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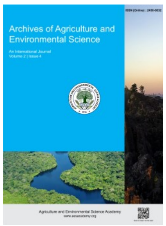

ORIGINAL RESEARCH ARTICLE

CrossMark

Heavy metal contamination and risk assessment on ecological and public health in a tropical estuarine river

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ABSTRACT

Heavy metals contamination of water is one of the most severe environmental and public health issues. The present study was conducted to assess the levels of lead (Pb), chromium (Cr), cadmium (Cd), arsenic (As), copper (Cu) and zinc (Zn) in surface water of the Pasur River estuary in Bangladesh along with their health risk through the ingestion and dermal exposure. The decreasing order of studied metals was Cr > Pb >Cu > Zn > As > Cd with the mean value of 0.050 > 0.024 > 0.021 > 0.014 > 0.012 > 0.006 mg/L respectively. Pb, Cr, Cd and Zn concentration in water samples exceeded the safe limits of drinking water and thereby not safe for drinking. The multivariate analysis identified the common anthropogenic source and existence of studied metals. Heavy metal pollution index (HPI) and heavy metal evaluation index (HEI) indicated significant contamination of water. The HQ and HI through ingestion and dermal contact were <1 except for the adult, whereas HQ (only for As) and HI value through ingestion was >1 indicating an unacceptable risk of non-carcinogenic effects on public health. Carcinogenic risk through ingestion (CR_{ing}) indicated that consumption of water from Pasur River estuary may develop cancer risk of Cd. Therefore, strict rules and regulations must be adopted to reduce water contamination of this tidal river from anthropogenic sources for improving the health of this riverine ecosystem.

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INTRODUCTION

Heavy metal contamination in the aquatic environment has attracted global attention owing to its abundance, persistence, and environmental toxicity. During transportation in the riverine system, heavy metals may undergo frequent changes due to dissolution, precipitation, and absorption phenomena (Abdel-Ghani and Elchaghaby, 2007), which ultimately affect their performance and bioavailability (Nicolau *et al*., 2006; Nouri *et al*., 2011). The investigation of trace metal in water provide useful information about the metal concentrations in water of any contaminated riverine ecosystem therefore, also assess the anthropogenic and industrial impacts and risks posed by waste discharges on the riverine ecosystems (Zheng *et al*., 2008; Saleem *et al*., 2015). Water is an essential requisite for all life forms on earth (Bytyci *et al*., 2018), and it is also known as the most important irreplaceable natural resource on which the sustainable development of a country depends to a large extent (Yıldız, 2017; Pobi *et al*., 2019). The main source of water are rivers, lakes, glaciers, rain water, ground water etc. Bangladesh is a riverine country consisting of more than 230 large and small rivers (Hasan *et al*., 2019). Rivers play an important role in the acceptance and transport of toxic pollutants by receiving waters of point source (industrial, mining) and non-point source (city lives, agriculture, atmospheric precipitation, etc.) pollutants with a view that the dilution strength is sufficient (Tenebe *et al*., 2017). Nowadays heavy metal pollution is a main problem in many developing countries like Bangladesh (Islam *et al*., 2015c). The unplanned urbanization and industrialization of Bangladesh have detrimental effects on the quality of water and sediment as well as other aquatic fauna. The disposal of urban wastes, untreated effluents from various industries and agrochemicals in the open water bodies and rivers has reached alarming situation in Bangladesh which are continually increasing the metals level and deteriorating water quality (Khadse *et al*., 2008; Venugopal *et al*., 2009; Islam *et al*., 2015a, c).

The Pasur River is a tidal, meandering, perennial river in southwestern Bangladesh with a considerable number of fisheries, dockyards, shipyards, and industries that are located along this river's bank. Various types of industrial wastes, solid waste, and hazardous pollutants are produced as a result of unrest production activities, and most of them are promptly discharged into the river without adequate treatment which contribute to the metal pollution. Several studies have attempted to assess the status of heavy metal contamination from Bangladesh's River and estuarine environment (Ali, *et al*., 2016; Bhuiyan *et al*., 2015; Chakraborty, 2022). But unfortunately, there is no significant scientific research on health risk posed by contaminated water of the concerned area so far. Therefore, the objectives of this present study were to determine the seasonal and spatial variation of metal concentration in the surface water; and to assess the ecological risk and potential human health risk of water contamination of the Pasur River estuary, Bangladesh.

MATERIALS AND METHODS

Study area and selection of sites

Pasur River estuary (PRE) is located close to the Sundarbans and Bay of Bengal, Bangladesh (Figure 1). It is one of the major and most important rivers in Khulna division that experiences upstream saltwater intrusion in the southwestern coastal zone of Bangladesh. It is the deepest river in Bangladesh. The total length of the river is about 142 km and the depths ranges from 3 -15 m. The approximate tidal area of Pasur River ranging between 1.5-3 m. Seven sampling sites were selected which were Mongla Ferry Ghat (22°28.272’ N, 89°36.028’ E), Koromjol (22°25.550’ N, 89°35.579’ E), Chila (22°24.371’N, 89°37.171’ E), Joymoni (22°21.038’ N, 89°37.800’ E), Harbaria (22°18.000’ N, 89°36.536’ E), Bhati khal (22°14.171’ N, 89°34.195’ E) and Mazhar point (22°11.591’ N, 89°33.123’ E) of Pasur River.

Sampling

Seasonal sampling was conducted within the period January to December 2022. For the physicochemical analysis, 500 ml of water sample was sampled from the selected sites. On site measurement of these physicochemical parameters were performed. For trace metal analysis, water samples from all seven sampling sites were collected at a depth of about 0.3 m below water surface into 500 ml plastic bottles. Prior to sampling, the bottles were cleaned with 10% nitric acid and rinsed with distilled water. The bottles were rinsed three times with the river water at the time of sampling. Samples were then collected by direct immersion of the sampling bottle into the river. Water samples were filtered immediately after collection using 0.45 µm filters, cellulose nitrate, Millipore filter paper into polypropylene tubes using a plastic syringe (BD Plastipak, 50 mL) for dissolved metal concentrations. Samples were acidified to 0.24 M with 2 ml nitric acid (HNO_3) to reduce absorption of metals onto the walls of the plastic bottles. Sample bottles were then labeled to indicate the sampling date and site. Samples were transported in an ice-box to the laboratory and stored at 4 °C awaiting analysis.

