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Assessment of landfill leachate effects on heavy metal contamination in water and sediments of Garan River, Marivan County, Iran

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ABSTRACT

This study assessed heavy metal contamination in the Garan River's water and sediments to evaluate the impact of landfill leachate. Pollution levels and ecological risks were quantified using established indices, including the contamination factor (CF), pollution load index (PLI), geoaccumulation index (Igeo), enrichment factor (EF), and potential ecological risk index (PERI). Spatially, peak dissolved metal concentrations in water occurred at points S1 and S4, while maximum sediment concentrations were found at S3, directly downstream of the leachate outfall. Water quality analysis indicated that arsenic posed both acute and chronic human health risks at all stations, and chromium posed an acute risk at all stations. Sediment assessment revealed that barium, nickel, and mercury consistently exceeded the threshold effect concentration (TEC). Furthermore, nickel and mercury surpassed the probable effect concentration (PEC) at 100% and 85.71% of stations, respectively, signaling a significant potential for adverse effects on aquatic life. Index analysis showed higher CF values in sediments than in water. The PLI trend increased in sediments but decreased in water, whereas EF was higher in water and spatially stable. The average Igeo for all metals remained below zero, classifying the area as unpolluted. The PERI indicated medium ecological risk in sediments only at stations S3, S6, and S8. While the collective indices suggest that severe, widespread pollution is not present, the identified contamination hotspots and specific risks from metals like arsenic, nickel, and mercury warrant vigilant attention. Consequently, periodic monitoring of leachate-derived metals and proactive preventive measures are critically recommended to preserve aquatic ecosystem quality.

Highlights

* Leachate contamination elevates Ba, Ni, and Hg in river sediments above safety thresholds.
* Arsenic in water poses acute and chronic human health risks at all sampling stations.
* Pollution indices confirm higher heavy metal accumulation in sediments than in the water column.
* Geoaccumulation index classifies the area as unpolluted despite localized contamination hotspots.
* Medium ecological risk from metals is identified in sediments at three downstream locations.

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1. Introduction

The natural and evolutionary balance of ecosystems, including rivers, has always been altered by human activities, and their natural environment is changing and deteriorating (Salati & Moore, 2010). Over the past half-century, pollutants from waste have been a growing trend in the formation of the structure of surface water pollution (Wieczorek et al., 2021).

Surface water is considered the primary source of drinking water in most countries worldwide. Therefore, preventing the pollution of freshwater resources such as rivers and streams, while providing water of optimal quality, in addition to reducing treatment costs and providing human benefits, also brings environmental and biodiversity conservation (Edokpayi et al., 2016). Of the amount of waste produced annually, only 2.3% is disposed of hygienically and without harming the environment (Yang & Xu, 2020). As a result of this mismanagement, highly contaminated leachate is produced from the landfill, which, by migrating and discharging directly or as runoff into water resources, leads to soil, surface, and groundwater contamination. Concentrations of leachate pollutants higher than appropriate standards endanger the natural balance of the environment, living organisms, and general human health, which indicates the need to assess potential risks and reduce adverse effects from landfill leachate in order to achieve sustainable development and management of the environment (Atta et al., 2015; Benfridja et al., 2021; Enitan et al., 2018; Hredoy et al., 2022; Maiti et al., 2015; Naveen et al., 2018; Nta et al., 2020).

NH_3--N, NO_3^-, BOD, COD, SS, and heavy metals are the main pollutants in river basins that can be exacerbated by landfill leachate. The exacerbated effects of these pollutants on surface and groundwater resources have been investigated and evaluated in various studies (Atta et al., 2015; Benfridja et al., 2021; Iresha et al., 2023; Noerfitriyani et al., 2018; Olivares-Rieumont et al., 2005; Shaylinda et al., 2020). Heavy metals enter surface waters and rivers either naturally (geogenic), including weathering of rocks, soil erosion, atmospheric precipitation, wind, and dissolution of soluble salts in water, or through human activities, including various industries, untreated wastewater, landfills, and agricultural activities (Ali et al., 2022; Olivares-Rieumont et al., 2005; Salati & Moore, 2010; Wardhani et al., 2024; Zaranyika et al., 1993). The entry of heavy metals from landfill leachate into groundwater and surface waters has faced developing countries with the most severe environmental challenges (Boateng et al., 2019). Non-biodegradability, bioaccumulation in microorganisms, aquatic plants, and animals, and subsequent circulation in the food chain, environmental stability, persistence, and biotoxicity properties have made heavy metal pollution in rivers a significant issue for both aquatic ecosystems and human health (Dwanga et al. 2024; Varol, 2011). Heavy metals in leachate are affected by this environment through entering the river's aquatic environment and causing pollution through mechanisms of dilution by rainwater during flushing events, adsorption on solid sediment/soil compounds, hydrolysis and also precipitation of newly formed phases (which incorporate these pollutants into their crystal structure), as a result of which most of the metals settle in bottom sediments (Ettler et al., 2006; Islam et al., 2015; Varol, 2011). The mobility, solubility, and bioavailability of heavy metals are changed in combination with dissolved organic matter in leachate and speciation in aquatic environments, causing toxic effects on the environment and human health (Alsaffar et al., 2016; Naveen et al., 2018; Parvin & Tareq, 2021). Heavy metals in carbonate, sulfide, and organic compounds are more toxic and pose greater risks due to their greater bioavailability (Karbassi et al., 2008). Excessive intake of essential heavy metals and excessive intake of toxic heavy metals through direct consumption of drinking water, consumption of agricultural products irrigated with contaminated water, and consumption of aquatic animals that have accumulated these metals in their bodies expose humans to various diseases (Alsaffar et al., 2016). For example, continuous consumption of arsenic-contaminated water can lead to chronic effects such as neurological problems, cancer, cardiovascular disease, and high blood pressure (Boateng et al., 2019). In addition, accumulation of heavy metals from continuous consumption of contaminated fish has caused Itai-Itai disease (caused by chronic cadmium poisoning) and Minamata disease (caused by chronic mercury poisoning) in humans (James, 1977).

Sediments are important ecological components in the river environment, transferring between 30 and 98% of the total metal load of a river in a sediment-dependent manner (Varol & Şen, 2012). Continuous discharge of leachate into the river flow and deposition of persistent pollutants, including heavy metals, in riverbed sediments by increasing their concentration in sediments, changes this part of the river that, under variable physical and chemical conditions, cause sediments to act as both a source of nutrition, a storage reservoir, and a secondary source of pollutants, and also cause disturbances in aquatic life (Karbassi et al., 2008; Sayadi et al., 2015a). For example, iron can cause problems for aquatic life by covering the sediments of the riverbed. In addition, some of these deposited metals are used by aquatic plants and animals, and after decay and decomposition, they re-enter the water. The composition of bed sediment, the composition of suspended sediment, and water chemistry determine the behavior of metals in the river and lead to different amounts of metals and concentration differences between the two environments of river water and sediment (James, 1977).

The Garan River in Marivan, west of Iran, is affected by the leachate discharged from the Garan landfill and changes its physicochemical and chemical properties, including the concentration of heavy metals in the river water and the accumulation of these metals in the riverbed sediments. Therefore, considering the agricultural and horticultural uses of the river water downstream and aquaculture activities in the riverbed receiving reservoir, the ecological importance of this water body, and the possibility of direct human uses, it is vital to assess the concentration of heavy metals in the river after the point of leachate entry into the river to ensure its ability to withstand the load imposed on the river. Considering the above, the present study was conducted to determine the spatial distribution of heavy metals in the two environments of water and sediments of the Garan Riverbed, to confirm or reject the human origin of the pollution, and to determine the intensity of heavy metal pollution in water and sediments using relevant pollution indices.

2. Materials and Methods

2.1 Study area

The Garan River, located in Marivan County, Kurdistan Province, Iran, has geographical coordinates of 46°14'16" to 46°28'54" East and 35°29'45" to 35°47'2" West, and is approximately 25 km long in this county. The rainfall in this area is 800 mm. Agricultural and horticultural activities, fishing and aquaculture, and recreational activities are among

the various uses of this river by humans. As a result of these activities, this river faces the problem of the entry of domestic wastewater flows, pollution with solid waste from residents of the river basin and waste from commercial and industrial activities, and the entry of landfill leachate. The study area is characterized by a Mediterranean climate, featuring hot, dry summers and wet, rainy winters. The average temperature in this area is 14 °C.

2.2 Data collection and sample analysis

In order to investigate the effect of Garan landfill leachate on the heavy metal content of Garan River water, 8 points were selected for sampling, as well as the heavy metal content of 7 points in river bottom sediments. These points included one point upstream of the stream (S1) to determine the background metal concentration as a control point, one point at the leachate entry point into the river, and 6 points downstream of the river (S3, S4, S5, S6, S7, S8). Samples taken downstream of the river were selected based on access to the river for sampling at intervals of approximately 1 km and to investigate the quality conditions of the river water after the incoming leachate mixed with the river water. At the second point, a leachate sample was taken from the drainage area for analysis, so there was no sediment to sample. The geographical coordinates of the sampling points were recorded using a GPS device and are presented in Table 1.

