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The spatio-temporal dynamics of suspended sediment sources based on a novel indexing approach combining Bayesian geochemical fingerprinting with physically-based modelling

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Abstract:	Applications of sediment source fingerprinting continue to increase globally as the need for information to support improved management of the sediment problem persists. In our novel research, a Bayesian fingerprinting approach using MixSIAR was used with geochemical signatures, both without and with informative priors based on particle size and slope. The source estimates were compared with a newly proposed Source Sensitivity Index (SSI) and outputs from the INVEST-SDR model. MixSIAR results with informative priors indicated that agricultural and barren lands are the principal sediment sources (contributing ~5 to 85 % and ~5 to 80% respectively during two sampling periods i.e. 2018-2019 and 2021-2022) with forests being less important. The SSI spatial maps (using % clay and slope as informative priors) showed > 78% agreement with the spatial map derived using the INVEST-SDR model in terms of sub-catchment prioritization for spatial sediment source contributions. This study demonstrates the benefits of combining geochemical sediment source fingerprinting with SSI indices in larger catchments where the spatial prioritization of soil and water conservation is both challenging but warranted.
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The spatio-temporal dynamics of suspended sediment sources based on a novel indexing approach combining Bayesian geochemical fingerprinting with physically-based modelling

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Editor – *Journal of Environmental Management*
AGU

Dear Professor Dr. Jason Michael Evans,

<u>Research paper:</u> The spatio-temporal dynamics of suspended sediment sources based on a novel indexing approach combining Bayesian geochemical fingerprinting with physically-based modelling.

We had <u>two referee reports and comments from editor</u> suggesting minor revision on this manuscript. We have carefully revised the manuscript with acute attention and utmost respect.

We have <u>attended all comments</u> from all two reports. Please find the revised manuscript with track changed mode and the final version along with author response to each referee comments.

We do hope that the referees and the Editor would find the revised manuscript more interesting and recommend a publication in JEMA very soon.

We look forward to hearing from you.

Yours sincerely

Dr Renji Remesan, IIT Kharagpur

Editor's comments

General Comment: Following this message are the reviews of the above-referenced manuscript. We'll be pleased to accept this paper for publication after it's been revised in accordance with the reviewers' comments. Please proofread it carefully for typographical and grammatical errors. With the revised manuscript, please provide a detailed response to the reviewers' comments, indicating how each comment is addressed in the revised manuscript. If you disagree with any of the reviewers' comments, please address them in a rebuttal.

Reply: We would like to express our gratitude for the timely and positive review reports and for an indication that you are inclined to accept our manuscript after the suggested modifications. We have diligently addressed all the review comments provided by both reviewers with great care and consideration. The significant changes made to the R1 manuscript are as follows:

- We have included a new figure, Figure 6, which presents the spatial distribution of selected elemental proportions in the soil samples, as recommended by the reviewer.
- We have made modifications to Figure 1 in accordance with the suggestions provided by both reviewers 1 and 2.
- We have thoroughly proofread the manuscript using a Grammarly premium account to ensure accuracy and clarity.
- To further improve the R1 manuscript, we have incorporated a selection of the latest references relevant to the sediment source fingerprinting topic.

Reviewer 1 comments

General Comment 1 : Reviewer #1: The research paper "The spatio-temporal dynamics of suspended sediment sources based on a novel indexing approach combining Bayesian geochemical fingerprinting with physically based modelling" by Das and co-authors, submitted to Journal of Environmental Research (JEMA-D-23-07126), combines interesting and pertinent methodological approaches to the complex interactions between of soil erosion processes - sediment source identification - sediment delivery and land use at catchment scale, an important issue for soil quality and water resource management at regional scale. Combination / comparison of physical modelling and statistical approaches of sediment fingerprinting for delivery and soil source contribution issues lead to fruitful but rather complicated investigation and outputs. Together with land use classification, slope information and sub-catchment connectivity determinations, the study uses geochemical and textural (sediment grain size distribution) measurements to assess a source sensitivity index integrating physical and sediment source apportionment at catchment's scale. The authors also put forward some of the limitations and necessary future investigations of their study that is really of valuable assistance.

Reply: *We would like to express our sincere gratitude for the reviewer acknowledging the relevance of our work. The authors deeply appreciate the positive comments, which endorse our novel methodological approach. We firmly believe that our proposed indexing approach, which combines the outcomes of sediment fingerprinting and physically based modelling, holds significant potential in providing valuable management insights at the catchment level.*

General Comment 2: Although commendable efforts have been made to provide a well written manuscript, this paper is rather difficult to follow for non-specialized. The relationships between "real data", i.e., direct comparison between geochemical / textural properties of sediments and source soils (1st order approach) are not displayed and the paper directly deals with outputs from data treatments (2nd order approach). Furthermore, soil source samples are made up composite aliquots at each sampling site, a procedure that likely reduces potential geochemical discrimination properties. However, the references cited by the authors aim to provide reliable support to this study's main goals and fills up the methodological gaps. Accordingly, I recommend publication of this manuscript with limited (-minor) additions / changes detailed in the following.

Reply: *We sincerely appreciate the valuable feedback and constructive comments on our draft manuscript. We have addressed these comments in the R1 m/s as follows:*

Point 1. ***Difficult to follow for non-specialized readers****: We have done minor modifications of the text and graphical abstract to ensure that key concepts and findings are presented in a concise and accessible manner to improve the manuscript's readability for non- specialised – please see ln 83-87 and 106-109 in R1 m/s.*

Line no. 83-87

"While catchment managers are interested in the interplay between areas of high risk erosion and sediment delivery, this interplay can be complex, especially in large river catchments, and the sediment fingerprinting technique is a powerful tool to enhance the

understanding of sediment contributions from different types of sources in the watershed (such as land use classes, geological units, and tributary sub-catchments)."

Line no. 106-109

"In order to better understand catchment-wide erosion and sediment delivery mechanisms, this study combines the RUSLE-based INVEST-SDR model with geochemical sediment source fingerprinting in the Konar catchment, India."

Point 2: ***2nd order approach:*** *As the reviewer pointed out, the manuscript does indeed focus on the outputs derived from data treatments (2nd order approach) as our primary objective was framed so. No additional content was incorporated into the R1 manuscript in connection with this as no specific alterations were suggested by the reviewer. However, we tried our best to justify this approach by citing recent studies adopting a similar methodology – please see ln 44-51in the R1 m/s.*

Line no. 44-51

"Information on the contributions of different land use groups can be particularly informative for watershed management, and the sediment fingerprinting method based on statistically robust geochemical signatures can, in some instances, be used for obtaining such information (Demiguel et al., 2005; Laceby & Olley, 2015; Tiecher et al., 2018). To link the signatures of the sampled target sediment to the signatures of the sources, fingerprinting investigations typically combines the selected tracers (e.g., geochemical) with statistical techniques for source discrimination and numerical unmixing models for source apportionment."

Point 3: ***regarding use of composite aliquots****: We understand your concern about potential reductions in geochemical discrimination while doing composite sampling. However, it is important to note that the composite sampling procedure is established internationally as part of state-of-the-art source fingerprinting procedures (see, for example, Collins et al. 2017 – Journal of Environmental Management) Adoption of the approach is necessary to account for spatial heterogeneity within the sampling sites and to ensure that sample numbers are managed in the context of study resources. We have added a few lines in the R1 manuscript to highlight these aspects. Please see ln 146-155 in the R1 manuscript:*

Line no. 147-156

"The soil sampling plan was designed to cover the spatial heterogeneity of the land use classes in the study catchment and Google Earth and topographic data were used to locate the sampling points (Boardman, 2016). Adopting a composite sampling design is a practical solution to the issue of collecting enough source samples for statistical reliability when applying the sediment fingerprinting approach (Collins et al., 2017; Collins & Walling, 2002; Williamson et al., 2023). To execute the composite sampling approach, 105 sites distributed throughout the study catchment and representative of the different land use were used. Figure 2 shows photographs taken during the sampling campaign for both the land use source classes and for target sediment in the Konar reservoir (details of sampling protocol are shown in supplementary Table T1)."

Specific Comments

Comment 1: I suggest that the authors provide some information on the soil source composition, i.e., add a map for a selected relevant / discriminant geochemical parameter (as in Fig. 1).

Reply: Agreed and amended. *As per the suggestion of the reviewer, we have added Figure 6 to show the spatial variation of elemental proportion among the soil samples collected from the 105 sampling sites. We have mainly depicted the spatial patterns of Fe, K, Ti and Ca.*

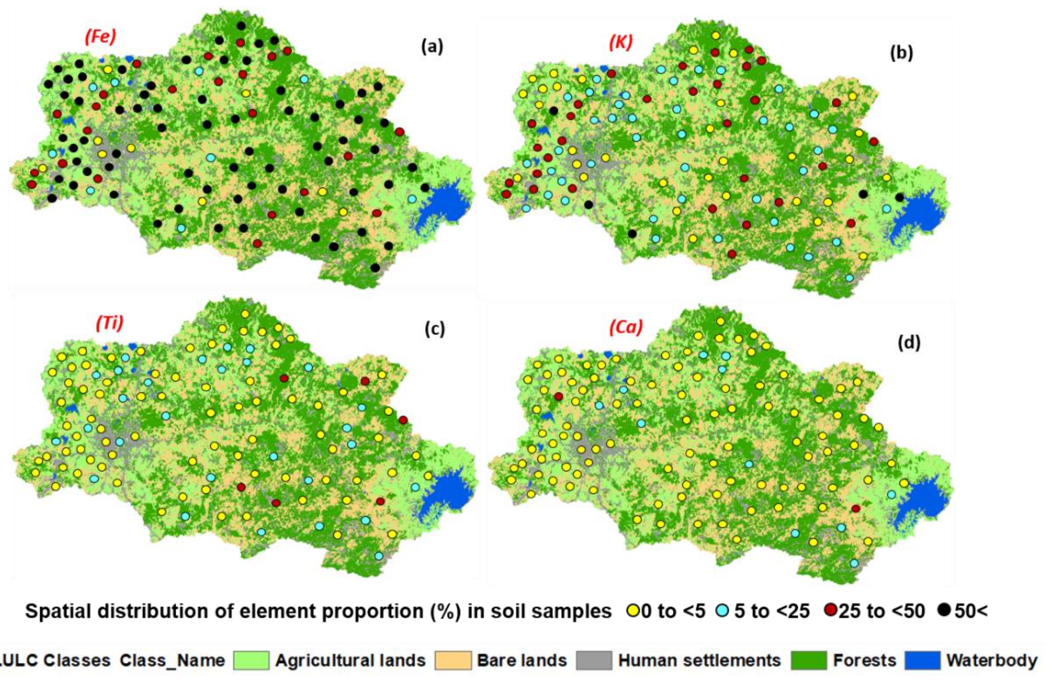


Figure 6 Spatial variation of elemental proportion (%) of (a) Fe, (b) K, (c) Ti and (d) Ca among the soil samples collected from the study catchment.

Comment 2: I also think that the authors should map the location of their 105 sample composites using one of their maps (i.e., Fig.1) so that the reader can visualize the distribution and representativeness of sampling.

Reply: Agreed and amended. *As per the suggestions, we have added the sampling locations in Figure 1c (i.e., on the DEM map of the study catchment). We have also added the gauging station and the inlet of the reservoir (location of target sediment sampling).*

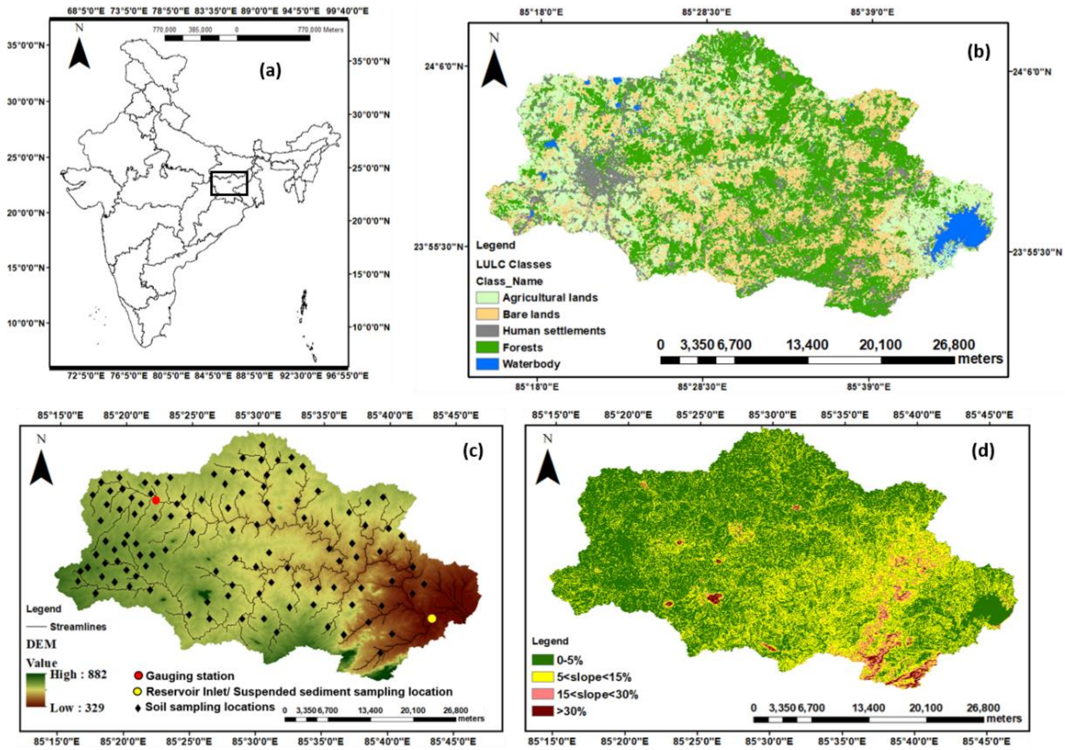


Figure 1. Information on the Konar study catchment characteristics: (a) location (b) land use (c) DEM (d) slope.

Comment 3: Are the results of this study (i.e., sediment export and export rates, section 3.4) supported by other nearby environments?

Reply: *Yes, the results of this study are in good agreement with the few other studies performed in this catchment. Specifically, one of our previous soil erosion and sediment yield studies (Das et al., 2022) conducted on this catchment, and the outcomes of the sediment fingerprinting results are in good agreement in identifying the crucial land use classes of the catchment. The mean annual sediment export identified in this study is computed to be 11tons/unit area as compared to 10 tons/ha/year for agricultural areas and ~25 tons/ha/year reported by Das et al., (2022) and Rajbanshi & Bhattacharya, (2020) respectively. We have* *discussed this aspect in the discussion section in the R1 m/s. Please refer to ln 345-357 in the R1 m/s:*

Line no. 345-357

"The highest human settlement contribution was estimated using the P0 model. However, the P0 model underestimated the contribution of barren lands drastically, and this land use has been reported to be a major sediment source by other studies (Das et al., 2022; Rajbanshi & Bhattacharya, 2020). The clay prior (P1) based model identified barren lands as the major sediment source (~20 to 70%) followed by agricultural lands (~10 to 70%) during both timeframes. Similar source estimates were generated using the slope based prior (P4) model. This suggests that the steepness of slope in the barren land areas is a major factor controlling sediment sources in the study catchment (Mishra et al., 2022). The silt based prior model (P2) predicted similar source contributions to the P0 model by identifying agricultural lands and human settlement areas as major sediment sources. The effects of silt concentration on geochemical properties were found to be negligible by Kraushaar et al., (2015) and this explains the lack of any significant difference between the source estimates using the silt based prior and no prior (P0) models."

The full reference details for the additional reference is now on ln 353 in the R1 m/s.

Rajbanshi, J., & Bhattacharya, S. (2022). Modelling the impact of climate change on soil erosion and sediment yield: A case study in a sub-tropical catchment, India. Modeling Earth Systems and Environment, 8(1), 689–711. https://doi.org/10.1007/s40808-021-01117-4

Minor comments

Most of my other "minor" requests refer to the "2. methodology section".

Comment 4: Soil and sediment preparation: I understand that samples were dry sieved at 63 µm after oven drying. Therefore, sieving involved aggregates of soil particles during drying. How the authors assume that there <63 µm size fractions were accurately separated? Sample preparation usually requires wet sieving and some preliminary "soft" grinding (to avoid overgrinding). Some precision is needed.

Reply: *We appreciate your insightful comment regarding the soil and sediment preparation in our study. However, we did take measures to minimize aggregation effects during the dry sieving process. To obtain the proportion of <63 µm particles in the soil samples we oven* *dried them for nearly 12 hrs and the soil samples were passed through a 63 µm sieve shaker for 24 hours. We also ensured that the sieving equipment used was of high quality with precise mesh sizes with prolonged oven drying and extended sieving duration to enhance the separation process. While wet sieving and preliminary grinding can be effective in certain contexts, we didn't adopt those in our study. We plan to consider trade-offs between different sampling and processing methods in future research.*

Moreover, we conducted particle size analysis on the soil samples to generate prior distributions for Bayesian modelling, and the results exhibited a substantial level of concordance with the proportion of fine soil particles that were extracted from the soil samples.

Comment 5: "the" instead of "he" in the figure legend

Reply: Agreed and amended.

Comment 6: Fig. 10: please improve horizontal and vertical scales by adding intermediate graduations

Reply: *Agreed and amended.*

Comment 7: Reference list:

Reply: *Agreed and amended.*

- *Burrough Jr… incomplete: Removed*
- *Palazon… duplicates: Modified accordingly*
- *Upadhhayay… duplicates: Modified accordingly*
- *Small et al… incomplete: Modified accordingly*
- *Stock et al… incomplete: Modified accordingly*

Specific Comments

General Comment: The manuscript Number: JEMA-D-23-07126 entitled "The spatio-temporal dynamics of suspended sediment sources based on a novel indexing approach combining Bayesian geochemical fingerprinting with physically based modelling" is well written. On the other hand, there are some essential comments authors should take into consideration.