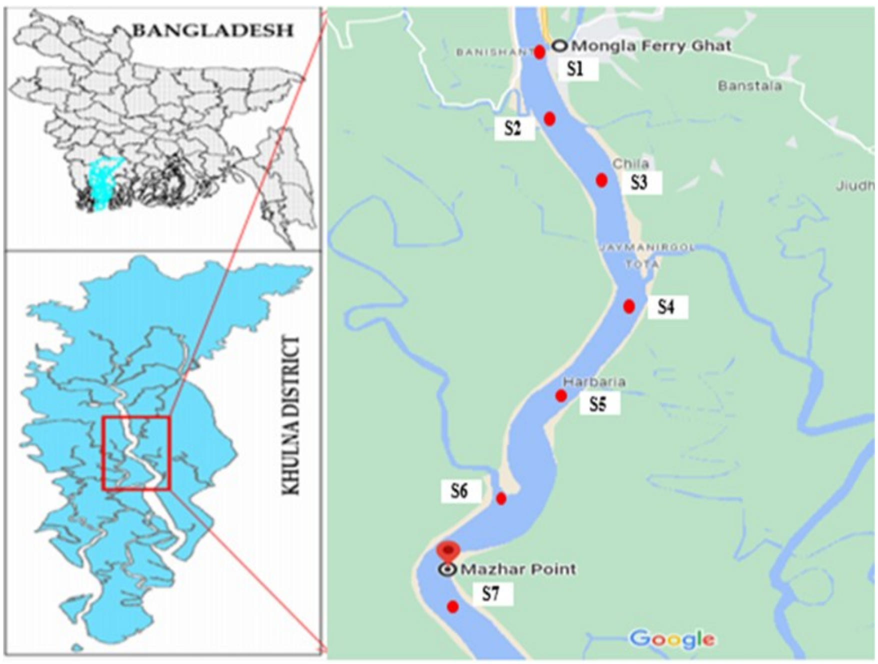

Figure 1. *Map showing the study area and sampling points in the Pasur river estuary, Sundarbans, Bangladesh.*

Determination of physicochemical parameters

The physicochemical parameters such as temperature, salinity, pH, DO, total alkalinity, TDS, NO_3-N and PO_4-P of Pasur River estuary were measured during the study period. Water temperature (°C) was estimated using a Centigrade thermometer. A HACH kit (model FF-2, No. 2430-01; Loveland, CO, USA) was used to assess the alkalinity of the collected samples. Water pH, salinity (ppt), DO (mg/L), and TDS (mg/L) were measured using a pH meter (Adwa AD12 waterproof pH tester); hand-held refractometer (TANAKA, New S-100, Adchi-ku, Japan); DO meter (PDO-519, Taipei, Taiwan) and TDS meter (Adwa AD31 waterproof TDS Testers), respectively. Nitrate-nitrogen (NO_3-N) and Phosphate-phosphorus (PO_4-P) were estimated with HACK Kit (DR-2020, Loveland, CO, USA) with high-range chemicals (Nitra Ver. 5 Nitrate Reagent Powder Pillows for 25 mL sample for NO_3-N and Phos. Ver. 3 Phosphate Reagent Powder Pillows for 25 mL sample for PO_4-P analysis).

Digestion of water samples for metal analysis

All standard solution for target element was supplied by Merck Germany with the highest purity level (99.98%). Ultra-pure HNO_3 was used for sample digestion. All other acids and chemicals were either supra pure or ultra-pure received form Merck Germany or Scharlau Spain. After collection, water samples were filtered through Millipore Filtration Assembly, using 0.45 µm membrane filter. Concentrated HNO_3 and hydrogen peroxide acid (5 ml) was added to 50 ml of filtrate water in a 100 ml beaker, and then heated on a bloc digester to boil until its volume reduced to 20 ml. Another 5ml of concentrated HNO_3 was added and then heated for 10 minutes and allowed to cool. About 5 ml of HNO_3 was used to rinse the sides of the beaker. After cooling, volume was made to desired level with deionized water passing through the Whatman no. 41 filter paper. A blank solution was similarly prepared.

Metal analytical technique

The determination of heavy metals (Pb, Cr, Cd, As, Cu and Zn) in the water samples were carried out by the Flame Atomic Absorption Spectrometer (Shimadzu, AA-6800) in central lab of University of Rajshahi, Rajshshi. Deionized ultrapure water was used for the experimental procedure. All glassware and containers were cleaned with 20% nitric acid, finally rinsed with deionized ultrapure water several times and oven-dried prior to use.

Risk assessment on ecology

Heavy metal pollution index (HPI)

The heavy metal pollution index is a convenient method for determining water quality concerning heavy metals. HPI value is used as a comprehensive instrument to find general water quality derived from heavy metals (Tokatli and Ustaoğlu, 2020). HPI was calculated using the following formulas (Mohan et al., 1996).

$$HPI = \sum_{i=1}^{n}(QiWi)\ \sum_{i=1}^{n}(Wi)$$

$$Q_i = C_i/S_i \times 100$$

$$W_i = K/S_i$$

Qi represents the subindex of each metal, C_i represents the detected concentration value of metals, the standard values of S_i parameters permitted by WHO (2011) as drinking water, W_i represents the unit weight of metals, and k represents a fixed value of "1". If HPI is <100, it indicates a slight level of heavy metal contamination and no adverse related health effects. HPI = 100 indicates threshold risk as well as potential adverse health effects. If HPI is > 100, water is not usable for drinking and not suitable for consumption (Saleh et al., 2019).

Heavy metal evaluation index (HEI)

The index of HEI was used as an indicator of heavy metal contamination in water. Hence, it helps the easy interpretation of the water pollution level (Edet and Offiong, 2002). HEI was computed according to the following formula:

$$HEI = \sum_{i=1}^{n} H_c / H_{MAC}$$

Here H_C stands for the value determined for each metal and H_{MAC} stands for the maximum allowed concentration value (MAC) of each metal (WHO, 2011). If HEI <10, it is interpreted as "low pollution"; if 10 <HEI <20, "medium pollution"; if HEI> 20, it is interpreted as "high pollution" (Saleh et al., 2019).

