



Assessment of metal contamination in sediments of a perennial river in India using pollution indices and multivariate statistics

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Abstract

The heavy metal contamination of river sediments by domestic, industrial, and agricultural waste is a major ecological and environmental problem. Therefore, heavy metal concentration and its associated risk were assessed in the sediments from the entry point of Sutlej River in Indian Punjab to its tail end when the river leaves India. The sediment samples ($N=47$) were collected from the Sutlej River during April, 2018 and analyzed for heavy metals (As, Cd, Cr, Co, Cu, Fe, Mn, Ni, and Zn). Results showed that the metal concentration in sediments was higher along the transboundary of the river. The average concentration of metals in sediments was in the order: Fe > Mn > Cu > Zn > Cr > Co > Ni > Cd > As. The concentration of Cd in the sediments only exceeded the threshold values of metal concentration. Principle component analysis indicated that geogenic sources are primarily associated with metal concentration in sediments before river's entry point in Punjab. In contrast, agronomic and industrial sources are related to the origin of metals in the river flowing in Indian state of Punjab. The contamination factor and potential ecological risk index results showed that Cd is the primary contaminant in the sediments of the Sutlej River. Spatial distribution maps of risk indices showed the higher risk values along the transboundary of the river in Indian Punjab. These results suggest that strategic management practices and policies across municipal, industrial, and agricultural sectors are required to reduce the metal concentration in the Sutlej River.

Keywords Heavy Metals · NMDS · Principal component analysis · Risk assessment · Sediments

Introduction

During the last few years, the river contamination is mainly due to global urbanization, industrialization and population explosion (Kumar et al. 2018; Sari et al. 2018). The release of waste water, waste dumping, combustion of fossil fuels, smelting and mining processes, and agricultural and transport activities are the primary sources of contamination of water bodies (Kumar et al. 2020b; Varol and Şen 2012). The metals enter into water bodies either by atmospheric

deposition or industrial wastewater effluents. As metals move into the hydrological system, they are accumulated in the sediments. The metals deposited in the sediments may re-enter into the water bodies by re-suspension and oxidation–reduction reactions. Sediments act as carriers as well as sink for pollutants indicating the pollution history and give a record of surrounding additions into the water bodies (Ahn et al. 2019; Gopalakrishnan et al. 2020; Kumar et al. 2020a). The assessment of metal contamination in river sediments is necessary since they provide shelter to many aquatic organisms (Haghnazar et al. 2021; Singovszka et al. 2017). In general, water quality of rivers is monitored without giving consideration to the sediments which are in contact with the water bodies (Duncan et al. 2018). Metals in river sediments enter either from point or non-point sources (Ali et al. 2016; Haghnazar et al. 2021). The point sources include industrial waste discharge, while non-point sources are silt-laden runoff from mined lands, and leachate from landfills responsible for increasing the metal concentration commonly released into the rivers (Kumar et al. 2018).

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In India, Punjab is one of the most leading states in terms of agriculture development. Its cropping intensity is 190% (Khush 2015; Singh et al. 2020) and the area under assured irrigation is 98% (Vatta 2019). The water of Sutlej River is one of the sources of irrigation in addition to groundwater. Due to the discharge of domestic sewage, agricultural runoff, and industrial wastes into the river and its adjoining areas, it is getting contaminated (Khurana et al. 2014; Setia et al. 2021). There are many studies on water quality of Sutlej river (Khurana et al. 2014; Verma et al. 2013), there are fewer studies on metal concentration in the sediments of the Sutlej River. The sediment contamination by domestic, industrial, and agricultural waste is now graduating into a significant environmental problem in the area. Bessa et al. (2018) and Mandeng et al. (2019) suggested that sediments could act as a screening tool to assess the heavy metal contamination in the surrounding environment. The transfer of heavy metals from sediments to aquatic biota and eventually into the food chain may pose health risks for humans. Therefore, a study was planned to assess the metal (As, Cd, Cr, Co, Cu, Fe, Mn, Ni and Zn) concentration in the sediments of Sutlej River and determine their sources using statistical methods. The specific objectives were (i) to assess the metal concentration in the sediments of the Sutlej River; (ii) to determine the sources and inter-relationships among metals in sediments using correlation and multivariate statistics; and (iii) to assess the contamination of sediments with metals using contamination factor, enrichment factor, and risk indices.