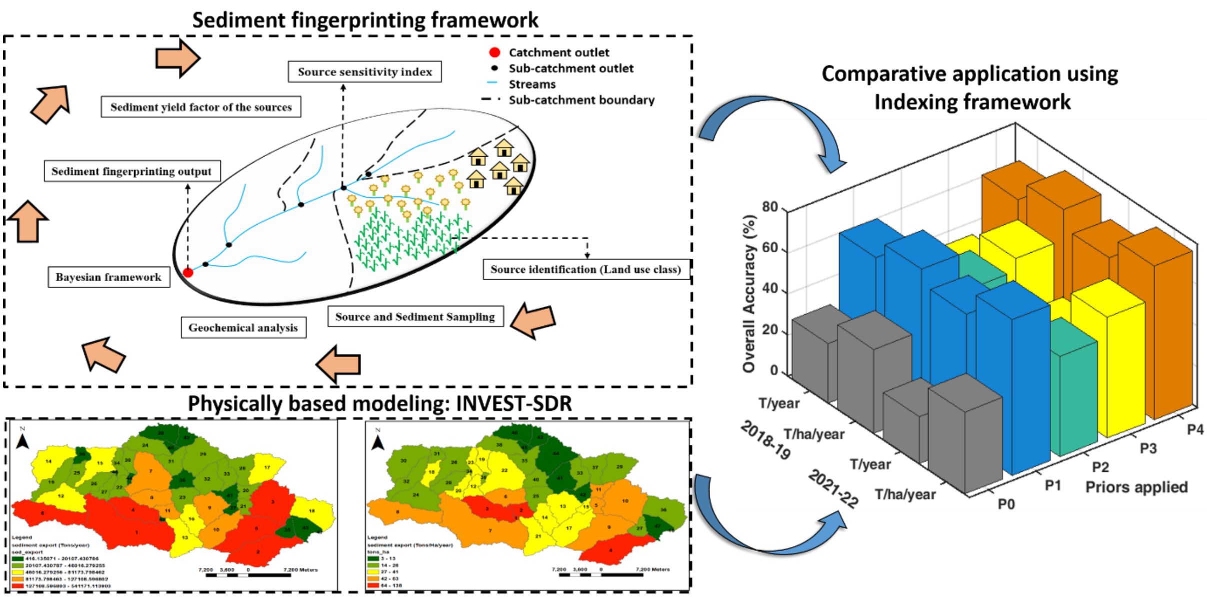
Reply: *We express our sincere gratitude to the reviewer for their favourable assessment of the novel methodology we employed for sediment fingerprinting. The integration of the indexing method and physically based modelling has provided valuable insights into the potential sediment production within our catchment. We have diligently addressed all the reviewer's comments in the subsequent responses, ensuring their inclusion in our R1 manuscript.*

Reviewer 2 Comments

Comment 1: Graphical abstract does not provide the visual interpretations of the manuscript.

Reply: Agreed and amended. *As per the recommendation, we have modified the graphical abstract to make it a more precise and proper representation of the work. Please see the following figure:*

Graphical abstract



Comment 2: More recent studies can be referred in the literature review.

Reply: Agreed and amended. *As per the comment, we have updated the R1 manuscript by citing the following recent references for sediment source fingerprinting work:*

Hirave, P., Nelson, D. B., Glendell, M., & Alewell, C. (2023). Land-use-based freshwater sediment source fingerprinting using hydrogen isotope compositions of long-chain fatty acids. Science of The Total Environment, 875, 162638. https://doi.org/10.1016/j.scitotenv.2023.162638 - - see ln 71 in the R1 m/s

Lake, N. F., Martínez-Carreras, N., Iffly, J. F., Shaw, P. J., & Collins, A. L. (2023). Use of a submersible spectrophotometer probe to fingerprint spatial suspended sediment sources at catchment scale. Science of The Total Environment, 873, 162332. https://doi.org/10.1016/j.scitotenv.2023.162332 - - see ln 43 in the R1 m/s

Liu, Y., Walling, D. E., Yang, M., & Zhang, F. (2023). Sediment source fingerprinting and the temporal variability of source contributions. Journal of Environmental Management, 338, 117835. https://doi.org/10.1016/j.jenvman.2023.117835 - - see ln 214 in the R1 m/s

Williamson, T. N., Fitzpatrick, F. A., & Kreiling, R. M. (2023). Building a library of source samples for sediment fingerprinting – Potential and proof of concept. Journal of Environmental Management, 333, 117254. https://doi.org/10.1016/j.jenvman.2023.117254 - - see ln 152 in the R1 m/s

Xu, Z., Belmont, P., Brahney, J., & Gellis, A. C. (2022). Sediment source fingerprinting as an aid to large-scale landscape conservation and restoration: A review for the Mississippi River Basin. Journal of Environmental Management, 324, 116260. https://doi.org/10.1016/j.jenvman.2022.116260 - - see ln 64 in the R1 m/s

Comment 3: Figure 1 needs a minor editing. The miles should be lower case Regarding this I would comment that the way of depicting units is incorrect.
According to the Bureau Internationale des Poids et des Mesures ' guidance, (https://www.bipm.org/documents/20126/41483022/SI-Brochure-9.pdf/fcf090b2-04e6-88cc-1149-c3e029ad8232) from the SI Brochure (PDF; see p. 147 for English) clause 5.2: They are printed in lower-case letters unless they are derived from a proper name, in which case the first letter is a capital letter. Few maps have the scale in miles while few are written in metres. Please check.

Reply: Agreed and amended. *As per the suggestions, we have modified all the maps by updating the scale formats. We have adapted 'Meters' to 'meters' in the scale.*

Comment 4: Proofreading at many places should be done. The paper needs to be thoroughly revised, and proper English writing skills should be applied.

Reply: Agreed and amended. *As per the suggestion, we have undertaken proofreading using a premium Grammarly account to correct any typographical and grammatical errors.*

Highlights

- Sensitivity of Bayesian sediment fingerprinting to particle size and slope explored
- Proposed a novel method to translate fingerprinting outputs to spatial information
- Method combines INVEST-SDR catchment modelling and Bayesian fingerprinting
- Combined approach revealed agricultural and barren regions as crucial sediment sources

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Abstract

Applications of sediment source fingerprinting continue to increase globally as the need for information to support improved management of the sediment problem persists. In our novel research, a Bayesian fingerprinting approach using MixSIAR was used with geochemical signatures, both without and with informative priors based on particle size and slope. The source estimates were compared with a newly proposed Source Sensitivity Index (SSI) and outputs from the INVEST-SDR model. MixSIAR results with informative priors indicated that agricultural and barren lands are the principal sediment sources (contributing ~5 to 85 % and ~5 to 80% respectively during two sampling periods i.e. 2018-2019 and 2021-2022) with forests being less important. The SSI spatial maps (using % clay and slope as informative priors) showed > 78% agreement with the spatial map derived using the INVEST-SDR model in terms of sub-catchment prioritization for spatial sediment source contributions. This study demonstrates the benefits of combining geochemical sediment source fingerprinting with SSI indices in larger catchments where the spatial prioritization of soil and water conservation is both challenging but warranted.

Keywords: Sediment fingerprinting, MixSIAR model, prior information, particle size distribution, INVEST model

1. Introduction

Water erosion is regarded as the most serious concern to global soil security, resulting in poorer agriculture yields and pollution of freshwater resources and estuaries (Das et al., 2022). A significant amount of research and policy attention is still directed towards reducing reservoir siltation and water pollution caused by water erosion; most notably, excess fine-grained (< 63 μm) sediment (Collins et al., 2020). Understanding water-induced soil erosion, sediment delivery and export, and sediment source patterns is crucial for targeted management of the impacts of human actions and natural processes on soil health and water resources. Though erosion models can be used for screening likely erodible areas in a catchment, critical information on sediment provenance can be obtained using sediment source fingerprinting (Lizaga et al., 2022). In particular, it is useful to identify the areas with disproportionately high erosion rates and connectivity with river channels, for developing optimal management strategies (Abban et al., 2016).

Several investigations have determined the relative contributions of surface and instream sources to sediment loads (Afshar et al., 2016; Boudreault et al., 2019; Carter et al., 2003; Collins & Walling, 2002, 2007; Lake et al., 2023). Geochemical fingerprinting of sediments is one of the most widely used approaches (Collins et al., 2020). Information on the contributions of different land use groups can be particularly informative for watershed management, and the sediment fingerprinting method based on statistically robust geochemical signatures can, in some instances, be used for obtaining such information (Demiguel et al., 2005; Laceby & Olley, 2015; Tiecher et al., 2018). To link the signatures of the sampled target sediment to the signatures of the sources, fingerprinting investigations typically combines the selected tracers (e.g., geochemical) with statistical techniques for source discrimination and numerical unmixing models for source apportionment. Various unmixing models have been proposed and used, including frequentist and Bayesian approaches (Collins, 2020; Collins et al., 2017; Davis & Fox, 2009; D'Haen et al., 2013). In order to determine the sources of target sediment, the Bayesian approach combines the likelihood of current sediment source data (geochemistry) with prior knowledge of sediment sources to form a posterior probability distribution of source contributions (Small et al., 2002). The assumption that tracers are adequately characterized for the potential source areas and the target sediment samples is eased when previous information is used for model parameterisation (Billheimer, 2001). Here, it is regarded by some investigators that Bayesian approaches are best for illustrating the uncertainty associated with estimated sediment source contributions.

Complex landscapes, however, make it difficult to pinpoint the origins of fine-grained sediment. As a result, new methods are required to provide additional insights into the interplay between catchment structure, surface cover, and land use practices for determining sediment source contributions (Tang et al., 2019; Xu et al., 2022). In support of this, Bayesian unmixing model frameworks can include prior information for relevant catchment characteristics (Stock et al., 2018; Upadhayay et al., 2017). In the existing literature, numerous possibilities are discussed concerning the applicability of prior information in Bayesian frameworks for understanding sediment dynamics at catchment scale. A study by Upadhayay et al., (2020), for example, applied a sediment connectivity index as prior information to identify the crucial land use classes of a study catchment. Similar attempts have also been made by other studies by using other catchment information such as land cover area (Hirave et al., 2023; Lizaga, 2021; Upadhayay et al., 2017, 2022). Beyond connectivity or land cover associated risks for erosion and sediment delivery, the effect of particle size selectivity on sediment source signals is widely recognised in many previous studies (Haddadchi et al., 2015; Gaspar et al., 2022). When it comes to rain-induced erosion, raindrops and slope controls the detachment and delivery of soil particles (Lu et al., 2016). In particular, the particle size distributions of sediment are useful background knowledge for elucidating soil erosion processes (Legout et al., 2005; Cheraghi et al., 2016; Kiani-Harchegani et al., 2019), making such understanding an effective form of prior information.

Despite the aforementioned importance of specific catchment characteristics, source apportionment studies have not, to date, explored the sensitivity of the results to the combination of mean slope and particle size.

While catchment managers are interested in the interplay between areas of high risk erosion and sediment delivery, this interplay can be complex, especially in large river catchments, and the sediment fingerprinting technique is a powerful tool to enhance the understanding of sediment contributions from different types of sources in the watershed (such as land use classes, geological units, and tributary sub-catchments). Here, combining sediment fingerprinting methods with physical erosion modelling and other indices has been shown to improve the efficacy of management decisions [e.g. (Palazón et al., 2014, 2016; Wilkinson et al., 2013)]. By combining the application of physically-based modelling and sediment fingerprinting methods at the catchment scale, it is possible to create novel indicators of the spatio-temporal variability of sediment sources [i.e. source sensitivity index (SSI)]. Previous studies have, for instance, shown the value of combining weathering indices with conventional geochemical tracers to gain further insight into sub-basin spatial suspended sediment sources (Nosrati et al., 2019). Integrating indices with sediment fingerprinting results can help to: (i) elucidate sub-catchment scale erosion processes spatially, (ii) improve the accuracy of sediment source fingerprinting, and; (iii) support comparisons between sediment fingerprinting results and physical modelling outputs as a weight-of-evidence approach to understanding catchment sediment dynamics. Developing SSI can address sampling uncertainties and the spatial limitations frequently associated with sediment fingerprinting results (Collins, 2020; Collins et al., 2017; Koiter et al., 2013). Previous research using the SWAT model has shown that the integration of physical modelling and tracer-based methods on large river systems greatly improves our understanding of erosion processes (Palazón et al., 2014, 2016; Wilk, 2022). Similar to the SWAT model, the INVEST-SDR model has been extensively applied to deal with a wide range of scales and issues related to sediment delivery modelling across various hydro climatic regions (Hamel et al., 2017; Vigerstol & Aukema, 2011). In order to better understand catchment-wide erosion and sediment delivery mechanisms, this study combines the RUSLE-based INVEST-SDR model with geochemical sediment source fingerprinting in the Konar catchment, India.

The specific objectives were:

a. To apply geochemical fingerprinting to apportion suspended sediment sources in the form of land use classes.
b. To apply a Bayesian mixing model with particle size distribution and mean slope as prior information and to examine the sensitivity of source apportionment estimates to such data.
c. To develop an innovative index (SSI) using the geochemical fingerprinting results to generate spatio-temporal soil erosion maps.
d. To assess and quantify the spatial distribution of sediment sources in the study catchment using the INVEST-SDR model and compare the outputs with the sediment fingerprinting and SSI results to evaluate the accuracy of the Bayesian sediment fingerprinting method.

2 Methodology

2.1 Study area characteristics

This study was carried out in eastern India's Konar catchment (990 km^2) of the Damodar River basin. The Konar catchment has diverse geo-physical terrains including high plains, moderate hills, and valleys. Elevations range between 329-882 m, with an eastern slope (Figure

1c). The climate is subtropical, with annual average rainfall of 1100-1300 mm distributed mostly (70-80%) between June and September. Summer temperatures can reach 46˚C compared with lows of 4˚C in the winter. The Hazaribagh district comprises more than 70 percent of the catchment area, and most of this territory is made up of forests and rocky soils. Overall, our land use categories are found in the study catchment: i.e., agricultural lands (38%), forests (36%), barren lands (14%), and human settlements (12%) (Figure 1b). Rice, groundnuts, and maize are the primary crops cultivated in the agricultural areas during the monsoon season, while wheat, mustard, and other vegetables are grown using terrace farming on uneven terrain during the off-monsoon season. Mixed deciduous and tropical dry forests predominate in the forest zones with several species of medicinal plants and timber trees including sal (*Shorea robust*) (Forest, Environment and Climate change Department, Government of Jharkhand; https://forest.jharkhand.gov.in). The three most common soil types are lithosols (46%), ferric luvisols (38%), and eutric nitosols (16%) (Supplementary Figure F1).

2.2 Soil and sediment sampling

One of the main challenges of the sediment fingerprinting approach is collecting sufficient source samples for statistical reliability (Collins & Walling, 2002). To study the temporal variation in the suspended sediment contributions from different sources, water samples were collected for six time periods from the inlet of Konar reservoir (shown in Figure 1c) for two alternate water years; i.e., July 2018-June 2019 and July 2021-June 2022. Three 2L swabs of suspended sediment were collected at a water depth of 0-10 cm and stored in high-density polyethylene bottles during these sampling periods (Wang et al., 2019). The soil sampling plan was designed to cover the spatial heterogeneity of the land use classes in the study catchment and Google Earth and topographic data were used to locate the sampling points (Boardman, 2016). Adopting a composite sampling design is a practical solution to the issue of collecting enough source samples for statistical reliability when applying the sediment fingerprinting approach (Collins et al., 2017; Collins & Walling, 2002; Williamson et al., 2023). To execute the composite sampling approach, 105 sites distributed throughout the study catchment and representative of the different land use were used. Figure 2 shows photographs taken during the sampling campaign for both the land use source classes and for target sediment in the Konar reservoir (details of sampling protocol are shown in supplementary Table T1). Composite sampling involved merging three to four sub-samples collected within a radius of 100 to 500 m, depending on accessibility (Collins et al., 2017). The upper 5 cm of soil was sampled at each source sampling location using a non-metallic trowel deployed in one extensive campaign. This one-off source sampling strategy assumed that lithological features remained constant through time (Tiecher et al., 2017).

2.3 Sample preparation and laboratory analysis

To extract suspended sediments from the bulk water samples, the samples were first centrifuged, then filtered, and finally oven dried at 70 ˚C for 12 hours. After 12 hours of oven drying, soil samples were passed through a 63 µm sieve shaker for 24 hours to avoid aggregation and extract the silt and clay fractions to improve the direct comparability of source and sediment samples (Collins & Walling, 2016). Prior to the sample processing, scanning of the soil and sediment samples was performed using a DP-6000 Delta Premium portable X-ray fluorescence (PXRF) machine equipped with an Rh X-ray tube operating at 15-40 keV. Using the instrument's Geochem Mode, the concentrations of V, Cr, Fe, Co, Ni, Cu, Zn, W, Hg, As, Se, Pb, Bi, Rb, U, Sr, Y, Zr, Th, Mo, Ag, Cd, Sn, Sb, Ti, Mn, Mg, Al, Si, P, S, Cl, K, and Ca were estimated (Sharma et al., 2014). The particle size characteristics of the source and target sediment samples were measured using a Malvern Pananalytical Mastersizer 3000.

2.4 Formulation of the Bayesian framework and priors for source apportionment modelling

Geochemical tracers were selected using a standard two-step process consisting of a Kruskal-Wallis H-test for inter-group differences, and a stepwise discriminant function analysis (DFA) for selecting a minimal set of tracers that maximises source discrimination (Collins et al., 1997). MixSIAR, a state-of-the-art Bayesian Isotope Mixing Model (BIMM) available as a free R package, was used to estimate sediment source apportionment (Guerrero & Rogers, 2020; Stock et al., 2018). The geochemical tracers were entered into a concentration-dependent MixSIAR model with and without the use of the informative priors (Upadhayay et al., 2020). The following settings were applied for the Markov Chain Monte Carlo (MCMC) iterations: number of chains = 3, chain length = 3,000,000, burn = 1,500,000, thin =500. Gelman-Rubin and Geweke diagnostic statistics were used to verify model run convergence (Stock et al., 2018). The deviance information criteria (DIC) was used to choose the best fitting model. Means and 95% Bayesian confidence intervals (CI) were provided from posterior distributions derived for sediment source contributions to estimate these contributions and their associated uncertainty (Upadhayay et al., 2020). Upadhayay et al., (2017) provide a comprehensive description of MixSIAR, and Stock et al., (2018) provides a comprehensive mathematical explanation of MixSIAR.