Risk assessment on human health

Human health risk assessment indices were calculated for both non-cancer and cancer risks from ingestion and absorption of studied metals for the child and the adults. The Average Daily Dose (ADD) intake was calculated according to Iqbal and Shah (2013) following the Eq. 1 and 2:

$$ADD_{ingestion} = Cw \times IR \times ED \times EF/BW \times AT \quad (1)$$

where, $ADD_{ingestion}$ (mg/kg/day) represents the exposure dose through ingestion, Cw is the mean concentration of the trace elements in water (mg/L); IR is both direct and indirect intake rate of drinking water (1 L/day for the child and 2 L/day for the adult), ED is the exposure duration (6 years for the child and 30 years for the adult), EF is the exposure frequency to pollutants (365 days/year), BW represents the total body weight (15 kg for the child and 70 kg for the adult), AT is equal to ED×365 for non-carcinogenic risk, which is 2190 and 10950 for the child and the adult, respectively. For carcinogenic risk, AT is the average life expectancy of people, which is 70×365 = 25550 for both the child and the adult:

$$ADD_{dermal} = Cw \times SA \times Kp \times ET \times EF \times ED \times CF/BW \times AT \quad (2)$$

where, ADDdermal (mg/kg/day/) is the average daily dose of heavy metal through dermal absorption. SA is the exposure area of skin (6600 cm^2 for the child and 18,000 cm^2 for the adults); Kp is the dermal permeability coefficient of pollutants in water (cm/h) in this study, 0.0001 cm/h for Pb, 0.002 cm/h for Cr, 0.001 cm/h for Cd, As and Cu and 0.0006 cm/h for Zn; ET is the exposure time (h/day), in this study, ET is 0.6 h/day; CF is unit conversion factor 0.001 L/cm^{-3} (Asare-Donkor et al., 2016).

The health risk from river water ingestion and dermal absorption was assessed in relation to its non-carcinogenic hazard quotient effects based on the Eq. 3:

Hazard quotient ($HQ_{ingestion/dermal}$) = $ADD_{ingestion/dermal}$/ $RfD_{ingestion/dermal}$ (3)

where, $ADD_{ingestion/dermal}$ and $RfD_{ingestion/dermal}$ are in mg/kg/day. RfD (reference dose) was taken from the United States Environmental Protection Agency, The Integrated Risk Information System (USEPA., 2016). According to Lim *et al.* (2008), HQ value greater than '1' indicates an unacceptable risk of adverse non carcinogenic effects and HQ value less than '1' indicates an acceptable level of risk for human health. However, the potential risk to human health through the mixture of all chemicals was assessed by Li *et al.* (2013) based on Eq. 4:

Hazard index ($HI_{ingestion/dermal}$) = $\sum_{i=1}^{n} HQ_{ingestion/dermal}$ (4)

where, $HI_{ingestion/dermal}$ is potential hazard through ingestion and dermal absorption of heavy metals, $HQ_{ingestion/dermal}$ is the hazard quotient through ingestion or dermal absorption, i is the pathways of exposure; n is the kinds of trace elements; HI>1 means an unacceptable risk and HI<1 means an acceptable level of risk of non-carcinogenic effects on health.

The carcinogenic risk is the multiplication of ADD (mg/kg/day) and Cancer Slope Factor (CSF) (mg/kg/day). Cancer risk due to ingestion of contaminated water with heavy metals was calculated according to Wongsasuluk *et al.* (2014) following the Eq. 5:

$CR_{ingestion} = ADD_{ingestion} \times CSF$ (5)

where, $CR_{ingestion}$ is cancer risk through ingestion of heavy metals contaminated water, $ADD_{ingestion}$ is average daily dose (mg/kg/day) of heavy metals and CSF is cancer slop factor (mg/kg/day). During the present study, the carcinogenic risk values were calculated for Pb, Cr and Cd according to Masok *et al.* (2017), and As according to USEPA (2012). The acceptable limit for lifetime exposure of CR varies from 10^{-6} to 10^{-4} (Yin *et al.*, 2015). When CR > 0.1, the cancer risk is very high; $10^{-3} < CR \le 0.1$ indicates a high risk; $10^{-4} < CR \le 10^{-3}$ indicates a moderate risk; $10^{-6} < CR \le 10^{-4}$ indicates a low risk; and CR ≤ 10−6 indicates an ignorable risk (Hu *et al.*, 2017).

Statistical analysis

Correlation coefficient test was used to determine whether there was a relationship among water quality parameters. The calculation of risk indices was done by Microsoft Office Excel, version 2010. Principle component analysis (PCA) and cluster analysis (CA) were performed to identify the sources of heavy metals in the water using Origin 2023 (Origin Lab, Northampton, MA).

RESULTS AND DISCUSSION

Physicochemical parameters

The physicochemical parameters of different seasons and sites of water measured during the study period are shown in Table 1. The values of water temperature during the study period ranged between 21.44±0.57 (Post-monsoon) to 31.39±0.39 °C (Pre-monsoon) with a mean value of 26.28±3.50 °C, which was close to the findings of Shefat *et al.* (2020), who reported that, water temperature in the Pasur river estuary varied between 20.69 and 33.65 °C. The mean value of water temperature was found within the permissible limits set by WHO (2004), which was between 25 and 30 °C. However, if we compare the mean water temperature with Meghna River estuary reported by Rahman *et al.* (2021), we found lower mean value, that is because Meghna River is a highly polluted river and its higher mean temperature (33.7 °C) were caused by the discharge of hot water effluent from industrial activities that causes dark and turbid water, which are responsible for capture of more heat content. The salinity value during the study period ranged from 5.35±0.10 (Site-1) to 18.82±0.11 (Site-7) in monsoon and pre-monsoon respectively which was close to the findings of Shefat *et al.* (2020) as they found salinity of the Pasur River estuary ranged from 8.47 to 16.15 PSU. Masoud *et al.* (2019) recorded the salinity values ranged between 8.0±3.0 to 29.0±0.6 ppt. in the Reju khal river estuary, Bangladesh which was much higher to the present findings. In the present study maximum pH of water was recorded at Site-1 (7.95±0.04) during post-monsoon whereas minimum was recorded at Site-7 (7.19±0.06) during monsoon with a mean value 7.57±0.23. Geetha *et al.* (2009) showed that the pH varies from 7.3 to 8.9 in the mangrove ecosystem along the Southwest coast of Kerala in India. Higher pH values can be attributed to the removal of CO_2 by photosynthesis through bicarbonate degradation, dilution of seawater by the freshwater influx, reduction of salinity and temperature, and decomposition of organic matter. The DO of Pasur River estuary during the study period ranged from 4.49 (pre-monsoon) to 10.12 mg/L (post-monsoon) with mean value of 7.08±1.78 mg/L. Shefat *et al.* (2020) reported that dissolved O_2 concentration of Pasur River estuary was 5.97 mg/L at pre-monsoon that is close to the current findings. The lowest value of DO was observed during summer and can be due to there being less or no rainfall and the increase in temperature that led to a decrease in DO resulting from the rate of oxygen consumption from aquatic organisms and the high rate of decomposition of organic matter. Maximum value of alkalinity was observed at site-7 (175.92±4.14) in pre-monsoon and minimum was observed at site-1 (72.93±3.19) in monsoon with a mean value of 129.34±28.78 mg/L. Ali *et al.* (2016) conducted similar observation and recorded highest range alkalinity (114.4 ± 3.782-189 ± 16.355 mg/L) from Karnaphuli River. However, the mean alkalinity recorded during the study period was in the productive limit as Huq (2002) narrated that for river water standard value of alkalinity for fisheries activities is 100-200 mg/L. The mean TDS value of Pasur River estuary during the study period was 145.19±29.48 mg/L and the value was below the WHO (2004) maximum allowable limit (500 mg/L) as well as DoE standard of Bangladesh (1000 mg/L). In the present study period significant variation was observed in NO_3-N concentration and maximum value was observed at post-monsoon (1.85±0.07 mg/L) and

