

## Contamination and Human Health Risk Assessment of Toxic Trace Elements in Drinking Water of Gilgit-Baltistan, Pakistan

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### ABSTRACT

This study investigated the contamination level and risk associated with toxic trace elements in springs' water from Gilgit-Baltistan, Pakistan. Toxic trace elements, including Hg, As, and Zn, were analyzed by metalizer, HM 2000 serial no. MY-011-006, while elements such as Cr, Al, B, Ni, Cu, Mn, and Fe were analyzed using Metalometer HM 2000 serial no. MM005-007, the United Kingdom. The mean concentrations of TTEs in water samples from Skardu were ordered as, Mn < Cu < Fe < Zn < Al < Cr < As < Ni < Hg, in Gilgit, Mn < Cu < Zn < Ni < B < Cr < Fe < As < Hg, in Ghizer Cu < Mn < Zn < Ni < Cr < Fe < As < Hg, while in Nagar the concentration of TTEs in water samples were ordered as Cu < Mn < Fe < Ni < Al < Cr < Zn < As < Hg. Results obtained from this study showed that the concentrations of As, Hg, Ni, Cr, Al, and Mn in some water samples were higher than the limits recommended by WHO and Pak-NDWQS. However, the chronic daily intake indices (CDIs) and health risk index (HRI) in all samples were found below the US-EPA standards. The correlation analysis revealed a positive association among

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different elements, which revealed that the sources of TTE<sub>s</sub> in water samples were mainly geological strata and anthropogenic activities.

*Keywords:* Correlation analysis, Gilgit-Baltistan, Pakistan springs water, risk assessment, toxic trace elements

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## INTRODUCTION

Naturally, heavy metals in the aquatic environment are very small amounts, mainly released from the natural weathering of rocks and soils (Dessie et al., 2021). However, it is clear that metal enters the aquatic system from multiple sources, both point and non-point and can be easily transported from the abiotic to the biotic system (Khan, 2011).

Currently, the effect of heavy metal pollution on human beings is becoming critical. Heavy metals are classified as essential and nonessential or toxic depending on their toxicity and nutritional value. Cu, Mn, Fe, Zn, and Co are needed in minute quantities for living beings' normal function and survival (Muhammad et al., 2019). Toxic trace elements can follow multiple pathways to enter the body, e.g., skin contact, taking foods, drinking, and breathing (Ali & Rubina, 2018). Though some trace elements are necessary for metabolic activities at small concentrations (Mohmand et al., 2015), a higher concentration may cause havoc on human health.

The most abundant toxic trace elements in the environment are Cd, Pb, Cr, As, Cu, Zn, Ni, and Pb, and prior studies have highlighted that their exposure can cause various poisoning influences on the human body because it can damage the kidney function, circulatory system, and nervous system (Yang & Massey, 2019; Zhang et al., 2015). Moreover, agglomeration of the toxic trace elements in the body brings about various ailments, while their synergistic may even cause death (Huma-Khan et al., 2016; Rasool et al., 2016). For example, zinc helps in body growth and is necessary for the normal functions of living organisms, and its deficiency causes poor wound healing, anorexia, hair loss, immune dysfunction, diarrhea, and dermatitis (Stephanie, 2010). In the same way, Alzheimer's and Manganism are the top diseases caused by prolonged exposure to a high concentration of Mn and Cu in drinking water (Dieter et al., 2005; Muhammad Tahir et al., 2020). Furthermore, the high concentration of Mn in drinking water is perilous to children's intellectual functions (Wasserman et al., 2006).

Several previous studies in different countries assessed the heavy metal contamination in different environmental samples such as water, sediment, soil, and foodstuff (Ali et al., 2019; Dessie et al., 2021; Jiang et al., 2018). Furthermore, the health risk assessment of heavy metals in tap water/drinking water has been conducted in different parts of the world, such as carcinogenic and non-carcinogenic health risk assessments in Iran (Alidadi et al., 2019; Mohammadi et al., 2019; Saleh et al., 2019), compositional and health risk assessment in Pakistan (Muhammad et al., 2011; Murtaza et al., 2020), and water quality

and human health risk assessment in China (Ji et al., 2020). Therefore, it is good to conclude by stating the two sides of the study outcomes: some with low risk and some with alarmingly high risk.

