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RESEARCH ARTICLE

Occurrence, distribution, and pollution indices of potentially toxic elements within the bed sediments of the riverine system in Pakistan

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Abstract

Potentially toxic elements (PTEs) are a major source of pollution due to their toxicity, persistence, and bio-accumulating nature in riverine bed sediments. The sediment, as the largest storage and source of PTEs, plays an important role in transformation of mercury (Hg), lead (Pb), nickel (Ni), chromium (Cr), copper (Cu), zinc (Zn), and other toxic PTEs. Several important industrial hubs that contain a large population along the banks of different rivers, such as Kabul, Sutlej, Ravi, Jhelum, and Chenab in Pakistan, are acting as major sources of PTEs. In this study, 150 bed sediment samples (n=30 from each river) were collected from different sites. Total (acid extracted) PTE (Hg, Cu, Cr, Ni, Zn, and Pb) concentrations in bed sediments were determined using inductively coupled plasma mass spectrometry (ICP-MS). Sediment pollution indices were calculated in the major rivers of Pakistan. The results demonstrated high levels of Hg and Ni concentrations which exceeded the guideline standards of river authorities in the world. The contamination factor (CF) and contamination degree (CD) indices for Hg, Ni, and Pb showed a moderate to high (CF≥6 and CD≥24) contamination level in all the selected rivers. The values of geo-accumulation index (I_{geo}) were also high (I_{geo}≥5) for Hg and Pb and heavily polluted for Ni, while Cr, Cu, and Zn showed low to unpolluted (I_{geo}) values. Similarly, the enrichment factor (EF) values were moderately severe (5≤EF≤10) for Hg, Pb, and Ni in Sutlej, Ravi, and Jhelum, and severe (10≤EF≤25) in Kabul and Jhelum. Moreover, Hg and Ni showed severe to very severe enrichment in all the sampling sites. The ecological risk index (ERI) values represented considerable, moderate, and low risks, respectively, for Hg (The ERI value should not be bold. Please unbold the ERI in the whole paper. It should be same like RI, CD and EF. 80≥ERI ≥160), Pb and Ni (40≤ERI ≤80), and Cr, Cu, and Zn (ERI ≤40). Similarly, potential ecological risk index (PERI) values posed considerable (300≤RI≤600) risk in Ravi and moderate (150≤RI≤300) in Kabul and Jhelum, but low (RI≤150) risk in Ravi and Chenab. On the basis of the abovementioned results, it is concluded that bed sediment pollution can be dangerous for both ecological resources and human beings. Therefore, PTE contamination should be regularly monitored and a cost-effective and environmentally friendly wastewater treatment plant should be installed to ensure removal of PTEs before the discharge of effluents into the freshwater ecosystems.

Keywords Potentially toxic elements · Major rivers · Bed sediments · Contamination · Ecological risk assessment

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Introduction

In recent decades, the riverine ecosystem has experienced serious threats from human activities such as industrial effluents, intensive agricultural practices, metropolitan waste management problems, and increase in urbanization (Meijide et al. 2018). Due to urbanization and rapid development in the industrial sector, potentially toxic element (PTE) contamination has drawn tremendous attention due to its adverse effects on human health and aquatic ecosystem (Krasnići et al. 2013; Jordanova et al. 2018; Ullah et al. 2019). Among the hazardous substances that enter aquatic ecosystems, PTEs have been enlisted as inorganic contaminants due to their persistent nature, toxicity, and their ability to transfer into the food chain, resulting in serious risk to human health and aquatic biota and causing a major health concern worldwide (Franco-Uría et al. 2009; Machado et al. 2017). PTEs are usually distributed in aquatic environment in different forms such as colloids, suspended, water-soluble species, and deposited segments. In some conditions, more than 99% of PTEs entering a river can be stored in river sediments in various forms (Salomons and Stigliani 2012). Sediment is a good indicator of PTEs in aquatic ecosystems because of its tendency to accumulate and hydrophobicity (Łuczyńska et al. 2018).

PTE pollution generates from both anthropogenic and natural sources; anthropogenic sources include mining wastewater, domestic sewage, coal burning, industrial wastewater, and agricultural fertilizer leachate, while natural sources include soil erosion, rock weathering, and volcanic activities (Pan and Wang 2012; Skordas et al. 2015; Nawab et al. 2015, 2016, 2017; Rashid et al. 2019; Liu et al. 2020). Over the past several decades, the main source of PTEs in the environment are human activities including rapid industrialization and urbanization (Islam et al. 2015a, 2015b). In river ecosystems, sediments are the main repository and source of PTEs (Superville et al. 2014; Wang et al. 2018). These PTEs enter the water through different pathways such as soil erosion, atmospheric deposition, scouring, wastewater discharge, leaching, and runoff (Yi et al. 2011).

The concentrations of these toxic PTEs are very high in sediments as compared to water column due to its deposition in the bottom of the rivers (Sultan and Shazili 2009; He et al. 2009; Nobi et al. 2010; Rezayi et al. 2011; Liu et al. 2019). These PTEs are toxic, persistent, and bio-accumulative in nature, while some of the PTEs which are carcinogenic in nature can bio-magnify and bio-accumulate in seafood (oysters, shrimps, mussels, fish) and through various pathways can be transferred to humans (Rahman et al. 2013; Fang et al. 2014). After the discharge of PTEs into river systems, contamination can be distributed in different components of the river systems such as biota, water, and sediments (Maanan et al. 2015; Ali et al. 2016). Consequently, a majority of PTEs are deposited in the sediments and only a small amount remains in the water column (Malvandi 2017).

River sediments are very suitable for manmade pollution monitoring because they do not act only as pollutants carriers, but also has the potential sources of secondary pollutants in the water system (Mekonnen et al. 2015; Ma et al. 2016). When these PTEs enter water systems, they become very dangerous to water ecosystems through a range of physiological and biochemical processes. PTEs can accumulate in river sediments and about 85% of PTEs accumulate in surface sediments (Bosch et al. 2016; Zhang et al. 2016). More specifically, these PTEs bind to sediments through multiple mechanisms, including ion exchange, co-precipitation, particle surface adsorption, and complexation with organic matter (Passos et al. 2010; Dong et al. 2014). Furthermore, some of the PTEs bound to sediments can be released into river water through desorption reactions, sediment resuspension, oxidation, or reduction reactions (Dong et al. 2012; Zhao et al. 2013).

After entering the water ecosystem, only a very small portion of free PTE ions stay dissolved in water as particularities, and the rest of the PTEs get deposited in the river sediments (Varol 2011; Zhuang and Gao 2015). This causes PTE pollution in river water systems, which results in a significant threat to water quality, as well as to the food chain stability and the whole ecosystem. Sediments can have a double role (i.e., source and sink) in PTE exchange with river water (Chen et al. 2018). Thus, for characterizing the influence of anthropogenic activities and natural sources, sediment quality serves as a useful parameter; additionally, the quality of sediments can provide anthropogenic effects evidence on ecosystems and provide management of policy for the surrounding areas (Wang et al. 2014; Xu et al. 2014).

Pakistan has been gifted with enough surface and groundwater resources but the main contributors to ground and surface water pollution include textile, fertilizers, mining, metals, pesticides, and others industrial chemicals (PCRWR n.d.). Water and sanitation agencies in Pakistan are mainly focusing on water quantity rather than water quality because of the increasing requirement rate. All these are related due to a lack of treatment technologies, awareness, trained personal, and quality monitoring (Aziz 2005). The polluted water discharge from municipal sewage, urban wastes, farms, and industries is carried by drain pipes and canals to river systems where it increases and deteriorates the water quality (Haq 2005; Tariq et al. 2006). Therefore, the river sediment analysis is a very suitable method to investigate PTE contamination in an area (Thuong et al. 2013; Islam et al. 2015b). The aim of this study is to provide useful information on the distribution of PTE pollution in the five main rivers (Kabul, Sutlej, Ravi, Jhelum, and Chenab) of Pakistan. In the current research work, an effort has been made to establish the role of bed sediments as indicators for evaluating the level of PTE contamination. The degree of pollution has been determined and relative mobility of different PTEs has been presented in this study. Moreover, a considerable number of different indexes have been developed to assess PTE pollution levels, source appointment, enrichment factor (EF), and geo-accumulation index (I_{geo}) (Müller 1979; Feng et al. 2011). Therefore, it is necessary to investigate the distribution of PTEs in bed sediments of the major rivers in Pakistan and assess the risk caused by these PTEs to protect the corresponding aquatic ecosystems.