2.4.1 Prior selection

Informative priors for sediment source proportions should be selected logically with proper physical meaning (Upadhayay et al., 2020). Research suggests that rainfall-induced soil erosion occurs in primarily three phases, each of which is particle size-based (Figure 3) (Sadeghi et al., 2017; Wang & Shi, 2015). The initial stage comprises the detachment of soil particles by raindrop splash - eroding mostly very fine particles and some fine particles (Figure3a). The second stage i.e. after a prolonged rainfall event, is characterized by an increased proportion of coarser particles and fine particles (Figure 3b). Finally, in the third stage, sediment transportation of very fine particles and lower quantities of fine and coarse particles takes place through the stream network (Figure 3c). The slope of the terrain plays a very important role in the erosion process (Reza Vaezi et al., 2020). In this study we explored the sensitivity of the source proportion estimates to the choice of particle size priors (clay <2 µm, silt 2-50 µm, and fine sand >50 µm) by adjusting the prior specification and observing the impacts on the posterior distributions generated by MixSIAR. Particle size priors were derived by determining the mean proportions of clay, silt, and very fine sand in the individual land use classes. Mean slopes of the land use groups were used to incorporate the slope priors.

2.5 Formulation of the source sensitivity index (SSI)

The sediment fingerprinting outputs provided by MixSIAR are relative and thereby do not provide information on the sediment yields from individual sources. To address this shortcoming, we developed an SSI using: (i) sediment yield factor for each land use class; (ii) proportion of area covered by each land use class in the overall catchment and the sub-catchments therein, and; (iii) information on the temporal variability of the source contribution obtained from the sediment fingerprinting analysis (Liu et al., 2023). An SSI value may be derived to show the severity of the sub-catchments by linking the area covered by the sub-catchment, the land use distribution within the sub-catchment, and the temporal variations in the total catchment's sediment production. Accordingly, the SSI was estimated as:

$$\boldsymbol{S_{ef}} = \frac{\boldsymbol{X_e}}{\boldsymbol{A_e}} \qquad (\boldsymbol{e = 1{:}m, f = 1{:}n}) \tag{1}$$

$$\boldsymbol{SSI_x} = \left(\sum_{f=1}^{n}\sum_{e=1}^{m}\frac{\boldsymbol{S_{ef}.A_{ex}}}{\boldsymbol{A_x}}\right).\boldsymbol{D_x.P_f} \tag{2}$$

$\boldsymbol{S_{ef}}$ = sediment yield factor for land use $\boldsymbol{e}$ in the month $\boldsymbol{f}$.

$\boldsymbol{X_e}$ = Source contribution (land use) to the catchment sediment yield obtained from the fingerprinting approach.

$\boldsymbol{A_e}$ = Area covered by respective land use class ($\boldsymbol{e}$) in the catchment.

$\boldsymbol{SSI_x}$ = Source sensitivity of sub-catchment $\boldsymbol{x}$.

$\boldsymbol{A_{ex}}$ = Area covered by land use class e in sub-catchment $\boldsymbol{x}$.

$\boldsymbol{A_x}$ = Area covered by sub-catchment $\boldsymbol{x}$.

$\boldsymbol{D_x}$ = Distance factor of the sub-catchment outlet (calculated as the ratio of longest flow path and distance of the sub-catchment outlet from the catchment sink where the target sediment samples were collected) (Supplementary Figure F2).

$\boldsymbol{P_f}$ = Proportion of annual sediment yield in the respective month.

Figure 4 depicts the analytical framework used to generate spatial maps based on SSI values in our study area. This figure also shows the whole methodology of this study employing physical modelling outputs to authenticate sediment fingerprinting outcomes through SSI generated spatial maps.

2.6 RUSLE-based hydrological model development: INVEST-SDR

Quantification and mapping of soil loss and sediment delivery in a landscape can be accomplished using the Integrated Valuation of Ecosystem Services and Tradeoffs (INVEST)- Sediment Delivery Ratio (SDR) model (Aneseyee et al., 2020). Compared to other models, such as SWAT, INVEST-SDR uses less input data (Table 1) and is more flexible; it can also be modified to a given scenario and used with locally and globally accessible data and exemplifies the hydrological connectivity concept proposed by Vigiak et al., (2012). To validate the INVEST SDR model, annual simulated sediment inflow data was compared to real data collected from Damodar Valley Corporation (DVC) using root mean square error (RMSE) and the coefficient of determination (R^2).

3 Results

3.1 Catchment sediment sources using geochemical source fingerprinting

Correct and reliable use of sediment fingerprinting requires statistical analysis and interpretation of isotope tracers from sediment source end members. One of the most important step is to identify a representative set of final geochemical tracers to use in a "composite signature" to determine the origin of fine sediments. From the laboratory analysis, 24 elements were detected in the soil and sediment samples collected from the Konar catchment. To identify the set of discriminatory geochemical tracers to be used as an input to the MixSIAR, all the elements were passed through a KW test followed by DFA (Figure 5). Based on this analysis, 21 elements were selected (Fe, K, Ti, Ca, Zn, Mn, Ba, Zr, Rb, V, Cr, Sn, Ni, Sr, Pb, Cu, Ga, Sb, Ag, A, and Br). A summary of the statistical analysis for source discrimination is provided in supplementary Table T2. Furthermore, we have also shown the spatial variability of the proportional (%) presence of few elements (Fe, K, Ti and Ca) in the soil samples collected from 105 locations of the catchment (Figure 6).

3.2 Bayesian modelling results for source apportionment

MixSIAR was run with both 'no priors (P0)' and priors based on particle size and slope information for the study catchment (details of P1 to P4 are given in Table 2). During both time periods, both agricultural and barren lands were the most important sediment sources (Figure 7). The Bayesian model with no priors (P0) and with the silt based prior (P2) identified agricultural lands as the major sediment source in most of the seasons (varying from ~30-45% during the water year 2018-19 and ~30 to 60 % during the water year 2021-22) (Figure 7 a1, a2). In contrast to the other prior-based models, these two models (P0 and P2) predicted a smaller contribution of suspended sediment from agricultural areas and a larger contribution from human settlements (Figure 7 b_1, b_2). The clay based prior (P1) and the slope based prior (P4) model outputs, both identified barren lands as the primary source of suspended sediment (varying from ~25-75% during the water year 2018-19 and ~20 to 60 % during water year 2021-22). In contrast, the outputs of the slope based prior model suggested negligible contributions from human settlement areas compared with the other land use classes (varying between 0-5% for both water years 2018-19 and 2021-22). The very fine sand prior (P3) based model predicted a much higher contribution from forests (varying from ~20-50% during the water year 2018-19 and ~25-50% during 2021-22). Overall, all the prior based models identified agricultural land as the primary source of sediment in July 2021 (Figure 7 a2).

3.3 SSI based Bayesian modelling results for source apportionment

This study investigated the potential utility of a new index [Source Sensitivity Index (SSI)] to help the policymakers better understand the relative impacts of sediment sources in the Konar catchment. It integrates sediment fingerprinting information with the physical controls of sediment deposition in the sub-catchments to generate an index value for ranking the sub-catchments to explain conservation urgency. To establish a link with physical modelling results, the sub-catchments were classified into five SSI classes [class1 (26-174), class2 (175-378), class3 (379-822), class4 (823-1267), class5 (1268-5266)] (Figure 8). Figure 9 displays the distribution of the sub-catchments across the SSI classes for the particle size and slope prior based model outputs, suggesting little disparity among the sub-catchments using the P0 and P2 priors based models. However, upon closer inspection, it can be seen that the number of sub-catchments with higher SSI classes is marginally greater during 2021–2022, compared with 2018–2019. This SSI based information and associated maps can be used for assessing the reliability of mixing model source apportionment results based on the comparison with physically-based model outputs. More sub-catchments under class 5 (C5) were discovered by P1 based model in both time frames, similar to the P4 based model. However, it is evident from the Bayesian model findings described in section 3.2 that a comparable number of sub-catchments come under the prior-based models P0 and P2. Importantly, the SSI based analysis demonstrated how effectively Bayesian sediment fingerprinting results can be converted into translating tool to spatially explain the sediment dynamics in the Konar catchment. This SSI based information and maps can be used as a criterion for assessing the reliability of mixing model source apportionment results in comparison with physically based sedimentation models spatial outputs.

3.4 INVEST-SDR model based results

The assessment of sediment export using INVEST-SDR model involved computing the annual soil loss, sediment connectivity, and sediment delivery ratio of the study catchment. According to the coefficient of determination (R^2) and root mean square error (RMSE) values of 0.81 and 6.85 t/ha/year for the Nagwan gauging station, the predicted sediment export from the catchment was in line with the available observed data over time (Supplementary Figure F3). Using the sediment connectivity index (SCI) proposed by Borselli et al., (2008) the mean sediment connectivity is estimated to be -6.326, ranging between -11.038 and 0.179 (Figure 10a). INVEST-SDR's estimate for soil loss ranged from 0 to 23321 t/year, with a mean soil loss rate of 23.65 tonnes per year (Figure 10b). The sediment delivery ratio was computed based on the connectivity index and ranged from 0 to 0.322, with a corresponding mean value of 0.082 (Figure 10c). The spatial variation in the Konar catchment's sediment export, ranging from 0 to 3490 t/year with a mean value of 11.16 t/year, is shown in Figure 10d. The Konar catchment was sub-divided into 47 sub-catchments to pinpoint the crucial hot spots of soil erosion for targeting preventative measures. All sub-catchments were ranked and classified based on the sediment export in t/year (Figure 10e) and t/ha/year (Figure 10f). Here, we used two different sub-catchment ranking methods. Firstly, the sub-catchments were ranked according to their annual sediment yield, and secondly, the sub-catchments were ranked according to annual sediment yield per unit area (i.e., specific sediment yield). The overall variance between the two ranking techniques is shown in Figure 11a. Sub-catchments ranked 18, 21, 29, 37, 40, and 47 based on annual sediment yield showed substantial disagreement with the ranks assigned using annual specific sediment yield (Figure 11b).

3.5 Examining the authenticity of the SSI with physically-based modelling

Sediment fingerprinting and the INVEST-SDR model were used to generate independent predictions. Here, the sediment fingerprinting based SSI technique generated estimates of relative source contributions for surface sediments collected in the reservoir, while the INVEST-SDR model generated sediment yield estimates for the four land use groups. Using results from INVEST-SDR as an independent evaluation, we computed the overall accuracy of the SSI ranking method by employing a confusion matrix for the sediment yield (Figure 12). Agreement between the annual specific sediment yield and the SSI approach was better than that with the annual sediment yield of the sub-catchments with all the prior based models. The P0 and P2 based models (i.e., no prior and silt-based priors) exhibited the lowest level of accuracy over both time frames, whereas models based on P1 and P4 (i.e., clay and slope priors, respectively) displayed the highest levels of accuracy (80% and 68% with t/ha/year and t/year, respectively) (Figure 12). The P3 (very fine sand-based prior) model predictions of the sub-catchment sediment output was ~65% accurate. Over the two time frames, the clay and slope-based priors performed the best.

4. Discussion

4.1 Multiple prior based geochemical sediment source fingerprinting

The DFA findings on the geochemical tracers showed some overlap between the source groups. Several other investigations have reported similar issues using geochemical data.

The PSD of target sediment can be affected by several factors, including the PSD of the sources, erosion patterns and intensity, and catchment slopes. It is therefore important to consider particle size carefully when using the sediment fingerprinting approach (Laceby et al., 2017; Koiter et al., 2018). Accordingly, particle size based priors were used with slope when applying the MixSIAR model. The Bayesian model with no priors (P0) identified agricultural lands as the most important sediment source (i.e. contributing ~40-55%), with human settlement contributing 8-15%, whereas the barren lands contributed 20 to 25%, and forest ~18 to 25%. The highest human settlement contribution was estimated using the P0 model. However, the P0 model underestimated the contribution of barren lands drastically, and this land use has been reported to be a major sediment source by other studies conducted in this region (Das et al., 2022; Rajbanshi & Bhattacharya, 2020, 2022). The clay prior (P1) based model identified barren lands as the major sediment source (~20 to 70%) followed by agricultural lands (~10 to 70%) during both timeframes. Similar source estimates were generated using the slope based prior (P4) model. This suggests that the steepness of slope in the barren land areas is a major factor controlling sediment sources in the study catchment (Mishra et al., 2022). The silt based prior model (P2) predicted similar source contributions to the P0 model by identifying agricultural lands and human settlement areas as major sediment sources. The effects of silt concentration on geochemical properties were found to be negligible by Kraushaar et al., (2015) and this explains the lack of any significant difference between the source estimates using the silt based prior and no prior (P0) models. The proportion of very fine sand is highest in the bare lands; however, the prior based model for this particle size fraction (P3), identified forests as the major sediment source (~20 to 50 %). A substantial contribution of forests to sediment yield has been reported by a few previous studies (Upadhayay et al., 2020).

4.2 Validation of sediment source fingerprinting with INVEST-SDR

The INVEST-SDR outputs were used as an independent evaluation of the sediment fingerprinting estimates. Previous studies have evaluated source fingerprinting using physically-based modelling. A study by Palazón et al., (2016), for example, reported good consistency between sediment fingerprinting results and SWAT modelling. The accuracy of the five prior based Bayesian model in prioritizing the sub-catchments indicates that the annual sediment yields for the sub-catchments agree less with the SSI results compared to annual specific sediment yield. Similar, apportionment patterns were established in a study conducted by Hamel et al., (2015) which prioritized the sub-catchments based on soil erosion alone, rather than erosion and sediment delivery. The clay and slope priors based models performed better than the other models with an overall accuracy of >78% (Figure 12). Previous studies on soil erosion have illustrated how catchment slope affects erosion patterns and the PSD of mobilised sediment particles (Lu et al., 2016; Vigiak et al., 2012; Wang & Shi, 2015). Further, some studies have reported how variable rainfall intensity and slope conditions have resulted in greater detachment of clay particles at the experimental scale (Kiani-Harchegani et al., 2018, 2019; Sadeghi et al., 2017; Zhang et al., 2018).

4.3 Limitations and future outlook

There are a few limitations that must be borne in mind while interpreting the results of this study. Our ability to sample the reservoir for target sediment beyond two water years was limited by time, money, and accessibility issues, and continuous monitoring was restricted in 2020-2021 due to COVID19 lockdowns. Even though, the results of the prior based sediment fingerprinting study performed well when compared to physical modelling, additional source sampling campaigns (annual or seasonal) may have shown greater temporal variation in catchment sediment source contributions. Since a single target sediment sampling location was deployed, the source fingerprinting estimates must be viewed as scale-dependent and longitudinal sampling along the stream network could be used to provide further insight into the sediment dynamics of the study catchment (Koiter et al., 2013). The target sediment samples were not age dated (Fatahi et al., 2022) and may therefore reflect sediment sources over recent times rather than the present day alone. Despite the aforementioned limitations, however, this novel research suggested that the prior based sediment fingerprinting procedure provides valuable information for understanding the spatial and temporal dynamics of fine sediment sources and delivery in the study catchment. Our study illustrates the benefits of combining sediment source fingerprinting with independent approaches such as physically-based modelling.

5 Conclusion

. The major findings of this study are as follows:

a) The geochemical fingerprints of the sources successfully discriminated between the surface soil samples collected from the land use classes of the Konar study catchment. The sensitivity of the Bayesian model predictions to priors based on particle size and slope was evaluated. The model comparisons suggested that agricultural and barren lands are the most important sediment sources.

b) The performance of INVEST-SDR was satisfactory using the observed datasets from the Nagwan gauging station (R^2=0.81 and RMSE = 6.85 tons/ha/Year). The two sub-catchment ranking scenarios using the outputs from INVEST-SDR showed disagreements in terms of the higher ranking sub-catchments. However, there were some similarities between the prioritization based on both ranking schemes.

c) Comparison between the two INVEST-SDR ranking schemes and the results of the SSI technique based on five prior based Bayesian models for prioritising the sub-catchments was informative. The overall accuracy of the SSI method considering all the models with the first ranking scheme (i.e., annual sediment yield) varied from 25-62%, whereas, based on the second scheme (i.e., annual specific sediment yield) it ranged between 68-82%. The performance of slope and clay prior based models performed best with > 78% accuracy.

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The spatio-temporal dynamics of suspended sediment sources based on a novel indexing approach combining Bayesian geochemical fingerprinting with physically-based modelling

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Abstract

Applications of sediment source fingerprinting continue to increase globally as the need for information to support improved management of the sediment problem persists. In our novel research, a Bayesian fingerprinting approach using MixSIAR was used with geochemical signatures, both without and with informative priors based on particle size and slope. The source estimates were compared with a newly proposed Source Sensitivity Index (SSI) and outputs from the INVEST-SDR model. MixSIAR results with informative priors indicated that agricultural and barren lands are the principal sediment sources (contributing ~5 to 85 % and ~5 to 80% respectively during two sampling periods i.e. 2018-2019 and 2021-2022) with forests being less important. The SSI spatial maps (using % clay and slope as informative priors) showed > 78% agreement with the spatial map derived using the INVEST-SDR model in terms of sub-catchment prioritization for spatial sediment source contributions. This study demonstrates the benefits of combining geochemical sediment source fingerprinting with SSI indices in larger catchments where the spatial prioritization of soil and water conservation is both challenging but warranted.

Keywords: Sediment fingerprinting, MixSIAR model, prior information, particle size distribution, INVEST model

1. Introduction

Water erosion is regarded as the most serious concern to global soil security, resulting in poorer agriculture yields and pollution of freshwater resources and estuaries (Das et al., 2022). A significant amount of research and policy attention is still directed towards reducing reservoir siltation and water pollution caused by water erosion; most notably, excess fine-grained (< 63 µm) sediment (Collins et al., 2020). Understanding water-induced soil erosion, sediment delivery and export, and sediment source patterns is crucial for targeted management of the impacts of human actions and natural processes on soil health and water resources. Though erosion models can be used for screening likely erodible areas in a catchment, critical information on sediment provenance can be obtained using sediment source fingerprinting (Lizaga et al., 2022). In particular, it is useful to identify the areas with disproportionately high erosion rates and connectivity with river channels, for developing optimal management strategies (Abban et al., 2016).