Around two million people of Gilgit-Baltistan are at risk due to the lack of safe drinking water, clear air, and hygienic food products. The number of prevailing diseases, including cardiovascular, upper respiratory tract index, cancer, hepatitis, and many more, are distressing to the public. The rapidly increasing contaminations in the northern part of Pakistan due to the growing population and unplanned infrastructures pose threats to the water quality requiring deep study.

According to our knowledge, there is limited data available on toxic trace element concentrations and their sources in the drinking water of Gilgit-Baltistan. Our studies on toxic trace element concentrations in drinking water sources are extremely important to prevent water contamination and its effects on humans in Pakistan and as well as in other countries. This study attempts to investigate the contamination level of toxic trace elements and their potential health risk in drinking water sources of Gilgit-Baltistan, Pakistan.

## MATERIALS AND METHODS

### Study Area

Gilgit-Baltistan is spread over an area of about 72,500 km<sup>2</sup> and is in northern Pakistan over the Himalayas, Karakoram, and Hindukush Mountains. The climate of Gilgit-Baltistan varies spatially; the complex topography of mountains is responsible for the substantial variability. The eastern part (western Himalayas) is relatively moist, while the climate becomes considerably drier in the Karakoram and Hindukush mount ranges (WWF, 2009). The tectonic setting of the area is highly complex, and the rocks are tremendously deformed. The Indian plate contributed mostly to the exposed rocks, made up of ultramafic and mafic rocks, i.e., greenschist, serpentinite, meta basalt, and schist (Arif et al., 2011; Muhammad et al., 2010). Due to the collision of the Indian and Eurasian plates, different metamorphosed rocks are formed. The primary rocks include peridotite, dunite, gabbros, gabbronorite, pegmatite, quartz vein, basalt, diorite, granodiorite, hornblendite, and granite. Metamorphic rocks are phyllite, schist, gneiss, quartzite, and granulite structurally (A. Khan et al., 2015). Rocks begin to change chemically at temperatures above 200°C. At these temperatures, the crystalline structures of the minerals in the rock are broken down and transformed using different combinations of the available elements and compounds (Pidwirny, 2006), thus creating minerals. Gilgit-Baltistan is divided into three divisions and ten districts, including Gilgit, Hunza, Nagar, Skardu, Shigar, Khapulo, Ghanche, Astore, Diamer, and Ghizer. The present study was conducted in four districts, including Gilgit, Skardu, Ghizer, and Nagar, as shown in Figure 1.

### Sampling Procedure

Four districts, i.e., Gilgit, Skardu, Nagar, and Ghizer, were selected for the springs' water sampling based on tectonic and geologic settings. Thirty-six triplicate samples were collected through random sampling: ten were collected from district Skardu, seven from Gilgit, twelve from Ghizer, and seven from Nagar in June, July and August 2018. The water samples were collected from the targeted sampling locations with a clean polyethylene plastic bottle. Before the sampling, bottles were pre-washed with 20% nitric acid (HNO<sub>3</sub>) and double-distilled water. Samples were filtered and acidified with HNO<sub>3</sub> before transferring them to the laboratory. All the samples were stored at 4°C in a refrigerator (USEPA, 2003). A geographical position system (GPS) was used to record the coordinates of sampling sites. The study area map and sampling sites are shown in Figure 1.