Experimental

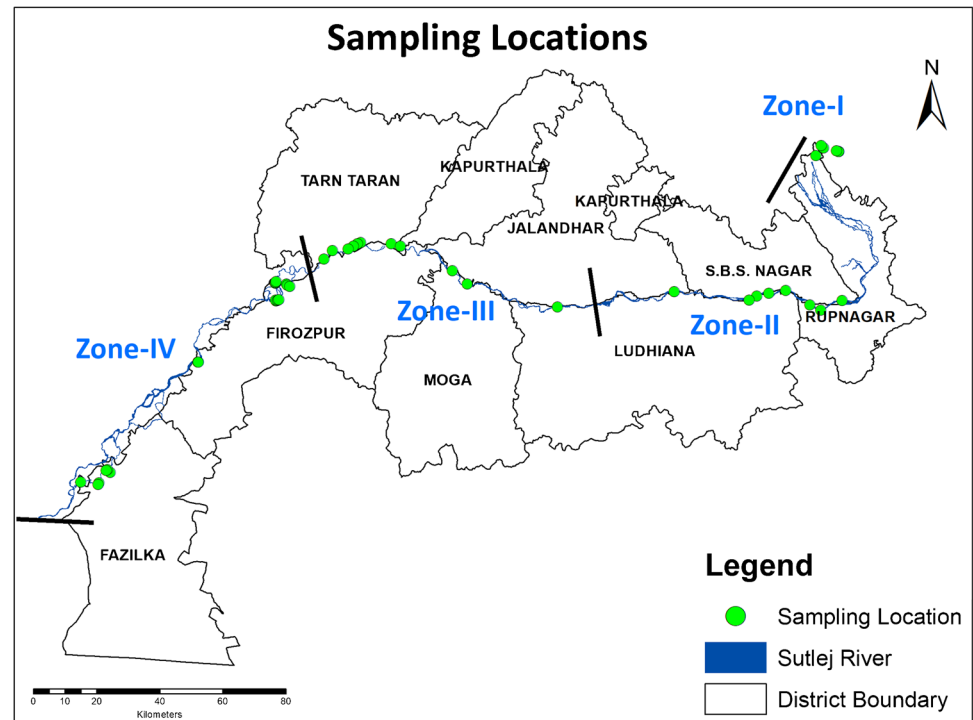
Study area

The sediment samples were taken from the Sutlej River which is one of the tributaries of the Indus River. Sutlej enters India near Shipki La Pass. It traverses through the many Himalayan valleys before it merges with Beas River in Punjab that connects with Chenab in Pakistan before meeting into the Indus River. In the Indian Punjab, the Sutlej River enters near Nangal and passes through Rupnagar and Ludhiana districts. Satluj River joins the Beas river at Harike wetland and crosses over to Pakistan after passing through districts of Indian Punjab.

Sampling and analysis of metals in sediments

Sediment samples ($N=47$) were collected (up to 50 cm depth) in zip lock polyethylene bags during April, 2018 (Fig. 1) and transported to the laboratory at low temperature ($<4\text{ }^{\circ}\text{C}$). The duplicate samples were taken from each site. For heavy metal analysis, 1 g of sediment sample was digested in di-acid mixture ($\text{HNO}_3:\text{HClO}_4:4:1$) until the sediments were digested completely as described in Khurana et al. (2014). The content in the digesting bottles was further diluted with 6 N HCl to make its final volume to 50 ml. The concentrations of Zn, Cu, Fe, Mn, Ni, Cd, Co, Cr, and As were determined using atomic absorption spectrophotometer

Fig. 1 Sediment sampling locations from Sutlej river in Indian Punjab



(Varian AAS FS 240 model) fitted with a graphite furnace. The atomic absorption spectrophotometer (AAS) was calibrated using traceable standards diluted to their appropriate concentrations ranges for each metal. The precision of tracing element measurements was checked by analysis of duplicate samples and the accuracy was checked using four standard samples. The instrument’s detection limit is 0.05 mg L⁻¹ for Zn, Cd, 0.1 mg L⁻¹ for Cu, and 0.5 mg L⁻¹ for Fe, Ni, Co, and Cr. The metal concentrations recorded from AAS were multiplied with their corresponding dilution factors and final results were interpreted as mg/kg.

