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Geo-accumulation and enrichment of trace metals in sediments and their associated risks in the Chenab River, Pakistan

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ABSTRACT

In this study the level of toxic metals (Mn, Zn, Pb, Cd, Co, Cu) was determined in sediment samples from the Chenab River, Pakistan. The potential toxicity of studied metals was determined by evaluating enrichment factor (EF), geo-accumulation index (I_{geo}) and metal pollution index (MPI). Considering the spatial distributional patterns, the metal concentrations were higher at Trimmu Headwork site followed by Pujnad, Khanki, Marala and Qadirabad Headwork sites. Unusual higher concentrations in the deeper sediments were observed, suggesting a historical deposition of the investigated metals in the area. The I_{geo} and EF values revealed that sediments in this study were considerably polluted by Cd and Pb and moderately polluted by other metals. Evaluation of metal toxicity based on mean probable effect concentration PEC quotient confirmed that the Chenab River is seriously contaminated with Cd and Pb. Results of the spatial distribution pattern revealed that rapid industrialization and urbanization nearby the study area were probable sources of metal pollution. Proper measures should be taken by industrial units to ensure appropriate treatment of wastewater before disposing the toxic effluents into nearby tributaries. Government authorities must ensure strict enforcement of the National Environmental Quality (NEQ) standards of municipal and industrial effluents to save the Chenab River from further degradation.

1. Introduction

Trace metal contamination is a serious threat in aquatic systems due to their toxicity, abundance, persistence in the environment and subsequent accumulation in aquatic habitats (Qadir and Malik, 2011; Waheed et al., 2013; Fu et al., 2014). It has been recently reported that contamination of water sources from trace elements includes geological weathering and erosion (Kaushik et al., 2009), atmospheric deposition (Demirak et al., 2006), disposal of treated and untreated liquid effluents (Zheng et al., 2008), metal containing fertilizers and pesticides (Iqbal and Shah, 2014), terrestrial run-off (Zahra et al., 2014) and chemicals originating from various urban, industrial and agricultural activities (Park and Presley, 1997; Suthar et al., 2009; Xiao et al., 2013). Sediments have a significant role in the aquatic ecosystem because they are the source of substrate nutrients, and micro- and macroflora and fauna that are the basis of support to living aquatic resources (Jain et al., 2004; Guo et al., 2010). Moreover, few studies have suggested that sediment quality could serve as an indicator for the pollution levels and sediments could act as a screening tool to fingerprint the historical as well as the recent contamination in the surrounding environment (Lin et al., 2008; Xiao et al., 2013; Zahra et al., 2014). Elevated levels of trace metals into aquatic sediments may pose a potential risk to human health due to their transfer into aquatic biota, and ultimately into the food chain (Salati and Moore, 2010; Varol and Şen, 2012).

In developing countries like Pakistan, inland water bodies and estuaries are often contaminated by the anthropogenic activities of the adjoining population and industrial establishments (Qadir and Malik, 2011; Eqani et al., 2012). Industrial wastewater (effluents) containing hazardous chemicals have a great influence on the pollution of the water bodies altering the physical, chemical and biological nature of the receiving water system (Eqani et al., 2012). In particular, toxic metal contamination in the river ecosystem of Pakistan is progressively increasing due to the uncontrolled disposal of increased volumes of industrial and municipal wastewater (Jabeen et al., 2012). Nevertheless, wastewater irrigation is also a common practice in Pakistan, in particular for food crops, which can result into an excessive accumulation of considerable amount of toxic metals in agricultural soils but also lead to elevated trace elements uptake by crops, and thus affect not only food quality but also

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human health (Muchuweti et al., 2006; Rehman et al., 2008). A number of carp species of the Ravi River (i.e. *Catla catla*, *Labeo rohita* and *Cirrhina mrigala*) have been reported to be extinct due to the increased metal toxicity, which created adverse growth conditions in the river and threatened ecological integrities (Azmat et al., 2012; Qadir and Malik, 2011). Recently, few studies have investigated the water quality and heavy metal pollution affecting the freshwater ecosystem in different areas of Pakistan (Bhowmik et al., 2015; Hussain et al., 2014; Zahra et al., 2014). However, the river sediment contamination has been studied extensively in Pakistan but until to date no information is available identifying the potential anthropogenic sources of trace metals into the sediments of Chenab River and its related tributaries. The main aims of the present study were to determine the levels of trace metals in the sediments from the Chenab River in order to identify their naturally enriched or anthropogenic sources using enrichment factor (EF) and geoaccumulation index (I_{geo}), as well as to assess the environmental risk of heavy metal in the investigated area by comparing the acquired metal values with standard sediment quality guidelines (SQGs). This spatial survey of metal concentrations in the sediments is also useful to assess pollution in the Chenab River and to provide basic information for the judgment of environmental health risks and management of urgent environmental pollution issues in the area.

2. Materials and methods

2.1. Study area and sampling strategy

The Chenab River is the major river in Pakistan which originates from Himachel Pardesh (India) and flows through the Jammu Region (India) into the plains of the Punjab Province (Pakistan), with an average annual water flow of 5.29 billion m^3 (BCM). The river basin measures 67,500 km^2, whose 38,500 km^2 in Pakistan. The Chenab River is joined on its way by the Jhelum River at Trimmu Barrage (Jhang district), 40 miles downstream of Trimun Barrage, and Ravi River in the Khanewal district. The Sutlej River joined Chenab at upstream of Punjnad Barrage and finally about 40 miles below Punjnad Barrage, River Chenab meets the Indus at Mithankot. Important engineering structures on the River Chenab are Marala Barrage, Khanki Barrage, Trimun Barrage and Punjnad Barrage, which provide irrigation water for the Punjab plains, known worldwide for the rice and cotton production. Moreover, the Chenab River passes through the major populated/industrial cities of Pakistan, including Sialkot, Gujrat, Gujranwala, Faisalabad, Jhang and Multan, whose wastewaters discharge into the river. The flows of the Chenab River are regulated through five major headworks: Marala, Khanki, Qadirabad, Trimmu and Punjnad, which were selected as sampling sites (Fig. 1) in this study. Head Marala (S1), located near Sialkot district, is a relatively polluted site, whereas Khanki (S2) and Qadirabad (S3) receive industrial and urban wastewaters from the surrounding cities of Gujrat, Gujranwala, and Sialkot via several small drains. Similarly, Trimmu headwork (S4) is located downstream the Chenab River and with its tributaries (i.e., Jhelum River) is considered to dilute the effects of upstream pollution in the Chenab River. On the other hand, Panjnad headwork (S5) is located downstream after the convergence of Ravi and Sutlej Rivers, and is also affected by the surrounding agricultural areas.