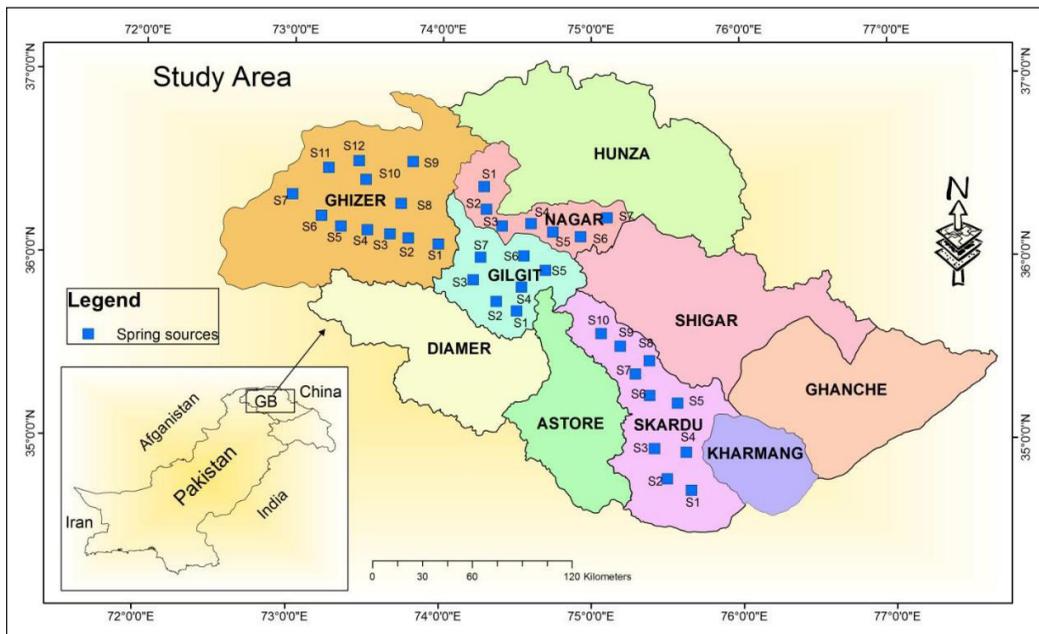


Figure 1. Study area map showing sampling sites in Gilgit-Baltistan, Pakistan

### Chemical Analysis

Water samples were analyzed in a laboratory at Gilgit-Baltistan Environmental Protection Agency (GB-EPA). The concentration of TTEs, including Hg, As, and Zn, were analyzed using Metalyzer HM 2000 Serial No. MY-011-006, while Cr, Al, B, Ni, Cu, Mn, and Fe were analyzed by Metalometer HM 2000 Serial No. MM 005-007, the United Kingdom. The lower and upper instrumental detection limits of Hg, As, and Zn were 5 µg/L and 500 µg/L, respectively. Similarly, the detection limits of Cr, Al, B, Ni, Cu, Mn, Fe were 0.02, 0.01, 0.1, 0.01, 0.05, 0.1, 0.02 mg/L and 2, 0.25, 2, 10, 5, 18, 3 mg/L, respectively.

## Human Health Risk Assessment

Among several pathways through which TTEs enter the body, oral intake is substantial (Lambert et al., 2000). Therefore, the chronic daily intakes (CDIs) of TTEs via water ingestion were calculated using Equation 1 (Krishna et al., 2009; Lambert et al., 2000).

$$CDI = C_m \times L_w / W_b \quad (1)$$

Where  $C_m$ ,  $W_b$ , and  $L_w$  are the TTEs concentration ( $\mu\text{g/l}$ ), average body weight, and daily water intake, respectively. An average  $L_w$  is 2 l/day (adult) or 1 l/day (child), whereas an average  $W_b$  is 72 kg (adults) or 32.7 kg (child), as reported (Alina et al., 2012; Sharma et al., 2008). The health risk index (HRI) was calculated using Equation 2 (Krishna et al., 2009).

$$HRI = CDI/RfD \quad (2)$$

Where  $RfD$  is the value of oral toxicity, reference dose ( $\mu\text{g/kg}^{-1}\text{day}^{-1}$ ). According to the US-EPA database, the  $RfD$  values for Zn, Cr, Mn, Cu, and Ni are  $3.0\text{E} + 02$ ,  $1.5\text{E} + 03$ ,  $1.4\text{E} + 02$ ,  $3.7\text{E} + 01$ , and  $2.0\text{E} + 01$ , respectively (Krishna et al., 2009). It is reported that if the HRI is  $< 1$ , it is considered a safe limit (Sharma et al., 2008).

