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TECHNICAL ARTICLE

Mercury Contamination in Water and Sediments and the Associated Health Risk: A Case Study of Artisanal Gold-mining

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Abstract

This study investigated mercury (Hg) contamination in the Gilgit-Baltistan, Pakistan. Water and sediment samples were collected from various sites having artisanal gold-mining along the Indus, Gilgit, and Hunza Rivers. Sediments were classified as wet sediment (WS, collected just after amalgamation), fresh dry sediment (FDS, collected after 1–2 h of amalgamation), and old dried sediment (ODS, collected after 1–2 days of amalgamation). Samples were analyzed for Hg by atomic absorption spectrophotometry with a mercury hydride system. Mercury mean concentrations were 2767 mg/kg in WS, 1917 mg/kg in FDS, and 191 mg/kg in ODS. These results revealed very high levels of Hg contamination in the WS and FDS. The Hg contamination of the spring (drinking water) did not exceed the permissible limits set by the World Health Organization (WHO) and Pakistan's environmental protection agency (Pak-EPA); however, the stream water surpassed these limits. Therefore, the Hg levels in the drinking water was used to assess the potential human health risk via the average daily dose (ADD) and hazard quotient (HQ). The ADD values were 0.03 and 0.06 µg/kg-day and HQ value of 0.9 and 2.0 for adults and children, respectively. This level of Hg contamination level is alarming and could be hazardous in future.

Keywords Average daily dose · Drinking water · Hazard quotient · River water · Pakistan

Introduction

Mercury (Hg) is one of the most potentially hazardous elements (Okpala et al. 2017). Mercury contamination in the environment is mainly (37%) released when it is used to scavenge gold from very fine-grained sediments in artisanal gold-mining activities (Riaz et al. 2018; UNEP 2013). During amalgamation process, the Hg is evaporated by roasting, leaving behind the raw gold (Bose-O'Reilly et al. 2008). The vaporized Hg eventually settles in the surrounding environment, including water, sediments, and soils (Biber et al. 2015; Gibb and O'Leary 2014; UNEP 2013). Due to dynamic interaction, the sediment in aquatic systems can acts as a potential sink and source in the biogeochemical cycling of Hg (Tong et al. 2013). The inorganic Hg can be mobilized and transformed by microorganisms into more mobile and toxic organic forms, including monomethylmercury (MMeHg; CH_3Hg^+) and dimethylmercury (DMeHg; $(CH_3)_2Hg$) (Leopold et al. 2010). The organic Hg is absorbed by water plankton, which is consumed by other zooplanktons and fish (Gibb and O'Leary 2014; WHO 2007). The Hg that deposits on the soil is absorbed by plants and can contaminate the aquatic and terrestrial food chain (Riaz et al. 2018; Zhong et al. 2018).

Unfortunately, artisanal gold mining is still practiced in developing countries (Olivero-Verbel et al. 2015) like Pakistan (Riaz et al. 2018). Environmental and health problems associated with artisanal gold mining have been reported (Fernández-Martínez et al. 2005; Gibb and O'Leary 2014; Tomicic et al. 2011). Human populations are very sensitive to inorganic and organic Hg contamination (Martínez-Salcido et al. 2018; Rodrigues et al. 2014). The inhalation of Hg can damage the kidneys, lungs, digestive, nervous, and

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immune systems and could be lethal (WHO 2007). Various studies had observed Hg as an agent of numerous human health problems (e.g. neurological, reproductive, cardiac, immunological, nephrological, and genetic) (Burch et al. 2014; Gibb and O'Leary 2014; Langeland et al. 2017). Other problems caused by exposure to higher Hg doses include damage to the lungs, kidneys, heart, brain, and immune system, and headaches, weakness, emotional changes, tremors, insomnia, changes in nerve responses and disturbances in sensations (Alizadeh et al. 2017; Kim et al. 2016; Turaga et al. 2014).