Table 1. Physicochemical parameters of Pasur River estuary at different seasons and different locations during the study period.

Season	Stations	Temperature (°C)	Salinity (ppt)	pH	DO (mg/L)	Total alkalinity (mg/L)	TDS (mg/L)	NO_3-N (mg/L)	PO_4-P (mg/L)
Pre-monsoon	S-1	29.86±0.08	14.32±0.13	7.78±0.06	5.98±0.17	121.54±4.09	145.23±6.50	1.41±0.05	2.24±0.05
	S-2	29.95±0.28	15.19±0.31	7.67±0.08	5.76±0.10	129.35±2.06	156.42±4.38	1.32±0.04	2.13±0.05
	S-3	30.42±0.60	15.85±0.29	7.58±0.07	5.54±0.08	137.83±2.73	163.87±6.89	1.25±0.04	1.97±0.13
	S-4	30.58±0.61	16.57±0.19	7.53±0.05	5.32±0.14	146.67±7.49	168.71±3.80	1.13±0.05	1.82±0.17
	S-5	31.39±0.39	17.23±0.31	7.45±0.10	4.80±0.17	154.41±5.46	174.53±8.06	0.95±0.07	1.75±0.08
	S-6	30.31±0.84	18.39±0.39	7.36±0.08	4.65±0.08	167.76±3.72	187.28±2.94	0.84±0.10	1.67±0.11
	S-7	30.14±0.69	18.82±0.11	7.27±0.06	4.49±0.07	175.92±4.14	198.65±5.83	0.73±0.05	1.58±0.13
Mon-soon	S-1	25.83±0.13	5.35±0.10	7.64±0.07	7.92±0.06	72.93±3.19	87.42±3.67	0.64±0.05	1.22±0.07
	S-2	26.19±0.24	5.81±0.28	7.57±0.07	7.73±0.10	80.19±3.17	95.74±5.37	0.57±0.06	1.09±0.10
	S-3	26.40±0.65	6.58±0.34	7.45±0.08	7.36±0.12	87.75±2.96	106.63±3.73	0.49±0.05	0.84±0.08
	S-4	26.52±0.81	6.95±0.30	7.39±0.06	6.78±0.14	96.52±4.20	121.47±3.47	0.38±0.04	0.71±0.12
	S-5	27.31±0.41	7.14±0.31	7.32±0.05	6.45±0.12	108.86±3.66	130.81±3.44	0.27±0.05	0.63±0.11
	S-6	26.47±0.98	7.62±0.37	7.24±0.06	5.83±0.36	117.63±2.85	136.93±3.83	0.19±0.06	0.49±0.08
	S-7	26.38±0.62	8.43±0.13	7.19±0.06	5.67±0.18	125.34±2.02	143.78±5.76	0.12±0.05	0.27±0.07
Post-monsoon	S-1	21.71±0.24	9.27±0.13	7.95±0.04	10.12±0.17	114.85±3.17	117.65±4.44	1.85±0.07	3.15±0.08
	S-2	22.15±0.42	9.64±0.20	7.90±0.06	9.78±0.19	122.64±3.27	125.87±4.64	1.69±0.06	2.91±0.12
	S-3	22.24±0.45	10.41±0.43	7.86±0.08	9.47±0.19	135.48±3.77	138.52±3.50	1.57±0.07	2.73±0.10
	S-4	22.49±0.60	10.89±0.28	7.79±0.05	9.21±0.19	143.29±2.98	146.39±4.70	1.45±0.07	2.61±0.11
	S-5	22.53±0.68	11.57±0.14	7.74±0.06	8.85±0.28	151.56±3.37	157.26±4.62	1.32±0.05	2.54±0.08
	S-6	21.62±0.88	11.85±0.44	7.68±0.04	8.63±0.15	159.91±2.84	164.78±5.90	1.23±0.04	2.43±0.08
	S-7	21.44±0.57	12.74±0.19	7.61±0.04	8.34±0.08	165.78±1.97	180.98±5.52	1.18±0.06	2.39±0.07
Mean		26.28±3.50	11.46±4.24	7.57±0.23	7.08±1.78	129.34±28.78	145.19±29.48	0.98±0.51	1.77±0.85

Table 2. Correlation matrix for physico-chemical parameters of Pasur River estuary.

	Temperature	Salinity	pH	DO	TA	TDS	NO_3-N	PO_4-P
Temperature	1							
Salinity	0.528**	1						
pH	-0.521**	-0.045	1					
DO	-0.916**	-0.529**	0.745**	1				
TA	0.077	0.820**	-0.037	-0.226	1			
TDS	0.313*	0.884**	-0.215	-0.471**	0.944**	1		
NO_3-N	-0.344**	0.358**	0.878**	0.526**	0.348**	0.186	1	
PO_4-P	-0.426**	0.364**	0.849**	0.561**	0.428**	0.249*	0.976**	1

**Correlation is significant at the 0.01 level (2-tailed); *Correlation is significant at the 0.05 level (2-tailed).

minimum was observed at monsoon (0.12±0.05 mg/L) season respectively. Rahaman *et al.* (2014) recorded the highest nitrate in winter and minimum at Karamjal station in monsoon in the Rupsha-Pasur River system of Bangladesh. The higher value of NO_3-N was recorded at site-1 in all seasons might be due to the higher amount fertilizers, municipal wastewaters, feedlots, septic systems in water which causes higher concentration of Nitrate (NO_3^-). During the study period, PO_4-P of Pasur River estuary water ranged between 0.27 to 3.15 mg/L with mean value of 1.77±0.85 mg/L. Shefat *et al.* (2020) also recorded maximum PO_4-P concentration (5.8 mg/L) from Pasur River estuary which was higher compared to the present findings. However, all the observed values for PO_4-P were found below the DOE standard (6 mg/L) for aquatic life during pre-monsoon, monsoon, and post-monsoon seasons. This means that the river water is quite safe in terms of PO_4-P pollution.