Materials and methods

Study area

Figure 1 represents the detail map of the major rivers (Chenab, Jhelum, Ravi, Sutlej, and Kabul) across Pakistan. River Kabul (34.1734° N, 71.5576° E) is approximately 700 km long and it emerges from the Hindu Kush Mountains Range of Afghanistan into Pakistan through the northern part of the Khyber Pass River Kabul joining the Upper Indus at Attock. The major tributaries of the Kabul River are the Logar, Panjshir, Alingar, Surkhab, Kunar, Bara, and Swat Rivers. The river Sutlej (N 30° 57' 26.838, E 74° 25' 55.362) is one of the longest rivers that flow through Punjab in northern India and Pakistan. It is located east of the Central Suleman Range in Pakistan. The water level of river Sutlej is on a decreasing rate. Wastewater is discharging to the river Sutlej from different anthropogenic activities along the riverbank sides. In Pakistan, the major sources of water contamination include both industrial and municipal effluents, solid wastes, and agricultural runoff (Azizullah et al. 2011; Ayesha 2012). Nearby, 90% of municipal and industrial wastes in Pakistan are dumped into open water bodies and reach the underground aquifers (Mustafa et al. 2013). Only 8% of urban sewage in the country is treated and the remaining is disposed untreated and released to freshwater bodies (Pakistan Economic Survey 2013).

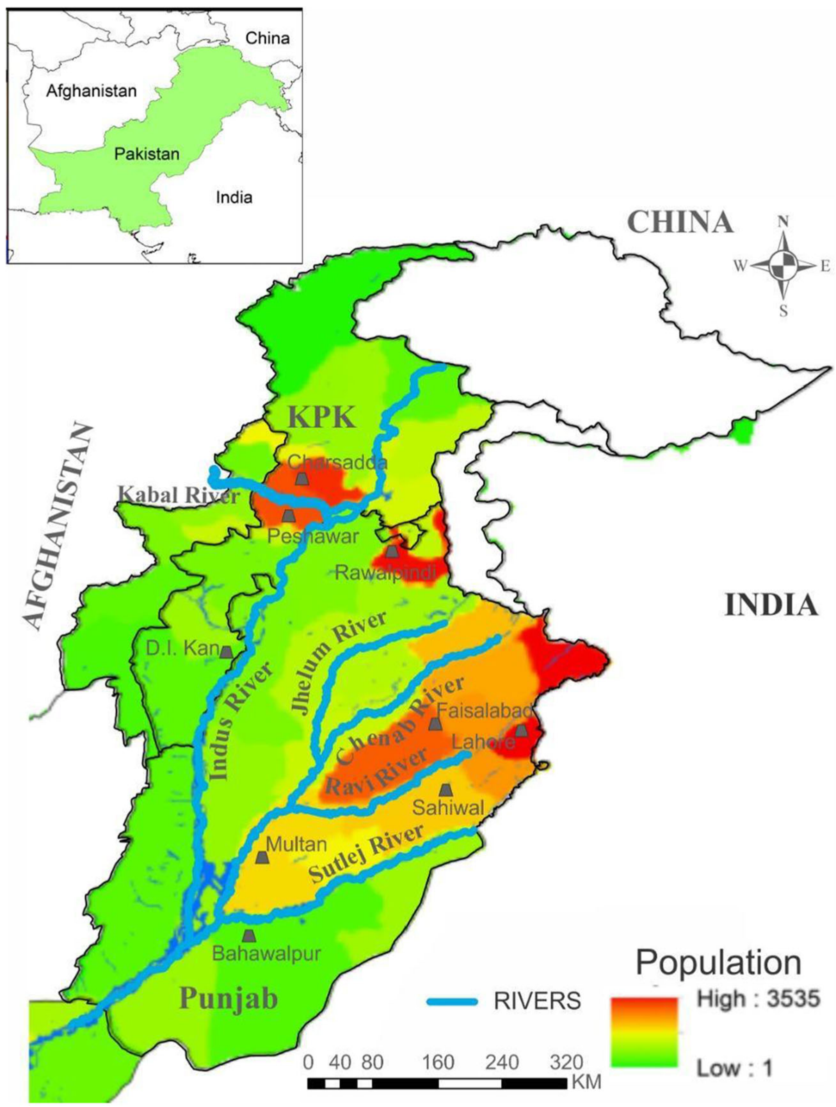

Fig. 1 Location map of the study area showing the major rivers of Pakistan

The Ravi River (N 31° 29' 2.2272, E 74° 9' 46.4292) is the transboundary river between India and Pakistan. It is an integral part of Indus river basin and forms the Indus basin headwaters. The Jhelum River (N 31°11'60.00, E 72°07'60.00) is a transboundary river in northern India and eastern Pakistan. It is the westernmost of the five rivers of the Punjab region, and passes through the Kashmir Valley. The river Chenab (N 32° 29' 50.0028, E 74° 32' 9.9960), which originates from the Himachel Pardesh in India, is another major river in Pakistan. Its total length is 1240 km and its course travels across Jammu Province into Pakistan's Sialkot District and Punjab Province, and then flows into the Indus. All the five rivers are the backbone of Pakistan irrigation, domestic, and drinking water supply system.

Sample collection and preparation

Samples were collected from the five rivers in August 2018 and their proper locations are shown in Fig. 1. The sample collection points were based upon densely populated areas. A total of 150 composite samples were collected from five rivers (n=30 from each river). Each composite sample consisted of three subsamples, and every subsample was collected about 150 m (approximately) from each other. The sediment samples were collected by a Petersen's grab with the depth of 0–15 cm. A GPS device was used for sample locations and coordinates. After collection, the sediment samples were stored in clean polyethylene zip bags and then were transferred immediately to the laboratory for further process. The sediment samples were air dried and then sieved with a 0.71-mm sieve to remove plant fragments, stones, and other debris from the samples. The samples were then ground and passed through a 0.20-mm nylon mesh. Finally, all the samples were then stored in the dark at 4°C before further analyses (Liu et al. 2007).

Analysis of sediment samples

In this study, the bed sediment samples were acid extracted for PTEs according to the method mentioned in the paper published by Nawab et al. (2018). The sieved sediment samples were weighted (0.5 g) into Teflon beakers. For acid digestion, 3 ml of concentrated hydrofluoric acid (HF) and 9 ml of ultra-pure nitric acid (HNO_3) in 1:3 were used and heated up to 120°C for 15 to 30 min on a hot plate. After that, the samples were cooled to 40°C and then 5 ml (HF), 5 ml (HNO_3), and 3 ml ($HClO_4$) were added and then the samples were heated on 160 °C for 1 h with a tight cap. After 1 h, the samples were heated at 140°C for another 1 h with an open cap. All the chemicals used in this study were of analytical grade (Merck).

Quality control and assurance

For accuracy and precision, at least one blank reagent, reference material, and sample replicate were used in each digestion batch to check for method efficiency and consistency. For quality control, standard reference material of sediments (GBW07314) was purchased from National Standard Material Center China. Throughout the analysis, blank experiments were also performed to validate the accuracy of the sediment extraction process. After digestion, the samples were then allowed to cool down and filtered with 0.45-μm filter paper. After filtration, the samples were diluted with DDW to 100 ml in a volumetric flask for PTE analysis. In the next step, ICP-MS (7500CX, Agilent Technologies, USA) was used to determine the concentrations of these six PTEs under proper quality control and assurance procedures in the Institute of Urban Environment, Chinese Academy of Sciences, Xiamen, China. All the samples in the study were analyzed in duplicate and the recoveries for the selected elements were 95 to 108%.

Contamination factor

The contamination factor (CF) technique is used to assess the degree of PTE contamination in sediment with respect to PTEs in the reference sediment (Mmolawa et al. 2010). The CF was calculated using the following Eq. (1):

$$CF = \frac{C_m \text{ sample}}{C_m \text{ reference}} \tag{1}$$

Where, C_m sample is the measured concentration of an element in the sediment sample and C_m is the concentration of the corresponding PTE in the reference sediment. The reference values for each metal were calculated using SEPA (1998) reference values in this study. The classified values for contamination factor are given in Table 1.