Artisanal gold-mining provides jobs to approximately 15 million people, including 4.5 million women and 600,000 children in more than 50 countries. Globally, 20–30% of gold production (500–800 tonnes) is carried out by artisanal mining practice (Veiga et al. 2004). Since Hg is a cheap and easily available scavenger for gold, it is most commonly used for artisanal mining in developing countries. Each gram of gold needs 1–2 g of Hg, which is dumped into the surrounding environment by the roasting process (Craw et al. 2015; Veiga et al. 2006). Artisanal gold-mining has been practiced along the Indus, Gilgit, and Hunza Rivers in Gilgit-Baltistan for the last four decades (Shah and Khan 2004). However, so far, no environmental study had been carried out to identify the level of Hg contamination in the area. Therefore, the present study was undertaken to determine the Hg contamination levels in water and sediments. Furthermore, the Hg level in drinking water was evaluated for its potential human health risk.

Materials and Methods

Study Area Description

Gilgit-Baltistan Province has a population of 1.44 million in an area of 72,971 km^2 (Riaz et al. 2018). The study area hosts mineral resources and is rich in biodiversity. It has a mean maximum temperature of 36 °C and minimum 16 °C. The monsoon winds are blocked by the high mountain terrains of Nanga Parbat, making the study area dry and rugged with a minimum precipitation of 2.1 mm in the month of November and a maximum of 28.3 mm in April (IUCN 2003). Gilgit-Baltistan is located up-gradient in northern Pakistan and supplies water to the Indus River and groundwater (aquifers). Gilgit-Baltistan is a remote and hilly area, where people use stream and spring water for drinking and domestic purposes.

Mineralogically, the area hosts stream sediments composed of rock fragments, quartz, feldspar, and various light and heavy minerals. The light minerals are mainly mica, chlorite, pyroxene, olivine, and glauconite, while the heavy minerals are mainly comprised of magnetite, garnet, tourmaline, apatite, amphibole, zircon, and traces of gold (Riaz et al. 2018; Zain 2010). The gold is extracted as panned concentrates from the stream sediments along the banks of the Hunza, Gilgit, and Indus Rivers. The grain size of the bulk sample ranged from < 64 mm to < 62 µm, while the panned concentrates and tailings grain size generally ranged from 2 mm to < 62 µm. There tailings have no acid generation potential as it is only sand to silt size sediment, which is usually washed out after panning and Hg amalgamation. The riverbanks are regularly washed during the summer high flow flood season.

In Gilgit-Baltistan province, hundreds of families are engaged in artisanal gold-mining/panning along the rivers and streams beds. These families, known as gold washers, live as nomads, moving from one place to another in search of gold traps. They place their tents wherever they find a gold trap along the riverbed and start panning the river sediments for gold. The mining and extraction practices peak during cold weather (November-to-February) due to the low river discharge, and completely stop during the summer months (June-to-August) due to high discharges coming from glaciers melting and rainfall. During the field visit, it was found that these gold washers were busy the whole day washing the gravel-size sediments along the riverbank through handmade screens and then panning the sediment in small pans. These panned concentrates (2 mm to < 62 µm) were subsequently hand-mixed with a few drops of Hg; the gold grains were amalgamated in Hg by rubbing and mixing the panned concentrates for about 10 min. The amalgamated panned concentrates were then washed several times and the gold-containing amalgam was roasted, evaporating the Hg and leaving behind the raw gold flakes. The gold washers sell the raw gold to goldsmiths, either in their shops or on site. The Hg-amalgamated panned concentrates are discarded as tailings material along the riverbank. In this way, hundreds of families of gold washers are contaminating the river water and sediments.

Sample Collection

Representative water and sediment (river bed and amalgam) samples were collected in a radius of 50–80 m from 25 selected sites in the study area (Fig. 1). Water samples ($n = 46$; 1 L each) were collected from various sources, including drinking water (springs and stream), river water, and wastewater (used for washing amalgam), and filtered and acidified with nitric acid (HNO_3). Sediment samples ($n = 52$;