Several investigations have determined the relative contributions of surface and instream sources to sediment loads (Afshar et al., 2016; Boudreault et al., 2019; Carter et al., 2003; Collins & Walling, 2002, 2007; Lake et al., 2023). Geochemical fingerprinting of sediments is one of the most widely used approaches (Collins et al., 2020)~~[Collins et al., (2020)]~~. Information on the contributions of different land use groups can be particularly informative for watershed management, and the sediment~~geochemical~~ fingerprinting method based on statistically robust geochemical signatures can, in some instances, be used for obtaining such information (Demiguel et al., 2005; Laceby & Olley, 2015; Tiecher et al., 2018). To link the signatures of the sampled target sediment to the signatures of the sources, fingerprinting investigations typically combines the selected tracers (e.g., geochemical) with statistical techniques for source discrimination and numerical unmixing models for source apportionment. Various unmixing models have been proposed and used, including frequentist and Bayesian approaches (Collins, 2020; Collins et al., 2017; Davis & Fox, 2009; D'Haen et al., 2013). In order to determine the sources of target sediment, the Bayesian approach combines the likelihood of current sediment source data (geochemistry) with prior knowledge of sediment sources ~~in the study catchment~~ to form a posterior probability distribution of source contributions (Small et al., 2002). The assumption that tracers are adequately characterized for the potential source areas and the target sediment samples is eased when previous information is used for model parameterisation (Billheimer, 2001). Here, it is regarded by some investigators that Bayesian approaches are best for illustrating the uncertainty associated with estimated sediment source contributions.

Complex landscapes, however, make it difficult to pinpoint the origins of fine-grained sediment. As a result, new methods are required to provide additional insights into the interplay between catchment structure, surface cover, and land use practices for determining sediment source contributions (Tang et al., 2019; Xu et al., 2022). In support of this, Bayesian unmixing model frameworks can include prior information ~~including that~~ for relevant catchment characteristics (Stock et al., 2018; Upadhayay et al., 2017). In the existing literature, numerous possibilities are discussed concerning the applicability of prior information in Bayesian frameworks for understanding sediment dynamics at catchment scale. A study by Upadhayay et al., (2020), for example, applied a sediment connectivity index as prior information to identify the crucial land use classes of a study catchment. Similar attempts have also been made by other studies by using other catchment information such as land cover area (Hirave et al., 2023; Lizaga, 2021; Upadhayay et al., 2017, 2022). Beyond connectivity or land cover associated risks for erosion and sediment delivery, the effect of particle size selectivity on sediment source signals is widely recognised in many previous studies (Haddadchi et al., 2015; Gaspar et al., 2022). When it comes to rain-induced erosion, raindrops~~,~~ and slope controls the detachment and delivery of soil particles (Lu et al., 2016). In particular, the particle size distributions of sediment are useful background knowledge for elucidating soil erosion processes (Legout et al., 2005; Cheraghi et al., 2016; Kiani-Harchegani et al., 2019), making such understanding an effective form of prior information.

Despite the aforementioned importance of specific catchment characteristics, source apportionment studies have not, to date, explored the sensitivity of the results to the combination of mean slope and particle size.

While catchment managers are interested in the interplay between areas of high risk erosion and sediment delivery, this interplay can be complex, especially in large river catchments, and the sediment fingerprinting technique is a powerful tool to enhance the understanding of sediment contributions from different types of sources in the watershed (such as land use classes, geological units, and tributary sub-catchments). ~~Though the sediment fingerprinting technique is a powerful tool to enhance the understanding of sediment contributions from various types of sources in the watershed (e.g., land use classes, geological units, and tributary sub-catchments), catchment managers are also interested in the interplay between areas of high risk erosion and sediment delivery and such interplay can be complex, especially in large river catchments.~~ Here, combining sediment fingerprinting methods with physical erosion modelling and other indices has been shown to improve the efficacy of management decisions [~~(~~e.g. (Palazón et al., 2014, 2016; Wilkinson et al., 2013)]. By combining the application of physically-based modelling and sediment fingerprinting methods at the catchment scale, it is possible to create novel indicators of the spatio-temporal variability of sediment sources [i.e. source sensitivity index (SSI)]. Previous studies have, for instance, shown the value of combining weathering indices with conventional geochemical tracers to gain further insight into sub-basin spatial suspended sediment sources (Nosrati et al., 2019)~~]~~. Integrating indices with sediment fingerprinting results can help to: (i) elucidate sub-catchment scale erosion

processes spatially, (ii) improve the accuracy of sediment source fingerprinting, and; (iii) support comparisons between sediment fingerprinting results and physical modelling outputs as a weight-of-evidence approach to understanding catchment sediment dynamics. Developing SSI can address sampling uncertainties and the spatial limitations frequently associated with sediment fingerprinting results (Collins, 2020; Collins et al., 2017; Koiter et al., 2013). Previous research using the SWAT model has shown that the integration of physical modelling and tracer-based methods on large river systems greatly improves our understanding of erosion processes (Palazón et al., 2014, 2016; Wilk, 2022). Similar to the SWAT model, the INVEST-SDR model has been extensively applied to deal with a wide range of scales and issues related to sediment delivery modelling across various hydro climatic regions (Hamel et al., 2017; Vigerstol & Aukema, 2011). In order to better understand catchment-wide erosion and sediment delivery mechanisms, this study combines the RUSLE-based INVEST-SDR model with geochemical sediment source fingerprinting in the Konar catchment, India.
~~Accordingly, this study combined application of the RUSLE-based INVEST-SDR model and geochemical sediment source fingerprinting in the Konar catchment, India, to explore the scope for improving understanding of catchment-wide erosion and sediment delivery processes.~~ The specific objectives were:

a. To apply geochemical fingerprinting to apportion suspended sediment sources in the form of land use classes.
b. To apply a Bayesian mixing model with particle size distribution and mean slope as prior information and to examine the sensitivity of source apportionment estimates to such data.
c. To develop an innovative index (SSI) using the geochemical fingerprinting results to generate spatio-temporal soil erosion maps.

d. To assess and quantify the spatial distribution of sediment sources in the study catchment using the INVEST-SDR model and compare the outputs with the sediment fingerprinting and SSI results to evaluate the accuracy of the Bayesian sediment fingerprinting method.

2 Methodology

2.1 Study area characteristics

This study was carried out in eastern India's Konar catchment (990 km^2) of the Damodar River basin. The Konar catchment has diverse geo-physical terrains including high plains, moderate hills, and valleys. Elevations range between 329-882 m, with an eastern slope (Figure 1c). The climate is subtropical, with annual average rainfall of 1100-1300 mm distributed mostly (70-80%) between June and September. Summer temperatures can reach 46–˚C compared with lows of 4–˚C in the winter. The Hazaribagh district comprises more than 70 percent of the catchment area, and most of this territory is made up of forests and rocky soils. Overall, our land use categories are found in the study catchment: i.e., agricultural lands (38%), forests (36%), barren lands (14%), and human settlements (12%) (Figure 1b). Rice, groundnuts, and maize are the primary crops cultivated in the agricultural areas during the monsoon season, while wheat, mustard, and other vegetables are grown using terrace farming on uneven terrain during the off-monsoon season. Mixed deciduous and tropical dry forests predominate in the forest zones with several species of medicinal plants and timber trees including sal (*Shorea robust*) (Forest, Environment and Climate change Department, Government of Jharkhand; https://forest.jharkhand.gov.in). The three most common soil types are lithosols (46%), ferric luvisols (38%), and eutric nitosols (16%) (Supplementary Figure F1).

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2.2 Soil and sediment sampling

One of the main challenges of the sediment fingerprinting approach is collecting sufficient source samples for statistical reliability (Collins & Walling, 2002). To study the temporal variation in the suspended sediment contributions from different sources, water samples were collected for six time periods from the inlet of Konar reservoir (shown in Figure 1c) for two alternate water years; i.e., July 2018-June 2019 and July 2021-June 2022. Three 2L swabs of suspended sediment were collected at a water depth of 0-10 cm and stored in high-density polyethylene bottles during these sampling periods (Wang et al., 2019). The soil sampling plan was designed to cover the spatial heterogeneity of the land use classes in the study catchment and Google Earth and topographic data were used to locate the sampling points (Boardman, 2016). Adopting a composite sampling design is a practical solution to the issue of collecting enough source samples for statistical reliability when applying the sediment fingerprinting approach (Collins et al., 2017; Collins & Walling, 2002; Williamson et al., 2023). To execute the composite sampling approach, 105 sites distributed throughout the study catchment and representative of the different land use were used. Figure 2 shows photographs taken during the sampling campaign for both the land use source classes and for target sediment in the Konar reservoir (details of sampling protocol~~details~~ are shown in supplementary Table T1). Composite sampling involved merging three to four sub-samples collected within a radius of 100 to 500 m, depending on accessibility (Collins et al., 2017). The upper 5~~five~~ cm of soil was~~were~~ sampled at each source sampling location using a non-metallic trowel deployed in one extensive campaign. This one-off source sampling strategy assumed that lithological features remained constant through time (Tiecher et al., 2017).

2.3 Sample preparation and laboratory analysis

To extract suspended sediments from the bulk water samples, the samples were first centrifuged, then filtered, and finally oven dried at 70 ˚C for 12 hours. After 12 hours of oven drying, soil samples were passed through a 63 µm sieve shaker for 24 hours to avoid aggregation and~~to~~ extract the silt and clay fractions to improve the direct comparability of source and sediment samples (Collins & Walling, 2016). Prior to the sample processing,

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scanning of the soil and sediment samples was performed using a DP-6000 Delta Premium portable X-ray fluorescence (PXRF) machine equipped with an Rh X-ray tube operating at 15-40 keV. Using the instrument's Geochem Mode, the concentrations of V, Cr, Fe, Co, Ni, Cu, Zn, W, Hg, As, Se, Pb, Bi, Rb, U, Sr, Y, Zr, Th, Mo, Ag, Cd, Sn, Sb, Ti, Mn, Mg, Al, Si, P, S, Cl, K, and Ca were estimated (Sharma et al., 2014). The particle size characteristics of the source and target sediment samples were measured using a Malvern Pananalytical Mastersizer 3000.

2.4 Formulation of the Bayesian framework and priors for source apportionment modelling

Geochemical tracers were selected using a standard two-step process consisting of a Kruskal-Wallis H-test for inter-group differences, and a stepwise discriminant function analysis (DFA) for selecting a minimal set of tracers that maximises source discrimination (Collins et al., 1997). MixSIAR, a state-of-the-art Bayesian Isotope Mixing Model (BIMM) available as a free R package, was used to estimate sediment source apportionment (Guerrero & Rogers, 2020; Stock et al., 2018). The geochemical tracers were entered into a concentration-dependent MixSIAR model with and without the use of the informative priors (Upadhayay et al., 2020). The following settings were applied for the Markov Chain Monte Carlo (MCMC) iterations: number of chains = 3, chain length = 3,000,000, burn = 1,500,000, thin =500. Gelman-Rubin and Geweke diagnostic statistics were used to verify model run convergence (Stock et al., 2018). The deviance information criteria (DIC) was used to choose the best fitting model. Means and 95% Bayesian confidence intervals (CI) were provided from posterior distributions derived for sediment source contributions to estimate these contributions and their associated uncertainty (Upadhayay et al., 2020). ~~(~~Upadhayay et al., (2017) provide a comprehensive description of MixSIAR, and Stock et al., (2018) provides a comprehensive mathematical explanation of MixSIAR.

2.4.1 Prior selection

Informative priors for sediment source proportions should be selected logically with proper physical meaning (Upadhayay et al., 2020). Research suggests that rainfall-induced soil erosion occurs in primarily three phases, each of which is particle size-based (Figure 3) (Sadeghi et al., 2017; Wang & Shi, 2015). The initial stage comprises the detachment of soil particles by raindrop splash - eroding mostly very fine particles and some fine particles (Figure3a). The second stage i.e. after a prolonged rainfall event, is characterized by an increased proportion of coarser particles and fine particles (Figure 3b). Finally, in the third stage, sediment transportation of very fine particles and lower quantities of fine and coarse particles takes place through the stream network (Figure 3c). The slope of the terrain plays a very important role in the erosion process (Reza Vaezi et al., 2020). In this study we explored the sensitivity of the source proportion estimates to the choice of particle size priors (clay <2 µm, silt 2-50 µm, and fine sand >50 µm) by adjusting the prior specification and observing the impacts on the posterior distributions generated by MixSIAR. Particle size priors were derived by determining the mean proportions of clay, silt, and very fine sand in the individual land use classes. Mean slopes of the land use groups were used to incorporate the slope priors.

2.5 Formulation of the source sensitivity index (SSI)

The sediment fingerprinting outputs provided by MixSIAR are relative and thereby do not provide information on the sediment yields from individual sources. To address this shortcoming, we developed an SSI using: (i) sediment yield factor for each land use class; (ii) proportion of area covered by each land use class in the overall catchment and the sub-catchments therein, and; (iii) information on the temporal variability of the source contribution obtained from the sediment fingerprinting analysis (Liu et al., 2023). An SSI value may be derived to show the severity of the sub-catchments by linking the area covered by the sub-catchment, the land use distribution within the sub-catchment, and the temporal variations in the total catchment's sediment production. Accordingly, the SSI was estimated as:

$$S_{ef} = \frac{X_e}{A_e} \qquad (e = 1{:}m, f = 1{:}n) \qquad (1)$$

$$SSI_x = \left(\sum_{f=1}^{n} \sum_{e=1}^{m} \frac{S_{ef} . A_{ex}}{A_x} \right) . D_x . P_f \qquad (2)$$

S_{ef} = sediment yield factor for land use e in the month f.

X_e = Source contribution (land use) to the catchment sediment yield obtained from the fingerprinting approach.

A_e = Area covered by respective land use class (e) in the catchment.

SSI_x = Source sensitivity of sub-catchment x.

A_{ex} = Area covered by land use class e in sub-catchment x.

A_x = Area covered by sub-catchment x.

D_x = Distance factor of the sub-catchment outlet (calculated as the ratio of longest flow path and distance of the sub-catchment outlet from the catchment sink where the target sediment samples were collected) (Supplementary Figure F2).

P_f = Proportion of annual sediment yield in the respective month.

Figure 4 depicts the analytical framework used to generate spatial maps based on SSI values in our study area. This figure also shows the whole methodology of this study ~~paper which~~ employ~~s~~ings physical modelling outputs to authenticate sediment fingerprinting outcomes through SSI generated spatial maps.

2.6 RUSLE-based hydrological model development: INVEST-SDR

Quantification and mapping of soil loss and sediment delivery in a landscape can be accomplished using the Integrated Valuation of Ecosystem Services and Tradeoffs (INVEST)- Sediment Delivery Ratio (SDR) model (Aneseyee et al., 2020). Compared to other models, such as SWAT, INVEST-SDR uses less input data (Table 1) and is more flexible; it can also be modified to a given scenario and used with locally and globally accessible data and exemplifies the hydrological connectivity concept proposed by Vigiak et al., (2012). To validate the INVEST SDR model, annual simulated sediment inflow data was compared to real data collected from Damodar Valley Corporation (DVC) using root mean square error (RMSE) and the coefficient of determination (R^2).

3 Results

3.1 Catchment sediment sources using geochemical source fingerprinting

Correct and reliable use of sediment fingerprinting requires statistical analysis and interpretation of isotope tracers from sediment source end members. One of the most important step is to identify a representative set of final geochemical tracers to use in a "composite signature" to determine the origin of fine sediments. From the laboratory analysis, 24 elements were detected in the soil and sediment samples collected from the Konar catchment. To identify the set of discriminatory geochemical tracers to be used as an input to the MixSIAR, all the elements were passed through a KW test followed by DFA (Figure 5). Based on this analysis, 21 elements were selected (Fe, K, Ti, Ca, Zn, Mn, Ba, Zr, Rb, V, Cr, Sn, Ni, Sr, Pb, Cu, Ga, Sb, Ag, A, and Br). A summary of the statistical analysis for source discrimination is provided in supplementary Table T2. Furthermore, we have also shown the spatial variability of the proportional (%) presence of few elements (Fe, K, Ti and Ca) in the soil samples collected from 105 locations of the catchment (Figure 6).

3.2 Bayesian modelling results for source apportionment

MixSIAR was run with both 'no priors (P0)' and priors based on particle size and slope information for the study catchment (details of P1 to P4 are given in Table 2). During both time periods, both agricultural and barren lands were the most important sediment sources (Figure

7~~6~~). The Bayesian model with no priors (P0) and with the silt based prior (P2) identified agricultural lands as the major sediment source in most of the seasons (varying from ~30-45% during the water year 2018-19 and ~30 to 60 % during the water year 2021-22) (Figure 7~~6~~ a1, a2). In contrast to the other prior-based models, these two models (P0 and P2) predicted a smaller contribution of suspended sediment from agricultural areas and a larger contribution from human settlements (Figure 7~~6~~ b_1, b_2). The clay based prior (P1) and the slope based prior (P4) model outputs, both identified barren lands as the primary source of suspended sediment (varying from ~25-75% during the water year 2018-19 and ~20 to 60 % during water year 2021-22). In contrast, the outputs of the slope based prior model suggested negligible contributions from human settlement areas compared with the other land use classes (varying between 0-5% for both water years 2018-19 and 2021-22). The very fine sand prior (P3) based model predicted a much higher contribution from forests (varying from ~20-50% during the water year 2018-19 and ~25-50% during 2021-22). Overall, all the prior based models identified agricultural land as the primary source of sediment in July 2021 (Figure 7~~6~~ a2).