Table 1 Geographic coordinates of sampling points

Sample Site	Zone	Elevation	Longitude	latitude
S1 (Upstream)	38 S	1343	617804	3932998
S2 (Conjunction)	38 S	1311	617894	3930353
S3 (Downstream)	38 S	1312	617300	3930441
S4 (Downstream)	38 S	1212	616930	3930829
S5 (Downstream)	38 S	1194	616567	3931304
S6 (Downstream)	38 S	1227	616684	3931730
S7 (Downstream)	38 S	1194	616730	3932147
S8 (Downstream)	38 S	1221	617697	3932817

Polypropylene containers were used for water sampling to prevent heavy metal contamination of the sample. The water and leachate sampling containers were first washed with nitric acid and then rinsed with distilled water. All containers were rinsed three times with river water before sampling and collecting the samples. At each station, 500 ml of sample was collected with three sampling times at 4-hour intervals, and real samples were extracted from them. 1 ml of pure nitric acid was added to each sample to reduce the pH to approximately 1 (to minimize precipitation and adsorption of metals on the container wall) (Bazrafshan et al., 2016; Islam et al., 2015). After recording the sample characteristics on the sampling containers, these containers were transported to the laboratory at 4°C for analysis. (Association, 1926). The standard protocol of the US Environmental Protection Agency was used to collect composite samples of riverbed sediment (US EPA, 2001). A portable Ekman sampler was used to sample riverbed sediments at a depth of 0 to 5 cm. Three composite cores (200 g) of sediment were collected from each sampling site. The top 2 cm of each core was removed from the center of the receiver with an acid-washed plastic spatula to eliminate any contamination from the metal parts of the corer. For the analysis of background metals, the sediment was cored using a percussion hammer corer (50–80 cm in length) (Schottler & Engstrom, 2006). Air-tight polyethylene bags were used to collect composite sediment samples in the field and then transported to the laboratory. The sediment samples were dried in an oven at 45°C for 48 h to a constant weight. The dried samples were then ground with a mortar and pestle and sieved using a 106-µm mesh. The smaller particle size fraction was homogenized by grinding in an agate mortar and stored in labeled glass bottles for chemical analysis.

2.3. Sample digestion and extraction of metals

In this study, all chemicals used were analytical grade reagents, and purified water (Milli-Q) (Elix UV5 and MilliQ models, manufactured by Millipore, USA) was used to prepare the solutions. Laboratory containers (Teflon and polypropylene) were first carefully cleaned, and after soaking in a 5% nitric acid (HNO_3) solution for more than 24 hours, they were washed with Milli-Q water and dried completely. To calibrate the devices, the required standard concentrations of heavy metals were prepared by diluting 1000 mL stock solutions (Merck Certificate AA standard (Merck)). For metal analysis, 20 mL of water sample and 0.5 g of sediment sample were mixed with 5 mL of HNO_3(69% purity) and 2 mL of hydrogen peroxide (30% purity) in a Teflon-capped vessel and digested in a microwave digestion system. After digestion, the resulting solution was filtered using a syringe filter (DISMIC® – 25HP PTFE model, 0.45 µm pore size, Toyo Roshi Kaisha, Japan) and stored in 50 mL polypropylene tubes (Nalgene, New York). Finally, the vessels were washed with Milli-Q water and dried by air flow.

2.4 Instrumental analysis and quality assurance

For the analysis of heavy metals in the samples, an inductively coupled plasma spectrometer (ICP-OES EOP) (SPECTRO ARCOS) was used. The calibration curve was prepared using the multi-element standard solution XSTC-13 (Spex CertiPrep®, USA) and the calibration curves with a coefficient of determination (R^2) greater than 0.999 were accepted for calculating the metal concentrations. Moreover, a multi-element solution (Agilent Technologies, USA) with a concentration of 1.0 µg/L was used as a tuning solution, which covered a wide range of element masses. The quality of the results was assessed using an internal quality control method and the experimental batches were confirmed if they met the internal quality control criteria (IQCs). In each experiment, blanks, certified reference materials (CRMs), and samples were analyzed twice to eliminate batch-specific errors. Relative standard deviation (RSD) of less than 5% was checked using the calibration solution, and then the analysis process was started. For quality control of the laboratory methods, reference materials NMIJ CRM 7303 and river sediment were used. Comparison of the measured mean values and standard deviations for elemental values in the reference materials with the verified values showed that the sample preparation conditions and instrument settings had the desired accuracy and precision.

2.5 Heavy metal evaluation indices in sediments

In order to analyze chemical and geochemical data, it is of great importance to determine the background values to estimate heavy metal pollution indices and perform

calculations. For this purpose, the concentrations measured at the first sampling station, which is located before the leachate enters the river, were considered as background values. Then, to determine the level of heavy metal pollution caused by the leachate entering the river, the contamination factor (CF), pollution load index (PLI), geoaccumulation index (I_{geo}), enrichment factor (EF), and potential ecological risk index (PERI) were calculated and evaluated.

2.5.1 Contamination Factor

The *CF* is used to show the sediment contamination with heavy metals (Eq. 1)

$$CF = C_m/C_b \quad (1)$$

Where C_m is the concentration of the metal of interest in the sediment at the sampling site, and C_b is the background concentration of the metal of interest in the sediment before the leachate enters the river. The *CF* is categorized into four levels: low contamination level ($CF<1$), moderate contamination level ($1<CF\leq3$), significant contamination level ($3<CF\leq6$), and very high contamination level ($CF>6$) to describe the contamination of each metal individually in the sediment (Schottler & Engstrom, 2006).

2.5.2 Pollution Load Index (PLI)

In order to determine the combined effect of each specific metal on the overall water quality and sediments in the water body, the pollution load index was calculated. Eq. 2 was used to calculate PLI.

$$PLI = \sqrt[n]{CF_1 \times CF_2 \times \ldots \times CF_n} \quad (2)$$

Where *n* represents the number of metals and CF represents the contamination factor associated with each metal. To describe this index, four pollution levels were defined: non-polluted level (PLI < 1), moderate pollution level (1 < PLI < 2), high pollution level (2 < PLI < 3), and very high pollution level (3 < PLI) (Zarei et al., 2014).

2.5.3 Geoaccumulation index

The geoaccumulation index (Igeo) was used to assess the level of heavy metal contamination from leachate entering the river at various concentrations of these metals in the river sediments. This index has been used by various researchers to investigate the contamination of soils and sediments. This method determines the level of contamination by comparing the current amount of heavy metals in samples with their background (natural) levels. To take into account natural changes in materials in the environment, as well as minor impacts of human activities, the background amount of each heavy metal is multiplied by a constant factor of 1.5. The I_{geo} value was calculated using Eq. 3.

$$I_{geo} = log_2(C_n/1.5B_n) \quad (3)$$

where C_n is the measured heavy metal concentration at the sampling sites in river sediments (mg/kg), B_n is the measured background heavy metal concentration in river sediments before leachate contamination (mg/kg), and 1.5 is a constant used to calculate the effect of lithological changes on heavy metal background values. The pollution level calculated with the I_{geo} index was divided into seven classes (0-6) as uncontaminated ($I_{geo} \leq 0$), uncontaminated to almost contaminated ($0 < I_{geo} < 1$), almost contaminated ($1 < I_{geo} < 2$), contaminated ($2 < I_{geo} < 3$), highly contaminated ($3 < I_{geo} < 4$), highly contaminated to very highly contaminated ($4 < I_{geo} < 5$), very highly contaminated ($I_{geo} \geq 5$) based on the Mueller classification (Sayadi et al., 2015b; VESALI et al., 2012).

2.5.4 Enrichment Factor

Enrichment factor (*EF*) is considered one of the useful tools for assessing the level of heavy metal pollution in rivers in environmental studies. The *EF* is calculated based on the comparison of the tested elements with a reference element. The reference element is selected in such a way that there are few changes in its presence in nature. Among conservative elements such as aluminum (Al), iron (Fe), or silicon (Si), Al and Fe are used as geochemical normalizing elements of metal data due to the insignificant contribution of human resources in their production compared with natural resources. Having the least variability in their presence and existence in very high concentrations plays a role in the selection of the reference element. These conditions neutralize the effect of minor changes in heavy metal concentration or interactions with other elements on the results. In the present study, iron was used to normalize metal pollutants to separate natural components from components caused by human activities due to its strong association with fine solid surfaces and geochemical behavior similar to many rare metals. To assess human impacts on the studied heavy metals, the *EF* value for each metal was calculated using Eq. 4.

$$\mathrm{EF} = \frac{(C_n/C_{Fe})_S}{(C_n/C_{Fe})_B} \quad (4)$$

Where $(C_n/C_{Fe})_S$ is the ratio of the concentration of the n^{th} heavy metal to the concentration of iron in the sample, and $(C_n/C_{Fe})_B$ is the ratio of the concentration of the nth metal to the iron in the background sample.