## Statistical Analysis

Descriptive statistical analysis was performed by using MS Excel 2016 and OriginLab, 2019. In addition, a statistical one-way ANOVA analysis was performed for the correlation among elements using SPSS software V-17. Finally, the spatial distribution of TTEs in water samples was estimated based on the inverse distance weighted (IDW) interpolation method by using ArcGIS.

## RESULTS AND DISCUSSION

### Mean Concentrations of TTEs

The mean concentrations of TTEs in water samples collected from Skardu were ordered as,  $\text{Mn} < \text{Cu} < \text{Fe} < \text{Zn} < \text{Al} < \text{Cr} < \text{As} < \text{Ni} < \text{Hg}$ , in Gilgit as,  $\text{Mn} < \text{Cu} < \text{Zn} < \text{Ni} < \text{B} < \text{Cr} < \text{Fe} < \text{As} < \text{Hg}$ , in Ghizer  $\text{Cu} < \text{Mn} < \text{Zn} < \text{Ni} < \text{Cr} < \text{Fe} < \text{As} < \text{Hg}$ , while in Nagar the concentrations were ordered as,  $\text{Cu} < \text{Mn} < \text{Fe} < \text{Ni} < \text{Al} < \text{Cr} < \text{Zn} < \text{As} < \text{Hg}$ . The mean concentrations of Hg in Skardu, Gilgit, Ghizer, and Nagar districts were measured as  $0.0107 \pm 0.0075$ ,  $0.0005 \pm 0.0002$ ,  $0.002 \pm 0.0007$ , and  $0.0007 \pm 0.0007$  mg/L, respectively as shown in Table 1. The higher concentration of Hg in spring water sources might be due to leaching or weathering of ultramafic and mafic rocks as well as mining/anthropogenic activities (Hussain et al., 2019). The mean concentrations of As in water samples collected from Skardu, Gilgit, Ghizer, and Nagar districts were measured as  $0.0296 \pm 0.0101$ ,  $0.011$

$\pm 0.0030$ ,  $0.011 \pm 0.0033$ , and  $0.01 \pm 0.0038$ , respectively. The highest concentration of As ( $0.18 \text{ mg/L}$ ) was measured in water samples from Skardu, which might be due to weathering/leaching of ultramafic and mafic rocks. The mean concentrations of Cr in Skardu, Gilgit, Ghizer, and Nagar districts were  $0.03 \pm 0.0557$ ,  $0.02 \pm 0.0054$ ,  $0.032 \pm 0.0050$ , and  $0.014 \pm 0.0041 \text{ mg/L}$ , respectively.

The highest concentrations of Cr were detected in Skardu and Ghizer ( $0.07 \text{ mg/L}$ ). Cr is used extensively in industrial activities, such as cement dyeing, metal cleaning, leather tanning, and electroplating. Moreover, agricultural activities and weak and corrosive plumbing may have added Cr into the environment (Kisku et al., 2011). It is reported that a certain quantity of Cr is necessary for growth and different functions; however, the high concentrations of Cr are genotoxic, carcinogens, and cause liver and kidney ailments (Huma-Khan et al., 2016). The mean concentrations of Ni in Skardu, Gilgit, Ghizer, and Nagar were as,  $0.029 \pm 0.0073$ ,  $0.057 \pm 0.0176$ ,  $0.075 \pm 0.0200$  and  $0.03 \pm 0.018 \text{ mg/L}$  respectively. The highest concentration of Ni  $0.3 \text{ mg/L}$  was found in water samples from Ghizer. The highest concentration of Ni in spring water sources could be because of dissolution from nickel ore-bearing rocks in the area. The mean concentrations of Mn were  $1.76 \pm 0.3438$ ,  $0.87 \pm 0.3139$ ,  $0.177 \pm 0.0408$ , and  $0.187 \pm 0.0643 \text{ mg/L}$ . The highest concentration of Mn was detected in water samples collected from Skardu ( $4.3 \text{ mg/L}$ ), which might be due to the direct interconnection between water ultramafic and mafic rocks in the area (Huma-Khan et al., 2016).