3.3 SSI based Bayesian modelling results for source apportionment

This study investigated the potential utility of a new index [Source Sensitivity Index (SSI)] to help the policymakers ~~to~~ better understand the relative impacts of sediment sources in the Konar catchment. It integrates sediment fingerprinting information with the physical controls of sediment deposition in the sub-catchments to generate an index value for ranking the sub-catchments to explain conservation urgency. To establish a link with physical modelling results, the sub-catchments were classified into five SSI classes [class1 (26-174), class2 (175-378), class3 (379-822), class4 (823-1267), class5 (1268-5266)] (Figure 8~~7~~). Figure 9~~8~~ displays the distribution of the sub-catchments across the SSI classes for the particle size and slope prior based model outputs, suggesting little disparity among the sub-catchments using the P0 and P2 priors based models. However, upon closer inspection, it can be seen that the number of sub-catchments with higher SSI classes is marginally greater during 2021–2022, compared with 2018–2019. This SSI based information and associated maps can be used for assessing the reliability of mixing model source apportionment results based on the comparison with physically-based model outputs. More sub-catchments under class 5 (C5) were discovered by P1 based model in both time frames, similar to the P4 based model. However,~~While~~ it is evident from the Bayesian model findings described in section 3.2 that a comparable number of sub-catchments come under the prior-based models P0 and P2. Importantly, the SSI based analysis demonstrated how effectively Bayesian sediment fingerprinting results can be converted into translating tool to spatially explain the sediment dynamics in the Konar catchment. This SSI based information and maps can be used as a criterion for assessing the reliability of mixing model source apportionment results in comparison with physically based sedimentation models spatial outputs.

3.4 INVEST-SDR model based results

The assessment of sediment export using INVEST-SDR model involved computing the annual soil loss, sediment connectivity, and sediment delivery ratio of the study catchment. According to the coefficient of determination (R^2) and root mean square error (RMSE) values of 0.81 and 6.85 t/ha/year for the Nagwan gauging station, the predicted sediment export from the catchment was in line with the available observed data over time (Supplementary Figure F3). Using the sediment connectivity index (SCI) proposed by Borselli et al., (2008) the mean sediment connectivity is estimated to be -6.326, ranging between -11.038 and 0.179 (Figure 10~~9~~a). INVEST-SDR's estimate for soil loss ranged from 0 to 23321 t/year, with a mean soil loss rate of 23.65 tonnes per year (Figure 10~~9~~b). The sediment delivery ratio was computed based on the connectivity index and ranged from 0 to 0.322, with a corresponding mean value of 0.082 (Figure 10~~9~~c). The spatial variation in the Konar catchment's sediment export, ranging from 0 to 3490 t/year with a mean value of 11.16 t/year, is shown in Figure 10~~9~~d. The Konar catchment was sub-divided into 47 sub-catchments to pinpoint the crucial hot spots of soil erosion for targeting preventative measures. All sub-catchments were ranked and classified based on the sediment export in t/year (Figure 10~~9~~e) and t/ha/year (Figure 10~~9~~f). Here, we used two different sub-catchment ranking methods. Firstly, the sub-catchments were ranked according to their annual sediment yield, and secondly, the sub-catchments were ranked according to annual sediment yield per unit area (i.e., specific sediment yield). The overall variance between the two ranking techniques is shown in Figure 11~~10~~a. Sub-catchments ranked 18, 21, 29, 37, 40, and 47 based on annual sediment yield showed substantial disagreement with the ranks assigned using annual specific sediment yield (Figure 11~~10~~b).

3.5 Examining the authenticity of the SSI with physically-based modelling

Sediment fingerprinting and the INVEST-SDR model were used to generate independent predictions. Here, the sediment fingerprinting based SSI technique generated estimates of relative source contributions for surface sediments collected in the reservoir, while the INVEST-SDR model generated sediment yield estimates for the four land use groups. Using results from INVEST-SDR as an independent evaluation, we computed the overall accuracy of the SSI ranking method by employing a confusion matrix for the sediment yield (Figure 12~~11~~). Agreement between the annual specific sediment yield and the SSI approach was better than that with the annual sediment yield of the sub-catchments with all the prior based models. The P0 and P2 based models (i.e., no prior and silt-based priors) exhibited the lowest level of accuracy over both time frames, whereas models based on P1 and P4 (i.e., clay and slope priors, respectively) displayed the highest levels of accuracy (80% and 68% with t/ha/year and t/year, respectively) (Figure 12~~11~~). The P3 (very fine sand-based prior) model predictions of the sub-catchment sediment output was ~65% accurate. Over the two time frames, the clay and slope-based priors performed the best.

4. Discussion

4.1 Multiple prior based geochemical sediment source fingerprinting

The DFA findings on the geochemical tracers showed some overlap between the source groups. Several other investigations have reported similar issues using geochemical data.

The PSD of target sediment can be affected by several factors, including the PSD of the sources, erosion patterns and intensity, and catchment slopes. It is therefore important to consider particle size carefully when using the sediment fingerprinting approach (Laceby et al., 2017; Koiter et al., 2018). Accordingly, particle size based priors were used with slope when applying the MixSIAR model. The Bayesian model with no priors (P0) identified agricultural lands as the most important sediment source (i.e. contributing ~40-55%), with human settlement contributing 8-15%, whereas the barren lands contributed 20 to 25%, and forest ~18 to 25%. The highest human settlement contribution was estimated using the P0 model. However, the P0 model underestimated the contribution of barren lands drastically, and this land use has been reported to be a major sediment source by other studies conducted in this region (Das et al., 2022; Rajbanshi & Bhattacharya, 2020, 2022). The clay prior (P1) based model identified barren lands as the major sediment source (~20 to 70%) followed by agricultural lands (~10 to 70%) during both timeframes. Similar source estimates were generated using the slope based prior (P4) model. This suggests that the steepness of slope in the barren land areas is a major factor controlling sediment sources in the study catchment (Mishra et al., 2022). The silt based prior model (P2) predicted similar source contributions to the P0 model by identifying agricultural lands and human settlement areas as major sediment sources. The effects of silt concentration on geochemical properties were found to be negligible by Kraushaar et al., (2015) and this explains the lack of any significant difference between the source estimates using the silt based prior and no prior (P0) models. The proportion of very fine sand is highest in the bare lands; however, the prior based model for this particle size fraction (P3), identified forests as the major sediment source (~20 to 50 %). A substantial contribution of forests to sediment yield has been reported by a few previous studies (Upadhayay et al., 2020).

4.2 Validation of sediment source fingerprinting with INVEST-SDR

The INVEST-SDR outputs were used as an independent evaluation of the sediment fingerprinting estimates. Previous studies have evaluated source fingerprinting using physically-based modelling. A study by Palazón et al., (2016), for example, reported good consistency between sediment fingerprinting results and SWAT modelling. The accuracy of the five prior based Bayesian model in prioritizing the sub-catchments indicates that the annual sediment yields for the sub-catchments agree less with the SSI results compared to annual specific sediment yield. Similar, apportionment patterns were established in a study conducted by Hamel et al., (2015) which prioritized the sub-catchments based on soil erosion alone, rather than erosion and sediment delivery. The clay and slope priors based models performed better than the other models with an overall accuracy of >78% (Figure 12~~11~~). Previous studies on soil erosion have illustrated how catchment slope affects erosion patterns and the PSD of mobilised sediment particles (Lu et al., 2016; Vigiak et al., 2012; Wang & Shi, 2015). Further, some studies have reported how variable rainfall intensity and slope conditions have resulted in greater detachment of clay particles at the experimental scale (Kiani-Harchegani et al., 2018, 2019; Sadeghi et al., 2017; Zhang et al., 2018).

4.3 Limitations and future outlook

There are a few limitations that must be borne in mind while~~when~~ interpreting the results of this study. Our ability to sample the reservoir for target sediment beyond two water years was limited by time, money, and accessibility issues, and continuous monitoring was restricted in 2020-2021 due to COVID19 lockdowns. Even though, the results of the prior based sediment fingerprinting study performed well when compared to physical modelling, additional source sampling campaigns (annual or seasonal) may have shown greater temporal variation in catchment sediment source contributions. Since a single target sediment sampling location was deployed, the source fingerprinting estimates must be viewed as scale-dependent and longitudinal sampling along the stream network could be used to provide further insight into the sediment dynamics of the study catchment (Koiter et al., 2013). The target sediment samples were not age dated (Fatahi et al., 2022) and may therefore reflect sediment sources over recent times rather than the present day alone. Despite the ~~above~~ aforementioned limitations, however, this novel research suggested that the prior based sediment fingerprinting procedure provides valuable information for understanding the spatial and temporal dynamics of fine sediment sources and delivery in the study catchment. Our study illustrates the benefits of combining sediment source fingerprinting with independent approaches such as physically-based modelling.

5 Conclusion

. The major findings of this study are as follows:

a) The geochemical fingerprints of the sources successfully discriminated between the surface soil samples collected from the land use classes of the Konar study catchment. The sensitivity of the Bayesian model predictions to priors based on particle size and slope was evaluated. The model comparisons suggested that agricultural and barren lands are the most important sediment sources.

b) The performance of INVEST-SDR was satisfactory using the observed datasets from the Nagwan gauging station (R^2=0.81 and RMSE = 6.85 tons/ha~~Ha~~/Year). The two sub-catchment ranking scenarios using the outputs from INVEST-SDR showed disagreements in terms of the higher ranking sub-catchments. However, there were some similarities between the prioritization based on both ranking schemes.

c) Comparison between the two INVEST-SDR ranking schemes and the results of the SSI technique based on five prior based Bayesian models for prioritising the sub-catchments was informative. The overall accuracy of the SSI method considering all the models with the first ranking scheme (i.e., annual sediment yield) varied from 25-62%, whereas, based on the second scheme (i.e., annual specific sediment yield) it ranged between 68-82%. The performance of slope and clay prior based models performed best with > 78% accuracy.

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Figures

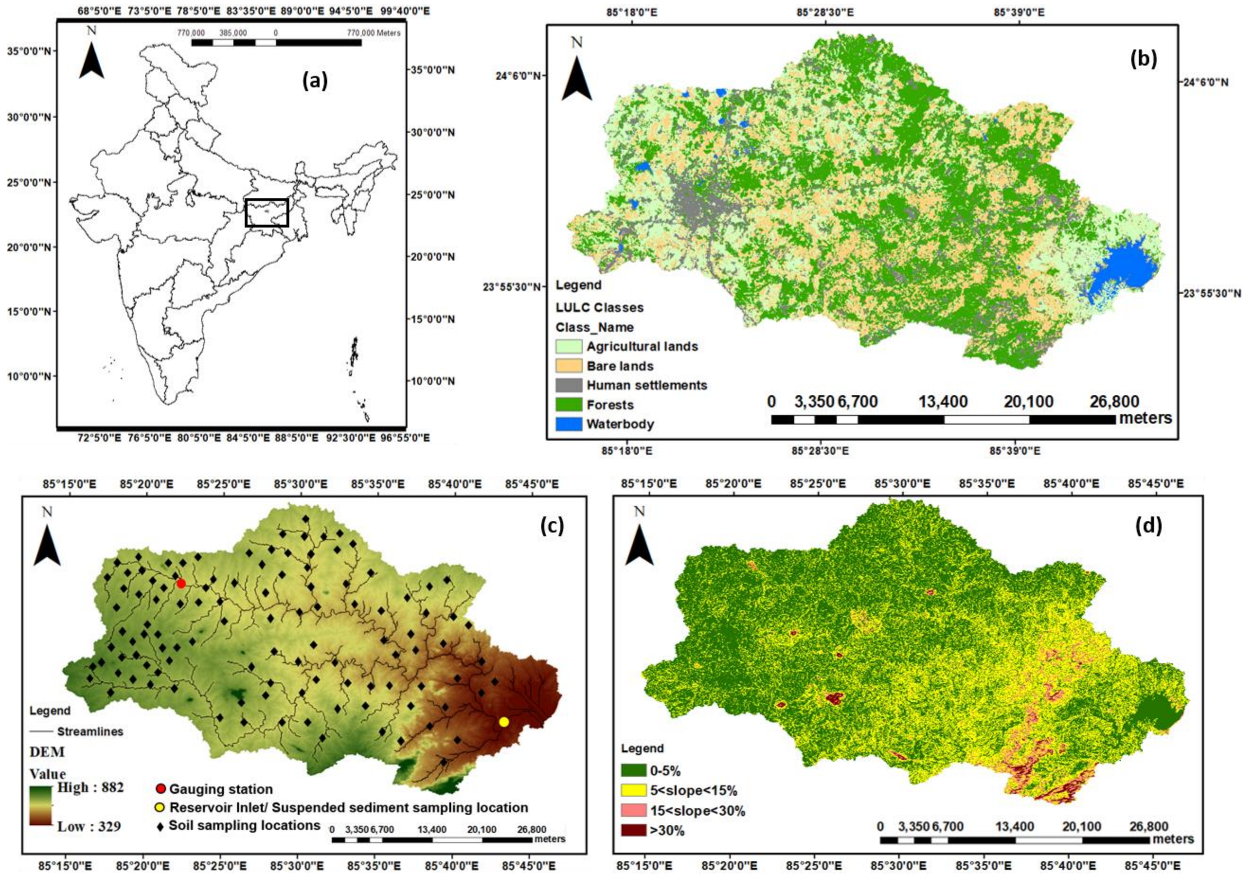

Figure 1. Information on the Konar study catchment characteristics: (a) location (b) land use (c) DEM (d) slope.

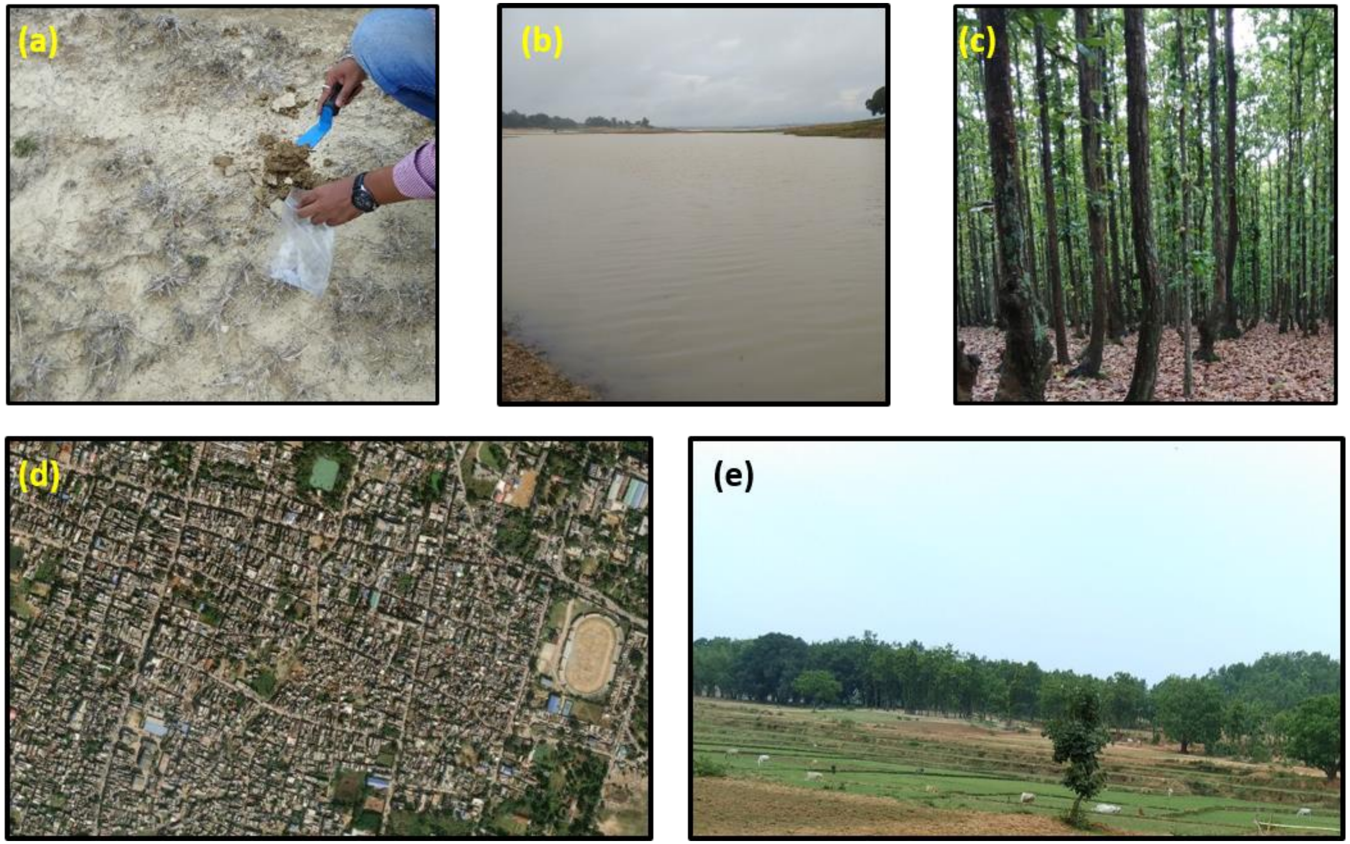

Figure 2 Land use classes of the catchment: (a) bare lands (b) Konar reservoir (c) forest (d) Human settlement (e) Agricultural fields undergoing terrace farming.