Geo-accumulation index

Geo-accumulation index (I_{geo}) is widely used to assess PTE contamination in the sediments (Muller 1969; Varol 2011). This method is helpful in evaluating the enrichment of PTEs in the sediments above baseline values. The I_{geo} values were calculated using Eq. (2):

$$I_{geo} = \log_2\left[\frac{C_m}{1.5B_n}\right] \tag{2}$$

Where C_m is the measured PTE concentration in the sediment sample and B_n is the geochemical reference value for metals of C_n in the reference values (Muller 1969; Yu et al. 2011). Due to lithogenic effects, the factor 1.5 is introduced to minimize the variation in the reference values. The classified values for geo-accumulation index in sediment are given in Table 1.

Table 1 Pollution indices of sediment quality and their classification used in the study area

Index							
Contamination factor (CF)[a,b,c]		Contamination degree (CD)[a,c]		Ecological risk index (ERI)[a,b,c]		Potential ecological risk (PERI)[a,b,c]	
CF value	Contamination level	CD values	Contamination level	ERI values	Ecological risk	RI values	Potential risk
CF < 1	Low	CD < 6	Low	ERI < 40	Low	RI<150	Low
1 < CF < 3	Moderate	6 < CD < 12	Moderate	40 ≤ ERI < 80	Moderate	150≤RI<300	Moderate
3 < CF < 6	Considerable	12 < CD < 24	Considerable	(80 ≤ ERI < 160	Considerable	300≤RI<600	Considerable
CF > 6	Very high	CD > 24	High	160 ≤ ERI < 320	High	RI>600	Very high
-	-	-	-	ERI ≥ 320	Very high	-	-
Enrichment factor (EF)[a,b,c]				Geo-accumulation (I_{geo})[a,b,c]			
Class	EF value	Contamination values		Class	I_{geo} value	Contamination values	
0	EF < 1	No enrichment		0	I_{geo}<0	Unpolluted	
1	1 < EF < 3	Minor		1	0≤I_{geo}<0	Unpolluted to moderate	
2	3 < EF < 5	Moderate		2	1 ≤ I_{geo}<2	Moderate	
3	5 < EF < 10	Moderately severe		3	2 ≤ I_{geo}< 3	Moderate to heavily	
4	10 < EF < 25	Severe		4	3 ≤ I_{geo}<4	Heavily	
5	25 < EF < 50	Very severe		5	4 ≤ I_{geo}<5	Heavily to extreme	
6	EF > 50	Extremely severe		6	I_{geo}>5	Extreme	

[a] Duodu et al. (2016)

[b] Malvandi (2017)

[C] Liu et al. (2020)

Contamination degree

The contamination degree (CD) represents the contamination index, which integrates total PTE sediment pollution in an area (Hakanson 1980). The contamination degree was calculated using the following Eq. (3):

$$CD = \sum_{i=1}^{n} CF \tag{3}$$

Where n is the number of PTEs and CF is the contamination factor for each heavy metal in the study area. The classified values for the degree of contamination are given in Table 1.

Enrichment factor

Enrichment factor (EF) is considered as an effective tool to evaluate PTEs in the environment (Hakanson 1980). The enrichment factor was calculated using Eq. (4):

$$EF = \frac{\left(C_n/C_{ref}\right)_{sample}}{\left(B_n/B_{ref}\right)_{background}} \tag{4}$$

Where, C_n (sample) is the measured concentration of an element and C_{ref} is the concentration of reference element for normalization, B_n is the concentration of metals in the crust, and B_{ref} is the concentration of the reference element used for normalization in the crust. Iron (Fe) value was used as the reference element at the crust in the study area. The classified values for the enrichment factor are given in Table 1.

Ecological risk index

The ecological risk index (ERI) techniques, as suggested by Hakanson (1980), were used for calculation which is considered an effective tool in environmental assessment of PTEs. The ecological risk index ERI was calculated using the following Eq. (5):

$$ERI = T_r^i \times C_f^i \tag{5}$$

Where T_r^i corresponds to the toxic response of PTEs, and C_f^i is the value obtained from the contamination factor for each metal. In the present study, the values used for T_r^i (toxic response) for each metal such as Hg, Ni, Cr, Cu, and Zn were 40, 5, 5, 2, 5, and 1, respectively (Hakanson 1980). The classified values for ERI are given in Table 1.

Potential ecological risk index

The potential ecological risk index (PERI) was calculated based on the sum of individual ecological risk index (ERI) in each river (Yuan et al. 2015). The PERI values were calculated using the following Eq. (6):

$$PERI = \sum_{i=1}^{n} ERI \tag{6}$$

Where n is the number of elements analyzed, and ERI is the individual element in each river. The classified values for ERI are given in Table 1.

Data analysis

Data analysis of the study was assessed using statistical software (statistic version 10) as a complementary software. Mean values (n=3 ± SD) and standard deviation were calculated using MS Excel 2016. Statistical analyses like principal component analysis (PCA) and Pearson's correlation were performed using XLSTAT version 2014 and Statistical Package for Social Sciences ver. 21 (SPSS Inc., Chicago, IL, USA).

Results and discussion

Potentially toxic elements in bed sediments

The total concentrations of Hg, Pb, Ni, Cr, Cu, and Zn in Kabul, Sutlej, Ravi, Jhelum, and Chenab sediments with mean, minimum, maximum, and standard deviation values are illustrated in Table 2. The concentrations of these six PTEs in the study area were compared with different international standards set for PTEs in sediments including the Swedish Environmental Protection Agency (SEPA 1998); Canadian Council of Ministers of the Environment (CCME (Canadian Council of Ministers of the Environment) 2002); National Oceanic and Atmospheric Administration (NOAA 2012); Taiwan EPA (2010); the Australian and New Zealand Environment and Conservation Council and Agriculture and Resource Management Council of Australia and New Zealand (Anzecc 2000); and China State Bureau of Quality and Technical Supervision (CSBTS 2002). The average concentration order of PTEs in study area was in the order of Zn>Ni>Pb>Cr>Cu>Hg. The concentrations of Hg, Pb, Ni, Cr, Cu, and Zn in river Kabul varied from 2.81–3.49, 4.32–26.27, 26.36–51.24, 33.67–47.54, 7.14–13.41, and 36.55–61.45 mg kg^{-1}, respectively. The mean values of the above metals in river Kabul were 3.17, 15.358, 39.8368, 45.934, 10.084, and 51.172 mg kg^{-1}, respectively. The PTE concentrations in the river Kabul were in the order of Zn>Cr>Ni>Pb>Cu>Hg.

The concentrations of Hg and Cr in the river Kabul were found to be higher as compared to all other countries' permissible limits as shown in Table 2. Furthermore, the concentrations of Cu and Zn were within the SEPA (1998), CCME (Canadian Council of Ministers of the Environment) (2002), NOAA (2012), Taiwan EPA (2010), Anzecc (2000), and CSBTS (2002) safe limits. However, only the concentration of Pb in the river Kabul exceeded SEPA (1998) standard values. Moreover, Cr concentration exceeded the SEPA (1998) limit, while Ni exceeded the SEPA (1998), NOAA (2012) and Taiwan EPA (2010), and Anzecc (2000) lower limits. Furthermore, the results of the river Kabul in comparison with those of Kayembe et al. (2018) showed similar results for Hg, Cr, Cu, and Ni, while Duodu et al.'s (2017) results were lower as compared to those for this study. The high concentration of Hg in the river sediments can be related to the untreated cosmetics industrial effluents, hospital drainage, municipal wastewater, and bleach used for cleaning purposes (Tshibanda et al. 2014; Kilunga et al. 2017). The long-term exposure to toxic PTEs may also be dangerous to the aquatic biota and local population (Álvaro et al. 2016).

In the river Sutlej, the concentrations of Hg, Pb, Ni, Cr, Cu, and Zn ranged from 1.23–1.46, 11.17–17.72, 11.41–27.54, 21.32–40.02, 22.41–76.02, and 111.86–132.57 mg kg^{-1}, respectively. The mean values of six PTEs in the river Sutlej were 1.36, 14.258, 20.51, 22.328, 37.98, and 123.486 mg kg^{-1} respectively as shown in Table 2. The PTE concentrations in river Sutlej were in the order of Zn>Cu>Cr>Ni>Pb>Hg, respectively. In the river Sutlej, the concentration of Hg was found to be higher than all the above countries' acceptable limits as shown in Table 2. The mean concentrations of Pb and Ni were found to be only higher than SEPA (1998) safe limits. The concentration of Cu in the river Sutlej was also found higher as compared to SEPA (1998), CCME (Canadian Council of Ministers of the Environment) (2002), NOAA (2012), and CSBTS (2002) lower limit standards.