The sediment sampling activities were scheduled by keeping in view the weather forecast in order to avoid the rainy days.

A total of 54 composite sediment samples were collected from all the selected sites and at each point, composite sediment samples were collected using standard protocol (USEPA, 2001). The river bed sediment samples were taken at four different depths (top, 2–10 cm, 10–20 cm and more than 20 cm) using a core sampler. In the laboratory, samples were air-dried to reduce the water content, sieved through a 10 mesh (2 mm) Nylon to remove gravel, organic debris and other dopants and stored in zip-bags at room temperature for further analysis.

2.2. Analytical aspect

For the measurement of the total metal concentrations, 1 g of each sediment sample was digested with a HNO_3 (5 ml), H_2O_2 (1 ml) and

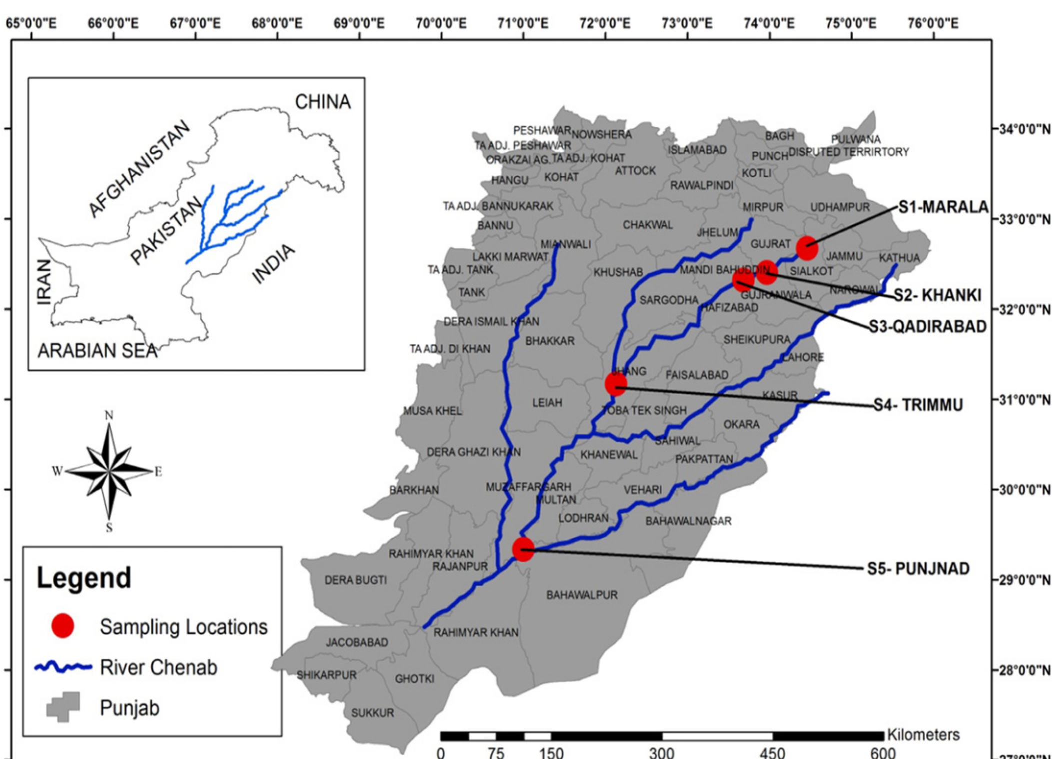

Fig. 1. Investigated area and sampling sites along the Chenab River, Pakistan.

$HClO_4$ (1 ml) mixture. Samples were kept overnight to degrade the organic content before digestion. Samples were digested at 200–250 °C by using automatic hotplate until the solution was evaporated to near dryness and become colorless and then the volume of digestate was raised up to 50 ml by deionized water. Afterwards, they were filtered through Whatman filter paper No. 1 and stored in polyethylene bottles for further analysis. The total concentrations of Cd, Cu, Co, Mn, Pb and Zn were determined by Flame Atomic Absorption Spectrophotometer (FAAS, Thermo SOLAAR M6). In Table S1 the instrumental conditions for the measurements of the analytes by FAAS are reported.

2.3. QA/QC

Analytical grade chemical solvents, purchased from Merck chemicals (Germany), were used for sample analysis. All solutions were prepared using double deionized water. All plastics and glassware were soaked in HNO_3 (10%) for at least 24 h and rinsed repeatedly with ultra pure water. Quality control (QC) samples were sediments fortified with test chemicals and were injected after every 15 samples to assess the instrumental stability. The variations of metal concentrations of QCs were <15%. Reagent blanks were also included in each batch of analyses to check any cross contamination of the different samples digested. Average values of three replicates were taken for each determination. The precision of analytical procedures was expressed as relative standard deviation (RSD), which ranged from 5 to 10% and was calculated from the standard deviation divided by the mean value. Calibration curves were prepared separately for each metal, using different concentrations (i.e. 0.5, 1, 2, 5 and 10 mg/L) of standard solutions. The working solutions were daily prepared by appropriate dilutions of standard stock solutions using a mixture of 65% (v/v) HNO_3, 30% (v/v) H_2O_2, $HClO_4$ and H_2O. The calibration curves, relative coefficients (r^2) of calibration curves and limit of detections (LOD) are reported in Table S1. The instrument was set to zero concentration for all samples, using a reagent blank. Each determination was based on the average values of three replicate measurements and all the values below the detection limit (<BDL) were expressed as zero.