Landsat 8 satellite imagery of the years 2016, 2017, and 2018 was visually interpreted to determine the spatial variations in contamination of Sutlej River water due to agricultural runoff, domestic, and industrial wastes. Based on the variations in visual interpretation elements (like tone, texture, and pattern, etc.), the sampling locations were classified into the following four zones:

- a. Zone-I (Gobind Sagar)
- b. Zone-II (from GobindSagar to the confluence of Buddha Nala and Sutlej River)
- c. Zone-III (from the confluence of Buddha Nala and Sutlej River to the transboundary)
- d. Zone-IV (Sutlej River along the transboundary)

There were 5 samples in Zone-1, 9 samples in Zone-II, 11 samples in Zone-III, and 22 samples in Zone-IV.

Metal contamination indices

The metal contamination in sediments was determined using the following two indices:

Table 1 Descriptive statistics of metals (mg kg⁻¹) in sediments collected from the Sutlej River in Indian Punjab and their comparison with different guidelines

	Zn	Fe	Cu	Mn	Ni	Cr	Co	Cd	As
Min	10.0	1698	11.2	102	3.80	4.20	5.30	1.10	0.64
Max	64.0	2019	70.4	393	26.2	41.0	22.3	8.70	1.19
Mean	22.8	1848	32.2	190	11.1	16.9	11.4	2.86	1.02
S.E	1.41	10.7	1.36	9.46	0.65	0.93	0.47	0.17	0.01
C.V	54.3	5.07	37.2	43.6	51.2	47.9	36.7	53.4	10.3
ERL ^a	120	-	70	-	30	80	-	5	33
ERM ^a	270	-	390	-	50	145	-	9	85
TEL ^b	123	-	35.7	-	35	37.3	-	0.59	5.9
PEL ^b	315	-	197	-	91.3	90	-	3.53	17
Australian Interim Sediment Quality Guidelines ^c	26.4–100	11,500–62,800	6.3–55.6	232–3,260	4.5–92.9	8.3–118	3.2–38.6	0.02–0.16	0.5–5.7

^aEffect range low (ERL) and effect range medium (ERM) for fresh water ecosystems (Bai et al. 2011)

^bThreshold effect level (TEL) or probable effect level (PEL) for freshwater ecosystems (MacDonald et al. 2000)

^cDeCaritat et al. (2011)

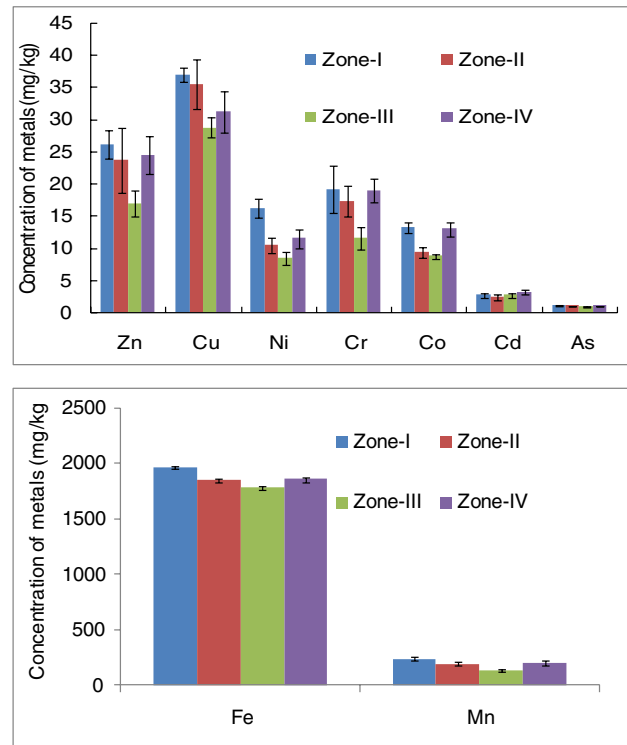


Fig. 2 Variations in metals concentration in sediments from different zones of Sutlej river in Indian Punjab

Contamination factor

The value of contamination factor (*CF*) is determined by dividing the concentration of each metal by its background value (Hakanson 1980). It was calculated using the following equation:

Table 2 Correlation among heavy metals in the sediments of Sutlej River

	Zn	Fe	Cu	Mn	Ni	Cr	Co	Cd	As
Zn	1.000	0.745	0.153	0.747	0.805	0.445	0.701	0.051	0.064
Fe	0.745	1.000	0.213	0.920	0.929	0.685	0.824	0.289	0.186
Cu	0.153	0.213	1.000	0.155	0.152	0.185	0.082	0.170	0.024
Mn	0.747	0.920	0.155	1.000	0.878	0.669	0.877	0.227	0.230
Ni	0.805	0.929	0.152	0.878	1.000	0.660	0.867	0.191	0.006
Cr	0.445	0.685	0.185	0.669	0.660	1.000	0.679	0.283	-0.032
Co	0.701	0.824	0.082	0.877	0.867	0.679	1.000	0.072	0.172
Cd	0.051	0.289	0.170	0.227	0.191	0.283	0.072	1.000	0.126
As	0.064	0.186	0.024	0.230	0.006	-0.032	0.172	0.126	1.000

The numbers in bold indicate significant correlation at $p < 0.05$

$$CF = \frac{\text{Concentration of each metal}}{\text{Background value of each metal}}$$

Enrichment factor

Enrichment factor (*EF*) is applied to determine the human and natural factors responsible for increasing the metal concentration (Delgado et al. 2010). Iron (Fe) was used as the reference element, and it is commonly used for normalization in many studies (Haghnazar et al. 2021; Setia et al. 2020; Setia et al. 2021). The *EF* was calculated using the following equation:

$$EF = \frac{\text{Concentration of metal in samples/Fe concentration in the samples}}{\text{Background values of metal/Background value of Fe}}$$

The background values were taken from Taylor and Mclennan (1995). The grades used to quantify contamination based on *CF* and *EF* values were given in Supplementary Table S1.

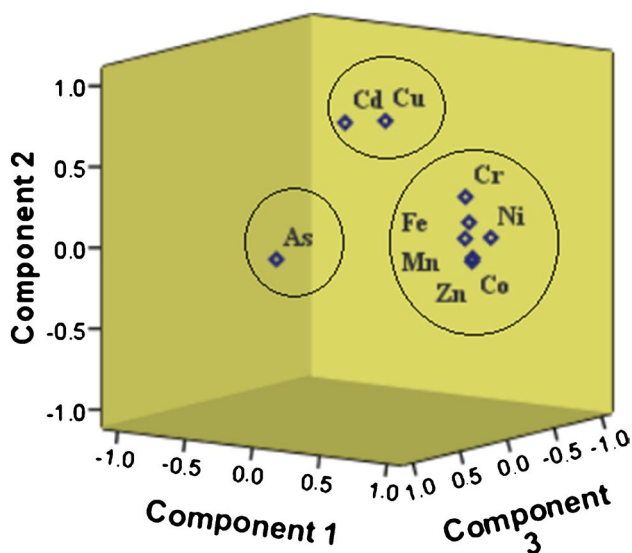
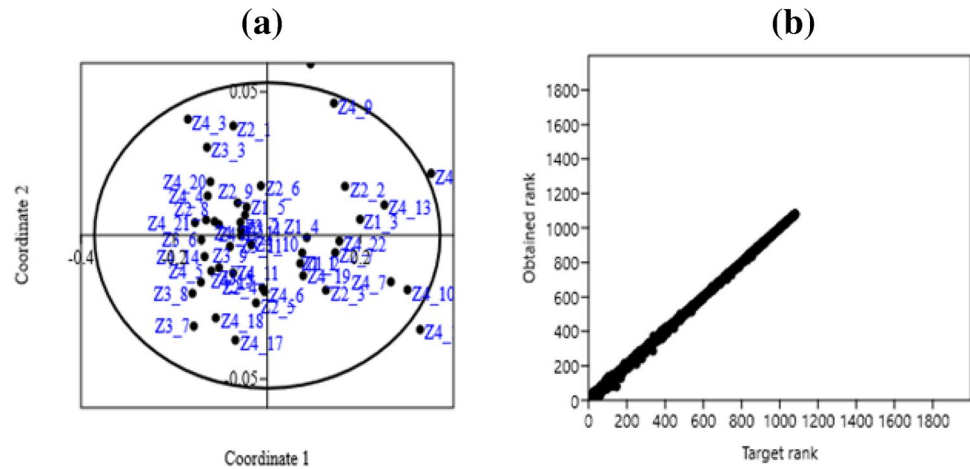