Heavy metal contamination was classified using the *EF* into seven categories: 1. no enrichment (< 1), 2. low enrichment (1-2.9), 3. moderate enrichment (3-4.9), 4. relatively severe enrichment (5-9.9), 5. severe enrichment (10-24.9), 6. very severe enrichment (25-49.9), and 7. very, very severe enrichment (> 50) (Babu et al., 2021; Zarei et al., 2014).

2.5.5 Potential Ecological Risk Index

The Potential Ecological Risk Index (PERI) is a qualitative assessment tool for assessing the ecological risks posed by pollutants in various environments, including sediments. This index was introduced by Hakanson in 1980 (Hakanson, 1980) and is used to assess the ecological risks of heavy metals and other pollutants in sediments. This index is based on the presence of toxic metals in the environment and the ecological responses to the toxicity of these metals in assessing the level of heavy metal contamination in sediments. This index was calculated according to Eq. 5.

$$PERI = \sum_{n=1}^{n}(T_n * CF_n) \quad (5)$$

where T_n is the toxic response index for the n^{th} heavy metal ((Zn = 1), (Cu = 5), (Cr = 2), (Pb = 5), (Mo = 15), (Ni = 2), (Cd = 30), (Hg = 40), (As = 10)) (Sheykhi & Moore, 2013) and CF_n is the calculated *CF* for the n^{th} heavy metal. Evaluation of sediment quality with the *PERI* of metals at four levels including: 1. low ecological risk level (PERI < 150) 2. medium

ecological risk level (150 < PERI < 300) 3. high ecological risk level (300 < PERI < 600), and 4. very high ecological risk level (PERI > 600) was investigated (Sayadi et al., 2015b; Wardhani et al., 2024).

3.1 Results and Discussion

3.1 Heavy metal concentration in water and sediment

The concentrations of heavy metals in water and sediments of the Garan River at the eight studied locations are presented in Tables 2 and 3, respectively. Heavy metal concentrations range for water (µg/l) and sediment (mg/kg) was 15.97-91.74, and 9400-13200 for Al, 1-1.06 and 5.56-14.99 for As, 10.25-12.39 and 45.76-173.91 for Ba, <0.042 and 0.52-0.72 for Be, <0.049 and 0.09-0.4 for Cd, <0.043 and 12.34-16.21 for Co, 1.88-3.13 and 21.72-35.91 for Cr, 18.95-21.36 and 18.22-28.27 for Cu, 13.71-135.21 and 7000-7100 for Fe, <0.351 and <20 for Hg, 5.73-6.31 and 11.73-22.46 for Li, 0.34-3.28 and 0.49-1.52 for Mn, <1.67 and <40 for Mo, 0.29-0.74 and 42.97-156.58 for Ni, <2.17 and 23.31-30.83 for Pb, <0.46 and <10 for Sb, 0.16-1.73 and <20 for Sn, 47.06-50.52 and 2.67-12.12 for Sr, <0.064 and 13.09-21.69 for V and 3.47-28.42 and <100 for Zn. According to Table 2, Be, Cd, Co, Hg, Mo, Pb, Sb, and V had the lowest concentrations in water samples at all sampling points, such that their values were lower than the detection limit of the device. The concentration of As was also lower than the detection limit of the device in all samples except for sample S5 (1.06 µg/L). The highest concentration of Al, Ba, Fe, Mn, and Zn was obtained in sample S5, and their lowest concentrations were obtained in samples S5, S8, S8, S8 and S7, respectively. The highest concentrations of Cr, Li, Ni, Sn, and St were also recorded in sample S4, and among these metals, the lowest values of Cr, Li, Sn, and St were recorded in samples S8, S7, S7 and S2, respectively, and the minimum concentration of Ni was estimated to be below the detection limit in 5 samples (samples S3, S5, S6, S7 and S8). The maximum and minimum concentrations of Cu were also recorded in samples S2 and S5, respectively. The results of heavy metal concentrations in Table 3 show that Hg, Mo, Sb, and Sn were below the detection limit in all samples. The minimum amounts of Al, As, Cd, Co, Cu, Li, Pb, and V were found in sample S1, and Be was also found in samples S1 and S4. Among these metals, the maximum amount of Be, although it did not change much along the sampling points, was found at point S8, and the maximum amount of Li was also recorded in sample S6; the rest of the heavy metals had their maximum amounts in sample S2. The lowest concentrations of Ba and Mn were found in sample S7, and Cr, Ni, and St were found in samples S4, S5, and S6, respectively. The concentration of Zn in all samples and Fe in all samples except sample S6 remained constant. The concentration of heavy metals was compared with the criteria for heavy metals in EPA river water and the California State Sediment Quality Assessment Guidelines (Table 4) (Florida Department of Environmental Protection, January 2003; U.S. Environmental Protection Agency; U.S. Environmental Protection Agency). As can be seen in Table 5, the As concentration exceeded the threshold effect concentration in 28.57% of sediment samples, posing a risk to sediment-dwelling organisms at these points and causing acute and chronic effects on human health through river water at all points. Ba, Hg, and Ni exceeded the threshold effect concentration in sediment at all points studied and exceeded the probable effect concentration at 28.57, 100, and 85.71% of points, respectively. Cr also had acute effects on human health only in water samples and at all points, and did not pose a risk to aquatic life. Adi Septiono and Roosmini investigated the distribution of heavy metals in the Citarum River. In this study, they found that the concentration of Cr in water at two points was higher than the relevant standard and was probably due to the discharge of wastewater from various industries. The higher-than-standard concentration of Cu was also attributed to the synergistic effect of agricultural and industrial activities. In sediments, the concentration of Cu exceeded the standard values compared, indicating moderate pollution levels at the first three points and severe pollution levels at the other points. This increase in Cu concentration was attributed to the deposition of suspended matter that absorbed Cu (Septiono & Roosmini, 2015). The order of heavy metal concentrations in the Cisadane River sediments in the study conducted by Sulistyowati et al. was Pb > Cr > Cd. The results of this study showed a positive and significant Pearson correlation between the metals, indicating a common origin of the metals (natural or anthropogenic). The concentration of metals increased as the river moved downstream, which was a result of point and non-point sources (soil leaching and urban and industrial wastewater discharge) (Sulistyowati et al., 2023). Those elements, having values of zero or non-calculable, are not shown in Table 5.

The speed and type of flow are hydrodynamic aspects of the river that contribute to the deposition of heavy metals in laminar flows and cause resuspension of these materials in turbulent flows. Additionally, the interaction between water and sediment, as well as the deposition and dissolution of heavy metals and their compounds, is influenced by factors such as the topography and geography of the region, as well as soil type, discharge, slope, and vegetation cover (Iresha et al., 2023).

3.2 Indices Analysis

3.2.1 CF

The results of calculating *CF* in river water and sediments are presented in Table 6, and Fig. 1 shows the ratio of sediment pollution to water. In the studied water samples, the *CF* for all metals in the background sample (S1), as well as for Be, Cd, Co, Hg, Mo, Pb, Sb, V, and As (except S5 with *CF* = 1.057 and medium pollution level), which were below the detection limit, was calculated to be 1. The metals of Al, Ba, Fe, Mn, and Zn had a CF less than 1 in all sampling points and were classified as low pollution level. The *CF* higher than 1 was obtained for three metals Cr (1.113), Li (1.016), and Ni (2.053), in S4, and were placed in the group with medium pollution level. Moreover, the three metals Cu, Sn, and St each had values higher than 1 and medium contamination levels at two points of S2 (1.061) and S8 (1.039), S4 (1.215) and S5 (1.136), and S4 (1.05) and S6 (1.001), respectively.