Furthermore, the concentrations of Cr were within the listed countries' safe limits and Zn slightly exceeded those of SEPA (1998). The results of the study conducted by Tabinda et al. (2013) in the river Sutlej showed a slightly high concentration of Ni while Zn, Cu, and Cr were found to be lower than those of this study. The pollution of PTEs in sediment system may be released due to the overlying water due to rapid environmental changes, causing serious risk to biota and the environment (Lintern et al. 2016).

The concentrations of Hg, Pb, Ni, Cr, Cu, and Zn in the river Ravi varied from 1.76–2.02, 114.64–151.33, 150.39–186.74, 3.41–8.2, 2.65–7.65, and 129.64–188.22 mg kg^{-1} respectively. The mean values of all PTEs in Ravi River are 1.9, 130.658, 165.252, 16.124, 14.806, and 159.7 mg kg^{-1} as listed in Table 2.

The PTE concentrations in river Ravi were in the order of Ni>Zn>Pb>Cr>Cu>Hg. The concentration of Hg was also found to be higher as compared to that in rivers Kabul and Sutlej than the given countries' acceptable limits. The concentrations of Pb with mean values were found to be higher than all the listed countries' permissible limits, although the concentrations of Cr and Cu were within the listed countries' safe limits. Furthermore, the concentration of Ni was found to be

Table 2 Mean, ranges, and standard deviation of the selected PTEs (mg kg $^{-1}$) in the studied rivers with comparison to international standards

River	Statistics	Hg	Pb	Ni	Cr	Cu	Zn	References
Kabul	Range	2.81–3.49	4.32–26.27	26.36–51.24	33.67–47.54	7.14–13.41	36.55–61.45	This study
	Mean ± SD	3.17±0.31	15.35±9.04	39.83±11.26	45.93±7.31	10.08±2.49	51.17±9.77	
Sutlej	Range	1.23–1.46	11.17–17.72	11.41–27.54	21.32–40.02	22.41–76.02	111.86–132.57	
	Mean ± SD	1.36±0.09	14.25±3.08	20.51±6.96	22.32±7.47	37.98±22.19	123.48±8.41	
Ravi	Range	1.76–2.02	114.64–151.33	150.39–186.74	3.41–8.2	2.65–7.65	129.64–188.22	
	Mean ± SD	1.9±0.11	130.65±15.31	165.25±15.25	6.12±1.85	4.80±2.11	159.7±23.11	
Jhelum	Range	1.32–2.64	110.4–133.47	8.01–12.7	11.52–22.12	37.42–64.4	138.27–196.43	
	Mean ± SD	2.48±0.53	122.12±9.77	10.42±1.73	17.77±4.39	55.27±13.76	161.26±24.24	
Chenab	Range	0.45–1.02	31.16–47.32	101.16–134.19	47.87–66.41	12.54–69.32	67.42–116.14	
	Mean ± SD	0.66±0.22	38.91±6.39	120.57±12.57	57.07±7.04	29.49±22.96	69.94±23.46	
Metal background guidelines	S. limits	0.8	5	10	160	15	100	SEPA 1998
Canadian sediment quality guidelines (TEL)	S. limits	0.13	30.2	*	37.3	18.7	124	CCME (Canadian Council of Ministers of the Environment) 2002
Canadian sediment quality guidelines (PEL)	S. limits	0.7	112	*	90	108	271	
Sediment quality guidelines (ERL)	S. limits	0.15	46.7	20.9	81	34	150	NOAA 2012
Sediment quality guidelines (ERM)	S. limits	0.71	218	51.6	370	270	410	
Taiwan EPA (upper limit)	S. limits	0.87	161	80	233	157	384	Taiwan EPA 2010
Taiwan EPA (lower limit)	S. limits	0.23	48	24	76	50	140	
ANZECC and ARMCANZ (low)	S. limits	0.15	50	21	80	65	200	Anzecc 2000
ANZECC and ARMCANZ (high)	S. limits	1	220	52	370	270	410	
Marine sediment quality (I)	S. limits	0.2	60	*	80	35	150	CSBTS 2002
Marine sediment quality (II)	S. limits	0.5	130	*	150	100	350	

*Not available, Swedish Environmental Protection Agency (SEPA 1998), Canadian Council of Ministers of the Environment (CCME 2002), National Oceanic and Atmospheric Administration (NOAA 2012), Taiwan EPA (2010), the Australian and New Zealand Environment and Conservation Council and Agriculture and Resource Management Council of Australia and New Zealand (ANZECC and ARMCANZ 2000), and China State Bureau of Quality and Technical Supervision (CSBTS 2002) higher than the given countries' allowable limits and Zn was found to be greater than SEPA (1998), CCME (Canadian Council of Ministers of the Environment) (2002), and NOAA (2012) permissible limits and lower than that of CSBTS (2002).

In comparison to Rauf et al. (2009), the Cr and Cu concentration showed similar results. The results of Javed (2005) stated higher concentrations of Zn, Pb, and Ni in the river Ravi as compared to those of this study. Clay and silt are mostly responsible for the absorbent of PTEs in river sediments (Sundaray et al. 2011). The content of organic matter can also increase the PTE adsorption on the surface sediments (Liang et al. 2018). In addition, the smaller size of grain indicates a higher concentration of metals including Cd, Pb, Cu, Ni, Zn, and Cr (Zhao et al. 2010).

In the river Jhelum, the concentrations of Hg, Pb, Ni, Cr, Cu, and Zn ranged from 1.32–2.64, 110.4–133.47, 8.01–12.7, 11.52–22.12, 37.42–64.4, and 138.27–196.43 mg kg^{-1} respectively. The mean concentration values of all PTEs in the river Jhelum are 2.48, 122.128, 10.428, 17.772, 55.276, and 161.264 mg kg^{-1} as summarized in Table 2. The PTE concentrations in the river Jhelum were in the order of Zn>Pb>Cu>Cr>Ni>Hg respectively. Furthermore, in the river Jhelum, the Hg value was also found to be higher than the different countries' permissible limits. The high concentration of Hg in the study area might be due to burning facilities, including open incineration in hospitals, municipal solid waste incineration, and coal-fired, as the main sources of Hg. The concentration of Cr is only found to be lower than the listed countries' safe limits as illustrated in Table 2. Furthermore, the concentration of Ni was found to be just slightly greater than SEPA (1998) while that of Cu exceeds only CCME (Canadian Council of Ministers of the Environment) (2002) lower limits. Moreover, the concentrations of Pb and Zn were found to be higher than all given countries' lower permissible limits.

Similarly, lower concentrations of Pb, Cu, Ni, and Cr have been also reported by Bhuyan et al. (2017) in Bangladesh. The high pollution level of Cu was related to the higher inputs of organic matter, which mostly originated from sedimentation of industrial and urban wastewater sediments (Xiao et al. 2015). The concentrations of Hg, Pb, Ni, Cr, Cu, and Zn in the river Chenab varied from 0.45–1.02, 31.16–47.32, 101.16–134.19, 47.87–66.41, 12.54–69.32, and 67.42–116.14 mg kg^{-1} respectively. The mean values of all PTEs in the river Chenab are 0.668, 38.918, 120.574, 57.07, 29.49, and 69.942 mg kg^{-1} as listed in Table 2. The PTE concentrations in the River Chenab were in the order of Ni>Zn>Cr>Pb>Cu>Hg respectively.

The concentration of Hg in the River Chenab is lower than NOAA (2012), Taiwan EPA (2010), and Anzecc (2000) safe limits and exceeded other listed countries' limits as shown in Table 2. The mean values of concentrations of Pb and Cr were also found to be higher than the SEPA (1998) and CCME (Canadian Council of Ministers of the Environment) (2002) permissible limits. Furthermore, the concentration of Ni was found to be higher than the stated countries' permissible limits while the Zn was found to be lower than all the countries' safety limits. The Cu concentration exceeded only SEPA (1998) allowable limits. A recent study by Nawab et al. (2018) also showed high concentrations of Ni and Cr in Chenab River. In Bangladesh, similar results were also reported (Ali et al. 2016). The concentration of Pb in the surface sediment may be due to the atmospheric deposition of lead, and emissions from industries, vehicles, and pesticides containing Pb used in agriculture (Hu et al. 2013).