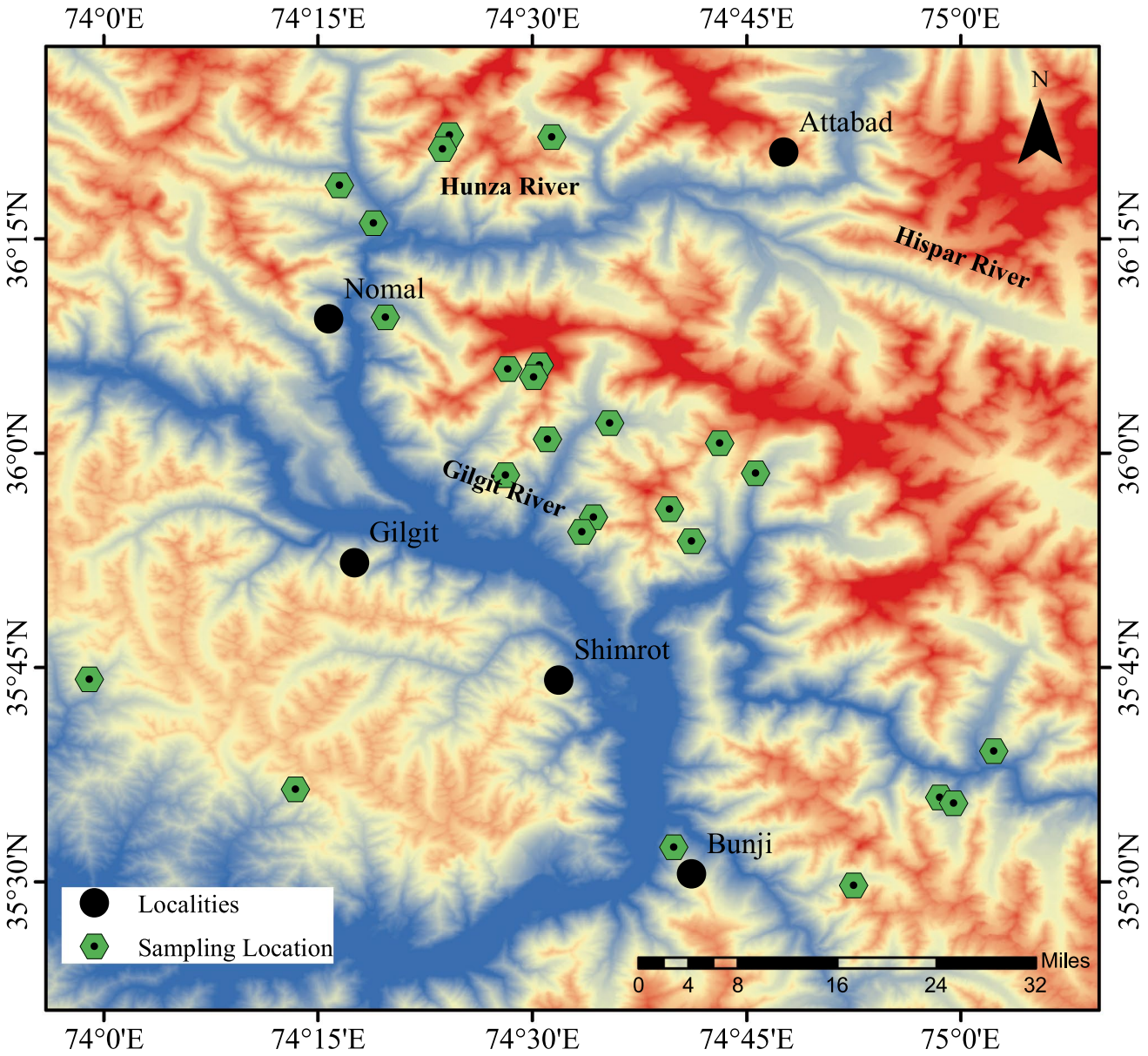

Fig. 1 Location map of the study area showing the gold extraction sites along with the rivers of Hunza, Gilgit and Indus modified from (Riaz et al. 2018)

2 kg each) were collected in polyethylene zip lock bags and classified as wet sediment (WS, collected just after amalgamation), fresh dry sediment (FDS, collected 1–2 h after the completion of amalgamation), and old dried sediment (ODS), collected after 1–2 d of amalgamation. In addition, sediment samples ($n = 25$; 2 kg each) were also collected from the beds of selected rivers. Geographic coordinates of each sampling site were measured by global positioning system (GPS) and used for distribution mapping of the study area. Sediment samples were transported, air dried in the shade to avoid Hg evaporation, and pulverized in a vibrating disc mill fitted with 200 µ mesh.