Table 2 Concentration of heavy metals in water samples

Water Sample	Descriptive	Al	As	Ba	Be	Cd	Co	Cr	Cu	Fe	Hg	Li	Mn	Mo	Ni	Pb	Sb	Sn	Sr	V	Zn
	Parameters	ppb	ppb	ppb	ppb	ppb	ppb	ppb	ppb	ppb	ppb	ppb	ppb	ppb	ppb	ppb	ppb	ppb	ppb	ppb	ppb
S1	Min	91.7	1.00	12.24	0.04	0.05	0.04	2.66	19.95	134.95	0.35	6.13	3.15	1.67	0.24	2.17	0.46	1.18	47.79	0.06	28.15
	Max	92	1.00	12.56	0.04	0.05	0.04	2.94	20.25	135.47	0.35	6.31	3.41	1.67	0.48	2.17	0.46	1.70	48.41	0.06	28.69
	Mean	91.7	1.00	12.40	0.04	0.05	0.04	2.80	20.10	135.21	0.35	6.22	3.28	1.67	0.36	2.17	0.46	1.44	48.10	0.06	28.42
	Stdev	0.48	0.00	0.23	0.00	0.00	0.00	0.20	0.21	0.37	0.00	0.13	0.18	0.00	0.17	0.00	0.00	0.37	0.44	0.00	0.38
S2	Min	38.67	1.00	10.82	0.04	0.05	0.04	2.13	21.29	42.68	0.35	5.61	0.96	1.67	0.27	2.17	0.46	0.76	46.71	0.06	8.74
	Max	39.13	1.00	11.18	0.04	0.05	0.04	2.47	21.51	43.06	0.35	5.99	1.42	1.67	0.41	2.17	0.46	1.04	47.41	0.06	9.10
	Mean	38.90	1.00	11.00	0.04	0.05	0.04	2.30	21.40	42.87	0.35	5.80	1.19	1.67	0.34	2.17	0.46	0.90	47.06	0.06	8.92
	Stdev	0.33	0.00	0.25	0.00	0.00	0.00	0.24	0.16	0.27	0.00	0.27	0.33	0.00	0.10	0.00	0.00	0.20	0.49	0.00	0.25
S3	Min	22.06	1.00	11.42	0.04	0.05	0.04	2.58	19.72	26.62	0.35	5.75	1.22	1.67	0.29	2.17	0.46	0.97	47.67	0.06	5.10
	Max	22.34	1.00	11.58	0.04	0.05	0.04	3.02	20.08	27.10	0.35	6.09	1.60	1.67	0.29	2.17	0.46	1.31	48.15	0.06	5.30
	Mean	22.20	1.00	11.50	0.04	0.05	0.04	2.80	19.90	26.86	0.35	5.92	1.41	1.67	0.29	2.17	0.46	1.14	47.91	0.06	5.20
	Stdev	0.20	0.00	0.11	0.00	0.00	0.00	0.31	0.25	0.34	0.00	0.24	0.27	0.00	0.00	0.00	0.00	0.24	0.34	0.00	0.14
S4	Min	23.71	1.00	12.16	0.04	0.05	0.04	2.92	18.99	23.88	0.35	6.18	0.26	1.67	0.60	2.17	0.46	1.47	50.33	0.06	4.99
	Max	24.09	1.00	12.44	0.04	0.05	0.04	3.28	19.41	24.22	0.35	6.44	0.46	1.67	0.88	2.17	0.46	2.01	50.71	0.06	5.31
	Mean	23.90	1.00	12.30	0.04	0.05	0.04	3.10	19.20	24.05	0.35	6.31	0.36	1.67	0.74	2.17	0.46	1.74	50.52	0.06	5.15
	Stdev	0.27	0.00	0.20	0.00	0.00	0.00	0.25	0.30	0.24	0.00	0.18	0.14	0.00	0.20	0.00	0.00	0.38	0.27	0.00	0.23
S5	Min	15.91	0.98	11.04	0.04	0.05	0.04	2.58	18.67	17.34	0.35	5.67	0.42	1.67	0.29	2.17	0.46	1.50	47.51	0.06	4.21
	Max	16.09	1.22	11.36	0.04	0.05	0.04	2.82	19.13	17.72	0.35	5.97	0.66	1.67	0.29	2.17	0.46	1.76	48.17	0.06	4.59
	Mean	16.00	1.10	11.20	0.04	0.05	0.04	2.70	18.90	17.53	0.35	5.82	0.54	1.67	0.29	2.17	0.46	1.63	47.84	0.06	4.40
	Stdev	0.13	0.17	0.23	0.00	0.00	0.00	0.17	0.33	0.27	0.00	0.21	0.17	0.00	0.00	0.00	0.00	0.18	0.47	0.00	0.27
S6	Min	22.43	1.00	11.09	0.04	0.05	0.04	2.62	19.96	16.49	0.35	5.64	0.30	1.67	0.29	2.17	0.46	0.20	47.98	0.06	3.54
	Max	22.77	1.00	11.51	0.04	0.05	0.04	2.78	20.24	16.91	0.35	6.00	0.46	1.67	0.29	2.17	0.46	0.36	48.26	0.06	3.82
	Mean	22.60	1.00	11.30	0.04	0.05	0.04	2.70	20.10	16.70	0.35	5.82	0.38	1.67	0.29	2.17	0.46	0.28	48.12	0.06	3.68
	Stdev	0.24	0.00	0.30	0.00	0.00	0.00	0.11	0.20	0.30	0.00	0.25	0.11	0.00	0.00	0.00	0.00	0.11	0.20	0.00	0.20
S7	Min	19.69	1.00	10.27	0.04	0.05	0.04	2.46	19.56	15.49	0.35	5.59	0.76	1.67	0.29	2.17	0.46	0.16	47.79	0.06	3.35
	Max	19.91	1.00	10.53	0.04	0.05	0.04	2.74	20.04	15.95	0.35	5.87	1.02	1.67	0.29	2.17	0.46	0.16	48.15	0.06	3.59
	Mean	19.80	1.00	10.40	0.04	0.05	0.04	2.60	19.80	15.72	0.35	5.73	0.89	1.67	0.29	2.17	0.46	0.16	47.97	0.06	3.47
	Stdev	0.16	0.00	0.18	0.00	0.00	0.00	0.20	0.34	0.33	0.00	0.20	0.18	0.00	0.00	0.00	0.00	0.00	0.25	0.00	0.17
S8	Min	19.07	1.00	10.10	0.04	0.05	0.04	1.74	20.77	13.53	0.35	5.67	0.25	1.67	0.29	2.17	0.46	0.21	47.47	0.06	3.67
	Max	19.33	1.00	10.50	0.04	0.05	0.04	2.06	21.03	13.89	0.35	5.87	0.43	1.67	0.29	2.17	0.46	0.43	47.79	0.06	3.97

	Mean	19.20	1.00	10.30	0.04	0.05	0.04	1.90	20.90	13.71	0.35	5.77	0.34	1.67	0.29	2.17	0.46	0.32	47.63	0.06	3.82
	Stdev	0.18	0.00	0.28	0.00	0.00	0.00	0.23	0.18	0.25	0.00	0.14	0.13	0.00	0.00	0.00	0.00	0.16	0.23	0.00	0.21

Table 3 Concentration of heavy metals (mg/kg) in sediment samples

Sediment Sample	Descriptive parameters	Al	As	Ba	Be	Cd	Co	Cr	Cu	Fe	Hg	Li	Mn	Mo	Ni	Pb	Sb	Sn	Sr	V	Zn
S1	Min	9396.0	5.5	55.4	0.4	0.1	12.2	27.2	18.1	7100.0	20.0	11.6	0.7	40.0	85.2	23.2	10.0	20.0	12.0	13.0	100.0
	Max	9404.0	5.7	55.8	0.6	0.2	12.5	27.5	18.3	7100.0	20.0	11.8	0.9	40.0	85.6	23.5	10.0	20.0	12.3	13.2	100.0
	Mean	9400.0	5.6	55.6	0.5	0.1	12.3	27.4	18.2	7100.0	20.0	11.7	0.8	40.0	85.4	23.3	10.0	20.0	12.1	13.1	100.0
	Stdev	5.7	0.2	0.3	0.1	0.0	0.2	0.2	0.2	0.0	0.0	0.2	0.1	0.0	0.3	0.2	0.0	0.0	0.2	0.2	0.0
S3	Min	13198.0	14.9	173.8	0.5	0.3	16.0	35.4	28.1	7100.0	20.0	19.8	1.4	40.0	62.4	30.7	10.0	20.0	3.4	21.5	100.0
	Max	13202.0	15.1	174.0	0.7	0.4	16.4	35.8	28.4	7100.0	20.0	20.0	1.7	40.0	62.7	30.9	10.0	20.0	3.8	21.8	100.0
	Mean	13200.0	15.0	173.9	0.6	0.4	16.2	35.6	28.3	7100.0	20.0	19.9	1.5	40.0	62.5	30.8	10.0	20.0	3.6	21.7	100.0
	Stdev	2.8	0.1	0.2	0.2	0.1	0.2	0.3	0.2	0.0	0.0	0.2	0.2	0.0	0.2	0.2	0.0	0.0	0.3	0.2	0.0
S4	Min	9594.0	8.2	51.0	0.5	0.1	12.6	21.5	20.7	7100.0	20.0	17.3	0.6	40.0	83.9	24.5	10.0	20.0	4.4	14.5	100.0
	Max	9606.0	8.4	51.2	0.6	0.2	12.9	21.9	20.9	7100.0	20.0	17.6	0.8	40.0	84.5	24.9	10.0	20.0	4.8	14.8	100.0
	Mean	9600.0	8.3	51.1	0.5	0.1	12.8	21.7	20.8	7100.0	20.0	17.5	0.7	40.0	84.2	24.7	10.0	20.0	4.6	14.7	100.0
	Stdev	8.5	0.2	0.2	0.1	0.1	0.2	0.3	0.2	0.0	0.0	0.2	0.1	0.0	0.4	0.3	0.0	0.0	0.3	0.2	0.0
S5	Min	11497.0	9.3	70.1	0.4	0.1	14.1	32.3	22.0	7100.0	20.0	15.8	0.7	40.0	42.8	27.0	10.0	20.0	7.0	16.9	100.0
	Max	11503.0	9.6	70.5	0.7	0.2	14.4	32.6	22.6	7100.0	20.0	16.1	0.9	40.0	43.1	27.5	10.0	20.0	7.2	17.1	100.0
	Mean	11500.0	9.4	70.3	0.6	0.1	14.2	32.4	22.3	7100.0	20.0	15.9	0.8	40.0	43.0	27.3	10.0	20.0	7.1	17.0	100.0
	Stdev	4.2	0.2	0.3	0.2	0.1	0.2	0.2	0.4	0.0	0.0	0.2	0.1	0.0	0.2	0.3	0.0	0.0	0.1	0.1	0.0
S6	Min	12799.0	12.3	46.5	0.5	0.3	15.3	31.9	22.6	6997.0	20.0	22.4	0.5	40.0	156.2	28.7	10.0	20.0	2.3	14.5	100.0
	Max	12801.0	12.7	47.0	0.7	0.4	15.7	32.2	22.9	7003.0	20.0	22.6	0.8	40.0	156.9	28.9	10.0	20.0	3.0	14.8	100.0
	Mean	12800.0	12.5	46.8	0.6	0.4	15.5	32.1	22.8	7000.0	20.0	22.5	0.6	40.0	156.6	28.8	10.0	20.0	2.7	14.6	100.0
	Stdev	1.4	0.3	0.4	0.2	0.1	0.3	0.2	0.2	4.2	0.0	0.1	0.2	0.0	0.5	0.2	0.0	0.0	0.5	0.2	0.0
S7	Min	10797.0	5.5	45.6	0.4	0.0	12.8	25.6	19.3	7100.0	20.0	18.3	0.4	40.0	132.1	24.8	10.0	20.0	6.0	14.8	100.0
	Max	10803.0	5.7	45.9	0.7	0.2	13.1	25.9	19.6	7100.0	20.0	18.7	0.6	40.0	132.5	25.1	10.0	20.0	6.3	15.2	100.0
	Mean	10800.0	5.6	45.8	0.6	0.1	13.0	25.7	19.5	7100.0	20.0	18.5	0.5	40.0	132.3	24.9	10.0	20.0	6.2	15.0	100.0
	Stdev	4.2	0.1	0.2	0.2	0.1	0.3	0.2	0.3	0.0	0.0	0.3	0.1	0.0	0.3	0.2	0.0	0.0	0.2	0.3	0.0
S8	Min	13098.0	9.1	57.6	0.6	0.3	15.1	35.8	21.9	7100.0	20.0	19.2	0.6	40.0	107.6	30.7	10.0	20.0	4.6	17.8	100.0
	Max	13102.0	9.4	57.8	0.9	0.5	15.4	36.0	22.4	7100.0	20.0	19.4	0.8	40.0	108.0	31.0	10.0	20.0	5.0	18.0	100.0
	Mean	13100.0	9.2	57.7	0.7	0.4	15.2	35.9	22.2	7100.0	20.0	19.3	0.7	40.0	107.8	30.8	10.0	20.0	4.8	17.9	100.0
	Stdev	2.8	0.2	0.1	0.2	0.1	0.2	0.2	0.3	0.0	0.0	0.2	0.2	0.0	0.3	0.2	0.0	0.0	0.3	0.1	0.0