Correlation matrix of physicochemical parameters

Pearson correlation reveals the relationships between the physicochemical parameters of Pasur River estuary with significance levels at $p < 0.01$ and $p < 0.05$ (Table 2). Water temperature has significant positive correlation with salinity at $p < 0.01$ level and TDS at $p < 0.05$ level while it has significant negative correlation with pH, DO, NO_3-N and PO_4-P (at $p < 0.01$ level. Salinity has significant positive correlation with TA, TDS, NO_3-N and PO_4-P at $p < 0.01$ level respectively and significant but inverse correlation with DO at $p < 0.01$ level. The pH has significant positive correlation with DO, NO_3-N and PO_4-P at $p < 0.01$ level respectively. The DO has significant negative correlation and alkalinity has significant positive correlation with TDS at $p < 0.01$ level while both DO and alkalinity has significant positive correlation with NO_3-N and PO_4-P at $p < 0.01$ level. TDS and NO_3-N has significant positive correlation with PO_4-P at $p < 0.05$ and $p < 0.01$ level respectively. Higher correlation between variables may indicate mutual dependence of these parameters in the surface water.

Table 3. Heavy metal concentration of water (mg/L) at different seasons and sites of Pasur River estuary during the study period.

Season	Stations	Pb	Cr	Cd	As	Cu	Zn
Pre-monsoon	S-1	0.046±0.002	0.091±0.003	0.010±0.001	0.019±0.003	0.036±0.002	0.023±0.002
	S-2	0.039±0.003	0.072±0.002	0.008±0.001	0.016±0.002	0.033±0.001	0.018±0.004
	S-3	0.028±0.004	0.057±0.002	0.008±0.001	0.013±0.001	0.025±0.002	0.014±0.003
	S-4	0.023±0.003	0.045±0.002	0.006±0.002	0.011±0.003	0.018±0.001	0.012±0.002
	S-5	0.017±0.004	0.033±0.001	0.005±0.002	0.009±0.003	0.014±0.002	0.010±0.003
	S-6	0.011±0.003	0.025±0.002	0.004±0.002	0.006±0.002	0.009±0.003	0.008±0.002
	S-7	0.006±0.004	0.017±0.003	0.003±0.001	0.005±0.002	0.008±0.001	0.007±0.002
Mean		0.024±0.014	0.049±0.025	0.006±0.003	0.011±0.005	0.020±0.011	0.013±0.006
Monsoon	S-1	0.025±0.001	0.052±0.003	0.003±0.001	0.014±0.002	0.026±0.003	0.016±0.001
	S-2	0.022±0.000	0.047±0.002	0.003±0.002	0.012±0.001	0.024±0.002	0.013±0.003
	S-3	0.021±0.001	0.043±0.004	0.002±0.001	0.010±0.003	0.021±0.002	0.010±0.004
	S-4	0.019±0.000	0.035±0.002	0.002±0.001	0.008±0.002	0.020±0.003	0.009±0.003
	S-5	0.015±0.001	0.030±0.002	0.001±0.001	0.006±0.002	0.015±0.002	0.006±0.002
	S-6	0.010±0.001	0.024±0.003	0.001±0.001	0.003±0.001	0.013±0.002	0.005±0.002
	S-7	0.008±0.001	0.015±0.003	0.001±0.001	0.002±0.001	0.009±0.002	0.004±0.001
Mean		0.017±0.006	0.035±0.013	0.002±0.001	0.008±0.005	0.018±0.006	0.009±0.005
Post-monsoon	S-1	0.050±0.001	0.118±0.015	0.017±0.002	0.023±0.002	0.036±0.002	0.028±0.004
	S-2	0.036±0.001	0.095±0.002	0.014±0.001	0.022±0.002	0.034±0.002	0.024±0.003
	S-3	0.034±0.000	0.074±0.002	0.011±0.001	0.020±0.002	0.032±0.002	0.022±0.001
	S-4	0.029±0.002	0.059±0.002	0.009±0.002	0.016±0.001	0.025±0.004	0.019±0.003
	S-5	0.025±0.001	0.047±0.003	0.007±0.002	0.014±0.001	0.019±0.003	0.014±0.003
	S-6	0.019±0.001	0.035±0.002	0.005±0.002	0.012±0.001	0.016±0.001	0.012±0.002
	S-7	0.014±0.002	0.026±0.003	0.003±0.002	0.008±0.003	0.013±0.002	0.011±0.003
Mean		0.030±0.011	0.065±0.032	0.010±0.005	0.017±0.005	0.025±0.009	0.019±0.007

Heavy metals in surface water

The results of toxic metal concentrations in surface water at different seasons and sites of Pasur River estuary during the study period are shown in Table 3. The average concentration of Pb was highest during post-monsoon (0.030±0.011) followed by pre-monsoon (0.024±0.014) and monsoon (0.017±0.006) season, which exceeded the WHO (2011) standard level for drinking water. Ali *et al.* (2018) also recorded that the Pb concentration of Pasur River estuary ranged from 0.02 and 0.0267 mg/L during the summer and winter seasons, respectively. The present concentration was higher than the findings of Rahman *et al.* (2021) at Meghna River estuary (0.009±0.003 mg/L). This may be due to presence of local pollution sources in the study area through river discharge. Cr concentration was 0.049±0.025, 0.035±0.013 and 0.065±0.032 mg/L in pre-monsoon, monsoon, and post-monsoon seasons, respectively. The present finding was close to the Cr level (0.045 mg/L) of Meghna River estuary (Rahman *et al.*, 2021) but lower than the Cr concentration (0.114 mg/L) of Buriganga River (Bhuiyan *et al.*, 2015). The maximum value of Cr was recorded at Site-1 (0.118±0.015 mg/L) in post-monsoon season, which was much higher than the recommended value of the WHO (2011) (0.05 mg/L) guidelines. The highest value of Cr at site-1 could be due to domestic sewage and jute, pharmaceutical and other industry effluents (Islam *et al.*, 2015b; Siddique *et al.*, 2021).