Fig. 3 PCA loading plots

Table 3 Principal component loadings of metals in the sediments of Sutlej River

Component	Initial Eigenvalues			Extraction Sums of Squared Loadings			Rotation Sums of Squared Loadings		
	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %
1	4.95	55.06	55.06	4.95	55.06	55.06	4.76	52.97	52.97
2	1.16	12.92	67.99	1.16	12.92	67.99	1.30	14.46	67.44
3	1.04	11.58	79.57	1.04	11.58	79.57	1.09	12.13	79.57
	Component matrix			Rotated component matrix			PC3		
	PC1	PC2	PC3	PC1	PC2	PC3			
Zn	.816	-.219	.006	.844	-.029	-.012			
Fe	.959	.045	.033	.925	.223	.132			
Cu	.224	.555	-.434	.092	.718	-.151			
Mn	.953	-.016	.120	.933	.132	.186			
Ni	.951	-.140	-.083	.958	.105	-.050			
Cr	.762	.076	-.243	.725	.325	-.124			
Co	.911	-.183	.111	.930	-.022	.106			
Cd	.267	.764	-.102	.089	.775	.240			
As	.158	.402	.866	.070	.030	.965			

Fig. 4 NMDS scatter plot (a) and Shepard 2-D plot (b) of sediment sampling sites from Sutlej river on the basis of heavy metals concentration using correlation as similarity measure with stress level 0.014 and R^2 for axis 1 = 0.914 and axis 2 = 0.0041



Ecological risk assessment due to heavy metals in sediments

The ecological hazard in the sediments was determined by applying potential and modified ecological risk indices. The ecological risk index (*RI*) was computed using *CF*, while modified ecological risk index (*MRI*) using *EF* values (Hakanson 1980). The following equations were used to calculate *RI* and *MRI*:

$$RI = \sum Er = \sum CF \times Tr$$

$$MRI = \sum mEr = \sum EF \times Tr$$

RIEr and *mEr* is the potential and modified potential ecological risk of each heavy metal, respectively. *Tr* is the toxic response factor of each metal taken from Kumar et al. (2020b). The grades used to categorize the ecological threat are provided in Supplementary Table S2.

The spatial patterns in *RI* and *MRI* in sediment samples were studied using inverse distance weighted method in ArcGIS 10.4.

Statistical analyses

The metal concentration was analyzed for minimum, maximum, mean, standard error, and coefficient of variation by using PAST v. 3.21. One-way ANOVA was performed to find the differences in metal concentration of the four zones using PAST v. 3.21. Pearson's correlation analysis was used to study the inter-relationships among metals in sediments and principal component analysis (PCA) was applied to find the sources of different metals. Non-metric multidimensional scaling (NMDS) was performed to find the similarity among sampling locations. The correlation analysis, PCA, and NMDS were performed using PAST v. 3.21.

Results and discussion

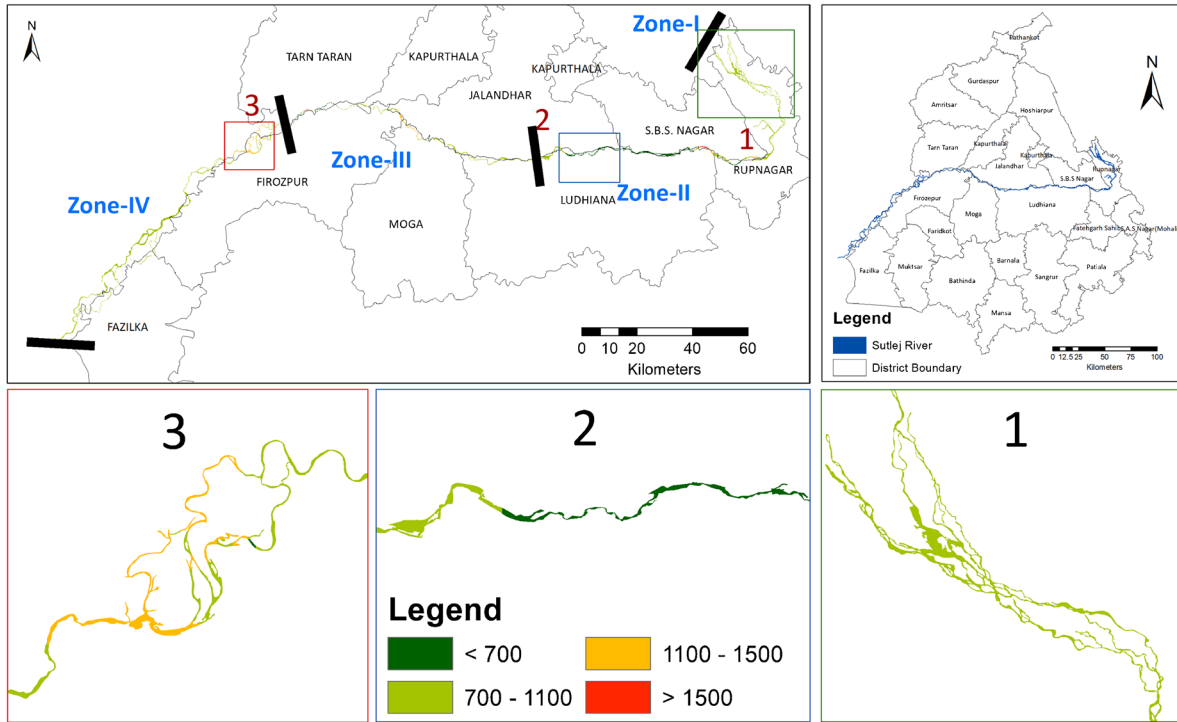
Descriptive statistics of metal concentration in sediments