Table 4 EPA River water heavy metal (µg/L) criteria for aquatic life and human health, and California State sediment quality assessment guidelines

Heavy metal	SQAG		WQC_ALC		WQC_HHC	
	TEC	PEC	acute	chronic	acute	chronic
Al	*	*	*	*	*	*
As	9.8	33	340	150	0.018	0.14
Ba	20	60	*	*	1000	*
Be	*	*	*	*	*	*
Cd	1	5	1.8	*	*	*
Co	50	*	*	*	*	*
Cr	43	110	16	11	total	*
Cu	32	150	*	*	1300	*
Fe	*	*	*	1000	*	*
Hg	0.18	1.1	1.4	0.77	*	*
Li	*	*	*	*	*	*
Mn	*	*	*	*	50	100
Mo	*	*	*	*	*	*
Ni	23	49	470	52	610	4600
Pb	36	130	65	2.5	*	*
Sb	*	*	*	*	5.6	640
Sn	*	*	*	*	*	*
Sr	*	*	*	*	*	*
V	*	*	*	*	*	*
Zn	120	460	120	120	7400	26000

Table 5 Percentage of samples exceeding EPA river water heavy metal criteria (µg/L) for aquatic life and human health, and California State Sediment Quality Assessment Guidelines

Heavy metal	SQAG		WQC_ALC		WQC_HHC	
	TEC	PEC	acute	chronic	acute	chronic
As	28.57	0	0	0	100	100
Ba	100	28.6	_	_	0	_
Cr	0	0	0	0	100	_
Hg	100	100	0	0	_	_
Ni	100	85.7	0	0	0	0

The *CF* of Hg, Mo, Sb, and Sn due to concentrations below the detection limit, Zn in all samples, and Fe in all samples (except S6, which faced a slight decrease in concentration and its *CF* decreased (0.99) were equal to 1 due to constant concentrations at all sampling points and therefore had a low contamination level in river sediments. The metals Al, Be, Co, Cu, Li, Pb, V, As, and Cd (except at S7, where their *CF* for As and Cd was ≤1) had *CF* values in the range of medium contamination levels. Cr in S4 and S7, Ba in S4, S6, and S7, and Ni in S3, S4, and S5 showed *CF* less than 1, indicating low contamination levels; in other locations, they had a medium contamination level. Mn also had *CF* > 1only in two points, S3 and S5, showing a medium pollution level; in the rest points, the contamination level was low. The maximum *CF* of Al, As, Ba, Be, Co, Cr, Cu, Mn, Pb, and V was recorded at S3 (the first point after leachate discharge), St at S5, Li, and Ni at S6, and Cd at S8. The other six heavy metals also had constant *CF* due to constant concentrations or below the detection limit. Mir Mohammad Ali et al. found in their study that the CF of Pb, As, and Cr was low in most of the sampling points in winter (<1), and only Cd had medium contamination level. In winter, Cr and Pb also showed medium contamination level in some points. The overall average value of *CF* in the two seasons was also Cd > Pb > Cr > As, respectively, which did not match the present study, which had the highest average for As (Ali et al., 2022). Varol also concluded in his study on the Tigris River that the highest *CF* values of the 10 metals studied were at the first point, which was the place of copper mine wastewater discharge. Different metals at this point showed different levels of contamination. Moderate contamination levels were also obtained for other metals at other points except for Pb. High *CF* values were also obtained at S3 and S7 due to the discharge of wastewater from different cities, which was consistent with the present study (Varol, 2011). Al, Cr, Fe, Co, Ni, and U had *CF* in the low contamination level range in the seven rivers studied in the study by Ustaoğlu and Saiful Islam, Pb and Cd had CF with significant contamination levels in three rivers, and Cu, Zn, As, Mn, Pb and Cd metals had *CF* values at moderate contamination levels in some rivers (Ustaoğlu & Islam, 2020).

3.2.2 PLI

PLI in river water varied between 0.59 (S8) and 0.78 (S2), which confirms the unpolluted level at all points along the river. This index in sediments also varied from 0.98 to 1.28, and at two points, S7 and S4, with PLI values of 0.98 and 0.99, respectively, there was an unpolluted level, and at other points with values higher than 1, it indicated an average level of pollution. The highest PLI value in sediments was also obtained at S3, the first point after the leachate discharge site. The average PLI value in sediments (1.1) indicates the average level of pollution caused by heavy metals in the river course. A comparison of the PLI values in the two water and sediment sections is shown in Fig. 2. According to Fig. 2, it can be seen that PLI in river water has a decreasing trend along the course, but in river sediments, the PLI value has gradually increased; this trend of changes could be due to the transfer of metals leached from upstream to downstream and their accumulation in the sedimentary environment of the river. In a study conducted by Amirul Islam on the Halda River, the value of this index was greater than 1 in all the studied points in sediments and had a moderate pollution level (1 < PLI < 2), which was consistent with other points except for S4 and S7 in this study (Islam et al., 2020). Saiful Islam et al. also found that the PLI was higher than 1 in all points and in both seasons of the study for heavy metals Cr, Ni, Cu, As, Cd, and Pb in the Buriganga River, and very high values of this index in points B4 to B8 were attributed to the possible effects of various industries and urban activities (Islam et al., 2018).