2.4. Statistical analyses

The descriptive statistics and correlation matrix were computed by using StatSoft Statistica (Version 5.0) software and data elaboration was performed by Microsoft Office Excel 2013. One-way ANOVA, followed by Tukey's HSD post hoc test was applied for multiple comparisons of mean metal concentrations. Spatial distribution maps of studied important metals were produced by using ArcGIS$^{(R)}$ (Version 10.1) Geostatistical Analyst-extension.

2.5. Geo-accumulation index, enrichment factor and metal pollution index calculation

The enrichment factor (EF) is widely used as an appropriate approach to discriminate between natural and anthropogenic sources and to reflect the status of environmental contamination, based on the use of a normalization element in order to alleviate the variations produced by heterogeneous sediments (Zhang et al., 2007; Zahra et al., 2014).

Metal concentrations were normalized to the textural characteristic of sediments with respect to Mn, used as reference material. According to Salati and Moore (2010), the EF of metals in the sediments at all the stations was calculated as follows:

$$EF = [(X_x)/(X_{Mn})]_s/[(X_x)/(X_{Mn})]_b$$

where $[(X_x) / (X_{Mn})]_s$ is the ratio of metal (X) and Mn concentrations of the sample and $[(X_x) / (X_{Mn})]_b$ is the ratio of metal and Mn concentrations of background. The background concentrations of Cd, Cu, Co, Mn, Pb and Zn were obtained from Turekian and Wedepohl (1961). The EF values were interpreted as reported in Table 1.

The geo-accumulation index (I_{geo}) for the metal concentrations was calculated by using the following formula (Eqani et al., 2016):

$$I_{geo} = Log_2\left(\frac{C_n}{(1.5)(B_n)}\right)$$

where C_n is the measured concentration of metal n (mg/kg) in sediment, B_n is the geochemical background value (mg/kg) of the element in the background sample and the factor 1.5 is introduced to minimize the effects of possible variations in the background values which may be attributed to lithogenic effects. The crustal abundance data of Turekian and Wedepohl (1961) were used as background data. Geoaccumulation index values were interpreted as reported in Table 1.

Metal pollution index (MPI) was used to assess the overall metal load at each site by using the equation as reported by Usero et al. (1997):

$$MPI_n = (C_1 \times C_2 \times \ldots.. \times C_n)^{1/n}$$

where, C_n is the concentration of the metal 'n' in the sample.

2.5.1. Data comparison with sediment quality guidelines

In Table 2 the heavy metal concentrations (mean; ppm) in the sediments of the Chenab River are reported and compared with the corresponding guideline quality values (MacDonald et al., 2000). Sediment quality guidelines (SQGs) are useful to screen sediment contamination by comparing sediment contaminant concentration with the corresponding quality guidelines (Caeiro et al., 2005). These guidelines evaluate the degree to which the concentrations of contaminants in the sediments might adversely affect aquatic organisms and are designed to assist the interpretation of sediment quality (Wenning et al., 2005). Two types of SQGs developed for freshwater ecosystems (MacDonald et al., 2000) were applied in the present study: threshold effect concentration (TEC) and probable effect concentration (PEC). TEC represents the concentration below which adverse effects are expected to occur infrequently/rarely. In contrast, the PEC represents the concentration above which adverse effects are likely/frequently expected.

3. Results and discussion

3.1. Occurrence and distribution of trace metals in the sediments of the Chenab River

The basic descriptive statistical values and spatial distributional patterns of the studied trace metals are presented in Table 2 and Figs. 1 and 2. On the average basis, the metals follow a decreasing concentration order Mn > Zn > Pb > Cd > Co > Cu. The comparison between metal concentration data found in this study to those reported in literature for other Pakistan rivers is presented in Table 3.

3.1.1. Lead (Pb)

Lead (Pb), a non-essential and toxic element, is released from natural and anthropogenic activities. Major sources include vehicular emissions, volcanoes, airborne soil particles, forest fires, waste incineration, effluents from leather industry, lead containing paints and pesticides (ATSDR, 2007; Eqani et al., 2016; Abdullah et al., 2015). Natural concentration of Pb in the earth's crust varied from 15 to 20 mg/kg (ATSDR, 2007). The current study reported that Pb concentrations (mg/kg) in the sediments ranged from 2.4 to 32.4. The comparison between Pb concentrations in the sediments of the Chenab River and those determined in other sites (see Table 3) showed that Pb levels in the Chenab River sediments had higher levels than those measured in the sediments of

Table 1
Classes of EF and I_{geo} in relation to enrichment and pollution levels, respectively.

EF classes	Enrichment level	I_{geo} value	I_{geo} Class	Pollution level
EF < 1	No enrichment	≤0	0	Unpolluted
EF = 1-3	Minor enrichment	0–1	1	Unpolluted to moderately polluted
EF = 3–5	Moderate enrichment	1–2	2	Moderately polluted
EF = 5–10	Moderately severe enrichment	2–3	3	Moderately to strongly polluted
EF = 25–50	Very severe enrichment	3–4	4	Strongly polluted
EF > 50	Extremely severe enrichment	4–5	5	Strongly to very strongly polluted
		>5	6	Very strongly polluted

EF, Ghrefat et al. (2011).
I_{geo}, Muller (1969).