Sample Extraction and Analysis

Sediments were extracted for Hg determination using hydrofluoric acid (HF), aqua regia (HNO_3 and hydrochloric acid, HCl, at a 1:3 ratio) as advised (Van Straaten 2000) with some modification. Briefly, 2 g of sediment was mixed with 10 mL of HF in a Teflon beaker and heated at 100–120 °C for one hour. Next, 20 mL of aqua regia solution was added and heated at the same temperatures until only a small portion was left. The solution was cooled at room temperature, filtered using filter paper of glass microfiber, and diluted to 30 mL with deionized water.

The water and dissolved sediment samples were analyzed for Hg using sodium tetrahydrobromate ($NaBH_4$) with diluted HCl as reducing agent by an atomic absorption spectrophotometer (AAS, Perkin Elmer-700) supported with a mercury hydride system (MHS-15).

For data precision and accuracy assessment, the sediment samples, blanks (without any sample), and standard reference material GBW07404 (GSS-4) were extracted and analyzed using the same procedures. The standard solutions were prepared by dilution of 1000 mg/L certified standard solution Fluka Kamica (Bucha, Switzerland) of Hg with 15% concentrated HNO_3 and a few drops of $KMNO_4$.

Health Risk Assessment

Human health risk assessment was measured through the exposure risk assessment, i.e. average daily dose (ADD) and chronic risk, i.e. hazard quotient (HQ). The exposure risk through ADD values was calculated using the USEPA (2002) equation:

$$\mathrm{ADD} = C_{\mathrm{Hg}} \times W_{\mathrm{V}}/B_{\mathrm{w}}, \quad (1)$$

where C_{Hg} represents the concentration of Hg, W_{v} represents an average volume of daily consumed water (2 L), and B_{w} represents the average body weight: 72 kg for adults (Muhammad et al. 2011) and 32.7 kg for children (Ullah et al. 2017).

The HQ values for estimation of chronic or non-carcinogenic risk were calculated using the USEPA (2002) equation:

$$\mathrm{HQ} = \mathrm{ADD}/\mathrm{RfD}, \quad (2)$$

where RfD represents the oral reference dose value set by USEPA (1995) for Hg (0.03 µg/kg/day) and values of HQ less than 1 is considered to be safe (Gul et al. 2015).

Questionnaire Survey

During sampling, questionnaires were filled out by individuals or their representative. Basic information such as age, weight, physical health, education, diseases, smoking or non-smoking, medications, eating patterns and habits, types of food, drinking water sources, exposure to Hg, and time period of mining workers along artisanal gold-mining and residents was collected. The questionnaire was developed from previous studies (Table 1; Basri et al. 2017; Chappells et al. 2015; Chen et al. 2011).

Data Analysis

Data manipulation such as means, standard deviation, and graphs were calculated and presented using Sigma plot (version 12.5, Systat Software, Inc.) and correlation by SPSS (SPSS ver. 21, Inc., Chicago, IL, USA).

Table 1 Questionnaire survey and socio-demographic data

Characteristics	Response	Quantity
Gender	Male	55%
	Female	45%
Age	1–15	43%
	16–65	57%
Education	None	32.1%
	Primary	47.8%
	Secondary	16.9%
	Tertiary	5.2%
Weight	Child	26.2 kg
	Adult	66.6 kg
Reported diseases		
Smoking		62%
Drinking water quantity		2 l
Drinking water source	Stream	42%
	Spring	58%
Safety measures awareness	Yes	45%
	No	55%
Safety measures use	Yes	33%
	No	67%
Income per season		1200–1500 $
Working hours		7–8 h