Table 6 Contamination factor for heavy metals in different river environments

Type Of Environment	Sampling Site	Al	As	Ba	Be	Cd	Co	Cr	Cu	Fe	Hg	Li	Mn	Mo	Ni	Pb	Sb	Sn	Sr	V	Zn	Ave
Water	S1	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
	S2	0.42	1.00	0.89	1.00	1.00	1.00	0.82	1.06	0.32	1.00	0.93	0.36	1.00	0.95	1.00	1.00	0.63	0.98	1.00	0.31	0.83
	S3	0.24	1.00	0.93	1.00	1.00	1.00	0.99	0.99	0.20	1.00	0.95	0.43	1.00	0.80	1.00	1.00	0.80	1.00	1.00	0.18	0.83
	S4	0.26	1.00	0.99	1.00	1.00	1.00	1.11	0.95	0.18	1.00	1.02	0.11	1.00	2.05	1.00	1.00	1.21	1.05	1.00	0.18	0.91
	S5	0.17	1.06	0.91	1.00	1.00	1.00	0.96	0.94	0.13	1.00	0.94	0.17	1.00	0.80	1.00	1.00	1.14	0.99	1.00	0.15	0.82
	S6	0.25	1.00	0.91	1.00	1.00	1.00	0.96	1.00	0.12	1.00	0.94	0.11	1.00	0.80	1.00	1.00	0.20	1.00	1.00	0.13	0.77
	S7	0.22	1.00	0.84	1.00	1.00	1.00	0.93	0.98	0.12	1.00	0.92	0.27	1.00	0.80	1.00	1.00	0.11	1.00	1.00	0.12	0.77
	S8	0.21	1.00	0.83	1.00	1.00	1.00	0.67	1.04	0.10	1.00	0.93	0.10	1.00	0.80	1.00	1.00	0.22	0.99	1.00	0.13	0.75
	Ave	0.35	1.01	0.91	1.00	1.00	1.00	0.93	1.00	0.27	1.00	0.95	0.32	1.00	1.00	1.00	1.00	0.66	1.00	1.00	0.28	0.83
Sediment	S1	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
	S3	1.40	2.69	3.13	1.13	2.56	1.31	1.30	1.55	1.00	1.00	1.70	1.92	1.00	0.73	1.32	1.00	1.00	0.30	1.66	1.00	1.43
	S4	1.02	1.49	0.92	1.05	1.01	1.04	0.79	1.14	1.00	1.00	1.49	0.90	1.00	0.99	1.06	1.00	1.00	0.38	1.12	1.00	1.02
	S5	1.22	1.69	1.26	1.12	1.03	1.15	1.19	1.22	1.00	1.00	1.36	1.04	1.00	0.50	1.17	1.00	1.00	0.58	1.30	1.00	1.09
	S6	1.36	2.24	0.84	1.13	2.54	1.25	1.17	1.25	0.99	1.00	1.91	0.79	1.00	1.83	1.23	1.00	1.00	0.22	1.12	1.00	1.24
	S7	1.15	1.00	0.82	1.09	0.67	1.05	0.94	1.07	1.00	1.00	1.58	0.62	1.00	1.55	1.07	1.00	1.00	0.51	1.14	1.00	1.01
	S8	1.39	1.65	1.04	1.37	2.80	1.24	1.31	1.22	1.00	1.00	1.64	0.87	1.00	1.26	1.32	1.00	1.00	0.40	1.37	1.00	1.24
	Ave	1.22	1.68	1.29	1.13	1.66	1.15	1.10	1.21	1.00	1.00	1.53	1.02	1.00	1.12	1.17	1.00	1.00	0.48	1.24	1.00	1.15

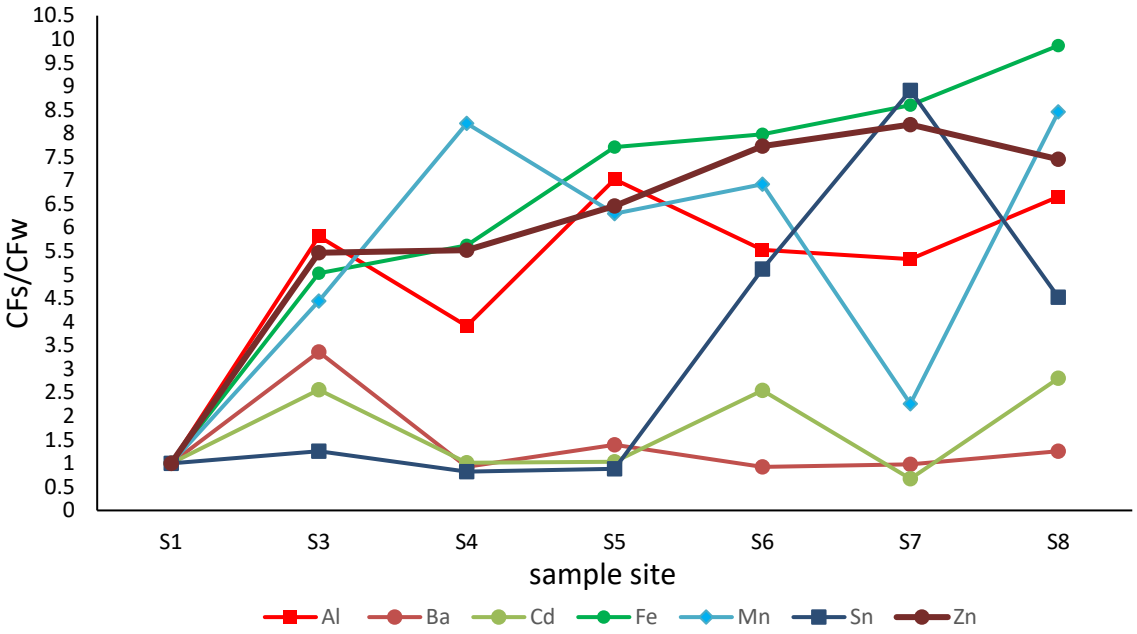


Fig. 1 Changes in the ratio of sediment to water contamination factor in the river for heavy metals

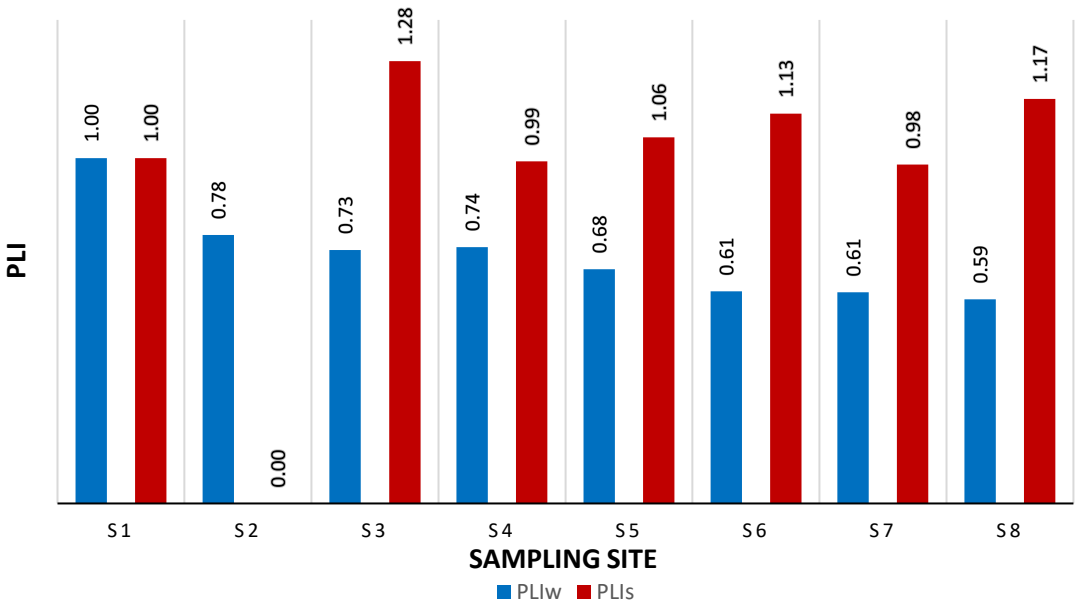


Fig. 2 Comparison of PLI index between water and sediment

3.2.3 EF

The *EF* value in S1 (background concentration) and also for Fe (reference metal) was calculated to be equal to 1. The lowest *EF* value in water samples was obtained for Al and Zn at S3, for Mn at S4, and for Sn at S7, and for other metals at the leachate entry point, i.e., S2. The maximum value of this factor was also recorded for Cr and Mn at S7, Sn at S5, and Ni at S4, and for other metals at S8. The trend of *EF* changes in the water at the studied sites is shown in Fig. 3. The *EF* value for all metals along the river course and at all points generally increased except for four metals: Cr, Mn, Ni, and Sn; among which the EF value of Cr decreased after S7 and Mn also showed a trend of consecutive increase and decrease and after reaching its maximum value at S7, it finally decreased at S8. The EF value of Ni also continued its gradual increase after a sudden increase and reaching a maximum value at S4, and then a sudden decrease at S5. The trend of changes in the *EF* of Sn was also reversed and was a gradual increase to a maximum value at S5, and then a sudden decrease and reaching a minimum value at S7, and an increase again at S8. The overall average *EF* of all metals at each point along the sampling route was S8 (7.41) > S7 (6.58) > S5 (6.31) > S6 (6.24) > S4 (5.09) > S3 (4.15) > S2 (2.63) > S1 (1.00). Accordingly, S1 (background) was considered without enrichment, S2 had low enrichment, S3 had moderate enrichment, and S4 to S8 had relatively severe enrichment.

