a vector f_q:

$$\hat{\mu}_j = \sum_{i=1}^{n} (f_i \times m_{j_{Source_i}}) \tag{2}$$

$$\hat{\sigma}_j = \sqrt{\sum_{i=1}^{n} (f_i^2 \times S_{j_{Source_i}}^2)} \tag{3}$$

where $m_{j_{Sourcei}}$ in Eq. (3) is the mean and $S_{j_{Sourcei}}{}^2$ in Eq. (4) is the variance of the jth sediment tracer and the ith tributary sub-basin source.

Based on the $\hat{\mu}_j$ and $\hat{\sigma}_j$ of each final composite fingerprint, the likelihood of the data given the proposed sediment mixture was calculated as:

$$L(x \mid \hat{\mu}_j, \hat{\sigma}_j) = \prod_{k=1}^{n} \prod_{j=1}^{n} \left[\frac{1}{\hat{\sigma}_j \times \sqrt{2 \times \pi}} \times \exp\left(-\frac{(X_{kj} - \hat{\mu}_j)^2}{2 \times \hat{\sigma}_j^2} \right) \right] \tag{4}$$

where X_{kj} represents the *j*th tracer property of the *k*th sediment sample.

Using a version of the sampling-importance-resampling (SIR) algorithm (Moore and Semmens, 2008), we generated 10^6 samples from the posterior distribution of the estimated target sediment mixture. This method establishes a threshold acceptance value prior to sampling and uses it simultaneously to resample, as the un-normalized posterior probabilities for each fq sample are calculated.

The two-sample Kolmogorov-Smirnov test was used to confirm statistically significant differences between the distributions of posterior proportional contributions computed for the three spatial sediment sources using each of the composite signatures selected by the different statistical approaches for source discrimination. This test was selected since it is sensitive to differences in the location and shape of predicted frequency distributions and has been used previously, to compare sediment mixing model outputs (Collins et al., 2010a). Use of the Kolmogorov-Smirnov test provided a means of comparing the predicted source proportions on the basis of the entire distributions of model outputs generated by uncertainty analysis.

Predicted source proportions based on the composite signatures selected using the different statistical approaches were also compared using the root mean square difference (RMSD). This analysis assessed the magnitude of the difference between the predicted source proportions for the three sources and for each of the composite signatures selected using the different statistical approaches (Eq. (5)):

$$RMSD = \sqrt{\frac{\sum_{i=1}^{n} (Y_{i1} - Y_{i2})^2}{n}} \tag{5}$$

where, Y_{i1} and Y_{i2} are the relative contributions of a specific source (i) based on the composite signatures selected using the different statistical approaches (e.g. Y_{11} is the relative contribution of sub-basin 1 based on KW-H tracers, and Y_{12} is the relative contribution of sub-basin 1 based on tracers selected using KW-H and DFA), and n is the number of sources ($n = 3$: sub-basin 1, sub-basin 2 and sub-basin 3).

3. Results and discussion

3.1. Final composite fingerprints

Table 1 compares the tracer concentrations in the sub-basin outlet (spatial sediment sources) and overall downstream outlet sediment samples. The results of the bracket test showed that Fe and Ca were non-conservative tracers and so seven remaining tracers were retained and tested using the KW-H test. Table 1 also presents the results of applying the KW-H test which indicated that six properties (Cu, K, Mg, Mn, Ni and organic carbon) showed a statistically significant difference between the tributary sub-basin spatial sediment sources. The tracer Na was unable to discriminate the sub-basin spatial sources and so was discarded from further analysis.