Results and Discussion

Mercury Concentrations in Sediments

The range of Hg concentrations in the WS, FDS, and ODS were 223–6403, 946–4185, and 432–629 mg/kg, with mean values of 2767, 1917, and 191 mg/kg, respectively (Fig. 2). The range of Hg concentrations in the Hunza, Gilgit, and Indus Rivers sediment were 0.22–6.40, 0.25–4.34, and 0.40–4.69 mg/kg with mean values of 3.70, 2.77, and 2.55 mg/kg, respectively (Fig. 2). Sediment

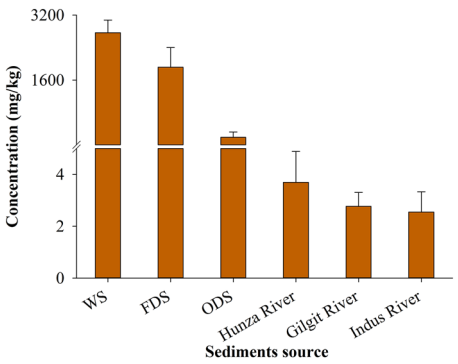

Fig. 2 Mercury concentration in sediments of the study area

are an important sink for the Hg in the aquatic ecosystem (Tong et al. 2013). Once the Hg contaminants are adsorbed/deposited on sediment, various microorganisms convert the less mobile and harmful inorganic Hg into the more highly mobile and toxic organic Hg species (AMAP/UNEP 2013; Gibb and O'Leary 2014), which can accumulate in fish, causing food chain contamination and ultimately target human consumers (Taylor et al. 2016). The Hg-enriched sediments also act as a continues source of contamination for the aquatic ecosystem and can cause ecological problems (Wang et al. 2017).

Mercury Concentrations in Water

The Hg concentrations ranged from 0.13–1.7, 0.91–2.63, and 0.07–159.0 µg/L with mean values of 0.91, 1.43, and 40.95 µg/L in the spring water, stream water, and wastewater, respectively (Fig. 3). The concentrations of Hg in the river water ranged from 0.73–159.0, 27.84–34.9, and 9.3–59.7 with mean values of 50.94, 31.37, and 28.8 µg/L in the Hunza, Gilgit, and Indus Rivers, respectively (Fig. 3). The results revealed that Hg concentrations exceeded the drinking water guideline values set by WHO (1 µg/L) and Pak-EPA (≤ 1 µg/L) (Pak-EPA 2008), however that of spring water is observed within the limits. Water contamination of the study area could be due to the release Hg from the sediments or direct deposition of the Hg that escaped during roasting (AMAP/UNEP 2013; Biber et al. 2015; Gibb and O'Leary 2014). Various environmental studies observed that higher levels of Hg in water and sediments can lead to contamination of drinking water (Gao et al. 2012) and the food chain in both terrestrial (Riaz et al. 2018; Zhong et al. 2018) and aquatic environments (Bravo et al. 2014), and ultimately pose a potential threat to the human population (Galimberti et al. 2016; Gil and Hernández 2015).