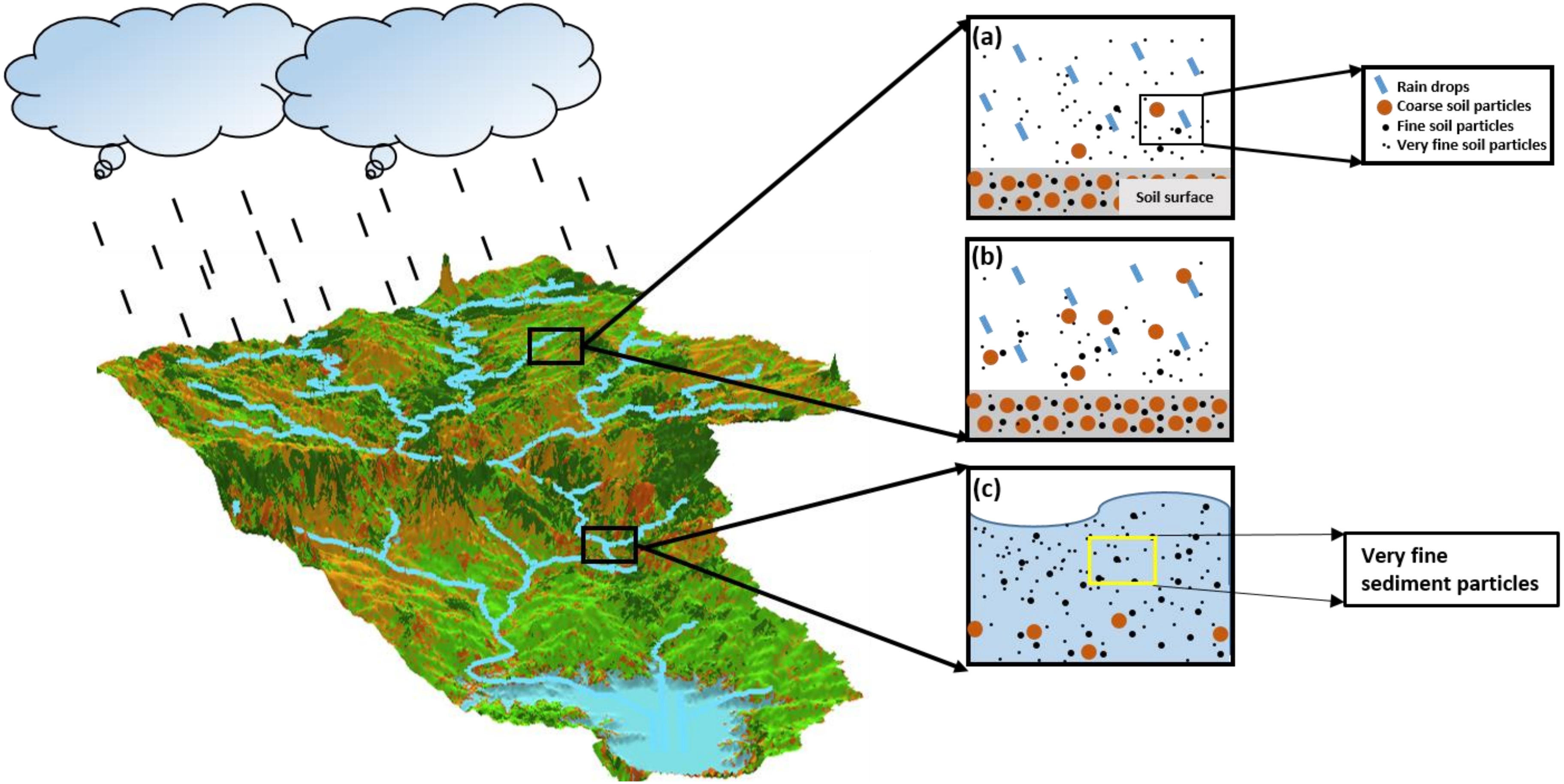

Figure 3 Conceptualisation of soil erosion scenario for prior development: (a) erosion scenario at the beginning of rainfall. (b) erosion scenario after prolonged rainfall. (c) Sediment transportation scenario.

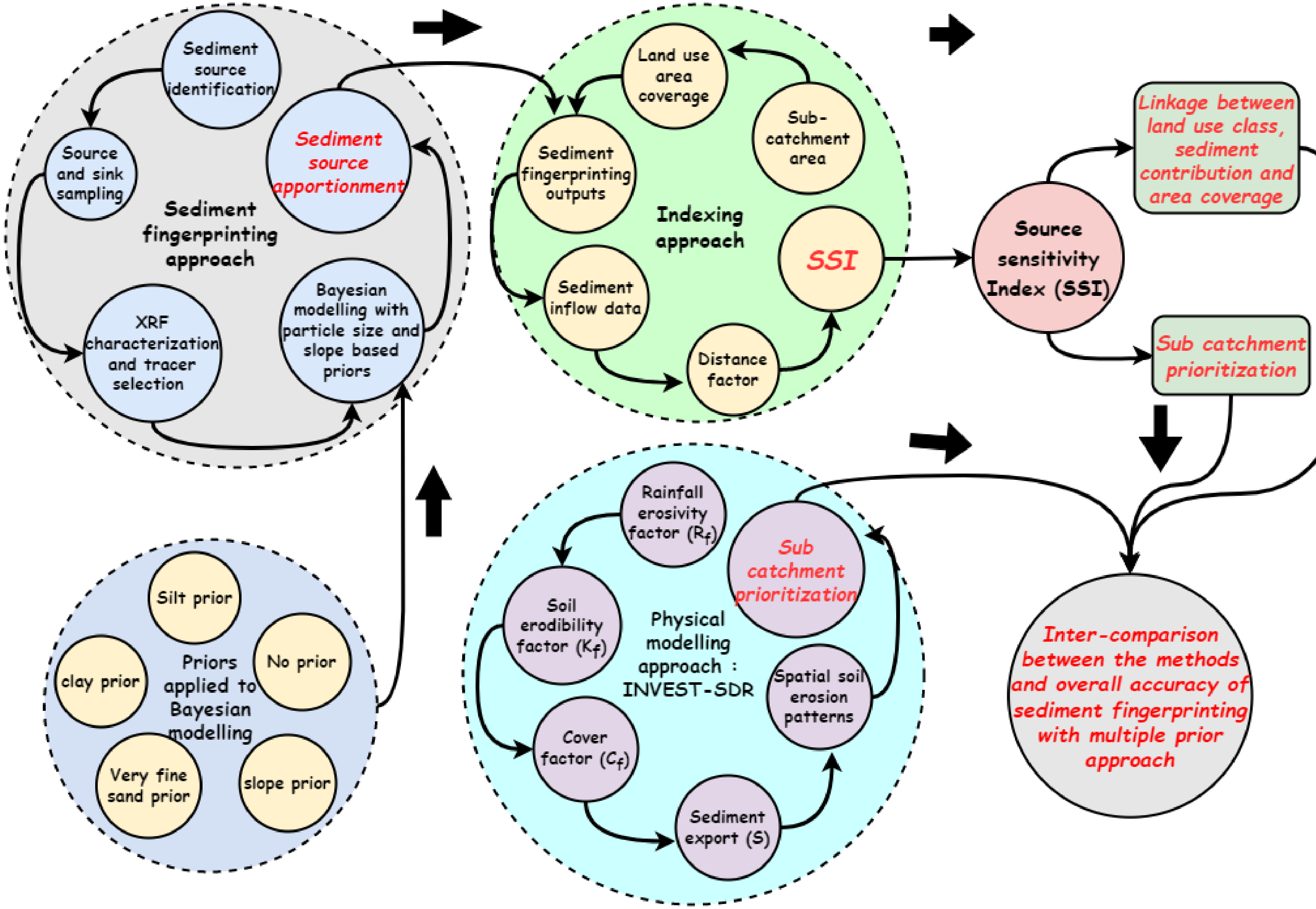

Figure 4 Methodological framework for developing the SSI.

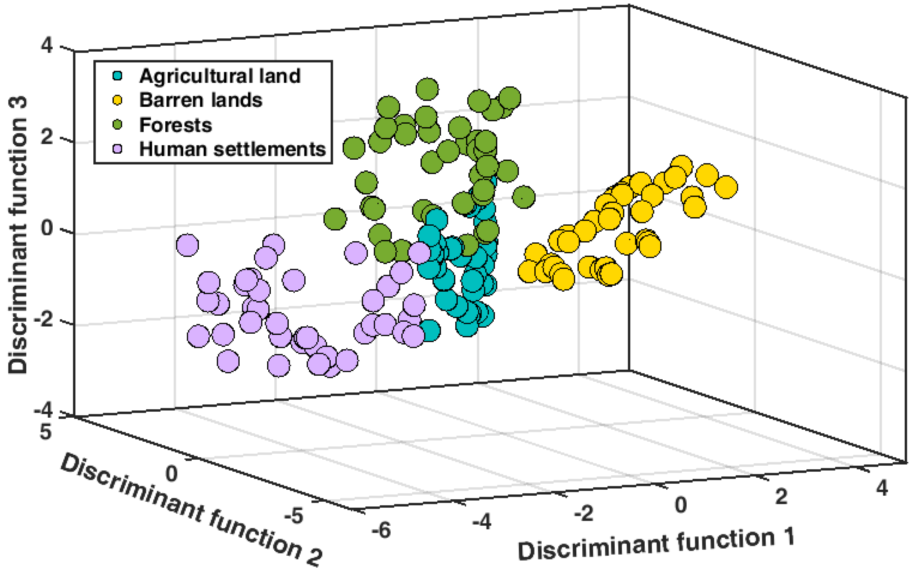

Figure 5 Results of Discriminant Function Analysis on the geochemical tracers of the source samples collected from the land use classes in the Konar study catchment.

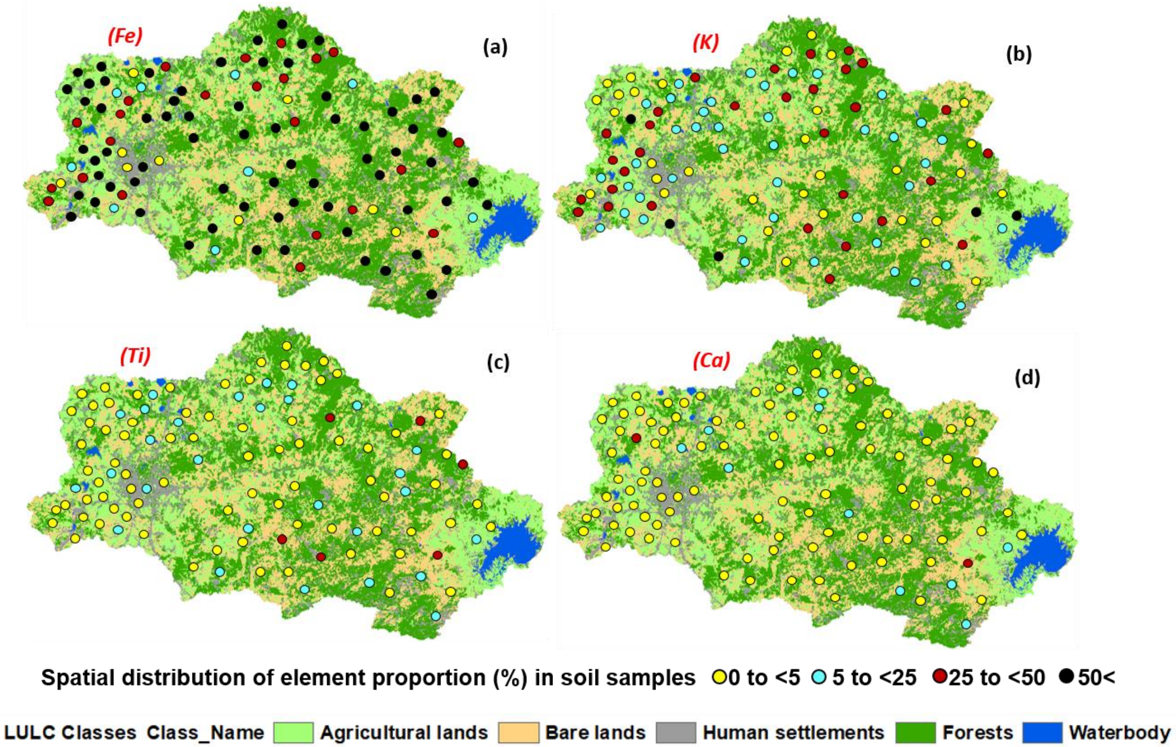

Figure 6 Spatial variation of elemental proportion (%) of (a) Fe, (b) K, (c) Ti and (d) Ca among the soil samples collected from the study catchment

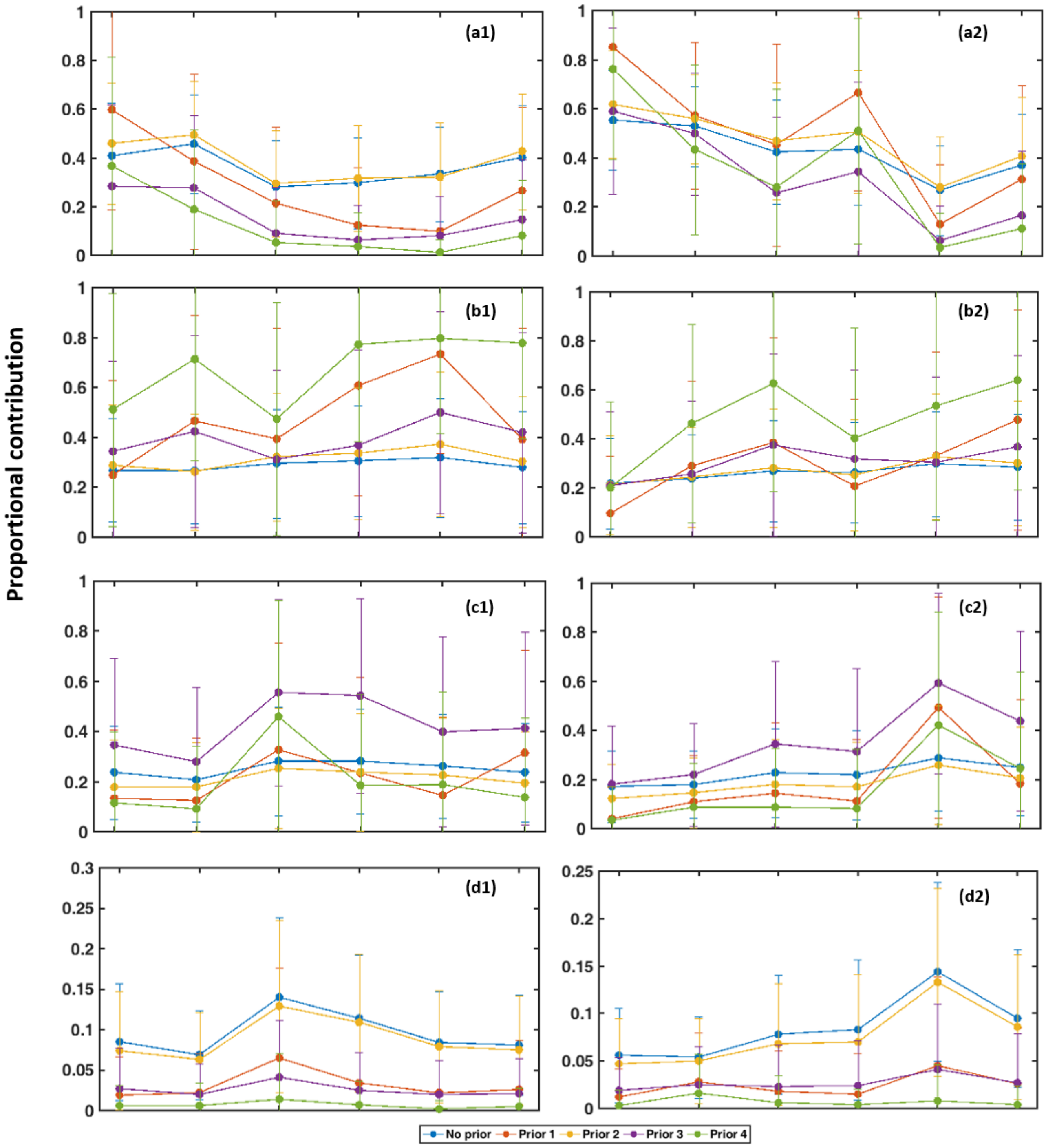

Figure 7 Temporal variation in suspended sediment source contributions (with 95% confidence intervals) using multiple priors (P0, P1, P2, P3, and P4) in MixSIAR. a1, b1, c1 and d1 represent the proportional contributions from agricultural lands, barren lands, forests and human settlements, respectively, in 2018-2019 (July, August, October, December, March and April). a2, b2, c2 and d2 represent the proportional contribution from the land use classes in 2021-2022.

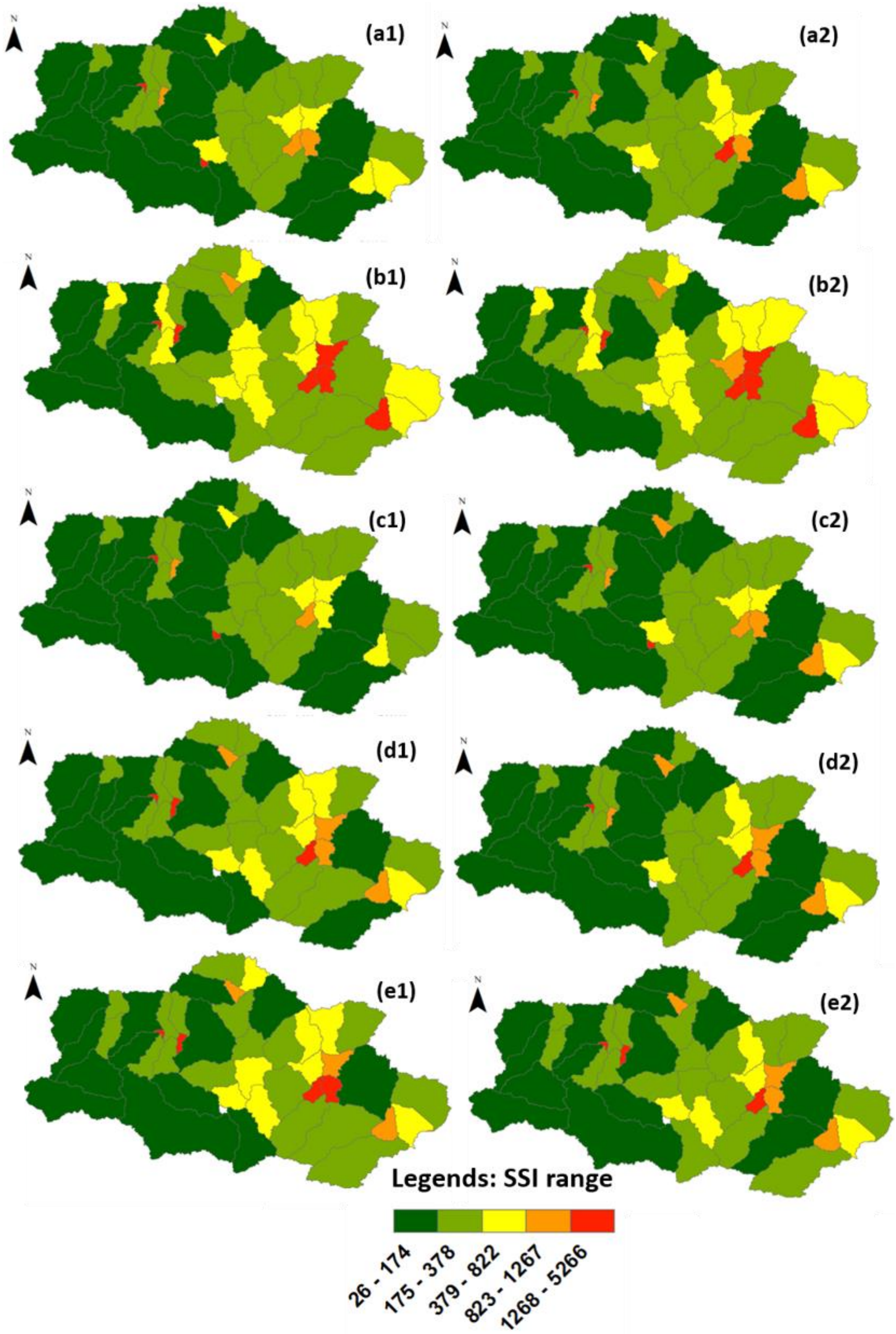

Figure 8 Spatial variation of the SSI calculated for the 47 sub-catchments in the Konar river basin. The SSI calculated using sediment fingerprinting results using multiple priors (i.e. P_0, P_1, P_2, P_3 and P_4) for the period of 2018-2019 is shown in a1, b1, c1, d1, e1 and for 2021-2022 in a2, b2, c2, d2, e2.