Cd concentration was maximum in (0.010±0.005 mg/L) post-monsoon compared to (0.006±0.003 mg/L) pre-monsoon and (0.002±0.001 mg/L) monsoon season respectively. Similar was also reported by Ali *et al.* (2018) and noted that the Cd concentration of Pasur River estuary ranged from 0.0012 and 0.00197 mg/L during the summer and winter seasons, respectively. The concentration of cadmium (Cd) was highest at Site-1 (0.017 ± 0.002 mg/L) in post-monsoon which exceeded the WHO (2011) (0.003 mg/L) guidelines for drinking water. Higher Cd concentration at site-1 might be attributed to the domestic sewage and effluents from the port area (Islam *et al.* 2015a). The average concentration of As in Pasur River estuary was 0.011±0.005, 0.008±0.005 and 0.017±0.005 mg/L in pre-monsoon, monsoon, and post-monsoon season respectively, which was like the WHO (2011) standard (0.01 mg/L) for drinking water. However, the present value recorded like the As concentration (0.024 mg/L) of Meghna River estuary reported by Rahman *et al.* (2021). During post-monsoon mean Cu concentration of Pasur River estuary was higher (0.025±0.009 mg/L) compared to pre-monsoon (0.020±0.011 mg/L) and monsoon (0.018±0.006 mg/L) season, respectively. The maximum value of Cu was recorded at Site-1 (0.036±0.002 mg/L) during pre-monsoon and post-monsoon season respectively which was lower than the WHO (2011) guidelines for drinking water (2 mg/L). The present finding was higher than the reported value of Uddin *et al.* (2019) at Karnaphuli River (0.0189±0.02531). Maximum Cu concentration was recorded at site-1 might be attributed to the extensive discharging of domestic sewage and urban runoff from extensively farmed areas (Islam *et al.*, 2014).

Zn concentration was highest at Site-1 (0.028±0.004 mg/L) in post-monsoon and the lowest was in monsoon at Site-7 (0.004±0.001 mg/L). The value of Zn recorded from Pasur River estuary during the present study period does not exceed WHO (2011) recommended value (3 mg/L) for drinking water. Zn concentration in water was lower during monsoon season because low level of water in pre-monsoon and post-monsoon increased the concentration of pollutants including raw waste materials from household and local market, which were responsible for higher value of Zn. The mean concentration of studied metals in water followed a decreasing order of Cr > Pb >Cu > Zn > As > Cd (Figure 2). Considering the standard level for drinking water proposed by WHO, among all the toxic metals, Cr and Cd, greatly exceeded the limit for safe water during the present study period, indicating that water from this river is not safe for drinking and/or cooking which is similar to the findings of Ali *et al.* (2018). The metals in the water were seasonally variable, where the post-monsoon season exhibited higher levels than in pre-monsoon and monsoon. The lower concentration of toxic metals during monsoon might be due to the dilution effect of water.

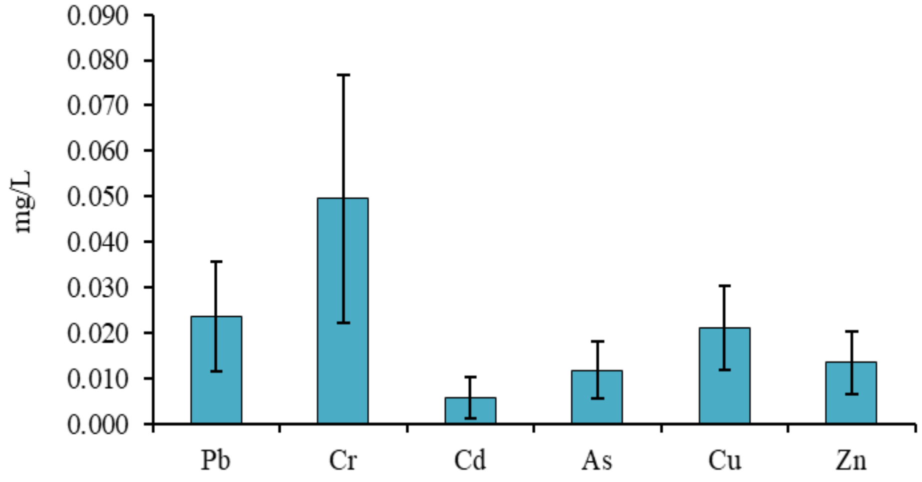

Figure 2. *Mean concentration of heavy metals in water of Pasur River during the study period.*

Table 4. *The health risk posed by the contaminated water of Pasur River estuary.*