Comparison with other countries

The concentrations of these six PTEs in the sediments of rivers Kabul, Sutlej, Ravi, Jhelum, and Chenab are compared with previously reported concentrations from different rivers as shown in Table 3. The average concentrations of Hg, Pb, Ni, Cr, Cu, and Zn of all rivers were 1.91, 64.26, 71.32, 31.84, 29.52, and 113.11 mg kg^{-1} respectively. The results of this study showed that some PTE concentrations are very high while some are very low as compared to those studies reported in Table 3. The concentration of Hg was found to be very high than that reported in Houjing, Taiwan, and Pearl River Estuary, China, by Vu et al. (2017) and Zhao et al. (2017).

Similarly, the Pb concentration is also found to be higher than that in Vu et al. (2017), Zhao et al. (2017), Larrose et al. (2010), and Ali et al. (2015), but only lower than that of Alyazichi et al. (2017) as listed in Table 3. Furthermore, the concentration of Ni showed similar results as compared to that in Vu et al. (2017) and lower than those of Ali et al. (2015), Larrose et al. (2010), Ilie et al. (2017), and other reference studies. Although the concentration of Cr was found to be lower than that in all the reported studies, it was just similar to M'kandawire et al. (2017) in all the rivers of this study as shown in Table 3. The concentration of Cu was found to be lower than the listed studies report, but just slightly similar to that in M'kandawire et al. (2017).

Moreover, the Zn concentration of this study as compared to that in Ali et al. (2015), Mkandawire et al. (2017), and Ilie et al. (2017) is found higher, but lower than that in all the other reported studies as shown in Table 3. The results of this study showed the concentrations of Hg, Ni, Pb, and Cu are higher as compared to those reported in Table 3, while some PTEs are very similar or lower than those in previously reported studies of other rivers as given in Table 3. Furthermore, it is expected that the exposure level of metals in sediments can trigger PTE accumulation in aquatic species and contaminate the environment (Fan et al. 2014). Mostly, the higher concentrations of PTE were found in the river Ravi followed by Chenab and Sutlej; the reason behind this may be due to the fact that these rivers receive large quantities of PTEs from industrial,

Table 3 Potentially toxic element concentrations (mg kg^{-1}) in the study area with comparison to other studies

River location	Hg	Pb	Ni	Cr	Cu	Zn	References
Kabul	3.17	15.35	39.83	45.93	10.08	51.17	This study
Sutlej	1.36	14.25	20.51	22.32	37.98	123.48	
Ravi	1.9	130.65	165.25	16.12	14.80	159.7	
Jhelum	2.48	122.12	10.42	17.77	55.27	161.26	
Chenab	0.66	38.91	120.57	57.07	29.49	69.94	
Average	1.91	64.26	71.32	31.84	29.52	113.11	
Houjing, Taiwan	0.1	52.23	71.17	53.11	432.29	341.48	Vu et al. 2017
Pearl River Estuary, China	0.1	44.61	*	55.19	42.89	135.87	Zhao et al. 2017
Gorges River, Australia	*	67	13	39	30	157	Alyazichi et al. 2017
Louro River, Spain	*	61.8	46	108	45	*	Filgueiras et al. 2004
Gironde Estuary, France	*	46.8	*	78.4	24.5	168	Larrose et al. 2010
Mamut River, Malaysia	*	23.48	170.74	*	583.38	61.36	Ali et al. 2015
Kafue, Zambia	*	*	20.2	31.1	*	33.3	M'kandawire et al. 2017
Danube, Romania	*	*	34.9	43.3	*	96.6	Ilie et al. 2017
Ravi, India	*	6.37	*	4.78	8.95	4.45	Kaur et al. 2019
River Jhelum Kashmir, India	*	0.12	0.007	0.009	0.11	0.116	Mehmood et al. 2017
Beas river basin, India	*	25.03	*	63.86	29.33	120	Singh and Kaur 2017

*Not available

agricultural, and sewage wastewater as they flow from upstream to downstream (Khan et al. 2012). Furthermore, Mehmood et al. (2017) evaluate the overall pollution load in the river Jhelum in Kashmir, India, which shows a higher concentration of PTEs in the summer season as compared to that in the autumn. The study revealed that Cu and Zn concentrations in sediments of river Jhelum were found enriched, while other PTEs show moderate pollution respectively. Furthermore, it was found that urban-located sites were more polluted as compared to rural sites. Higher concentrations of these PTEs in Pakistan rivers may be due to the long distance these rivers travel from the source of origin until the point of end. As the distance of a river increases, it receives more effluents as compared to origin source. Similarly, Kaur et al. (2019) reported a high concentration of Pb in the river Ravi at the Indian side. The Beas river basin that joins Sutlej River at Kapurthala Punajb, India, also shows higher concentrations of Pb, Cr, Cu, and Zn in sediment samples collected from Beas river areas respectively (Singh and Kaur 2017).

Contamination factor and contamination degree

The spatial distribution of contamination factor (CF) values in the rivers is presented in Fig. 2. The overall results showed that the downstream portions in terms of PTEs were more contaminated as compared to upstream portions in all the rivers (Fig. 2). Moreover, from the CF values, it can be observed that downstream high CF values may be due to the natural and anthropogenic inputs as the rivers flow from upstream to downstream (Liu et al. 2019). The contamination in the downstream portions can be transported through individual flood events, threatening farmlands and livestock which leads to increased damage to the environment (Foulds et al. 2014). Additionally, the overall CF and degree of contamination (CD) values in the rivers Kabul, Sutlej, Ravi, Jhelum, and Chenab are summarized in Table 4. The contamination factor (CF) values in the river Kabul (3.96) are considerable (3>CF>6) for Hg respectively as listed in Table 4. The CF values for Pb and Ni were (3<CF<6) considerable, while Cr, Cu, and Zn show low (CF<1) contamination. Respectively, the CF values for Pb in the river Sutlej were also very high.

Furthermore, the rivers Ravi and Chenab showed also very high CF (CF>6) values for Pb and Ni, while the river Jhelum shows very high values for Pb and Cu only, although the CF values for Hg, Ni, and Zn were moderate (1<CF<3) in the river Sutlej. Moreover, only Cr showed a low contamination factor in the river Sutlej. Furthermore, only Zn showed moderate contamination factor (1< CF<3) values while Cr and Cu showed low contamination factors in the river Ravi. The CF values for Cu were considerable (3<CF<6) and moderate for Ni and Zn, while Cr shows low CF (CF<1) values in the river Jhelum. In the river Chenab only, Cu showed moderate CF values while Cr and Zn showed low contamination factors (CF<1) as listed in Table 4. The CF values were very high for Hg in all the rivers followed by Pb and Ni, and the rest of the selected PTEs showed low to considerable contamination factors in the study area.