The trend of changes in *EF* of the studied heavy metals in sediments is shown in Fig. 4. In sediments, the *EF* of Hg, Mo,

Sb, and Sn due to concentrations below the detection limit and Zn due to constant concentrations at all sampling points and also the stability of the amount of Fe present in the sediments as a reference metal (except at S6, which was accompanied by a slight decrease and caused the recording of the maximum *EF* for these metals) was equal to 1. The maximum EF in sediments was obtained for St in S5 and for Be, Cd, Cr, and Pb at S8, Ni, and Li at S6, and for other metals at S3. The minimum *EF* value was obtained for Li Ni at S5 and for St at S6.

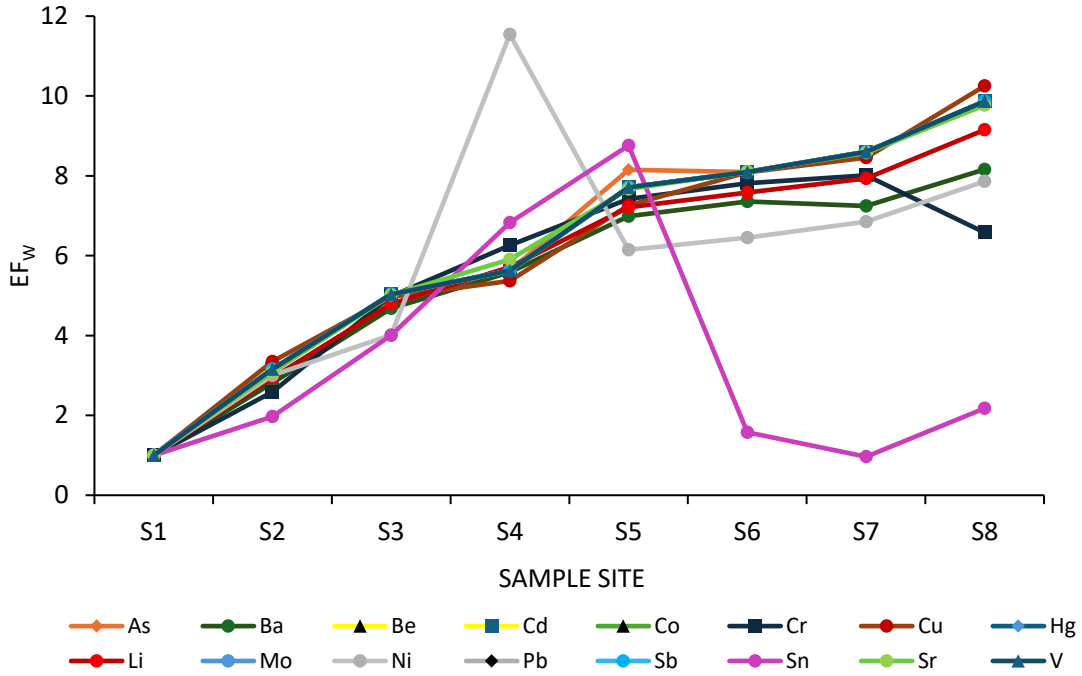


Fig. 3 EF Change trends for heavy metals in river water

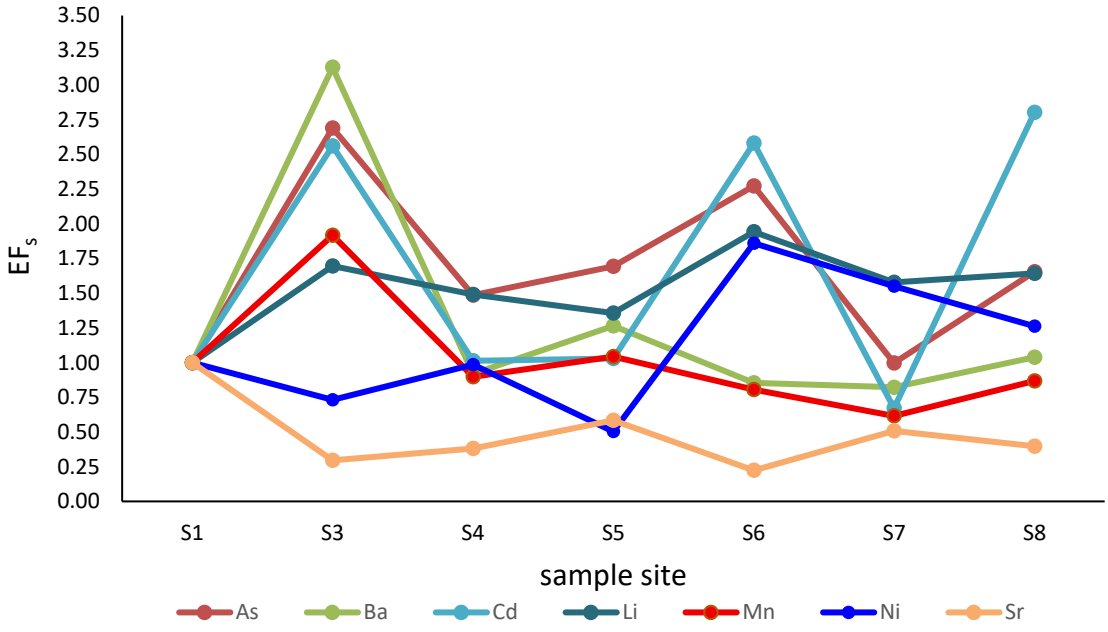


Fig. 4 EF change trends for heavy metals in river sediment

The *EF* value for As, Ba, Cd, Cu, and Mn was also recorded at the minimum value at S7, and the other metals had the minimum *EF* value at S4. The *EF* values for different metals in river sediments followed different patterns. Some metals, such as arsenic and barium, initially had the maximum *EF* value (at the point of leachate entry) and then followed a decreasing trend along the river course. Some other metals, such as Cr and Pb, had the opposite trend, and their *EF* values changed from low to high. Others, such as Ni and Li, first increased and then decreased. The average *EF* of total metals at different sampling points for sediments was S3 (1.43) > S6 (1.26) > S8 (1.24) > S5 (1.09) > S4 (1.02) > S7 (1.01) > S1

(1.00), respectively. Therefore, S1 was without enrichment, and all other points had low metal enrichment. EF values greater than 1.5 indicate metal contamination as a result of human activity, and values around 1 indicate the natural concentration of metals and the effect of weathering processes and entry due to crustal compounds (Islam et al., 2015). *EF* study of both aquatic and sedimentary environments shows that most heavy metals in water have human sources, while the amount of this factor in sediments for most metals is less than 1.5 in most places, indicating the natural origin of these metals. The contradictory trend of this factor for some metals and its increase and decrease in sampling points can indicate the presence of polluting sources downstream of the leachate entry point. Field studies showed that in the study route, there are rural areas, cultivated lands, asphalt plants, and sand and gravel plants. Therefore, EF changes can be caused by the entry of rural wastewater into the river or the entry of agricultural wastewater containing agricultural toxins and pesticides. It is also possible that the asphalt plant and sand and gravel plants play a major role in causing *EF* changes because sand and gravel extraction activities can cause the release and washing of heavy metals, or the wastewater from their processing can enter water sources. Asphalt production through asphalt binder, asphalt additives, and recycled concrete aggregate can also introduce various metals, including V, Ni, Zn, Mo, Cu, Fe, Ag, and As into the river through the resulting effluent (Park, 2024; Rentier & Cammeraat, 2022; Spreadbury et al., 2021; Von Gunten et al., 2020). In their study on Karun River sediments, Keshavarzi et al. examined EF of nine heavy metals and concluded that Cd, Cr, As, and Cu were in the pollution-free range at most sampling points. Moderate enrichment (2<EF<5) of Ni in all samples, moderate enrichment of Pb (caused by municipal wastewater and oil refinery), and Zn in most samples, as well as severe to very severe enrichment due to Hg in most samples were other results of this study that were attributed to the discharge of municipal and industrial wastewater containing these metals. Although these results were in conflict with the present study, they can confirm the effect of human activities on the increase of heavy metals in river sediments (Keshavarzi et al., 2015). Varol and Şen found in their study in the upper reaches of the Tigris River that the discharge of metal-containing wastewater from point sources (copper mining) into the river can increase the concentration and, consequently, the enrichment factor of metals such as Cd, Co, Cu, Ni, Pb and Zn in sediments at downstream points of the receiving river (Varol & Şen, 2012).

3.2.4 I_{geo}

I_{geo} of Hg, Mo, Sb, and Sn was recorded as constant and negative due to concentrations below the detection limit in all sample points and Zn in all points and Fe in all points except S6 (although the value of this index was negative at this point too) due to constant concentrations in the sediments, which indicates the lack of contamination of the sediments by these metals. Al, Be, Cr, Co, Pb, and St were negative at all points and therefore were free of contamination in the river sediments. I_{geo} values of Ba (1.06), Cu (0.05), Mn (0.35), and V (0.14) were positive, and the highest value at S3, and Ba was in the almost contaminated group, and the other three metals were in the non-contaminated to almost contaminated group. Cd at S3 and S8 and Ni at S6 and S7 had values between 0 and 1 and were in the non-contaminated to almost contaminated group.