The six properties identified by the KW-H test were then entered into the stepwise DFA (Table 2). The first function of Wilk's lambda value (0.02) indicated that 98% of the total variance among the sub-basin spatial sediment sources was explained by the tracers. The canonical correlation value was 0.96 and indicated a strong correlation between the discriminant scores and the source groups. Wilks' lambda was used to indicate the statistical significance of the discriminatory power of the variables in the DFA model. Three properties were selected using Wilks' lambda and the values indicated that the discriminatory power of Cu and Mg is perfect. Partial Wilks' lambda is the Wilks' lambda for the unique contribution of the respective variable to the discrimination between source groups. The smaller the Partial Wilks' lambda, the greater the contribution to the overall discrimination. The Partial Wilks' lambda values indicated that Cu contributed the most, Mg second most and K the least to the overall discrimination (Table 2).

The squared Mahalanobis distance showed that the sediment sources were well separated by a significant distance (Table 3). The backward stepwise DFA yielded classification matrices assigning 100% of the cases correctly with three (Cu, Mg, K) tracers (Table 3). A scatterplot using the first and second discriminant functions calculated using backward DFA confirmed that the sediment samples collected from the outlets of the different sub-basin spatial sediment sources are well separated (Fig. 4).

Tracers passing the KW-H test (Cu, K, Mg, Mn, Ni and OC) were also tested using PCCA. All tracers were then further explored as an alternative means of reducing the number of tracers and problems of multicollinearity. The results of PCCA showed that the first two principal components (PCs) with eigenvalues > 1 accounted for > 75% of the variability among the tracer values for the source groups (Table 4). The PC corresponding to the largest eigenvalue (2.8) accounted for approximately 46% of the total variance. The second PC corresponding to the second eigenvalue (1.8) accounted for approximately 29% of the total variance. All meaningful loadings (i.e. loadings > 0.70) were included in the interpretation of PCs.

The highly-weighted tracers under PC1 with absolute values within 10% of the highest tracer loading (the loading of selected tracers should be larger than 0.80) were Mg and Mn. Both were retained for the final composite signature because they were not strongly correlated ($r = -0.54$). Under PC2, the highly-weighted tracers with absolute values within 10% of the highest tracer loading (the loading of selected tracers should be larger than 0.83) were Cu and OC. Both were retained for the final composite signature because Cu was not strongly correlated with OC ($r = -0.58$).

The plot of factor coordinates of tracers for the first two PCs showed that the four tracers were represented by the current coordinate system (the range of correlation coefficients; -1 to $+1$) (Fig. 5a). Because PCCA was based on correlations, the closer a tracer in this plot is to the unit circle; the better is its representation by the current coordinate system. Thus, the set of selected tracers (i.e. composite fingerprint) clearly provided discrimination between the three potential sub-basin sediment sources (Fig. 5b). The results illustrated that PCCA can be used as a tool for identifying important dimensions in a set of tracers and to identify those sediment sources with similar or dissimilar characteristics.

PCs scores were calculated using the resulting component score coefficient matrix and tested for significant differences between the sediment sources (Table 4). PC scores for both PCs varied significantly

Table 1
Tracer concentration data for the different sub-basin spatial sediment sources and downstream main stem outlet sediment samples and the Kruskal-Wallis H-test results for discriminating the sub-basin spatial sediment sources. The units of all elements are mg kg^{-1} except OC which is expressed as g kg^{-1}.