The minimum, maximum, mean, standard error, and coefficient of variation of the metals (Zn, As, Mn, Cd, Cr, Co, Cu, Fe, and Ni) analyzed in sediments are given in Table 1. The mean concentration of metals was in the order: Fe > Mn > Cu > Zn > Cr > Co > Ni > Cd > As. A comparison of the mean concentration of metals in sediments with effect range low (ERL), effect range medium (ERM) (Bai et al. 2011), and probable effect level (PEL) (MacDonald et al. 2000) for the freshwater ecosystem, it was found that the average concentration of metals in sediments was lower than the threshold values defined by MacDonald et al. (2000). In comparison with threshold effect level (TEL) for the freshwater ecosystem, it was found that the concentration of all the metals in sediments was lower except for Cd. When the mean concentration of metals was compared with Australian sediment quality guidelines, only Cd concentration in sediments exceeded the threshold values (de Caritat et al. 2011). The highest coefficient of variation was found for Cd followed by Zn and Ni signifying that these metals' distribution in the sediments is more heterogeneous than the other metals. There are mainly electroplating and dyeing units in the catchment area of the river and many Cd, Zn, and Ni compounds are used in these units. Therefore, anthropogenic and industrial activities may be the sources of the large variations in Cd, Zn, and Ni concentration in sediments of the river (Cai et al. 2015).

The metal concentration in the four zones of Sutlej River showed the metal concentration was significantly ($p < 0.05$) higher in Zone-I followed by Zone-IV, Zone-II, and Zone-III (Fig. 2). The higher concentration of metals in Zone-I may be due to geogenic sources and little human activity in

(a)

Risk Index (RI)



(b)

Modified Risk Index (MRI)