Table 7 Geoaccumulation index of heavy metals in river sediment

Sampling site	1	3	4	5	6	7	8
Al	-0.58	-0.10	-0.55	-0.29	-0.14	-0.38	-0.11
As	-0.58	0.84	-0.01	0.17	0.58	-0.59	0.14
Ba	-0.58	1.06	-0.71	-0.25	-0.83	-0.87	-0.53
Be	-0.58	-0.40	-0.52	-0.42	-0.41	-0.46	-0.13
Cd	-0.58	0.77	-0.57	-0.54	0.76	-1.16	0.90
Co	-0.58	-0.19	-0.53	-0.38	-0.26	-0.51	-0.28
Cr	-0.58	-0.20	-0.92	-0.34	-0.36	-0.67	-0.19
Cu	-0.58	0.05	-0.39	-0.29	-0.26	-0.49	-0.30
Fe	-0.58	-0.58	-0.58	-0.58	-0.61	-0.58	-0.58
Hg	-0.58	-0.58	-0.58	-0.58	-0.58	-0.58	-0.58
Li	-0.58	0.18	-0.01	-0.15	0.35	0.07	0.13
Mn	-0.58	0.35	-0.74	-0.52	-0.92	-1.28	-0.79
Mo	-0.58	-0.58	-0.58	-0.58	-0.58	-0.58	-0.58
Ni	-0.58	-1.03	-0.60	-1.58	0.29	0.05	-0.25
Pb	-0.58	-0.18	-0.50	-0.36	-0.28	-0.49	-0.18
Sb	-0.58	-0.58	-0.58	-0.58	-0.58	-0.58	-0.58
Sn	-0.58	-0.58	-0.58	-0.58	-0.58	-0.58	-0.58
Sr	-0.58	-2.34	-1.98	-1.36	-2.77	-1.56	-1.92
V	-0.58	0.14	-0.42	-0.21	-0.43	-0.39	-0.13
Zn	-0.58	-0.58	-0.58	-0.58	-0.58	-0.58	-0.58

As was found to be uncontaminated to almost contaminated at S3, S5, S6, and S8, and at uncontaminated levels at other sites. Li was also found to be negative at S4 and S5 and at uncontaminated to almost contaminated levels at other sites. The order of I_{geo} of total metals at the sampling sites was S3 (-4.57) > S8 (-7.14) > S6 (-8.2) > S5 (-10.02) > S1 (-11.7) > S4 (-11.98) > S7 (-12.25), and thus the I_{geo} of total metals at all sites was found to be uncontaminated. The values of this index for the studied elements are presented in Table 7.

In a study conducted by Karbasi et al., it was concluded that the values of the I_{geo} calculated for the average concentration of Pb (3.78), Cd (0.57), Zn (4.36), and Cu (10.01) in the sediments of the Shoor River were in the range of highly polluted, unpolluted to almost polluted, highly polluted to very very polluted, and very very polluted, respectively, which was in contrast to the results of the present study (Karbassi et al., 2008). Salati and Mori also found in a study conducted on heavy metals (Cr, Mn, Ni, Cu, Zn, Pb, and Cd) in the dry River that except for manganese, which had negative values at all sampling points and was at the unpolluted level, and Cd, which had values less than zero only at S1, the other metals showed different levels of contamination with this index at other sampling points (Salati & Moore, 2010). Nur Fadlillah et al., in their study, assessed the ecological risk of heavy metals Pb, Cu, Cd, Cr, Fe, and Al in the Winongo River and concluded that the land accumulation index for aluminum and iron had different levels of contamination, and the remaining metals were at a level without contamination (Fadlillah et al., 2023).

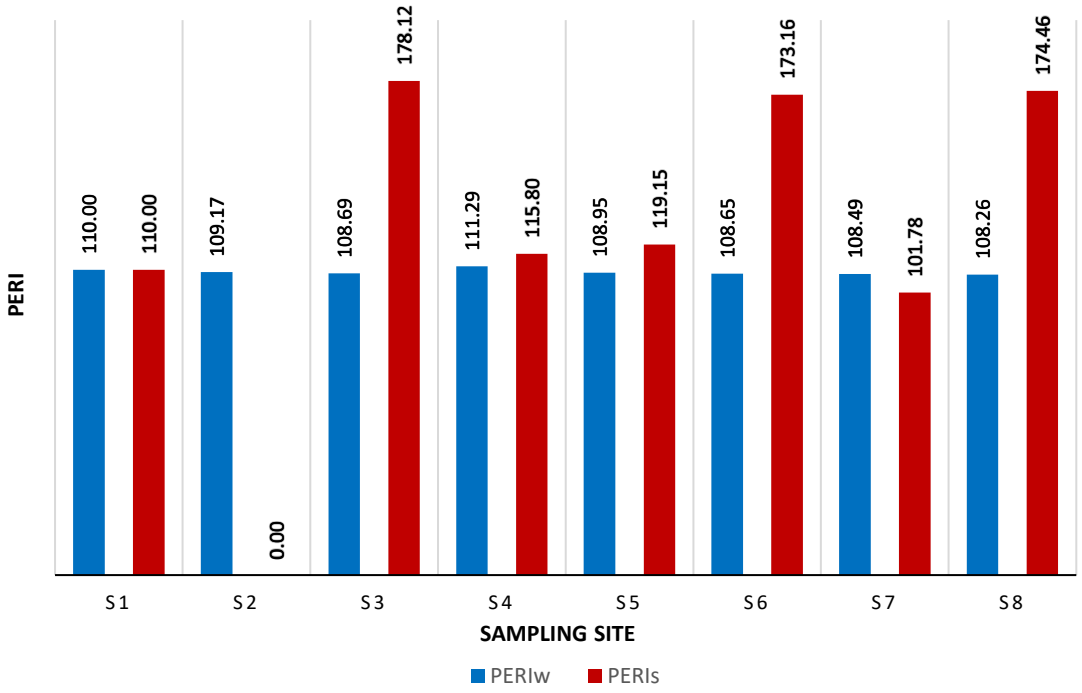


Fig. 5 Comparison of PERI index between the water and sediment of the river

3.2.5 PERI

The comparison of the results of PERI calculations in the two environments of water and river sediments is presented in Fig. 5. In performing this parameter, values lower than the detection limit of metals were considered equal to the detection limit. According to this figure, the PERI at all points in the river water was recorded in an almost constant range (108.26 - 111.29) and was placed in the group with a low ecological risk level. However, the level of PERI in river sediments at S3 (the highest value), S6, and S8 increased and were placed in the range with a medium ecological risk level. Sheykhi and Moore, in a study they conducted on the Kor River, found that river sediments at points 4, 5, 8, and 9 had a very high ecological risk degree and at points 2 and 6 had a high ecological risk, which did not correspond to the findings of the present study (Sheykhi & Moore, 2013). Banu et al. also concluded in their study on the Turag River that the PERI of the five metals studied was low in the first four points and moderate in only point 5 (Banu et al., 2013). In the four rivers affected by mining activities studied by Jimenez-Oyola et al., the PERI from heavy metals was high in the Fermı´n River and very high in the other rivers, with the highest contribution of As and then Cd (Jiménez-Oyola et al., 2021).

4. Conclusion

The present study investigated the effects of landfill leachate on heavy metal pollution in the water and sediments of the receiving water body, the Garan River, in Marivan County, Iran. Using heavy metal pollution indices (CF, PLI, I*geo*, EF, and PERI), it was found that although the overall level of heavy metal pollution in water and sediments of the river is not very high, some metals such as arsenic and chromium in water have acute and chronic effects on human health. In addition, arsenic in sediments exceeded the threshold concentration, and barium, nickel, and mercury exceeded the threshold concentrations, indicating possible effects and potential ecological risks for aquatic organisms. The trend of changes in pollution indices such as CF, PLI and EF indicates the gradual

accumulation of metals in sediments and their transfer to the downstream of the river. PLI and PERI indices showed that sediments in some places are exposed to moderate ecological risk. Given the ecological importance of the Garan River and its human uses, periodic monitoring and preventive measures are recommended to reduce the entry of pollutants and maintain the quality of the river. Given the increasing trend of PLI in sediments and EF in river water in the downstream reaches of the river, further metal surveys are recommended at further distances, including at the Garan Dam.

Statements and Declarations

Ethical considerations

Ethical issues (Including plagiarism, informed consent, misconduct, data fabrication and/or falsification, double publication and/or submission, redundancy, etc.) have been completely observed by the author.

Data availability

Data will be made available on request.

Conflicts of interest

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

Author contribution

B. Ahmadi: Writing the main Draft; H. Mohammadi, H. Salehzadeh, B. Ghaderi, and P. Yonessi: Conceptualization; Writing – Review & Editing; and B. Shahmoradi: Conceptualization, Methodology, Review, and Final Proof.

AI Use Declaration

During the preparation of this manuscript, the authors used AI tools for designing the Graphical Abstract. All content has been carefully reviewed and revised by the authors, who take full responsibility for the final version of the manuscript.

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