***** P_0 represents the no prior condition of the Bayesian modeling study.***

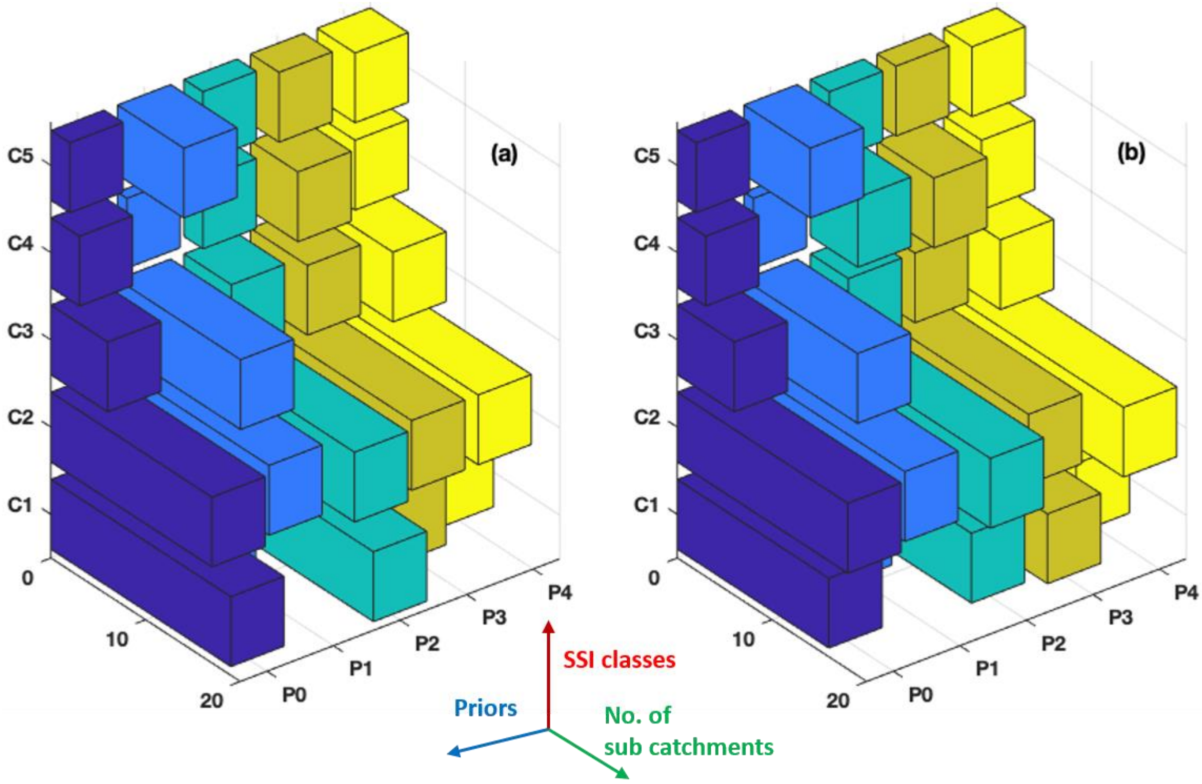

Figure 9 Distribution of numbers of sub-catchments based on respective SSI classes for each prior based Bayesian source apportionment model outputs in the two sampling time frames: (a) 2018-2019, and (b) 2021-2022.

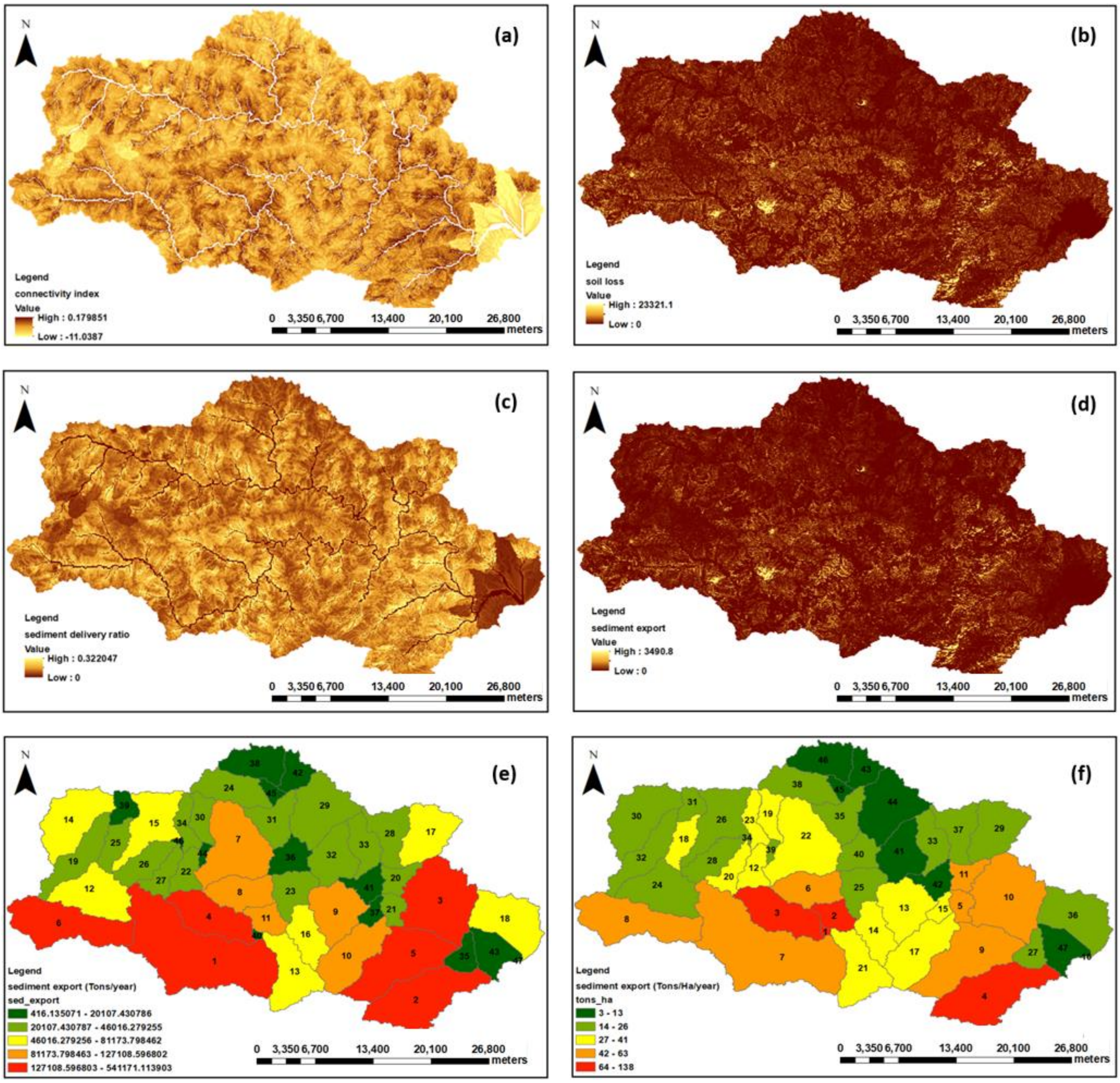

Figure 10 INVEST-SDR model outputs for the Konar study catchment, depicting the spatial variability in: (a) the connectivity index; (b) the annual soil loss (tons/year); (c) the sediment delivery ratio; (d) the annual sediment export (tons/year); (e) ranking and prioritization of the sub-catchments based on annual sediment export, and; (f) ranking and prioritization of the sub-catchments based on specific annual sediment yield. Note: the numbers on the sub-catchments (Figures e and f) represents their ranking based on the criteria.

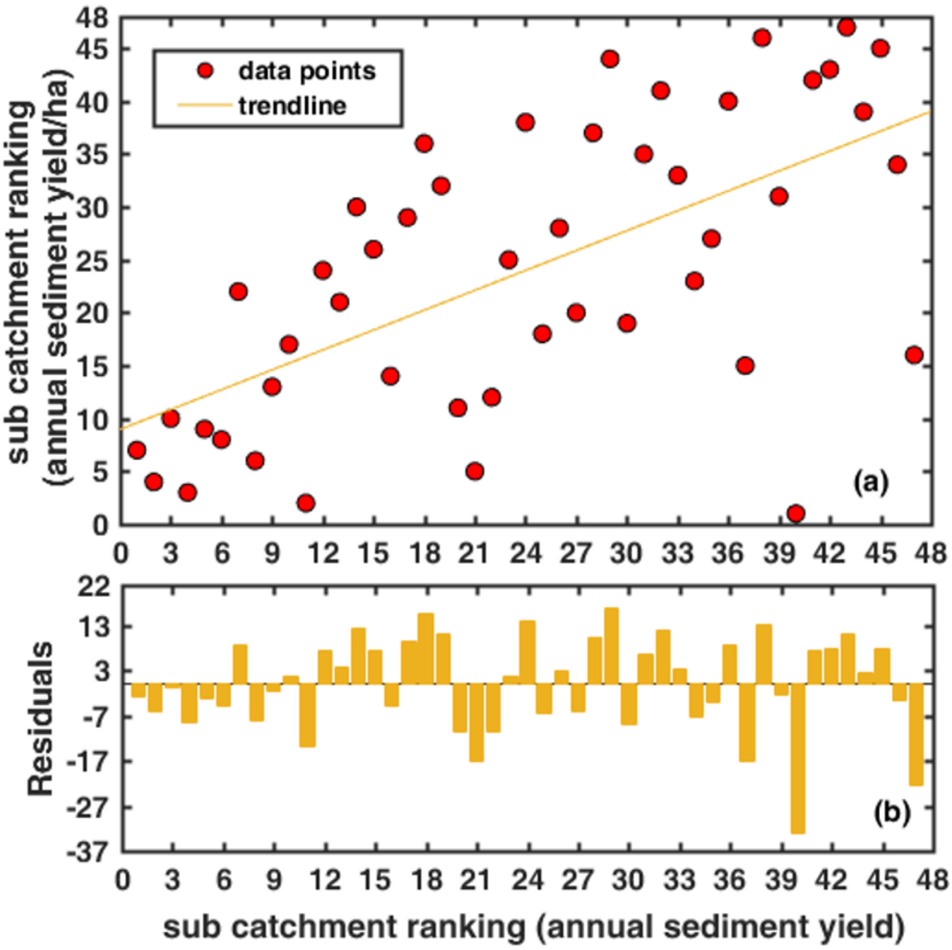

Figure 11 (a) Comparison between sub-catchment rankings based on annual sediment yield and annual specific sediment yield generated using the INVEST-SDR model. (b) Residuals of the ranking formats.

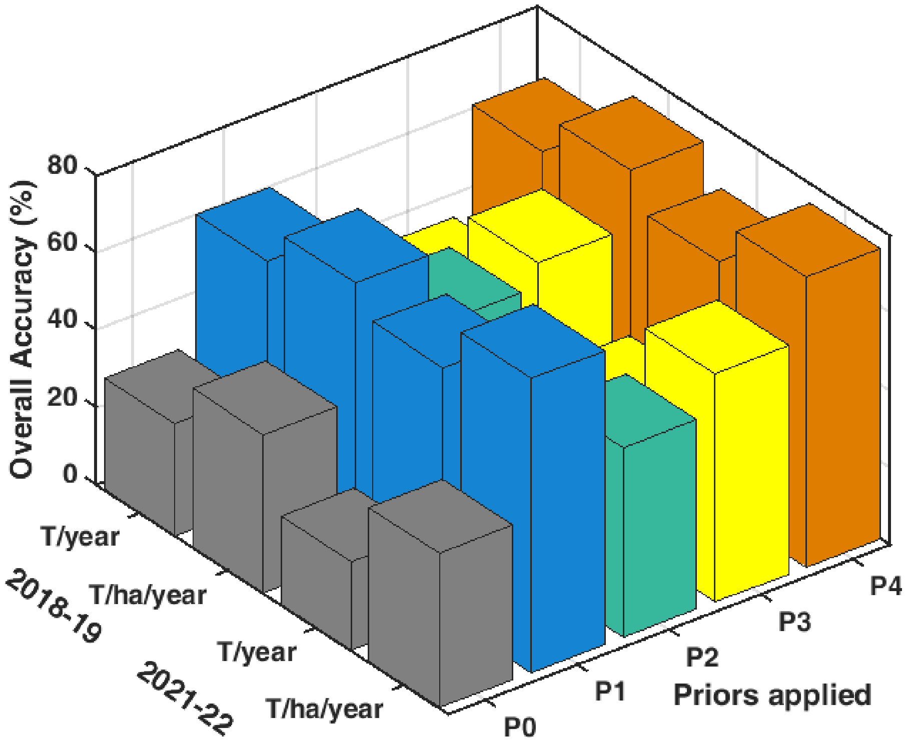

Figure 12 Overall accuracy (%) of the multiple prior based sediment fingerprinting SSI results compared with the INVEST-SDR predictions using confusion matrices.

The spatio-temporal dynamics of suspended sediment sources based on a novel indexing approach combining Bayesian geochemical fingerprinting with physically-based modelling

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Supplementary Information

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- **Table T2 Summary information on the geochemical properties of the source and target sediment samples**

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- **Figure F2 Distance factor for each sub-catchment used for developing the SSI**
- **Figure F3 INVEST-SDR model performance for sediment yield in the Konar river catchment**

Table T1 Sampling details for source and reservoir target suspended sediment samples Jharkhand

Period of sampling: 27^{th} June – 1^{st} August (2018)							
Tools used: Trowel, Polythene ziplock bags							
Steps	**Explanation**	**Sites Information**					**Supportive references**
		Location Code	**Locations**	**Land use classes present**	**Geographical Coordinates**	**Weather conditions during sampling**	
Site selection	The sampling sites (n = 105) were selected on the basis of land use classes. The sampling sites were selected **randomly** so that they should be able to represent the heterogeneity of land use across the watershed.	1.1	Sultana	Hs, A, Ba	23 59 28/85 15 16	Cloudy	**(Bahadori et al., 2019; Gaspar et al., 2019)**
		1.2	Banadag	FA	23 58 0/85 18 17	Cloudy	
		1.3	Hazaribagh town	FHs	23 58 33/85 15 48	Clear	
		1.4	Panchsheel colony	A	23 57 35/85 21 8	Clear	
Soil sample collection	All the soil samples were collected in zipped bags using a metallic trowel which was washed after every use to avoid contamination. The samples were collected from a depth of **0-5 cm.**	1.5	Chehla, churchu road	F, Ba	23 55 49/85 25 25	Clear	**(Nosrati and Collins, 2019)**
		1.6	Rehda/ dumar	has	23 58 57/85 28 32	Clear	
		1.7	Chalchalia	F	23 55 18/85 31 29	Clear	
		1.8	Mangarpatta	FA, Ba	23 58 48/85 35 37	Clear	
		2.1	Sulmi	FHsA	24 4 56/85 19 12	Clear	
Composite Sampling	To make the analyses more affordable, composite samples were collected. Each composite sample consisted of a mixture of **four to five sub- samples collected in a radius of 50 to 100 meters.**	2.2	Jalima/bartua	FHsA	24 2 14/85 18 25	Clear	**(Collins et al., 2010b)**
		2.3	Moktama	FA, Ba	24 4 33/85 29 19	Clear	
		2.4	Vinod bhave university	FHs	24 1 16/ 85 22 0		
		2.5	Tepsa/saram	FHsA	24 3 49/85 24 24	Clear	
		2.6	Hazaribagh national park	FF, Ba	24 0 58/85 24 10	Clear	
		2.7	Barwa	HsA	24 4 5/85 26 52	Clear	

Processing	All source samples were oven dried for around **12 hours** to remove the moisture content. Finally, after oven drying, the samples were passed through a **63µm sieve**. The sieving undertaken to minimize contrasting grain sizes between the soil and target sediment samples.	2.8	Pundri/bandua	FF, Ba	24 1 51/85 29 16	Cloudy	**(Tiecher et al., 2019)**
		3.1	Bakra khurd/jagra	HsA	24 6 20/85 28 10	Cloudy	
		3.2	Darha	FHsA, Ba	24 6 36/85 30 51	Clear	
		3.3	Madhu kunj	FA	24 3 49/85 30 42	Clear	
		4.1	Khaira	FHsA, Ba	24 5 6/85 33 42	Clear	
		4.2	Basobar	FA	24 1 44/85 32 54	Clear	
		4.3	Hatwe	FA, Ba	24 0 47/85 36 23	Clear	
		5.1	Khamwa	FHsA, Ba	24 3 32/85 37 41	Clear	
		5.2	Siju	HsA	24 3 31/85 40 20	Clear	
		5.3	Holang	HsA, Ba	24 1 26/85 38 29	Drizzle	
		6.1	Kutse	FA, Ba	23 54 48/85 34 40	Clear	
		6.2	Hatwal	F, Ba	23 56 32/85 35 55	Clear	
		6.3	Alkhari khurd	FHsA	23 58 34/85 43 11	Clear	
		7.1	Belwara	F	24 0 52/85 40 48	Rainy	
		7.2	Hanuman temple near Belwara	FA	23 59 13/85 40 44	Rainy	
		7.3	Ambadih	FA, Ba	23 57 36/85 38 47	Clear	
		7.4	Gobindpur kalan	HsA, Ba	23 57 10/85 41 58	Rainy	
		7.5	Karidih	FHs, Ba	23 55 51/85 38 33	Clear	
		7.6	Banda/bhitia	F	23 54 13/85 36 49	Clear	