		Ca	Cu	Fe	K	Mg	Mn	Na	Ni	OC
Sediment sources										
Sub-basin 1		535.7	0.51	548.0	5.0	220.7	15.3	2.1	0.42	1.1
		571.4	0.51	599.3	6.5	250.8	16.1	2.5	0.52	1.2
		571.4	0.51	548.0	5.7	268.7	14.5	2.3	0.42	1.4
		607.1	0.49	414.5	5.4	223.1	11.5	11.4	0.44	1.5
		571.4	0.49	486.2	5.8	215.8	14.3	1.9	0.52	1.2
		500.0	0.54	570.4	4.9	203.6	15.8	2.0	0.50	1.4
		500.0	0.51	509.9	5.5	205.2	14.8	1.4	0.42	1.5
	Mean	551.0	0.51	525.2	5.6	226.8	14.6	3.4	0.46	1.3
	SD	40.5	0.02	61.4	0.5	24.2	1.5	3.6	0.05	0.2
Sub-basin 2		571.4	0.60	510.5	11.3	168.6	18.4	3.0	0.34	1.2
		571.4	0.62	515.1	12.9	171.0	15.8	5.2	0.46	0.8
		714.3	0.62	515.8	15.5	184.0	16.8	5.5	0.50	0.6
		642.9	0.64	621.7	11.5	174.3	20.2	1.6	0.44	1.2
		642.9	0.59	640.8	11.7	167.8	18.6	2.7	0.32	0.8
		571.4	0.58	545.4	10.7	169.4	17.3	2.1	0.38	1.1
		571.4	0.55	988.2	13.0	166.1	25.0	2.6	0.36	1.2
	Mean	612.2	0.60	619.6	12.4	171.6	18.9	3.2	0.40	1.0
	SD	56.2	0.03	171.0	1.6	6.1	3.0	1.5	0.07	0.3
Sub-basin 3		500.0	0.49	646.1	7.4	159.6	17.6	3.3	0.36	1.4
		571.4	0.49	618.4	11.2	156.4	20.4	5.1	0.34	1.2
		571.4	0.46	491.4	7.7	161.2	19.1	2.2	0.42	2.9
		571.4	0.47	514.5	7.8	170.2	17.9	2.1	0.40	2.1
		571.4	0.47	492.8	7.4	157.2	15.1	2.2	0.38	1.4
		571.4	0.49	475.7	8.0	156.4	17.1	3.0	0.44	2.6
		724.5	0.50	477.6	10.1	155.5	15.6	1.9	0.34	1.2
	Mean	583.1	0.48	530.9	8.5	159.5	17.5	2.8	0.38	1.8
	SD	67.8	0.01	70.8	1.5	5.1	1.9	1.1	0.04	0.7
KW-H test	Chi-Square	n. c.	16.5	n. c.	17.5	16.5	10.5	1.7	6.1	9.9
	p-Value	n. c.	< 0.001*	n. c.	< 0.001*	< 0.001*	0.005*	0.42	0.04*	0.007*
Sediment samples										
S-1		642.9	0.51	498.0	7.3	206.8	13.3	2.6	0.42	1.7
S-2		571.4	0.49	499.3	7.6	176.7	13.8	5.6	0.44	1.2
S-3		607.1	0.49	660.5	7.0	200.3	14.8	3.2	0.4	1.8
S-4		750.0	0.51	430.9	9.7	188.1	14.5	2.2	0.32	1.4
S-5		642.9	0.50	544.7	7.7	204.4	15.3	3.0	0.38	1.7
S-6		607.1	0.50	621.1	6.8	197.1	15.1	2.0	0.4	1.1
S-7		678.6	0.49	492.1	8.5	178.3	14.0	5.9	0.38	1.2

* Critical *p*-value = 0.05. KW-H test, Kruskal-Wallis H-test; n. c., non-conservative tracer.

Table 2
Summary of the backward Discriminant Function Analysis (DFA).

Tracer	Wilks' lambda	Partial lambda	F-remove	*p*-Level	Tolerance
Cu	0.040	0.319	17.0	< 0.001	0.98
Mg	0.039	0.327	16.4	< 0.001	0.96
K	0.024	0.544	6.7	0.01	0.94

Table 3
Summary of the backward Discriminant Function Analysis (DFA) using the seven composite sediment samples collected from each tributary sub-basin.

DFA parameters	Result
Sediment sources samples classified correctly (%)	
Sub-basin 1	100
Sub-basin 2	100
Sub-basin 3	100
Total	100
Squared Mahalanobis distance	
Sub-basin 1 × Sub-basin 2	62.9
Sub-basin 1 × Sub-basin 3	33.8
Sub-basin 2 × Sub-basin 3	35.4
Squared Mahalanobis F-value	
Sub-basin 1 × Sub-basin 2	65.3
Sub-basin 1 × Sub-basin 3	35.1
Sub-basin 2 × Sub-basin 3	36.7

with sediment sources (Table 4). Thus, the tracers related to these PCs provided a basis for selection of an alternative composite signature (Mg, Mn, Cu, OC).