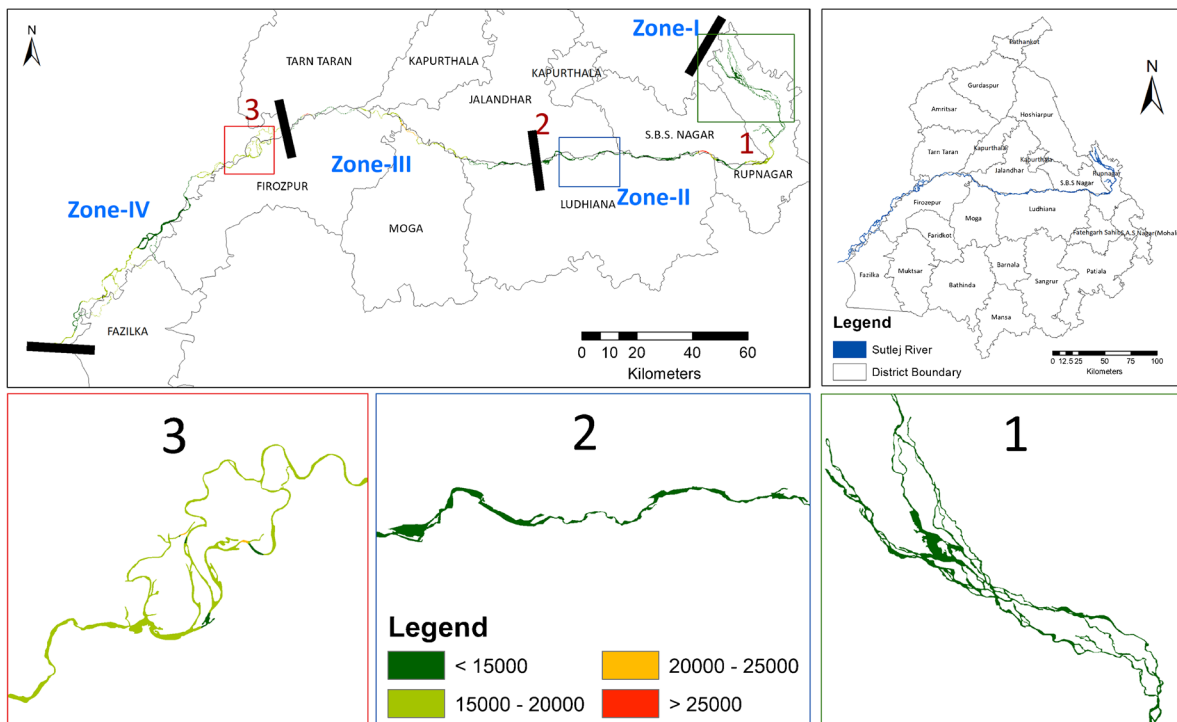


Fig. 5 Spatial variations in (a) potential risk index and (b) modified potential risk index of metals in sediments collected from different zones of Sutlej river in Indian Punjab

this zone. In contrast, the contaminants from industry, sewage sludge from domestic wastes, fertilizers, and pesticides may be the sources of contaminants in Zones II, III, and IV. Setia et al. (2020) found that the concentration of metals in water of the Gobind Sagar lake (Zone-I) was lower than transboundary of the Sutlej river (Zone-IV).

Correlation among metal concentration in sediments

There was a positive correlation among metals showing similar source for metal concentration in sediments of Sutlej River (Table 2). Among the metals, the correlation coefficient was significant and positive among Zn, Fe, Mn, Ni, Co, and Cr (Table 2). Marrugo-Negrete et al. (2017), Patel et al. (2018), and Yan et al. (2018) also found that the agronomic practices (like fertilizers and pesticides) are responsible for these metals in the river’s sediments besides geogenic and parent rock material. Previous studies have also found similar correlations among metals in sediments of the river (Alghobar and Suresha 2015; Aschale et al. 2017; Duodu et al. 2016).

Source apportionment of metals in sediments

The source apportionment of metals in sediments was studied using PCA (Fig. 3). The first three components showed 79.4% (53.0%, 14.4%, and 12.1%) of the variance with eigenvalues greater than 1 (Table 3). PC1 is dominated by

Fe, Mn, Zn, Co, Ni, and Cr indicating the contribution of geogenic and antropogenic factors (industrial and domestic wastes) to this PC (Figs. 4). Tian et al. (2017) also found that these metals are also originated from geogenic origin. PC2 is controlled by Cd and Cu (Table 3) which are mainly added through fertilizers (Tian et al. 2017). PC3 is controlled by As, and the natural processes are responsible for this PC.

Non-metric multidimensional scaling (NDMS) ranks alterations among the metals in multifaceted space (2- or 3-D space) using Euclidean distance. The NMDS scatter plot (Fig. 4) with 95% eclipse showed that all the metals are included in the eclipse and it indicates that all the samples are of similar origin except only two samples of the Zone-IV. This location-specific sample was taken from the point where there was a confluence of the highly contaminated local drain with the river. The results showed a stress of 0.01, signifying that data fit well to the model (Kaur et al. 2018).

Assessment of risks due to metals in sediments of Sutlej river

The maximum CF value was found for As, whereas the minimum for Cu (Table 4).

The CF values were classified into four grades based on the categories suggested by Hakanson (1980): $CF < 1$ indicates low, $1 \leq CF < 3$ moderate, $3 \leq CF < 6$ considerable, and $CF \geq 6$ high contamination. It was found that CF values for Cu, Zn, and Ni indicated low contamination in the study area, but its value for As indicated the high contamination. The CF values of Zn and Mn showed no contamination. The CF results of Cu, Ni, Cr, and Co indicated that 68%, 10.6%, 4.2%, and 46.8% samples had moderate contamination,

Table 4 Contamination factor (CF), enrichment factor (EF), potential (Er) and modified potential ecological risk (mEr) of metals in the sediments collected from the Sutlej River