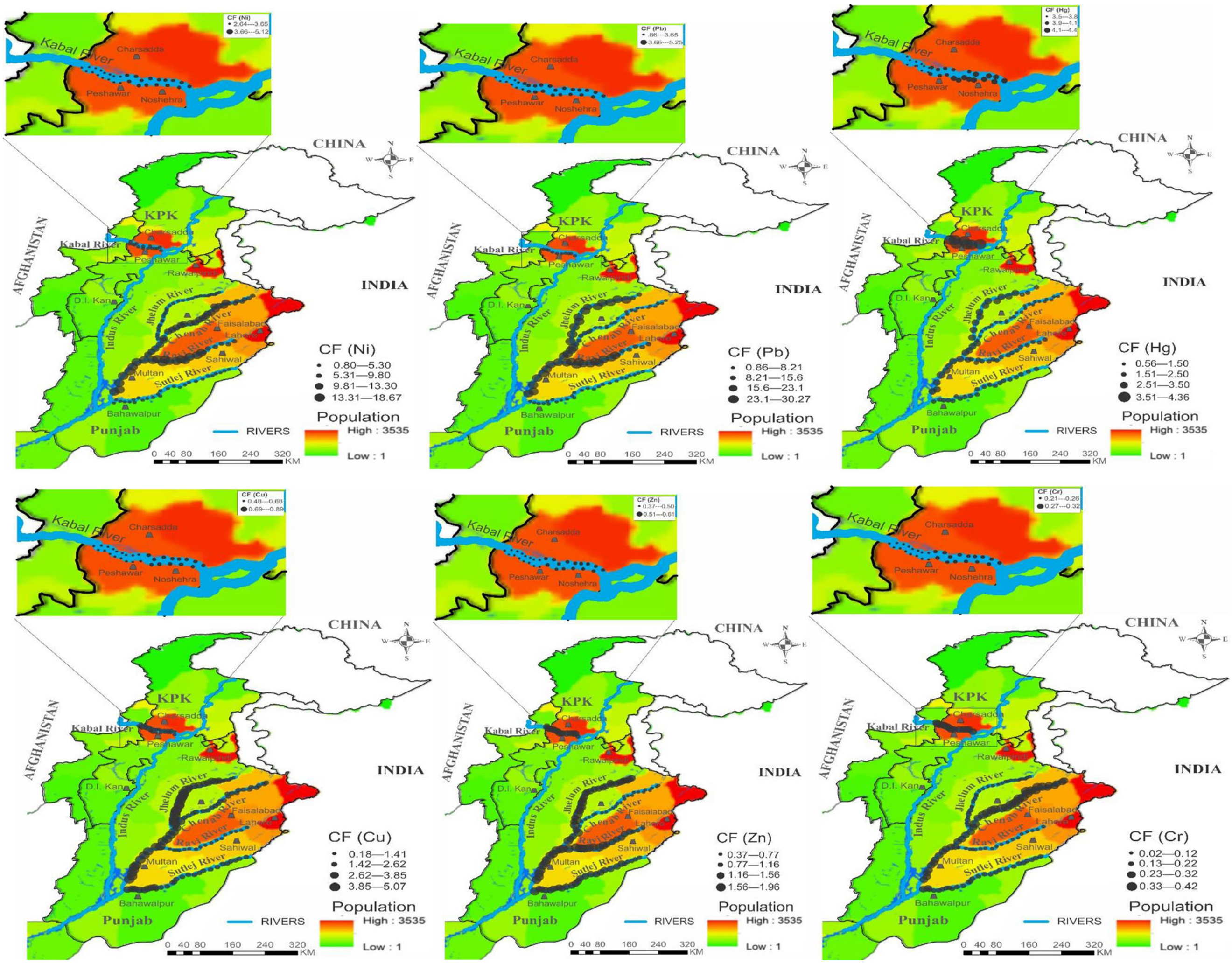

Fig. 2 The contamination factor of PTEs in the major rivers of Pakistan

Respectively, the CD values in the rivers Kabul, Sutlej, Ravi, Jhelum, and Chenab are 12.48, 10.50, 47.71, 33.97, and 23.69 which showed considerable CD values (12 < CD < 24) for rivers Kabul and Chenab and high (CD > 24) for rivers Ravi and Jhelum, but a moderate degree of contamination (6 < CD < 12) for the river Sutlej as shown in Table 4. The order of CD values was Ravi>Jhelum>Chenab>Kabul>Sutlej. As previously reported by Alahabadi and Malvandi (2018),

Table 4 Geo-accumulation, contamination factor, and degree of contamination in the bed sediments of major rivers in Pakistan

Indices	River	Hg	Pb	Ni	Cr	Cu	Zn	Contamination degree
Geo-accumulation	Kabul	7.95	0.61	0.79	0.05	0.13	0.10	
	Sutlej	3.41	0.57	0.41	0.02	0.50	0.24	
	Ravi	4.76	5.24	3.31	0.02	0.19	0.32	
	Jhelum	6.22	4.90	0.20	0.02	0.73	0.32	
	Chenab	1.67	1.56	2.41	0.07	0.39	0.14	
Contamination factor	Kabul	3.96	3.07	3.98	0.28	0.67	0.51	12.48
	Sutlej	1.70	2.85	2.05	0.13	2.53	1.23	10.50
	Ravi	2.37	26.13	16.52	0.10	0.98	1.59	47.71
	Jhelum	3.10	24.42	1.04	0.11	3.68	1.61	33.97
	Chenab	0.83	7.78	12.05	0.35	1.96	0.69	23.69

rivers in Iran showed also high CF values as compared to those of this study. Kayembe et al. (2018) and Duodu et al. (2016) reported similar results of high CF and CD values.

Geo-accumulation index

The geo-accumulation index (I_{geo}) values in the rivers Kabul, Sutlej, Ravi, Jhelum, and Chenab are illustrated in Table 4. The geo-accumulation indices were calculated based on the local background values of the study area. The I_{geo} values in the river Kabul for Hg, Pb, Ni, Cr, Cu, and Zn were 77.95, 0.61, 0.79, 0.05, 0.13, and 0.10 respectively. The I_{geo} values in the river Kabul were extreme (I_{geo}≥5) for Hg while Pb, Ni, Cr, Cu, and Zn showed unpolluted to moderate I_{geo} values. The I_{geo} values in the river Sutlej for Hg, Pb, Ni, Cr, Cu, and Zn are 3.41, 0.57, 0.41, 0.02, 0.50, and 0.24 respectively as shown in Table 2. The geo-accumulation indices in the river Sutlej for Hg were heavy to extreme (4<I_{geo}≤ 5), but those for Pb, Ni, Cr, Cu, and Zn showed unpolluted to moderate (0≤I_{geo}≤1) values. Similarly, the Cr, Cu, and Zn I_{geo} indices were unpolluted to moderate (I_{geo}) in the rivers Ravi, Jhelum, and Chenab respectively.

Furthermore, the Ni values in the rivers Ravi and Chenab are moderately to heavily (2<I_{geo}≤3) polluted as listed in Table 4, while for Hg in the Ravi River showed heavily extreme (4<I_{geo}≤5) pollution and in the River Jhelum extreme (I_{geo}≥5). Moreover, in the Chenab River, Ni showed moderate (1<I_{geo}≤2) and Pb in the Jhelum River showed heavy to extreme pollution respectively. In comparison to those reported by others, Zhuang et al. (2018) and Zhang et al. (2018) also showed high I_{geo} and CF values.

Enrichment factor

The enrichment factor (EF) values in the rivers Kabul, Sutlej, Ravi, Jhelum, and Chenab are summarized in Table 5. The EF values in the river Kabul for Hg 13.87 and Ni 12.20 show severe (10< EF<25) enrichment and Cr showed minor, while Pb, Cu, and Zn show no (EF<1) enrichment respectively, although the EF values for Hg and Ni in the river Sutlej show moderately severe (5<EF<10) while Cu and Zn show minor (1<EF<3) enrichment. Similarly, in the river Sutlej, Pb and Cr showed no enrichment respectively. The calculated EF values in the river Ravi showed that Hg was moderately severe (5<EF<10) and Ni was extremely severe while Pb showed moderate and Zn minor enrichment.

Furthermore, the EF values for Hg were severe in the Jhelum River, while in Chenab Ni showed a very severe enrichment factor. Respectively, the EF values for Pb and Ni in the Jhelum River showed moderate (3<EF<5) enrichment while Cu and Zn showed minor (1<EF<3) enrichment. Chenab River showed minor (1<EF<3) EF values for Hg, Pb, and Cr, but no (EF<1) enrichment for Cu and Zn respectively. The EF values ranged from low enrichment (EF<1) to extremely severe (EF>50) for some PTEs in the study areas. The results revealed that the study area is highly enriched with Hg and Ni followed by Pb as shown in Table 5. In comparison with those of Duodu et al. (2016) and Feng et al. (2019), the results of EF in this study presented similarity to those reported in China.

Potential and ecological risk index

The two most useful techniques ((i) ecological risk factor (ERI) and (ii) potential ecological risk index (PERI)) were used to assess PTEs threatening the sediment biota of the study area. The ERI technique was used to illustrate the risk of individual metals and PERI was used to illustrate the risk caused by all the PTEs in the study area.