	Non-carcinogenic risk of adult				Non-carcinogenic risk of child				Carcinogenic risk of adult		Carcinogenic risk of child	
	ADDing	ADDderm	HQing	HQderm	ADDing	ADDderm	HQing	HQderm	CRing	CRderm	CRing	CRderm
Pre-monsoon												
Pb	6.81E-04	3.55E-07	4.86E-01	8.46E-04	2.18E-04	1.05E-06	1.56E-01	2.50E-03	5.79E-06	3.02E-09	1.85E-06	8.91E-09
Cr	1.35E-03	7.07E-07	4.51E-01	9.43E-03	4.33E-04	2.09E-06	1.44E-01	2.78E-02	6.77E-04	3.53E-07	2.17E-04	1.04E-06
Cd	1.59E-04	8.29E-08	1.59E-01	1.66E-02	5.08E-05	2.45E-07	5.08E-02	4.89E-02	1.00E-03	5.23E-07	3.20E-04	1.54E-06
As	3.13E-04	1.63E-07	1.04E+00	9.61E-03	1.00E-04	4.82E-07	3.34E-01	2.84E-02	4.70E-04	2.45E-07	1.50E-04	7.23E-07
Cu	5.79E-04	3.02E-07	1.45E-02	2.52E-05	1.85E-04	8.91E-07	4.63E-03	7.43E-05	5.79E-06	3.02E-09	1.85E-06	8.91E-09
Zn	3.36E-04	1.75E-07	1.12E-03	2.92E-06	1.08E-04	5.18E-07	3.59E-04	8.63E-06	6.77E-04	3.53E-07	2.17E-04	1.04E-06
HI			2.16E+00	3.65E-02			6.90E-01	1.08E-01				
Monsoon												
Pb	4.52E-04	2.36E-07	3.23E-01	5.62E-04	1.45E-04	6.97E-07	1.03E-01	1.66E-03	3.85E-06	2.01E-09	1.23E-06	5.92E-09
Cr	9.61E-04	5.02E-07	3.20E-01	6.69E-03	3.07E-04	1.48E-06	1.02E-01	1.97E-02	4.80E-04	2.51E-07	1.54E-04	7.40E-07
Cd	5.95E-05	3.11E-08	5.95E-02	6.21E-03	1.90E-05	9.16E-08	1.90E-02	1.83E-02	3.75E-04	1.96E-07	1.20E-04	5.77E-07
As	2.07E-04	1.08E-07	6.90E-01	6.36E-03	6.63E-05	3.19E-07	2.21E-01	1.88E-02	3.11E-04	1.62E-07	9.94E-05	4.78E-07
Cu	5.03E-04	2.63E-07	1.26E-02	2.19E-05	1.61E-04	7.75E-07	4.03E-03	6.46E-05	3.85E-06	2.01E-09	1.23E-06	5.92E-09
Zn	2.56E-04	1.34E-07	8.53E-04	2.23E-06	8.19E-05	3.94E-07	2.73E-04	6.57E-06	4.80E-04	2.51E-07	1.54E-04	7.40E-07
HI			1.41E+00	1.98E-02			4.50E-01	5.85E-02				
Post-monsoon												
Pb	8.05E-04	4.20E-07	5.75E-01	1.00E-03	2.58E-04	1.24E-06	1.84E-01	2.95E-03	6.84E-06	3.57E-09	2.19E-06	1.05E-08
Cr	1.84E-03	9.60E-07	6.13E-01	1.28E-02	5.89E-04	2.83E-06	1.96E-01	3.78E-02	9.20E-04	4.80E-07	2.94E-04	1.42E-06
Cd	2.85E-04	1.49E-07	2.85E-01	2.98E-02	9.13E-05	4.39E-07	9.13E-02	8.79E-02	1.80E-03	9.38E-07	5.75E-04	2.77E-06
As	4.39E-04	2.29E-07	1.46E+00	1.35E-02	1.41E-04	6.76E-07	4.68E-01	3.98E-02	6.59E-04	3.44E-07	2.11E-04	1.01E-06
Cu	7.20E-04	3.76E-07	1.80E-02	3.13E-05	2.30E-04	1.11E-06	5.76E-03	9.24E-05	6.84E-06	3.57E-09	2.19E-06	1.05E-08
Zn	4.99E-04	2.61E-07	1.66E-03	4.34E-06	1.60E-04	7.69E-07	5.33E-04	1.28E-05	9.20E-04	4.80E-07	2.94E-04	1.42E-06
HI			2.96E+00	5.71E-02			9.46E-01	1.68E-01				

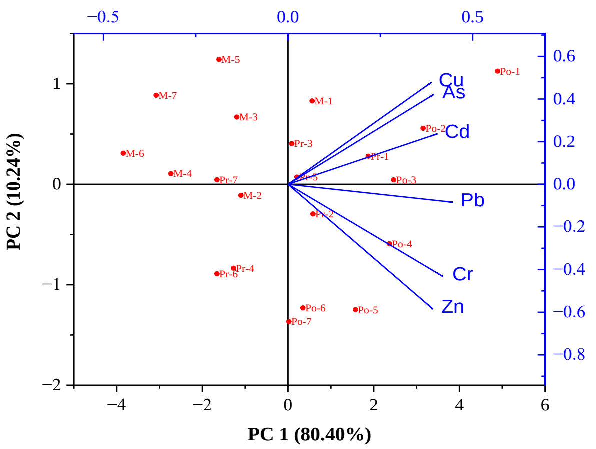

Figure 3. *Principal component analysis of studied metals in the Pasur River* estuary.

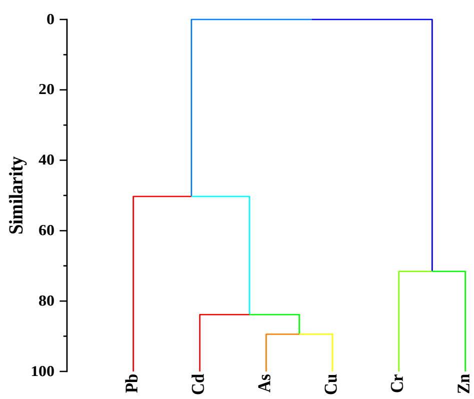

Figure 4. *Hierarchical cluster of studied metals in the Pasur River estuary.*

Source identification of heavy metals

The multivariate analyses (PCA and CA) are applied to know the source, distribution and movement of studied heavy metal in the Pasur River estuary during the study period (Figures 3, 4). This study successively used PCA and CA for precisely finding the studied heavy metal sources. PCA is the simplest of the eigen-vector-based multivariate analysis. The first principal component (PC1) contributed 80.40% of the total variance. Based on the eigen-value of PCA analysis, it was observed that all the metals analyzed in the present study might be originated from the similar source. However, cluster-based analysis showed two distinct cluster representing Pb, Cr, As and Cu in one cluster and Cr and Zn in another cluster. The sources of the studied metals in the study sites may be coming by direct discharging of industrial waste such as electrical, pigments and paints, varnish cosmetics and especially using of copper arsenate, arsenic sulfide for wood processing plant, leachates from defused Ni-Cd batteries and Cd plated items, as well as surface runoff from agricultural land due to excess uses of As rich pesticides (Fu *et al*., 2014; Islam *et al*., 2014, 2015c). Ultimately, the result of the multivariate analysis indicates that anthropogenic activities are dominating compared to geogenic sources, which support the previous findings of Ali *et al*. (2016) and Proshad *et al*. (2019).