Indus and Pakistan (Tariq et al., 1996) rivers, but lower concentrations than those measured in the sediments from the Tigris River, Turkey (Varol and Şen, 2012), Almendres River, Cuba (Olivares-Rieumont et al., 2005), Indian rivers (Singh et al., 2005; Suthar et al., 2009), Second Songhua River, China (Lin et al., 2008), Rimac River, Peru (Mendez, 2005) and Nile, Egypt (Rifaat, 2005). Most likely, the concentration of Pb reported in this study originated from both natural and anthropogenic sources including discharge of tanneries effluents from Sialkot (144502–215036 gallons/day) and different other cities i.e., Gujarat, Faisalabad, Jhang, Multan which drain out their waste from many industries like textile, dying, chemical, petro chemical, pulp and paper, hosiery, soap and detergent manufacturing plants, oil refineries, sugar and flour mills, distilleries, synthetic material plants for drugs, fibers, rubbers, plastics, and hosiery etc. into the river ecosystem of the Chenab River (Eqani et al., 2012; Ali et al., 2013). Moreover, use of fertilizers and pesticides is also a common practice in the catchment areas of the Chenab River, which could contribute significantly to the presence of Pb in the river. Moreover, open burning of waste products and dispose-off of sewage and car batteries in the river ecosystem by local communities might be another source of Pb contamination.

3.1.2. Cadmium (Cd)

Cadmium (Cd) is a non-essential element that negatively affects plant growth and development. Genotoxicity and ecotoxicity of Cd in animals have been also reported. Cd is released into the environment by power stations, metal working industries, natural weathering processes, atmospheric deposition, use of phosphate fertilizers, incineration of municipal solid waste, toxic effluents discharge from industrial facilities and sewage treatment plants (WHO, 2010; Ghrefat et al., 2011; ATSDR, 2012a, 2012b; Abdullah et al., 2015). It is recognized as an extremely significant pollutant due to its high toxicity and large solubility in water. Natural Cd concentration found in the earth's crust is in the range 0.1–0.5 mg/kg (ATSDR, 2012a, 2012b). Cd concentrations (mg/kg) ranged between nd to 7.6, and were analogous to those reported from, Indus River, Pakistan (Tariq et al., 1996), Hindon River, India (Suthar et al., 2009) and Almendras River, Cuba (Olivares-Rieumont et al., 2005), Nevertheless, these values are lower than those measured in the sediments from Ravi River, Pakistan (Rauf et al., 2009), Tigris River, Turkey (Varol and Şen, 2012) and Gomti River, India (Singh et al., 2005), Rimac River, Peru (Mendez, 2005) and from the paddy rice fields of Pakistan (Abdullah et al., 2015). High Cd values in the sediment may suggest an anthropogenic contribution due to the runoff from agricultural areas using phosphate fertilizer (Qadir and Malik, 2011; Waheed et al., 2013; Abdullah et al., 2015; Ullah et al., 2014). It can also be suggested that electronic waste from various industries in Gujranwala and Gujrat, effluents from dye and pigment industries in Faisalabad and manufacturing plants of phosphate fertilizers in Multan and Sheikhupura might be the possible sources of Cd contamination in the Chenab River. Similarly, Eqani et al. (2012) suggested that toxic effluents from industries in Faisalabad, Hafizabad and Gujranwala have been reported as major contributors towards high Cd pollution levels in the investigated river.

3.1.3. Copper

Copper (Cu) is an essential micronutrient for aquatic life in freshwaters and sediments but it becomes toxic at higher level. It is released to the environment from natural sources such as volcanic eruptions, decaying vegetation, forest fires, and sea spray etc. up to 50 mg/kg (ATSDR, 2004a, 2004b; Saleem et al., 2013) and anthropogenic activities, including municipal and industrial wastewater (ATSDR, 2004a, 2004b; Eqani et al., 2016). Cu has low solubility in aqueous solution and is easily adsorbed on water-borne suspended particles. After a series of natural processes, the water-borne Cu finally accumulates in the sediment and the quantity of Cu contained in the sediment reflects the degree of pollution of the water body. The average concentration values (mg/kg) were ranging from 5.8 and 9.4, relatively lower than those found in previous studies in the paddy fields sediments from Punjab, Pakistan (Abdullah et al., 2015), in the sediments of Ravi (Rauf et al., 2009) and Indus (Tariq et al., 1996) rivers and surface soil of Sialkot, Pakistan (Abdullah et al., 2015). Compared with the global studies it was found that the Tigris River, Turkey (Varol and Şen, 2012), Almendres River, Cuba (Olivares-Rieumont et al., 2005), Indian rivers

Table 2
Heavy metal concentrations in the sediments collected in the Chenab River (mg/kg) and comparison with the sediment quality guidelines (SQGs).

Sites		Zn	Mn	Cu	Cd	Co	Pb
Marala (n = 9)	Mean ± SD	23.3 ± 20.9	375 ± 308	6.0 ± 1.9	0.4 ± 1.0	2.0 ± 2.9	11.9 ± 2.3
	Median (Min–Max)	21.4 (11.7–50.5)	324 (269–851)	7.4 (5.8–9.4)	0.11 (nd–1.7)	1.1 (1.8–5.8)	14.7 (4.1–18.4)
Khanki (n = 12)	Mean ± SD	28.1 ± 8.1	436 ± 143	8.7 ± 0.7	0.5 ± 0.42	10.4 ± 14.6	13.2 ± 10.2
	Median (Min–Max)	27.9 (18.4–38.3)	457 (245–586)	8.9 (7.8–9.4)	0.19 (nd–0.9)	5.4 (1.4–31.0)	12.3 (3.9–24.4)
Qadirabad (n = 9)	Mean ± SD	22.0 ± 15.6	319 ± 215	5.4 ± 3.6	0.3 ± 0.6	5.6 ± 7.9	4.0 ± 5.4
	Median (Min–Max)	25.9 (13.7–36.2)	401 (400–474)	6.9 (6.5–7.6)	0.14 (nd–1.2)	2.8 (0.89–16.7)	2.4 (1.4–11.4)
Trimmu (n = 12)	Mean ± SD	38.4 ± 4.5	611 ± 137	8.1 ± 0.6	0.65 ± 0.14	8.9 ± 8.4	24.8 ± 5.04
	Median (Min–Max)	40.2 (31.8–41.4)	655 (416–721)	8 (7.6–9.1)	0.57 (0.8–1.9)	7.7 (1.3–20.1)	25 (18.7–30.7)
	Mean ± SD	40.1 ± 5.1	482 ± 177	8.3 ± 0.4	0.84 ± 0.6	8.8 ± 4.3	27.6 ± 6.05
Punjnad (n = 12)	Median (Min–Max)	39.1 (35.3–46.8)	443 (337–705)	8.3 (7.9–8.8)	0.74 (2.1–7.6)	8.8 (3.6–14.0)	29.6 (18.8–32.4)
SQGs	TEC	121[a]	460[b]	31.6[a]	0.99[a]	50[b]	35.8[a]
	PEC	459[a]	1,10[b]	149[a]	4.98[a]	NG[b]	128[a]