3.2. Spatial sediment source contributions

Using priors and estimates of uncertainty associated with the un-mixing model inputs (source proportions and means and corresponding variance for each tracer in any given composite signature for each potential tributary sub-basin sediment source), a Modified MixSIR model run of 10^6 iterations resulted in convergence (i.e. further model runs will not alter the results) on the posterior source contributions from the sub-basin spatial sources using three different composite signatures (Fig. 6). The model resampled a total of 6325, 5779 and 6297 posterior draws with no duplicates for tracers selected for the three independent composite signatures identified using KW-H, a combination of KW-H and DFA and a combination of K-HW and PCCA. The maximum importance ratio values (calculated by determining the ratio of the maximum un-normalized posterior probability resample to the sum of all un-normalized posterior probability resamples) were < 0.01 indicating that the un-mixing model was effective in estimating the true posterior density. Additionally, likelihood distribution plots were drawn based on the relative likelihood values (calculated as the ratio of the re-sampled un-normalized posterior probability values to the single largest value in the set of posterior) and these demonstrated that the model placed appropriate weights on the tails of the posterior

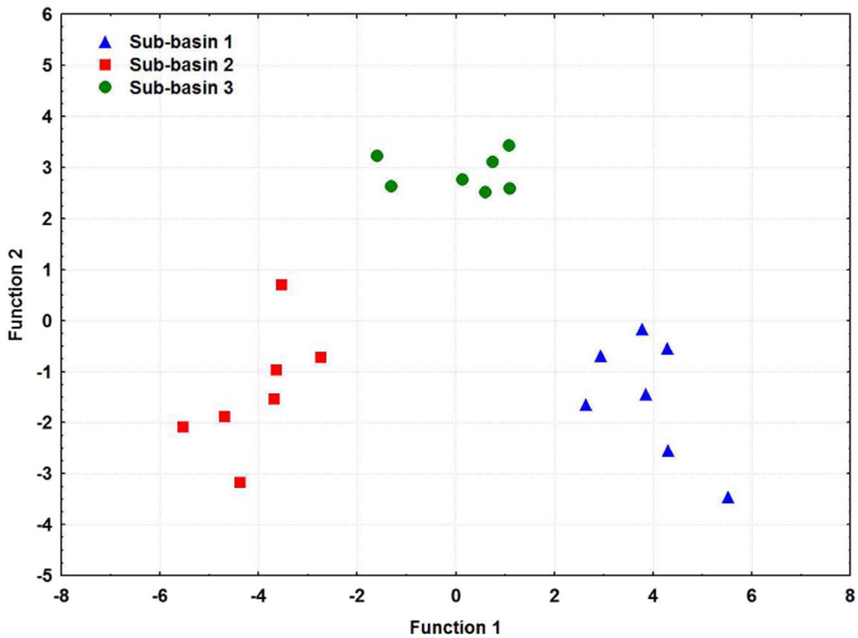

Fig. 4. Scatterplot of the first and second discriminant functions calculated using backward DFA associated with selection of the composite signature comprising (Cu, Mg and K).

Table 4
Principal component and classification analysis (PCCA) factor coordinates of the variables and the eigenvalues of the correlation matrix.