Based on the spatial distribution of ERI for each PTE in the selected rivers are presented in Fig. 3. However, the results show that ERI values were not consistent in all the rivers due

Table 5 Enrichment factor, ecological risk, and potential ecological risk in the bed sediments of major rivers in Pakistan

Indices	River	Hg	Pb	Ni	Cr	Cu	Zn	
Enrichment factor (EF)	Kabul	13.87	0.42	12.20	1.56	0.28	0.57	
	Sutlej	5.95	0.38	6.28	0.76	1.07	1.39	
	Ravi	8.31	3.57	50.62	0.54	0.41	1.80	
	Jhelum	10.85	3.34	3.19	0.60	1.56	1.82	
	Chenab	2.92	1.06	36.93	1.94	0.83	0.79	
								Potential ecological risk (PERI)
Ecological risk (ERI)	Kabul	158.5	15.35	19.91	0.57	3.36	0.51	198.22
	Sutlej	68	14.25	10.25	0.27	12.66	1.23	106.68
	Ravi	95	130.65	82.62	0.20	4.93	1.59	315.01
	Jhelum	124	122.12	5.21	0.22	18.42	1.61	271.60
	Chenab	33.4	38.91	60.28	0.71	9.83	0.69	143.84

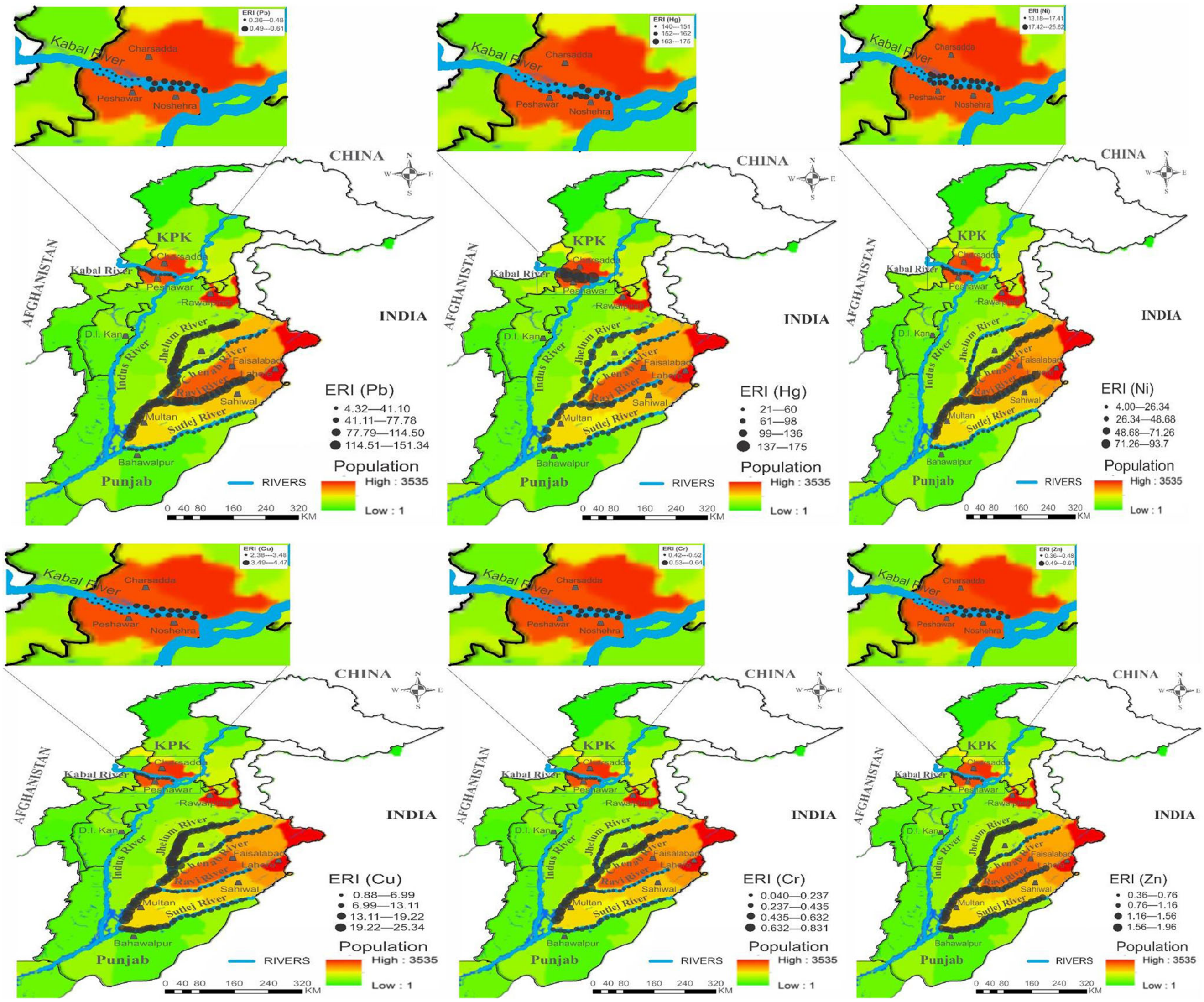

Fig. 3 The ecological risk index of PTEs in the major rivers of Pakistan

to the different pollution sources in each river. Moreover, it can be observed in Fig. 3 where Ravi River shows high ERI values for all the PTEs. Furthermore, the ERI values in all rivers indicated that the downstream section shows more risk in terms of PTEs as compared to the upstream section. Therefore, PTE impact on the upstream region becomes less significant as compared to that on downstream. In urban areas, river, canal, and lake downstream regions are mostly polluted (Nguyen et al. 2016). Industrial and populated adjacent river regions pose more ecological risk as compared to low-populated areas because of both anthropogenic and natural factors that influence river pollution (Nawab et al. 2018).

Moreover, the overall ERI and PERI values in the rivers Kabul, Sutlej, Ravi, Jhelum, and Chenab are illustrated in Table 5. The ERI values in river Kabul for Hg showed considerable (80ERI ≥160) risk while Pb, Ni, Cr, Cu, and Zn expressed low (ERI <40) risks respectively. Similarly, the ERI values for Hg in rivers Ravi and Jhelum also indicated considerable (80ERI ≥160) risk. Furthermore, the ERI values for Pb and Ni indicated considerable (80≤ ERI <120) risk in the river Ravi while Cr, Cu, and Zn showed low (ERI <40) risk in all the rivers.

Similarly, in the river Jhelum, Pb showed considerable and Ni indicated moderate ecological risk in river Chenab as stated in Table 5. The PERI values for rivers Kabul, Sutlej, Ravi, Jhelum, and Chenab were 198.22, 106.68, 315.01, 270.60, and 143.84 as shown in Table 5. The PERI values revealed considerable (300RI≥600) risk in all sites of river Ravi and those of other rivers range from low to moderate (150≤RI<300), but only the river Chenab shows low risk among other rivers as shown in Table 5. The overall results displayed that the all the rivers have very high risk to aquatic biota in the study area. Previous research work reported by Lin et al. (2016), Duodu et al. (2016), and Feng et al. (2019) also showed high PERI values in their study areas.

Principal component analysis

Principal component analysis (PCA) results aimed to identify the sources with high loadings of fewer components after varimax rotation, which may influence the river water sediments (Dragović et al. 2008). PCA results of the PTEs in the different riverine systems of Pakistan are shown in supporting information (SI table 1). The Kaiser-Meyer-Olkin (KMO) value for water was 0.364, and the significance of Bartlett's sphericity test was 0.01. PCA results of selected PTEs were obtained in the form of components. PCA of all river sediments data exhibits a maximum of three components responsible for 98.15, 91.26, 99.19, 97.63, and 93.52% of the total variance and having eigenvalues greater than 1 ((λ) > 1.0) of Kabul River, Sutlej River, Ravi River, Jhelum River, and Chenab River sediments, respectively.

The PC1 described 53.48, 45.31, 45.47, 54.65, and 47.98% of total variance with eigenvalues > 1 for Kabul River, Sutlej River, Ravi River, Jhelum River, and Chenab River sediments, respectively. PC1 shows strength of the Hg, Pb, Ni, and Zn, having moderate factor loading contributions of 0.68, 0.70, 0.78, and 0.86 for Kabul River sediments. The other river sediments also showed high contribution of loading factors of Pb (0.90) and Cr (0.87) for Sutlej River, Pb (0.88) and Zn (0.98) for Ravi River, Pb (0.72), Ni (0.96), and Zn (0.84) for Jhelum River, and Hg (0.73), Pb (0.72), and Ni (0.74) for Chenab River sediments, respectively. PC1 showed the high contribution of moderate and strong positive loadings for all river sediments, demonstrating that aforementioned PTEs could be highly influenced by geogenic and anthropogenic sources in the study area. The moderate and high loadings of PC1 for Hg, Pb, Ni, and Zn in Kabul, Jhelum, and Chenab Rivers suggest the mixed sources of anthropogenic and geogenic origins including erosion and weathering of igneous and mafic-ultramafic rocks in the study area (Ahmad et al. 2020), while strong factor loadings of Cr, Ni, Cu, and Pb in other river sediments could be attributed to anthropogenic sources of industrial wastewater discharge, mining, and agrochemicals (Ullah et al. 2019). Thus, PC1 showed the mixed sources of both anthropogenic and geogenic origins for river sediments in the study area.