[a] MacDonald et al. (2000).
[b] Persaud et al. (1993); NG = no guideline.

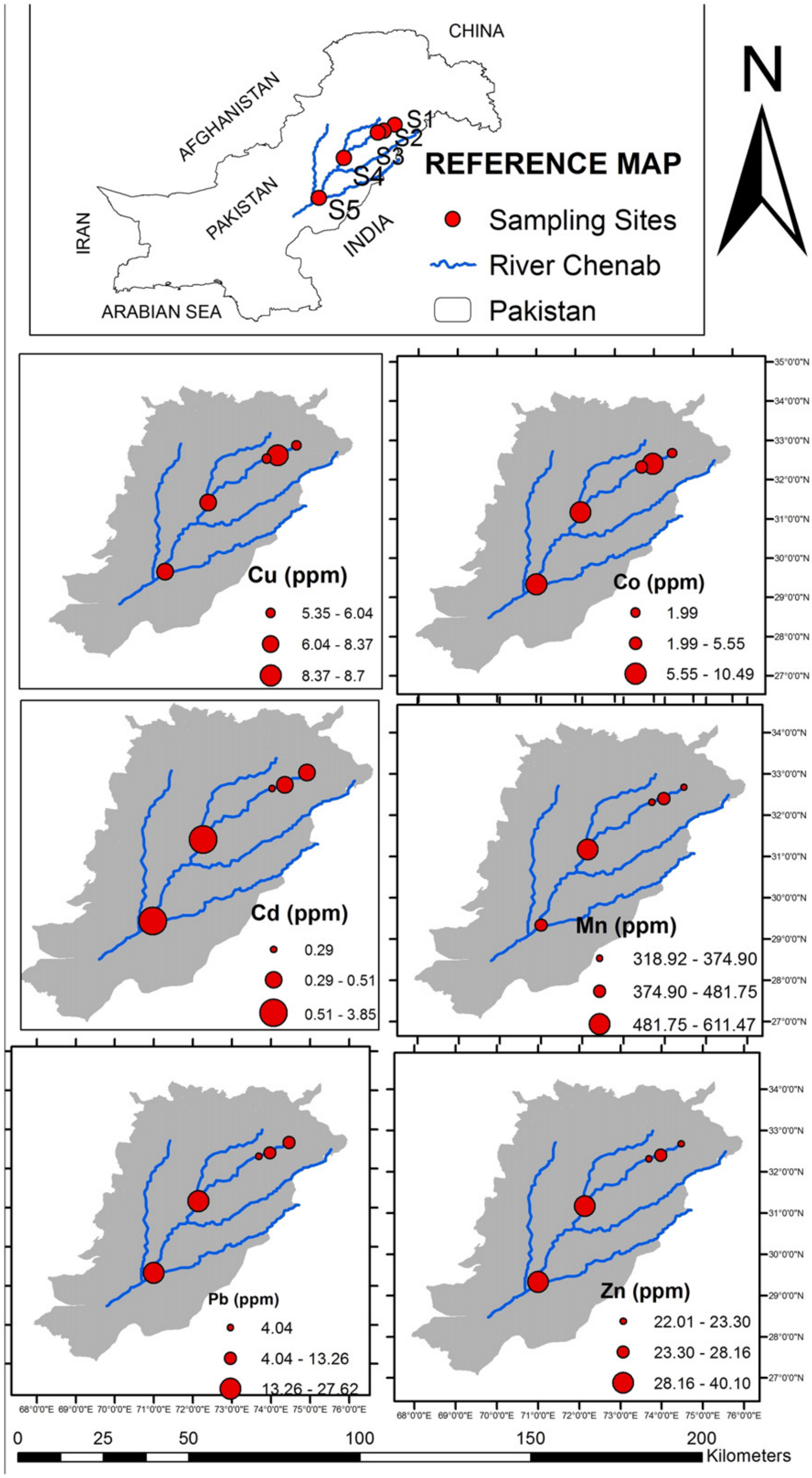

Fig. 2. Mean concentrations of heavy metals (mg/kg) and their spatial distribution pattern in the sediments at selected sites of the Chenab River (Marala Headwork (S1), Khauri Headwork (S2), Qadirabad Headwork (S3), Trimmu Headwork (S4), Punjnad Headwork (S5)).

(Singh et al., 2005; Suthar et al., 2009), Second Songhua River, China (Lin et al., 2008) and Rimac River, Peru (Mendez, 2005) also reported higher Cu levels (see Fig. 3). However, possible sources of Cu toxicity in the sediments of the Chenab River might include the dumping of municipal waste, domestic wastewater discharge, combustion of fossil fuels, copper wires manufacturing industry, steel manufacturing industry etc from the different industrial cities of Punjab, Pakistan.