	PC 1	PC2
Tracer		
Cu	0.12	**0.92**
K	0.62	**0.71**
Mg	**− 0.89**[1]	0.01
Mn	**0.80**	0.19
Ni	**− 0.75**	0.08
Organic C	0.17	**− 0.85**
Eigenvalue	2.8	1.8
% Total variance	46.0	29.2
Cumulative % variance	46.0	75.2
Mean scores of the three sediment sources		
Sub-basin 1	− 1.3 a[2]	− 0.91 a
Sub-basin 2	0.54b	− 0.24 a
Sub-basin 3	0.69b	1.15 b
ANOVA results		
F-value	34.7	29.8
p-Value	< 0.0001	< 0.0001

[1] Bold values indicate strong (> 0.7) loadings.
[2] Different small letters indicate that scores are significantly different at the 5% level, based on the Tukey HSD post hoc test.

distributions (Fig. 6).

Using KW-H (Table 5), the relative contributions from the sub-basin spatial sources 1, 2 and 3 were estimated as 45.6%, 3.8% and 50.6%, respectively. Using the composite signature selected by combining KW-H and DFA (Table 5), the corresponding respective contributions from the sub-basins were estimated as 46.8%, 18.8% and 34.4%. On the basis of the composite signature selected using a combination of KW-H and PCCA (Table 5), the relative contributions from sub-basins 1, 2 and 3 were estimated as 61%, 2.5% and 36.5%, respectively. The predicted spatial source contributions were therefore sensitive to the composite fingerprint used (cf. Collins et al., 2012; Palazón and Navas, 2017).

Pairwise comparisons of the distributions of posterior proportional contributions predicted for the three sub-basin spatial sediment sources using the three different composite signatures selected by different statistical approaches are presented in Table 6. The Z statistic in Kolmogorov-Smirnov test is a product of the combined sample size and the largest absolute difference between the two distributions being

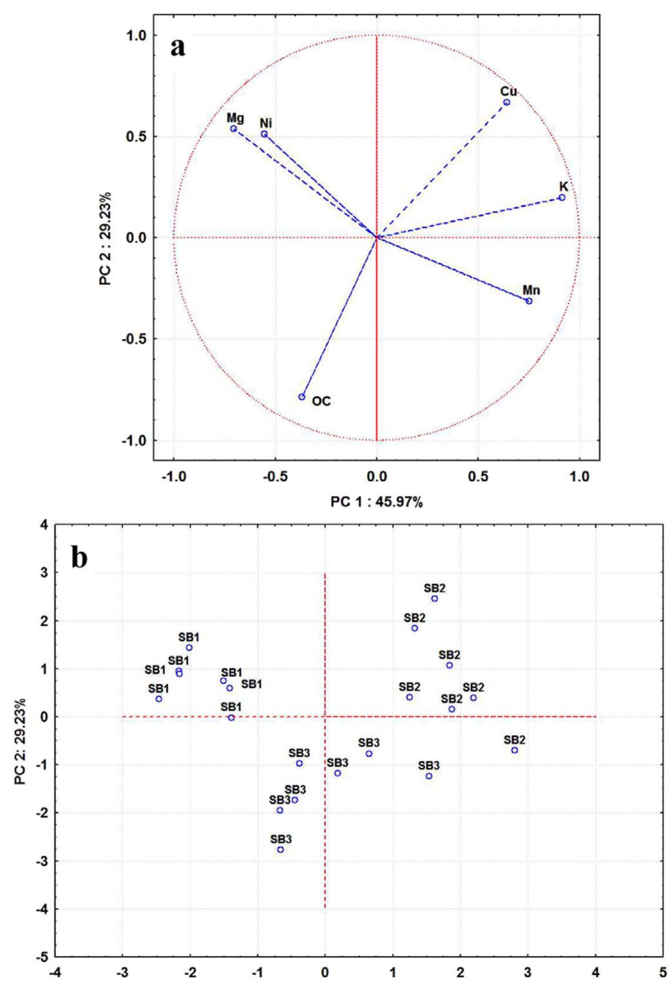

Fig. 5. (a) Projection of the optimum composite tracers on the factor-plane using PCCA, (b) Projection of the cases on the factor-plane using PCCA; SB1: sub-basin 1; SB 2: sub-basin 2; SB 3: sub-basin 3.