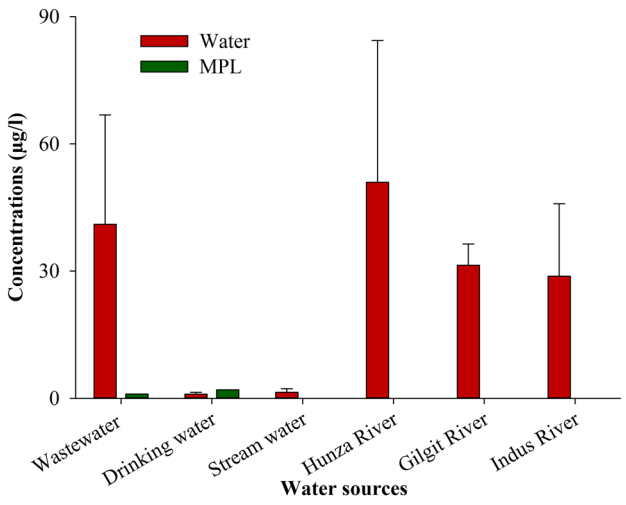

Fig. 3 Mercury concentration in water of the study area

Health Risk Assessment

The ADD values of exposure risk assessments through consumption of Hg in drinking water were 0.03 and 0.06 µg/kg-day for adults and children, respectively (Fig. 4), due to higher Hg consumption by children than adults. The HQ values were 0.9 and 2.0 for adults and children, respectively (Fig. 4). The HQ values through consumption of Hg in drinking water surpassed the threshold limit for children (HQ ≤ 1) and were very close to the limit for adults. The higher potential risk to children could be due to their higher consumption rate, susceptibility, and low body weight. The potential health risk through Hg consumption in drinking water was less than has been reported by Cobbina et al. (2011) for drinking water consumption in the upper east region, of Ghana, rice and fish in China (Liang et al. 2015), and food crops in Pakistan (Riaz et al. 2018).

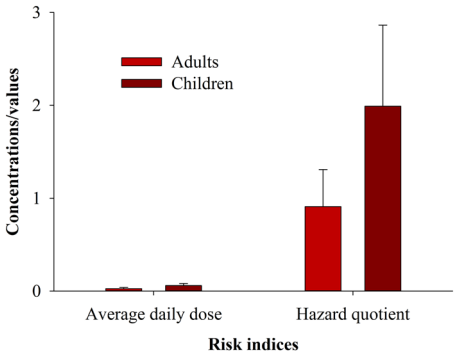

Fig. 4 Risk assessment via mercury consumption in drinking water of the study area

Table 2 Major health problems (%) occupational and non-occupational group ($n = 80$) in the study area, where n represents the number of people and NR means not reported

Health problems	Occupational		Non-occupational	
Gender	Male	Female	Male	Female
Kidney diseases	67	54	10	08
Stomach problems	65	75	32	25
Neck pain	45	58	05	02
Belly pain	87	46	NR	NR
Joint problems	51	46	09	15
Skin burn	89	98	NR	NR
Stunted growth in children	50	45	NR	NR
Heart problems	45	35	02	10
Respiratory problems	76	86	01	NR

Table 3 Pearson correlation of Hg concentrations between the water and sediments ($n=86$)

	Wastewater	Drinking water	River water	FDS	ODS	WS
Wastewater	1.000					
Drinking water	0.041	1.000				
River water	0.934[a]	0.196	1.000			
FDS	− 0.136	− 0.28	0.51	1.000		
ODS	0.534	0.008	0.996[a]	0.667[a]	1.000	
WS	0.293	0.148	0.55	− 0.161	0.242	1.000

[a]Correlation is significant at the 0.01 level (2-tailed)

[b]Correlation is significant at the 0.05 level (2-tailed)

Health Problems

Exposure to higher levels of Hg contaminations through water and food consumption or any other source can threaten the exposed ecological system (Jonsson et al. 2014) and human population (Cobbina et al. 2011; Mieiro et al. 2016). To learn the health situation in the study area, a questionnaire survey was arranged (Tables 1 and 2). The most common health impacts in workers include belly pain in both males and females, skin problems in females, and intellectual and growth problems in children. Other problems included neck and joint pain, fever, breathing problems, and kidney, stomach, and cardiovascular problems. Exposure to Hg contaminations through drinking water consumption could be the cause of all these problems. Burned skin of miners may resulted from long term exposure to solar radiation.

Correlation Matrix

Table 3 summarizes the Pearson correlation between the Hg concentrations in the water and sediment samples. The concentrations of Hg in the wastewater, drinking water, river water and ODS were positively correlated ($r=0.934$ and $r=0.996$, respectively). Significant positive correlation of the Hg in the wastewater and river water suggests that both are due to the artisanal gold mining practices in the study area. The upstream Hunza River showed higher levels of contamination than the other tributaries and the Gilgit River.

Conclusion

This study observed elevated Hg contamination levels in both the water and sediments. The Hg contamination levels in the drinking water caused a potential risk of HQ > 1 for children, due to their higher consumption rate. The observed high rate of health problems typically associated with Hg contamination could be linked to exceeding the health threshold limit.

Correlation analysis of Hg in the wastewater, stream water, and sediment suggest a common source of contamination. Government and private agencies should provide alternative techniques to avoid Hg contamination and protect the ecological environment and human population in the study area.

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