PC2 explained 25.69, 28.63, 39.22, 25.64, and 29.11% of the total variance, with eigenvalues > 1 for Kabul River, Sutlej River, Ravi River, Jhelum River, and Chenab River sediments, respectively. PC2 was mainly participated by Pb and Hg with moderate loadings of 0.68 and 0.78 in the Kabul River and Sutlej River sediments, respectively. Hg and Cu had loadings of 0.58 and 0.89 for Ravi River, and Pb and Cr showed moderate loadings of 0.67 and 0.69 for Jhelum River, while Pb and Ni exhibit intermediate loadings of 0.61 and 0.64 for Chenab River sediments. PC2 exhibits relatively less contribution with low loadings of PTEs, in comparison with PC1. The moderate and strong loadings of PTEs in PC2 indicate the natural resources of rock weathering, surface runoff, and riverbank erosion (Yarahmadi and Ansari 2018). Therefore, PC2 exhibits the natural sources of PTEs for river sediments.

PC3 described 18.99, 17.32, 14.49, 17.35, and 16.43% of the total variance, with eigenvalues > 1 for all the river (Kabul, Sutlej, Ravi, Jhelum, and Chenab) sediments respectively. PC3 described the influence of Cu and Ni, while their loading values were observed to be 0.75 and 0.71 for Kabul and Sutlej Rivers, respectively. Hg and Cu had loadings of 0.85 and 0.63, and Hg and Cr showed low loadings of 0.54 and 0.55 for Ravi and Jhelum Rivers, respectively, and Cr and Cu also had strong positive loadings (0.81 and 0.91) for Chenab River sediments. PC3 showed low to moderate positive loadings of PTEs for all river sediments, demonstrating the natural origin of parent rock materials and mineralized ore deposits due to weathering and erosion (Lenart-Boroń and Boroń 2014), but strong loadings of Cr and Cu in Chenab River sediments, demonstrating the anthropogenic sources that could result from agrochemicals and industrial wastewater sources (Ullah et al. 2019). Hence, PC3 also showed the mixed sources of natural and anthropogenic origins. The results of PCA were found in agreement with the previous studies of PTEs in sediments of the Hunza River and its tributaries, conducted by Wasim et al. (2013) and Ahmad et al. (2020).

Correlation analysis of PTEs

Pearson's correlation analysis is a useful technique to provide the elemental relationship information about the sources of selected PTEs (Guo et al. 2015). Pearson correlation analysis was carried out to explore the correlation between each pair of PTEs. Correlation coefficient value <0.5 is considered weak, the values that ranged from 0.5 to 0.75 as moderate, and >0.75 as strong correlation. A number of significant positive correlations were observed among PTEs presented in supporting information (SI table 2). The significant correlation coefficient values in the present study support the PCA results. Moderate and strong positive correlations were notably found between Ni-Pb (0.85), Zn-Pb (0.72), Zn-Ni (0.58), and Zn-Cu (0.63) in Kabul River sediments. For Sutlej River sediments, Cr and Pb showed a strong positive correlation (0.90). Similar results of strong positive correlation were identified between Cu-Pb (0.79) and Zn-Pb (0.87) for Ravi River sediments. The strong positive correlation pairs were observed between Ni-Pb (0.60) and Zn-Pb (0.71) in Jhelum River sediments, while Ni was correlated with Pb (0.89), Cu and Cr (0.60), and Zn-Cu (0.51) in Chenab River sediments. The significant contribution of positive correlations among PTEs in river sediments proposed the common and multiple anthropogenic sources like improper disposal of wastes, industrial wastes discharge, agricultural practices, poor sanitation, and organic decomposition in the study area (Howladar 2017). On the contrary, the negative correlation pairs were noted between Cr-Hg (−0.93) and Cr-Ni (0.58) in Kabul River, and Cu-Hg (−0.52), Cu-Ni (0.69), Zn-Pb (−0.58), and Zn-Cr (−0.54) in Sutlej River sediments. Furthermore, moderate negative correlations were recorded between Cu-Hg (−0.59), Cu-Pb (−0.70), Cu-Ni (−0.77), and Zn-Cu (−0.53) in the Jhelum River, and Cu-Hg (−0.65) and Zn-Ni (−0.65) in Chenab River sediments, whereas the same trends of negative correlations were observed among Cr, Hg, Zn, and Cr in Ravi River sediments, respectively. These negative loading factors demonstrate that the quality of river sediments is not significantly affected by the variables. Cu, Zn, and Cr indicated strong contributions of positive correlations among PTEs that could be associated to weathering of mafic and ultramafic rocks, dissolution of minerals, excess use of pesticide application, and industrial emissions that lead to the potential sources of sediment contamination in the study area (Rashid et al. 2019). Moderate and strong correlations of PTEs in Kabul, Jhelum, and Chenab River sediments could be attributed to the common natural geogenic source of parent rock materials in the regions and mineralized ore deposits due to erosion and weathering (Lenart-Boroń and Boroń 2014). Moreover, the PTEs may have different geochemical behaviors in river sediments due to their moderate and strong correlations in Kabul, Jhelum, and Chenab River sediments. Similar results of PTEs and their correlations in the present study were found to be in agreement with the previous study of sediments of Zhob River, Loralai River, and other tributaries of Baluchistan, in Pakistan (Ullah et al. 2019).

Conclusions

The results of this study revealed that all of the rivers (Kabul, Sutlej, Ravi, Jhelum, and Chenab) are highly contaminated with Hg followed by Pb. The concentrations of Hg, Ni, and Pb were found to be higher in the selected rivers; Cu exceeded the SEPA (1998), CCME (Canadian Council of Ministers of the Environment) (2002), NOAA (2012), and CSBTS (2002) limits, while Cr and Zn exceeded only few stated permissible limits. According to the results of CF values, all the rivers showed a very high contamination level (CF > 6) with Ni and Pb. Furthermore, the overall CF values in the study area showed a low to very high contamination level. The CD values showed a high degree of contamination in all the rivers. The I_{geo} values in the study area were extreme ($I_{geo} \geq 5$) for Hg and heavily polluted for Pb and Ni, while Cr, Cu, and Zn showed low unpolluted (I_{geo}) values in all the rivers. The EF values were high for Hg and Ni, and moderate for Pb and Cu in all the rivers which ranged from no enrichment (EF < 1) to severe (10 < EF < 25) enrichment. The ERI values for Hg, Ni, and Pb showed moderate (40≤ ERI <80) risk, while Cr, Cu, and Zn showed low (ERI <40) risk. The PERI values showed considerable (300≤RI<600) to moderate considerable (150≤RI<300) risk in all the rivers but only the river Chenab showed low (RI≤150) risk. The overall results showed that the downstream portions were more contaminated in terms of PTEs as compared to upstream portions in all the rivers and represent risk to aquatic biota due to natural and manmade activities like improper disposal of industrial and municipal discharges and haphazard use of agrochemicals in farmlands which are the key features contributing to the worsening of sediment quality in the study area. Based on the ecological risk of the study, it can be suggested that proper quality monitoring should be assessed on a regular basis to evaluate PTE contamination and identify the sources and pollution hotspots in these major rivers present in Pakistan.

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Author contribution Javed Nawab, Zia Ud Din, Riaz Ahmad, Sardar Khan, Shah Faisal, Waleed Raziq, and Hamza Khan collected the samples from different rivers and analyzed and interpreted the data regarding the PTEs. Mazhar Iqbal Zafar, Ziaur Rahman, Muahammad Qayash Khan, Javed Nawab, and Abid Ali helped in data analysis and reviewed the paper. Mr Sajid Ullah, Zia Ud Din, and Abdur Rahman performed the PTE analysis and calculated the values. Javed Nawab, Zia Ud Din, Mazhar Iqbal Zafar, and Sardar Khan play a major role in writing, revising, and polishing the manuscript. All the authors read and approved the final manuscript.

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