3.1.4. Zinc (Zn)

Natural background levels of zinc (Zn) are usually found up to 100 mg/kg (dry weight) in sediments (WHO, 2001). The concentrations (mg/kg) of Zn in the present study ranged from 11.7 to 50.5, which are lower than those reported for the sediments of Tigris River, Turkey (Varol and Şen, 2012), Almendres River, Cuba (Olivares-Rieumont et al., 2005), Indian rivers (Singh et al., 2005; Suthar et al., 2009), Second

Table 3
Comparison of metal levels (mg/kg) determined in this study with those reported in the literature.

Location	Mean concentrations (mg/kg)						
	Zn	Mn	Cd	Cu	Co	Pb	Reference
River Chenab, Pakistan	33.7	494	1.67	8.16	7.95	18.1	This study
River Ravi, Pakistan	–	–	3.17	159.79	18.53	–	Rauf et al. (2009)
River Indus, Pakistan	54.3	215	1.62	33.2	–	2.71	Tariq et al. (1996)
Tigris River, Turkey	1061	1682	7.9	2860	516	66	Varol and Şen (2012)
Rimac River, Peru	8076	–	31	796	24	2281	Mendez (2005)
Gomti River, India	99.4	320	7.90	35.7	–	92.2	Singh et al. (2005)
Second Songhua River, China	403	–	–	78.9	14.7	124	Lin et al. (2008)
Hindon River, India	85	202	3.47	195	–	59.1	Suthar et al. (2009)
Almendares River, Cuba	709	–	4.3	421	–	189	Olivares-Rieumont et al. (2005)
Nile River, Egypt	221	2810	–	81	–	23.2	Rifaat (2005)

Songhua River, China (Lin et al., 2008), Nile River, Egypt (Rifaat, 2005) and Rimac River, Peru (Mendez, 2005). However, these Zn levels were similar to those previously found in the sediments of the Indus rivers, Pakistan (Tariq et al., 1996) (Fig. 3). The sources of Zn are natural processes and human activities. Application of fertilizers to agricultural crops is a common practice in the catchment area of the Chenab River, which could contribute to the Zn concentrations in the sediments throughout the Punjab, Pakistan. Similarly, Abdullah et al. (2015) reported the use of salts in the tanneries of Sialkot, which resulted in higher concentrations of Zn in the surrounding environment including the ecosystem of the Chenab River.

3.1.5. Cobalt (Co)

The average concentrations of Cobalt (Co) present in the earth's crust are about 20–25 mg/kg (ATSDR, 2004a, 2004b). The concentrations (mg/kg) of Co found in the sediments of the Chenab River ranged from 0.89–31.0. Compared with previously published results, Co levels were lower than those reported for Tigris River, Turkey (Varol and Şen, 2012), Second Songhua River, China (Lin et al., 2008) Rimac River, Peru (Mendez, 2005) and Ravi River (Rauf et al., 2009) (see Fig. 3). Plastic manufacturing units in Gujranwala, Faisalabad and Sargodha as well as non-point sources, like vehicular emissions, might be responsible of the presence of Co in the sediments of the Chenab River.

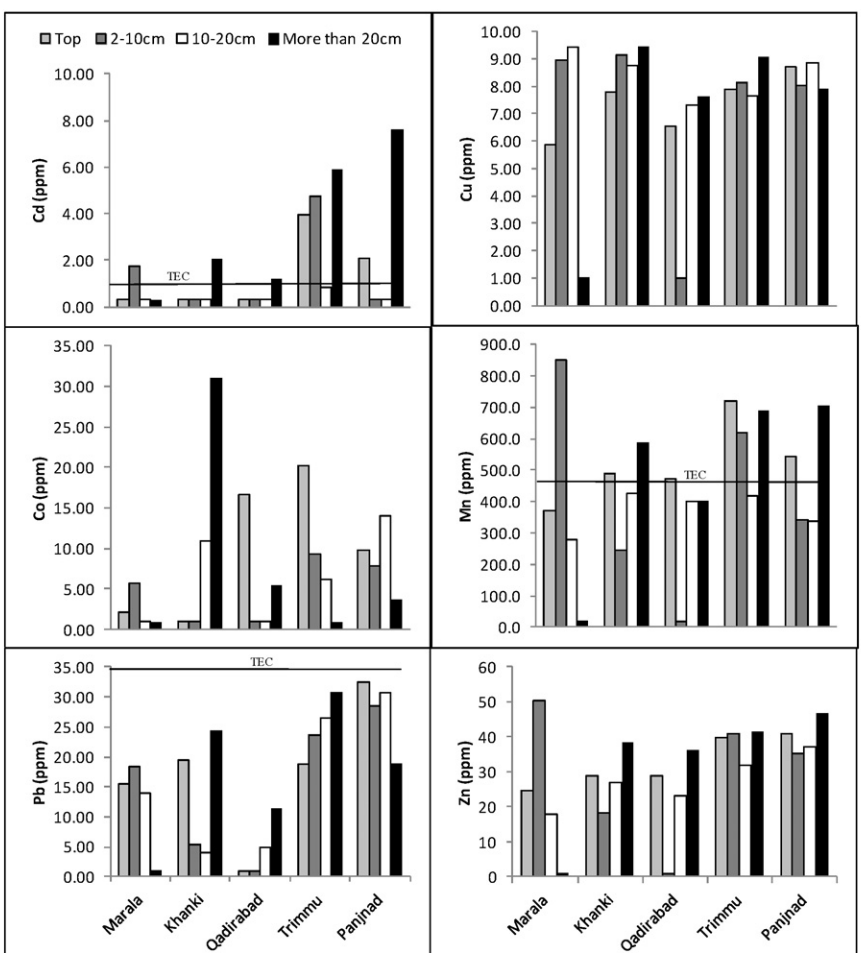

Fig. 3. Heavy metals concentrations (mg/kg) in the sediment samples of the Chenab River.