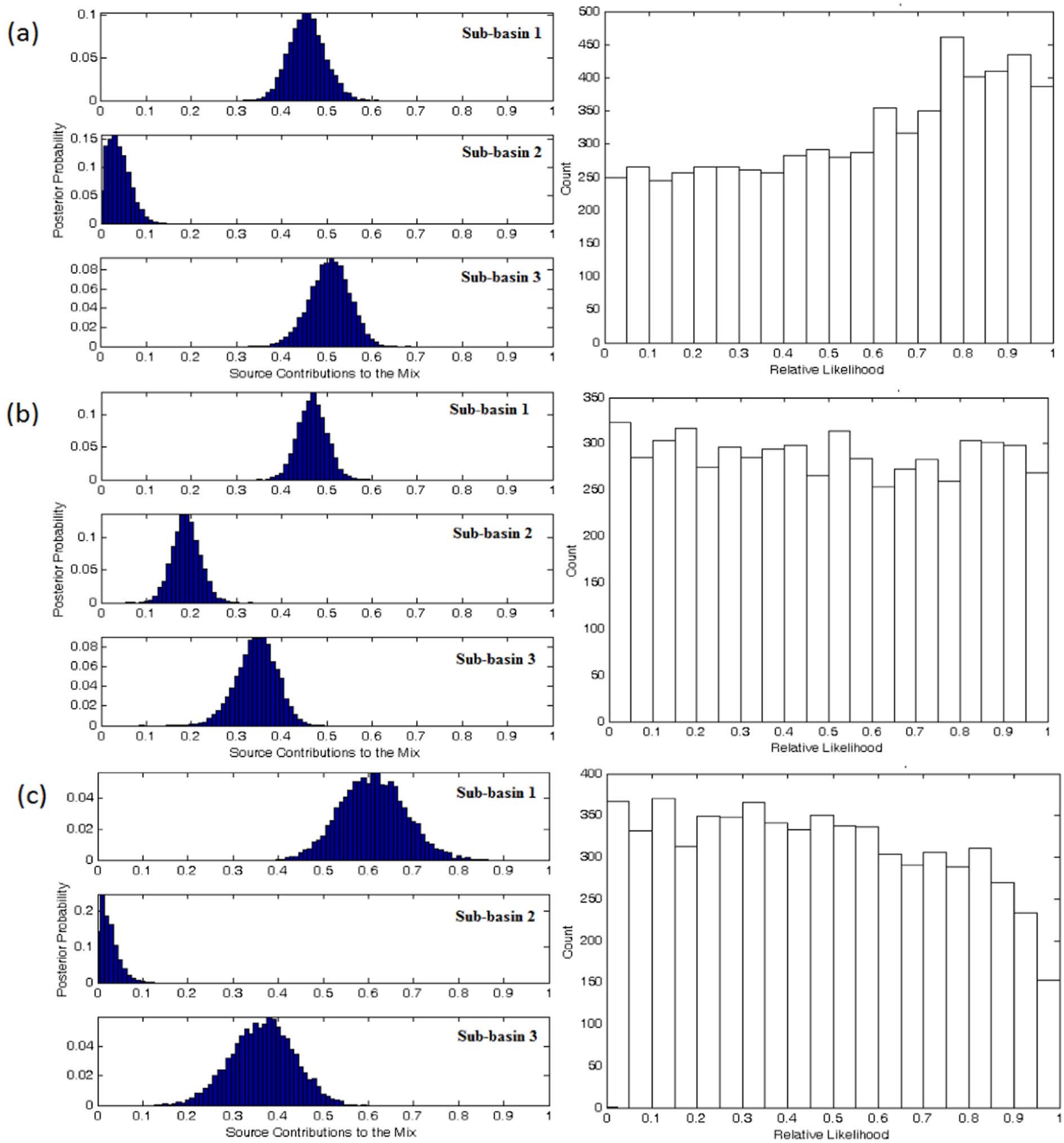

Fig. 6. Estimation of source contributions using data from (a) KW-H, (b) combined KW-H as the first step and discriminant function analysis (DFA) as the second step, and (c) combined KW-H as the first step and principal component & classification analysis (PCCA) as the second step. The histograms represent the relative likelihoods for Bayesian un-mixing model runs based on estimation of the source contributions to the target sediment samples collected from the overall outlet.