Location	**Sediment sampling Information**			**Supportive reference**
	Dates of sampling	**Seasons**		

Inflow point of the reservoir. (Magha [23 55 08/ 85 43 17])	July (2018)	Monsoon	All the target sediment samples were collected from a depth of **0-10 cm** from the surface using High Density Polyethylene bottles. All bottle was washed prior to being used.	EPA (operating procedure) for surface water sampling & (Wang et al., 2019)
	August (2018)	Monsoon		
	October (2018)	Post monsoon		
	December(2018)	Post-monsoon		
	March (2019)	Pre-monsoon		
	June (2019)	Pre-Monsoon		
	July (2021)	Monsoon		
	August (2021)	Monsoon		
	October (2021)	Post monsoon		
	December(2021)	Post-monsoon		
	March (2022)	Pre-monsoon		
	June (2022)	Pre-Monsoon		

Table T2 Summary information on the geochemical properties of the source and target sediment samples

	Soil samples collected from the land use classes																			
Soil properties (%)/ Land use details	**Agricultural lands**				**Forests**				**Barren lands**				**Human settlements**				**Sediment samples**			
	Mean	**Median**	**Min**	**Max**	**Mean**	**Median**	**Min**	**Max**	**Mean**	**Median**	**Min**	**Max**	**Mean**	**Median**	**Min**	**Max**	**Mean**	**Median**	**Min**	**Max**
Fe	52.4452443	53.8707585	36.7766686	61.8031957	53.7937426	55.2559103	35.8310485	64.0976394	48.4143684	49.7303193	34.5008208	65.1953114	65.5776417	51.5765574	35.7816638	67.6156874	59.5936819	46.8701965	32.516587	61.4457559
K	26.6267432	24.124644	18.4477141	43.7865306	27.3113833	24.7449489	17.9733773	45.4121056	24.580245	22.270454	17.3061156	46.1897879	33.2941347	30.1655046	23.141998	56.3155106	30.2560449	27.4129023	21.0302907	51.1767202
Ti	8.22615988	8.07292072	5.55015957	14.2070847	8.43767502	8.2804957	5.40745109	14.7345227	7.59390752	7.45244613	5.20669947	14.9868514	10.2860073	10.0943967	6.9624768	18.2722682	9.34740913	9.17328303	6.32715079	16.6049237
Ca	6.78911047	6.0491585	3.50763235	13.4732246	6.96367547	6.20469749	3.41744235	13.9734181	6.26730792	5.58422774	3.2905698	14.2127129	8.48911774	7.56388028	4.40019941	17.328423	7.71448575	6.8736762	3.99868121	15.7472044
Zn	1.39453264	1.02223172	1.0023	4.70556781	1.43038956	1.04851585	0.9871	4.88026201	1.28735061	0.94366427	0.74915	4.9638365	1.74372649	1.27820065	0.896523	6.05200846	1.58461145	1.16156484	1.00258	5.49976269
Mn	1.21850666	1.1110612	0.63872095	2.11310978	1.24983752	1.13962936	0.62229784	2.19155898	1.12485377	1.02566642	0.59919503	2.22908942	1.52362325	1.38927321	0.8012526	2.71775028	1.38459263	1.26250203	0.7281383	2.46975557
Ba	1.18539249	1.11783569	0.69374696	1.66690676	1.21587189	1.14657804	0.67590899	1.72879067	1.0942847	1.03192024	0.65081587	1.75839621	1.48221721	1.39774406	0.87028076	2.14387173	1.34696489	1.27019991	0.79086764	1.94824343
Zr	0.99539159	0.96756093	0.41107096	2.14517571	1.0209856	0.99243935	0.4005013	2.22481535	0.91888704	0.89319541	0.38563269	2.26291532	1.2446397	1.20984019	0.51567383	2.75899149	1.13106633	1.09944227	0.46861859	2.50723351
Rb	0.25882955	0.21189495	0.14579645	0.66251757	0.26548471	0.2173433	0.14204766	0.68711353	0.23893624	0.19560897	0.13677414	0.69888035	0.32364101	0.26495388	0.18289644	0.85208886	0.29410876	0.24077684	0.16620714	0.77433575
V	0.17936387	0.15922419	0.02008556	0.71901064	0.18397576	0.16331824	0.01956911	0.74570391	0.16557819	0.14698642	0.01884261	0.75847409	0.22427695	0.19909426	0.02519662	0.92474674	0.20381168	0.18092691	0.02289743	0.8403636
Cr	0.14868678	0.15507829	0.05145774	0.2285192	0.15250989	0.15906574	0.05013464	0.23700297	0.1372589	0.14315917	0.04827339	0.24106165	0.18591826	0.19391022	0.0645519	0.29390717	0.16895322	0.17621591	0.05866154	0.26708814
Sn	0.15069462	0.15335216	0.10483635	0.1853196	0.15456936	0.15729523	0.10214074	0.19219959	0.13911242	0.1415657	0.09834877	0.19549101	0.18842886	0.19175186	0.13151345	0.23834653	0.17123473	0.1742545	0.11951285	0.21659741
Ni	0.1035817	0.10588205	0.05958265	0.1296483	0.10624504	0.10860454	0.05805063	0.1344615	0.09562054	0.09774409	0.0558955	0.13676415	0.12951877	0.13239513	0.07474431	0.16674558	0.11770018	0.12031408	0.06792389	0.15153004
Sr	0.09478018	0.09375682	0.00849491	0.1420046	0.09721722	0.09616754	0.00827649	0.14727652	0.0874955	0.08655079	0.00796922	0.14979863	0.11851334	0.11723372	0.01065657	0.18263748	0.10769899	0.10653614	0.00968415	0.16597181
Pb	0.04710794	0.04277717	0.0238035	0.09706644	0.0483192	0.04387708	0.02319145	0.10067003	0.04348728	0.03948937	0.02233047	0.102394	0.05890386	0.05348866	0.02986064	0.12484081	0.05352888	0.04860782	0.02713586	0.11344909
Cu	0.04505262	0.04319631	0.03183113	0.08447427	0.04621103	0.04430699	0.03101267	0.08761037	0.04158993	0.03987629	0.02986132	0.0891107	0.05633389	0.05401275	0.03993101	0.10864555	0.05119342	0.04908409	0.03628731	0.09873164
Ga	0.02146313	0.02066514	0.01368761	0.04344121	0.022015	0.0211965	0.01333567	0.04505397	0.0198135	0.01907685	0.01284058	0.04582552	0.02683755	0.02583974	0.01717062	0.05587138	0.02438862	0.02348186	0.0156038	0.05077312
Sb	0.0413516	0.04238063	0.00257	0.07617307	0.04241485	0.04347034	0.02147	0.07900099	0.03817337	0.03912331	0.025871	0.08035388	0.05170612	0.05299283	0.02715	0.09796906	0.04698794	0.04815723	0.015736	0.08902938

Ag	0.018 08107	0.016 44419	0.010 71677	0.034 27788	0.018 54598	0.016 86701	0.010 44122	0.035 55045	0.016 69139	0.015 18031	0.010 05359	0.036 15925	0.022 60861	0.020 56184	0.013 44381	0.044 08608	0.020 54558	0.018 68557	0.012 21706	0.040 06322
As	0.007 01544	0.007 70575	0.004 32	0.013 33029	0.007 19583	0.007 90389	0.002 79	0.013 82517	0.006 47624	0.007 1135	0.001 596	0.014 06193	0.008 77212	0.009 63529	0.004 597	0.017 14458	0.007 97167	0.008 75607	0.004 15	0.015 58014
Br	0.002 91026	0.003 34191	0.001 27	0.011 40418	0.002 98509	0.003 42784	0.001 87	0.011 82756	0.002 68658	0.003 08506	0.000 982	0.012 03011	0.003 639	0.004 17873	0.001 93	0.014 66735	0.003 30694	0.003 79742	0.001 058	0.013 32895

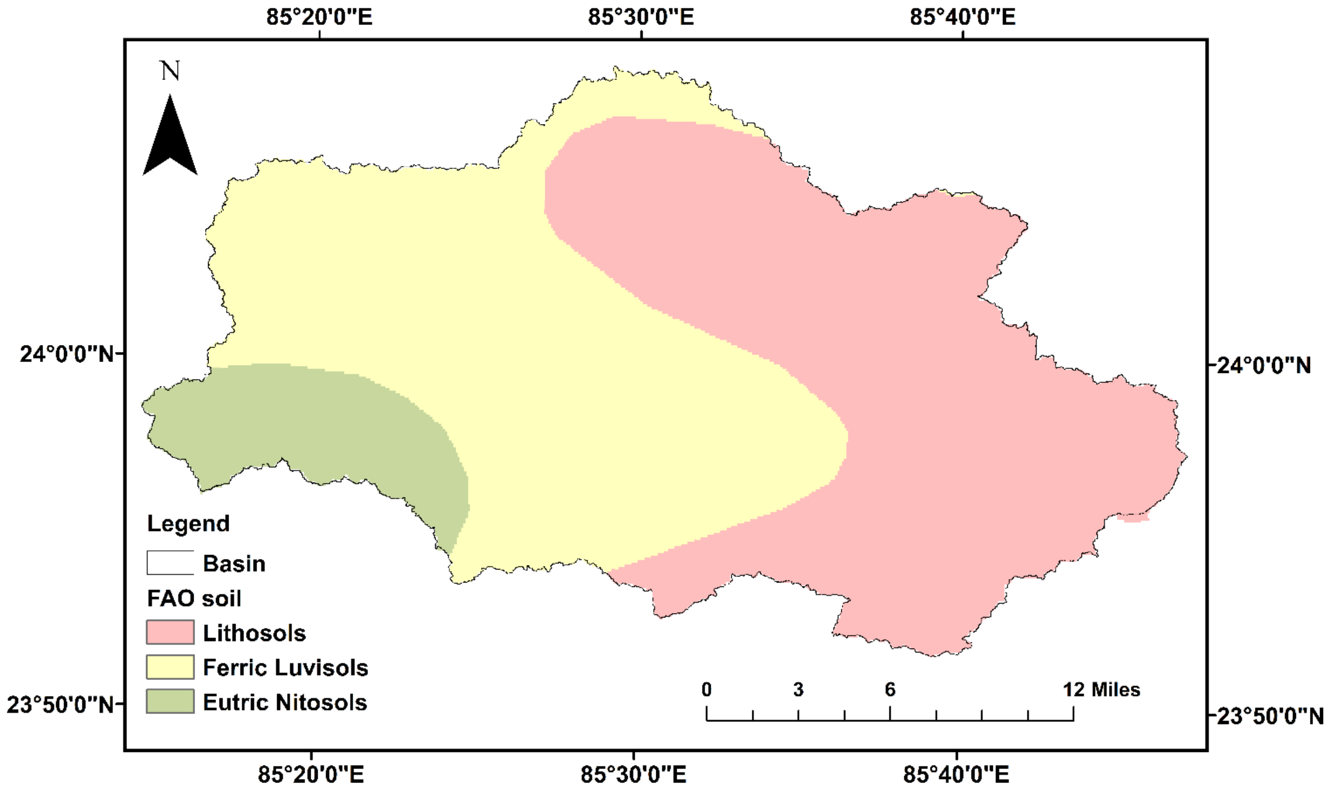

Figure F1 FAO based soil map of the Konar River basin

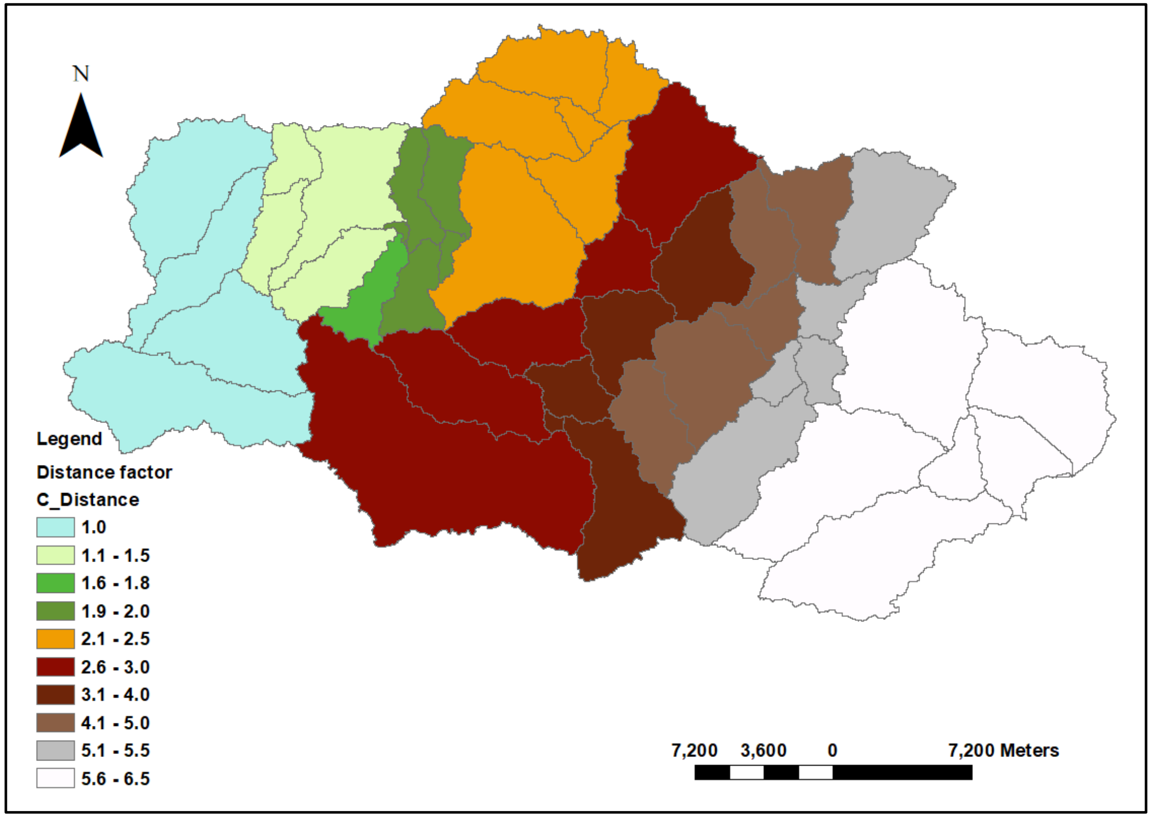

Figure F2 Distance factor for each sub-catchment used for developing the SSI

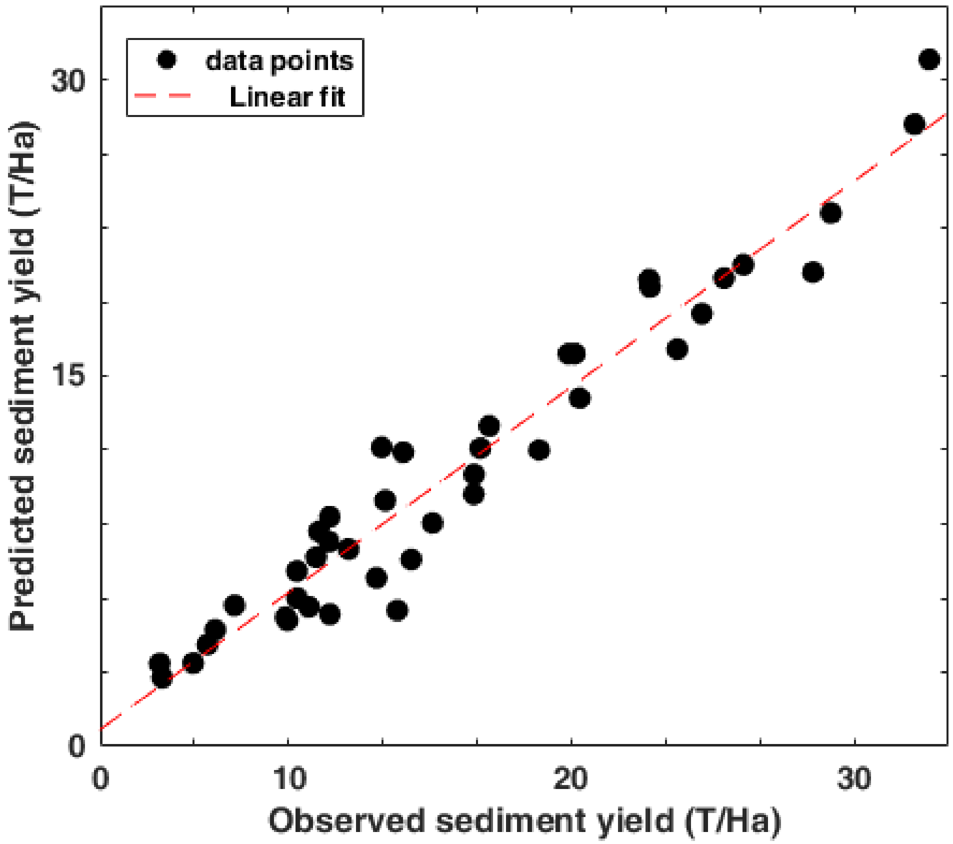

Figure F3 INVEST-SDR model performance for sediment yield in the Konar river catchment

Arnab Das: Modelling, Data curation, Writing- Original draft preparation, Visualization

Renji Remesan: Supervision, Conceptualization, Methodology, Writing

Adrian L. Collins: Supervision, correction and second draft preparation, funding

Ashok Kumar Gupta: Supervision, Validation, Writing- Reviewing and Editing,
All authors reviewed the results and approved the final version of the manuscript

Declaration of interests

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