3.1.6. Manganese (Mn)

Manganese (Mn) concentration in the earth's crust ranged between 40 to 900 mg/kg (ATSDR, 2012a, 2012b). In the current study Mn concentration (mg/kg) ranged from 245 to 851, lower than those reported previously from Tigris River, Turkey and Nile River, Egypt (Varol and Şen, 2012; Rifaat, 2005). However, these results are similar and/or higher than those reported for Indian rivers i.e. Gomti and Hindon (Singh et al., 2005; Suthar et al., 2009) and Indus River, Pakistan (Tariq et al., 1996). Municipal wastewater, sewage sludge, waste from mining and mineral processing units, emissions from alloy, steel, and iron production facilities and combustion of fossil fuel might be the source of Mn into sediments of the Chenab River.

3.2. Heavy metal distribution trends in sediment profile

In order to study the extent and long-term variation of trace metal contamination in aquatic ecosystem, investigation of samples at various depths provides pollution record of a particular area (Park and Presley, 1997). Fig. 3 shows the vertical distribution of trace metals from selected sites of the Chenab River. In previous studies, a decreasing trend of metal accumulation from top to bottom layers has been reported (Li et al., 2000; Yin et al., 2011). However, in the current study, sediment samples exhibited a distinct trend of metal accumulation in sediment profiles where, the concentrations of Pb, Cd and Zn were generally higher at 10–20 and >20 cm depths. Mn at Khanki (S2) and Cu at Punjnad (S5) and Trimmu (S4) also exhibited higher concentrations in the deeper sediment layers, which reflected the historical input of these contaminants. On the other hand, we also observed few exceptions with higher trace metal levels in the top layer showing their recent deposition from the surrounding environment; for example for the case of Pb and Mn (at (S5) Punjnad and (S2) Khanki), and Co (at (S2) Khanki and (S4) Trimmu). In the last few years (before 2009), Chenab River has been documented to be polluted from different industries and surface-run from its catchment area, and the lower flow due to less rainfall and the construction of several dams in the upstream areas in India aggravate its pollution level (Eqani et al., 2012). However, during the last three years (2010–2013), heavy rainfall and high water flow might have washed out the contaminants from upper surface layers and thus the lower extent of contamination may also be attributed to this dilution factor (Eqani et al., 2012). Similar trend has also been reported by Xiao et al. (2013) for sediments from Pearl delta River, China. Our results are also consistent with other studies (Zahra et al., 2014) conducted in the region, which reported the effect of flooding and heavy rainfall on the distribution of environmental contamination.

3.3. Spatial distribution of heavy metals in the sediments of River Chenab

Fig. 1 shows the sampling locations in the Chenab River, which followed the pattern based on the order of the pollution magnitude as (see Table 2): Trimmu Headwork (S4) > Punjnad Headwork (S5) > Khanki Headwork (S2) > Qadirabad Headwork (S3) > Marala Headwork (S1). Among these sites, Trimmu site (S4) is the most polluted site receiving the largest amount of the industrial discharges from Faisalabad and Jhang city. Moreover, Trimmu site receives toxic waste from the upstream flow, urban run-off, and the surrounding agricultural fields. Among the examined trace metals, Cd and Pb at Trimmu Headwork (S4) exceeded the background levels by 2.3 and 1.2 times, respectively. According to Eqani et al. (2011) industrial cities donate a huge flux of industrial and municipal waste into the water resources of Punjab Province. Faisalabad, industrially productive (279 industrial units) district of Punjab Province, disposes approximately 436 cusecs of industrial wastewater and 842 cusecs of municipal wastewater. While, Sargodha (14 industrial units) and Jhang (19 industrial units) were found to dispose 99 and 21 cusecs of effluents, respectively. Cd and Pb concentrations at Punjnad Headwork site (S5) were also higher than the background levels by 1.6 and 1.38 times, respectively. The possible reason for these high levels could be that Multan city disposes about 235 cusecs effluents into the Chenab River (GoP, 2007). Khanki Headwork site (S2), the third polluted site of the study area also exhibited Cd levels above the background concentrations, which are mainly associated with pollution burden from different tributaries, including Choai Nullah, Doara Nullah, Halsai and Dulli Nallah and Upper Jhelum canals which meet the Chenab River before Kanki site (S2). Moreover, Halsai, Dulli and Upper Jhelum canals pass through the Gujrat city, which is a well known industrial city of the country and dispose its toxic wastewater into these tributaries. On the other hand, Nullah Aik and Palkhu receive 547–814 m^3/day tanneries and surgical industrial wastewater and dispose this waste to the Chenab River in the upstream area of Khanki site (Qadir et al., 2008). Similarly, Marala (S1) and Qadirabad (S3) sites were also polluted, but the extent of pollution was not higher than those of other studied sites. In a few cases, Cd concentration from Marala Headwork and Qadirabad Headwork also exceeded the permissible benchmark, which is attributed to the low water flow, ultimately resulting into deposition of different trace metals in sediments on these sites of the Chenab River (Jain et al., 2004). Besides the anthropogenic sources, including urban run-off, industrial discharges, and agricultural inputs; the natural sources of these contaminants, released by natural weathering processes, cannot be neglected because they play a significant role in adding pollutants burden into the Chenab River.

3.4. Estimation of pollutant indicators

3.4.1. Enrichment factor (EF) and index of geo-accumulation (I_{geo})

EF values reflect the levels of metal pollution in a specific environment and their origin of pollution (Feng et al., 2004; Chen et al., 2007). Even if EF should be considered with prudence, EF values < 1 indicate a depletion of the element and reflect the crustal source of the elements in the sediment, whereas values >1 reflect highest levels of anthropogenic pollution (Zhang and Liu, 2002). The EF values of trace element for this study are presented in Table 4. The results show that EF for all metals except for Cd and Pb were <1.5. In many cases (i.e., >80%), values for Cd indicated considerable metal contamination, of which at Khanki site was the most significant (S2). The high EF values for Cd suggested that the Cd input was due to anthropogenic activities. This trend can be explained as due to anthropogenic activities which can release certain amounts of Cd into the environment and river systems. Cd has high geochemical activity in the environment and may be transported in a river system for a long distance because of its higher mobility and water solubility compared with other heavy metals. The variation in EF for different metals from different sites was possibly due to the difference of metal input or difference in the removal rate of each metal from sediments (Ghrefat et al., 2011).