compared. A significance value of < 0.05 indicates that the two distributions are significantly different. The results of the Kolmogorov-Smirnov test suggested that the distributions computed for the three sub-basin spatial sediment sources using the three different composite signatures selected by different statistical approaches were statistically significant for all pairwise comparisons. Depending on the composite signature used: the root mean square difference was ca. 12% (Table 6).

3.3. Limitations and uncertainties

The source apportionment results must be interpreted in the context of some limitations and uncertainties. Source sample numbers collected by any tracing investigation are inevitably constrained by available budgets as well as practical considerations including those associated with the mountainous terrain of the study area and rarely, if ever, satisfy statistically-based probability sampling. Given the challenges of deploying a more traditional slope-based source sampling strategy in the mountainous study area, a tributary-based sampling strategy was deployed as an alternative approach. The collection of sub-samples which are bulked into composite samples for laboratory analysis improves representativeness by taking account of micro-scale spatial variations within the river channel. Here, however, it is important to bear in mind that source estimates are scale dependent in that they can differ for different sampling locations along a channel network (Koiter et al., 2013). Target river sediment for source apportionment was collected from a single downstream location on the main stem of the study river. The estimated source proportions therefore relate to this sampling site. Sediment sampling also needs to be temporally representative and the approach adopted by this study was to sample immediately post a major flood event which could be assumed to have transported a

Table 5
Relative contributions from the three sub-basin spatial sediment sources to the sediment samples collected downstream at the overall outlet using composite signatures selected by different statistical approaches.

Statistical approaches for selecting composite fingerprints	Spatial sediment sources					
	Sub-basin 1 (%)		Sub-basin 2 (%)		Sub-basin 3 (%)	
	Mean	SD	Mean	SD	Mean	SD
KW-H (Tracers: Cu, K, Mg, Mn, Ni and OC)	45.6	3.9	3.8	2.5	50.6	4.4
Combination of KW-H and DFA (Tracers: Cu,Mg and K)	46.8	3.2	18.8	3.0	34.4	4.4
Combination of KW-H and PCCA (Tracers: Cu, Mg, Mn and OC)	61.0	7.1	2.5	2.0	36.5	6.9