	Zn	Cu	Mn	Ni	Cr	Co	Cd	As
CF								
Minimum	0.14	0.45	0.19	0.19	0.12	0.53	11.2	6.56
Maximum	0.90	2.82	0.73	1.31	1.17	2.23	88.8	12.1
Mean	0.32	1.29	0.35	0.56	0.49	1.14	29.1	10.4
EF								
Minimum	2.48	7.58	3.35	3.44	2.17	9.14	204	7.61
Maximum	13.9	45.1	11.1	20.3	18.0	34.2	1407	12.8
Mean	5.28	21.5	5.79	9.09	8.00	18.7	486	11.4
Er								
Minimum	0.14	2.24	0.19	0.95	0.24	2.65	337	13.1
Maximum	0.90	14.1	0.73	6.55	2.34	11.2	2663	24.3
Mean	0.32	6.43	0.35	2.78	0.97	5.68	874	20.9
mEr								
Minimum	2.48	37.9	3.35	17.2	4.35	45.7	6126	15.2
Maximum	13.9	225	11.1	101	35.9	171	42,232	25.7
Mean	5.28	107	5.79	45.5	16.0	93.7	14,590	22.8

respectively. The *EF* values (Table 4) showed that Cd is the primary contaminant in the study area and all the values had extreme enrichment. The *EF* values of Zn, Cu, Mn, Ni, Cr, Co, and As showed substantial enrichment in 34%, 38.2%, 100%, 100%, 100%, 65.9%, and 100% of the sediment samples collected from the river, respectively. Furthermore, 34% and 59.5% of the samples showed high enrichment of Co and Cu. The values of *CF* and *EF* in the present study are within the ranges of previous studies. For example, Ali et al. (2016) found that the average *CF* values of As, Cd, Cr, and Pb were 1.56, 0.40, 6.60, and 2.16 in the water and sediments of Karnaphuli River (Bangladesh), respectively. Mandeg et al. (2019) found that *CF* values of Cd, Pb, Cu, Ni, and Zn were 0.38, 0.30, 0.59, 1.86, and 0.51 in the Abiete-Toko gold district (Southern Cameroon), respectively.

Ecological risk assessment

The ecological risk assessment in the sediments of Sutlej River was determined from the ecological risk index (*RI*) and modified potential ecological risk index (*MRI*). The results of contamination factor (*CF*), enrichment factor (*EF*), potential (*Er*), and modified potential ecological risk of a metal (*mEr*) are given in Table 4. The potential ecological threat due to metals (*Er*) showed a low risk in the area due to Co, Ni, Cu, Zn, Fe, Mn, and Ni. The *Er* values of Cd for all the samples showed a very high ecological threat, whereas 23.4%, 44.6%, 21.2%, and 8.5% of the sediment samples showed low, modest, significant, and high risk due to As, respectively. The *RI* results showed a considerable risk in 27.6% of the samples and high risk in 78.7% of the sediment samples.

The *mEr* results showed the modest, significant and high risk in 34%, 59.5%, and 6.3% of the samples due to Co in sediments of the river, respectively. However, the *mEr* values of Ni indicated a modest and considerable risk in 46.8% and 8.5% of the samples, respectively. It was also found that modest, significant, and high risk due to Cu was found in 23.4%, 65.9%, and 8.5% of the sediment samples, respectively. The *MRI* results showed that all the samples exhibited very severe risk of metals in the sediments of the Sutlej River. The *mEr* values of Cd showed very high risk. Our results on *mEr* values on Cd find support from previous studies in India and different parts of the world (Abdullah et al. 2015; Kumar et al. 2020a; Soliman et al. 2015; Yin et al. 2011). They also reported modest to extreme threat in the sediments due to Cd.

Spatial distribution of *RI* and *MRI* (Fig. 5) showed that metal contamination was higher along transboundary of the river followed by the sites from the confluence of contaminated drain with the river to transboundary and localized spots in the river flowing in Indian Punjab (Fig. 5).

Conclusions

This study showed that only Cd concentration in sediments exceeded the TEL and Australian sediment quality guidelines. The correlation analysis, CA and PCA suggested that geogenic aspects, agronomic, and mixed geogenic and agronomic aspects contributed to the concentration of metals in the sediments. The *CF*, *EF*, *Er*, and *mEr* values indicated that Cd contributed towards contamination in sediments and an ecological threat. Since the importance of sediment management and water quality are recognized, the findings of this work may be applied to assess the spots with high contamination which require taking strategic management measures to improve the water quality of the river.

Supplementary Information The online version contains supplementary material available at <https://doi.org/10.1007/s12517-021-08524-y>.

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Declarations

Conflict of interest The author(s) declare that they have no competing interests.

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