The geo accumulation index values were interpreted with support of the classification of Abrahim and Parker (2008), reported in Table 1, consisting of seven grades or classes. The I_{geo} classes for 6 studied trace metals for each sampling site are reported in Table 5. The mean I_{geo} index (mg/kg) ranged from 0.01 to 4.91. The I_{geo} values for Cu, Co, Mn, Pb and Zn were 1 for all stations, which indicated that the

Table 4
Enrichment factor (EF) and average shale (mg/kg) for heavy metals in sediments of River Chenab.

Sites	Cd	Co	Cu	Mn	Pb	Zn
Marala	1.5	0.2	0.02	1	1.4	0.56
Khanki	5.7	1.1	0.02	1	1.3	0.58
Qadirabad	4.7	0.8	0.02	1	0.5	0.62
Trimmu	3.7	0.7	0.12	1	1.7	0.56
Punjnad	4.9	0.8	0.10	1	2.4	0.74
Mean	4.7	0.7	0.1	1.0	1.5	0.6
Average Shale[a]	0.3	19	45	850	20	95

[a] Turekian and Wedepohl (1961).

Table 5
Geoaccumulation index (I_{geo}) of heavy metals along with their classes for sediments of selected sites of River Chenab.

Sites	Cd	Class	Cu	Class	Co	Class	Mn	Class	Pb	Class	Zn	Class
Marala	4.05	5	0.002	1	0.02	1	0.09	1	0.12	1	0.05	1
Khanki	5.87	6	0.00	1	0.11	1	0.10	1	0.13	1	0.06	1
Qadirabad	3.58	4	0.00	1	0.06	1	0.08	1	0.04	1	0.05	1
Trimmu	5.47	6	0.02	1	0.09	1	0.14	1	0.25	1	0.08	1
Panjnad	5.60	6	0.01	1	0.09	1	0.11	1	0.28	1	0.08	1
Mean	4.91		0.01		0.08		0.10		0.16		0.06	

sediments in these stations were uncontaminated to moderately contaminated by these metals. The I_{geo} values revealed that the valued of Cd in most of the stations fall in class 6 except Marala (in class 5) and Qadirabad (in class 4) headwork sites.

3.4.2. Metal pollution index (MPI) and comparison with sediment quality guidelines (SQGs)

MPI calculated for the investigated metals exhibited the highest level (mg/kg) at Trimmu headwork site (23.4) whereas the smallest metal input was found at Marala headwork site (9). MPI from Punjnad headwork site was 21.4, followed by Khanki Headwork site (14) and Qadirabad Headwork site (10.6).

Two interpretation criteria were used to assess sediment quality with regard to trace metals. The TEC and PEC reference values for sediments are reported in Table 2. In terms of TEC, the Co, Cu, Pb and Zn concentrations were below the reference values in all sampling sites, suggesting a low probability of adverse effects to the local aquatic biota. In contrast, Mn and Cd exceeded up to 50 and 44.4% of the samples the threshold effect level, suggesting that the concentrations of Mn and Cd are likely to cause harmful effects on the benthic fauna.

In this study, an index of toxicity risk, PEC quotient was also calculated according to the definition reported in MacDonald et al. (2000). The mean probable effect concentration (PEC) quotients calculated for this study are provided in Table 6. Mean PEC quotients ranged from 0.002–1.76 mg/kg, while the lowest PEC quotient was reflected by Qadirabad headwork (S-3) whereas the highest by Khanki headwork (S-2). PEC for Cd in all sampling sites and PEC for Mn in Trimmu Headwork site was greater than 0.5, suggesting a potential toxicity of these two metals in the river sediments. The total PEC quotient calculated from each site of the study area followed the decreasing order as follows: Trimmu > Punjnad > Khanki > Marala > Qadirabad.

4. Conclusions

The aims of the present study were to evaluate the level risk assessment, spatial distribution and geo-accumulation of toxic metals in the sediments of the Chenab River (Pakistan). The distribution of these metals in the sediments is not uniform over the whole section of the river and the change in concentration was due to the release of these metals from different anthropogenic sources.

In the present investigation concentrations of Cd were higher than the safe recommended values, which suggested that Chenab River is polluted by Cd and might create an adverse effect on the river ecosystem.

Table 6
PEC quotients of heavy metals for sediments of selected sites of Chenab River.

	Qadirabad	Khanki	Marala	Trimmu	Punjnad
Zn	0.05	0.06	0.05	0.08	0.09
Mn	0.29	0.40	0.34	0.56	0.44
Cd	1.08	1.76	1.21	1.64	1.68
Cu	0.002	0.003	0.003	0.026	0.016
Pb	0.03	0.10	0.09	0.19	0.22
Mean	0.29	0.47	0.34	0.50	0.49

The I_{geo} and EF values revealed that sediments in this study were considerably polluted by Cd and Pb and moderately polluted by other metals.

Evaluation of metal toxicity based on PEC quotient revealed that the Chenab River is seriously contaminated with Cd and Pb. Heavy metal levels and distribution was found higher at that sites which were in the vicinity of industrial and urban areas. Results of the spatial distribution pattern revealed that rapid industrialization and urbanization nearby the study area were probable sources of metal pollution. Proper measures should be taken by industrial units to ensure appropriate treatment of wastewater before disposing the toxic effluents into nearby tributaries. Government authorities must ensure strict enforcement of the National Environmental Quality (NEQ) standards of municipal and industrial effluents to save the Chenab River from further degradation.

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