significant proportion of the annual sediment load. It was assumed that any tracer property transformation during transit to, and through, the river channel system, was not significant enough to impact on the predicted source proportions. Although tracer properties were tested for significant transformation using the standard bracket test, this does not confirm a complete absence of tracer transformation and again, this should be borne in mind. A number of factors can potentially influence sediment tracer conservation during the delivery continuum from source to outlet. These factors include biogeochemical processes such as adsorption or desorption (Forstnei and Salomons, 1980), as well as physical factors such as particle size sorting (Grygar and Popelka, 2016; Horowitz, 1991). The propensity for mobilised sediment to undergo some storage at intermediate locations in river basins (Fryirs, 2013; Skalak and Pizzuto, 2010) also provides scope for tracer transformation (Foster et al., 2008; Grygar and Popelka, 2016; Hudson-Edwards et al., 1998; Palmer and Douglas, 2008). Some previous research has examined the issue of tracer conservation experimentally (e.g. Motha et al., 2002) and both past (Motha et al., 2004) or more recent (Sherriff et al., 2015) work has included explicit assessment of tracer transformation in sediment mixing modelling. Attempts have also been made to pre-screen tracers before laboratory analyses using expert opinion on tracer behaviour (Kraushaar et al., 2015). Despite such work, however, there remains no consensus as to the most pragmatic way to assess tracer conservative behaviour in more detail and the bracket test thereby remains a standard component of fingerprinting methodological decision-trees. In the study reported herein, the sampling of deviate tracer values during the apportionment modelling, using tracer distributions constructed on the basis of the sediment samples collected and analysed, provides an additional means of taking some account of potential tracer transformations using physically-grounded data from replicate samples. To improve robustness here, main channel sediment samples could be collected from more than one reach to explore any potential scale-dependency associated with sediment tracer transformation during conveyance through the system. Tributary-based source sampling did not cover all confluences, but instead, those associated with the major tributaries draining the most erodible and well-connected portions of the study basin. This was confirmed by field surveys examining the connectivity of slopes to local river networks to identify potential major sub-basin spatial sediment sources for informing the final selection of tributaries. The most downstream sediment signature from the overall outlet is therefore unlikely to have been distorted by contributions from non-sampled tributaries since, on the basis of visual evidence and discharge monitoring, these were not discharging significant sediment loads to the main stem of the channel network. Equally, there was no well-developed bank or gully erosion along the main stem between the lowest confluence sampling site and the overall outlet sampling site. As a result, the signatures of the target sediment collected from the overall downstream outlet are unlikely to have been modified significantly by localised sediment inputs independent of the representative spatial zones included in the confluence sampling strategy (Stone et al., 2014). In the absence of using definitive mineralogy, the composite signatures identified in this study on the basis of geochemistry represented statistical solutions for source discrimination and further work is needed to validate the predicted source proportions. The three statistical procedures used herein to select composite signatures addressed the need to explore the sensitivity of predicted source apportionment to different composite signatures. Various statistical tests have been combined in this manner by previous work (e.g. Collins et al., 2012; Palazón et al., 2015). The guiding principle here should be that there is no fixed combination of tests, but rather, those applicable to the dataset in question and which are based on different mathematical procedures, should be applied in order to ensure that this particular aspect of sensitivity is explored explicitly. Where resources permit, a greater number of statistical tests might be included.

4. Conclusions

A modified Bayesian mixing model was used to estimate the relative contributions, with corresponding uncertainties, from three lithologically representative sub-basin sediment sources in a mountainous study catchment in north-western Iran. The findings provide some preliminary information to support the spatial targeting of conservation works which could include sediment dams to catch fine sediment being delivered to the main stem from the sampled tributaries. The success of the tributary sub-basin fingerprinting procedure in generating information on the relative importance of representative spatial sediment sources in this mountainous catchment, with obvious sampling challenges, demonstrates the potential for using this procedure in other mountain environments where conventional monitoring of sediment export from tributaries is absent, frequently in association with poor accessibility and problems for maintaining monitoring equipment. The confluence-based source tracing methodology also provides a more pragmatic solution where the collection of spatially distributed soil

Table 6
Kolmogorov-Smirnov test pairwise comparisons of the probability density functions computed for the predicted contributions from the three sub-basin spatial sediment sources based on composite signatures selected by different statistical approaches. The RSMD column shows an additional pairwise comparison of the source proportions predicted using the different composite signatures.

Paired statistical approaches in selecting tracers	Spatial sediment sources						Root mean square difference (RMSD; %)
	Sub-basin 1		Sub-basin 2		Sub-basin 3		
	Z	Sig.	Z	Sig.	Z	Sig.	
KW H-test vs. Combination of KW-H-test and DFA	17.2–18.2	< 0.0001	65.8	< 0.0001	65.8	< 0.0001	12.8
KW-H-test vs. Combination of KW-H-test and PCCA	68.2	< 0.0001	31.1	< 0.0001	67.7	< 0.0001	12.1
Combination of KW-H-test and DFA vs. Combination of KW-H test and PCCA	65.4	< 0.0001	65.8	< 0.0001	19.4	< 0.0001	12.6

samples for characterising potential sources is challenging due to steep mountainous terrain by, instead, focussing on the collection of sediment samples from a limited number of representative tributaries.

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