Risk assessment on ecology

Heavy metal pollution (HPI) and evaluation index (HEI)

The integrated effect of the studied heavy metals viz. Pb, Cr, Cd, As, Cu and Zn on water quality of Pasur River estuary was determined by HPI and HEI (Figure 5). Global standard values were used when calculating HPI and HEI results (WHO 2011). The average HPI values according to the stations were calculated as 310.03, 261.34, 221.66, 185.75, 139.51, 102.98 and 71.31 from the upstream to the downstream, respectively, while the HEI values were recorded as 10.95, 9.14, 7.71, 6.46, 4.98, 3.67 and 2.56. Similarly, seasonal mean values of HPI were recorded as 189.84, 83.96, and 280.17 in pre-monsoon, monsoon and post-monsoon respectively, while HEI were recorded as 6.57, 3.80 and 9.12. Rahman *et al*. (2022) recorded the mean value of HPI was 760.538 and ranged between 5.497 and 3462.89 from the surface water of a remote island Nijhum Dweep, northern Bay of Bengal which, was too much higher compared to our present study. Both spatial and seasonal values of HPI were >100 except the values found in station-7 (71.31) and that recorded during monsoon (83.96) season which indicate that there was significant heavy metal contamination in the Pasur River estuary. If we compared the mean HEI in water with other rivers it was found that, the present value recorded was higher than HEI (1.94 to 2.76) of Yağlıdere Stream and Aksu Stream respectively (Ustaoğlu and Aydın, 2020) but lower than the HEI (2.175 to 96.598) of surface water of a remote island Nijhum Dweep, northern Bay of Bengal (Rahman *et al*., 2022). Both spatial and seasonal values of HEI were <10 except the values found in station-1 (10.95) which showed that this station was contaminated with metal pollution.

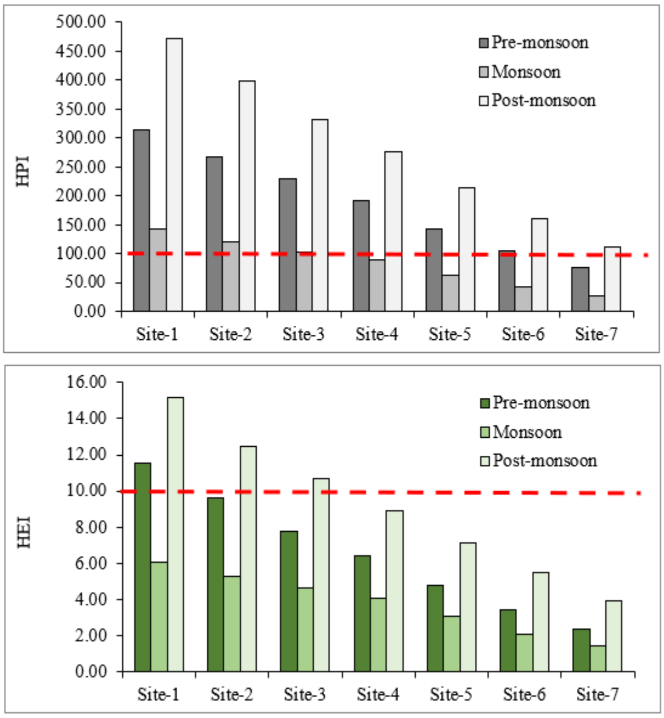

Figure 5. *Heavy metal pollution (HPI) and evaluation index (HEI) of the selected heavy metals from the Pasur River estuary.*

Risk assessment on human health

Direct ingestion and dermal absorption (excluding inhalation through the mouth and nose) are considered to be the two most common exposure pathways of trace elements in river water for human beings (Li *et al*., 2010). The non-carcinogenic and carcinogenic risk of heavy metals from the surface water of Pasur River due to ingestion and dermal contact was analyzed for adults and children (Table 4). Among all the studied metals Cr showed highest ADD_{ing} and ADD_{derm} value during post-monsoon for both adult (1.84E-03, 9.60E-07) and child (5.89E-04, 2.83E-06) respectively. The HQ_{ing} and HQ_{derm} for individual metals were revealed that no calculated metal (except As) in the surface water of Pasur River estuary poses any non-carcinogenic risk (as HQ_{ing} and HQ_{derm} values are below 1) to adults. However, only the concentration of As during pre-monsoon and post-monsoon was found risky for adults, where the HQ_{ing} value of As was found 1.04E+00 and 1.46E+00, respectively, which ultimately increased the overall HI values >1 for adults. Furthermore, HQ_{derm}, HQ_{ing} and HI values are below 1, so the drinking and potable water of Pasur River estuary does not pose a potential hazard for child health. Rahman *et al*. (2022) found that, HI values for adults were more than the children in all sampling locations of a remote island Nijhum Dweep, northern Bay of Bengal, indicating that adults are more vulnerable than children which support our present findings. CR stared at the chances of evolving cancer over the course of a lifetime after being exposed to significant carcinogens (Traina *et al*., 2019). CR_{derm} value found less than 10^{-6} during all the seasons for adult and only monsoon season for child indicated an ignorable risk. Furthermore, Pb, As, Cu during pre-monsoon and Pb and Cu during post-monsoon also showed ignorable cancer risk for child. Ustaoğlu *et al*. (2021) found CR_{dermal} values for children (5.68E-07) are higher than adults (2.56E-07) in Terme River, indicates that when children are exposed to the same environment as adults, they are relatively more susceptible than adults which is similar to our present findings. CR_{ing} for all the metals for child and adult except for Cd indicated low rick of carcinogen. Similarly, Cr, Cd, Zn during pre-monsoon and Cr, Cd, As, Zn during post-monsoon for dermal exposure were also showing low carcinogenic risk. However, carcinogenic risk was only evident for Cd for adult during pre-monsoon and post-monsoon season. The present findings suggest that water of Pasur River estuary is not safe for consumption for all consumers where, adult consumers were highly susceptible to the risk exposure than the child.

Conclusion

In the present study contamination of six toxic metals in the surface water of Pasur River estuary was investigated where the concentrations of Pb, Cr, Cd and As were found higher than the safe values indicating that water from this river is not safe for drinking and/or cooking. The overall pollution load was remarkably higher in post-monsoon compared to pre-monsoon and monsoon season. PCA and CA indicate the common anthropogenic sources of the studied metals in water. Both spatial and seasonal values of HPI exposed that the water were significantly contaminated by metals. Surface water of Pasur River estuary did not poses any non-carcinogenic risk (as HQ_{ing} and HQ_{derm} <1). However, CR indicated ignorable to low risk for all the consumers except for Cd, which showed carcinogenic risk for adult through ingestion during pre-monsoon and post-monsoon season. Therefore, the present study recommends that point sources of heavy metals in the surface water of the Pasur River water should be strictly monitored to reduce metal contamination of water and improved the health of the riverine ecosystem.

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