The concentration of Al in water samples from the Gilgit and Ghizer districts was not detected. In contrast, the mean concentration of Al in water samples collected from the Skardu and Nagar districts were  $0.045 \pm 0.0167 \text{ mg/L}$  and  $0.018 \pm 0.0138 \text{ mg/L}$ , respectively. Similarly, the concentration of B in water collected from Skardu, Ghizer, and Nagar was not detected, whereas, in water samples collected from the Gilgit district, the mean concentration of B was detected as  $0.042 \pm 0.0244 \text{ mg/L}$ . The mean concentrations of Fe and Cu in the study area are shown in Table 1, which were found within the permissible limits recommended by Pak-EPA. The mode of occurrence of Fe and Cu in spring water source samples are due to geogenic and anthropogenic activities in the form of mining, weathering, leaching of rocks, water passing through geological strata, agriculture activities like the use of pesticides and industrial flushing (Hussain et al., 2019).

Similar to this study's results, the study of Kosovo showed the highest mean concentration levels for Fe, Al, and Mn heavy metals; these three metals exceeded the safety limit for drinking water in some samples (Malsiu et al., 2020). Also, in Tamil Nadu, India, among sixteen analyzed heavy metals, Fe, AL, B, Cu, Cd, Ag, Pb, Ni, and Mn concentration levels were higher than their permissible limits (Vetrimurugan et al., 2017). On the other hand, the results of some studies conducted in Germany, Jordan, Malaysia, the USA, and Turkey highlighted that the heavy metal concentration in their drinking water

was lower than allowed and permissible limits (Alidadi et al., 2019) and higher than the standard limits from Pakistan (Ahmed et al., 2019; Habib et al., 2020; Islam et al., 2020; S. Khan et al., 2016), Nigeria (Maigari et al., 2016), India (Vetrimurugan et al., 2017), Ghana (Bortey-Sam et al., 2015), and Iran (Sadeghi et al., 2020).

Table 1

Mean concentrations (mg/L) of selected metals in drinking water samples in the study area ( $n^a = 36 \times 3$ )

Parameter	Statistics	Skardu n=10	Gilgit n=7	Ghizer n=12	Nagar n=7	PAK <sup>b</sup> - NDWQS	WHO <sup>c</sup> Guidelines
Hg	Range	0-0.07	0-0.003	0-0.01	0-0.004	≤0.001	0.001
	Mean	<b>0.0107</b>	0.0005	<b>0.002</b>	0.0007		
	Std Error	±0.0075	±0.0002	±0.0007	±0.0007		
As	Range	0-0.19	0-0.05	0-0.062	0-0.05	≤0.05	0.05
	Mean	0.0296	0.011	0.011	0.01		
	Std Error	±0.0101	±0.0030	±0.0033	±0.0038		
Zn	Range	0-0.23	0-0.41	0-0.37	0-0.07	5.0	3
	Mean	0.047	0.077	0.128	0.013		
	Std Error	±0.0122	±0.0306	±0.0441	±0.0049		
Cr	Range	0-0.08	0-0.07	0-0.09	0-0.05	≤0.05	0.05
	Mean	0.03	0.02	0.032	0.014		
	Std Error	±0.0557	±0.0054	±0.0050	±0.0041		
Ni	Range	0-0.1	0-0.3	0-0.4	0-0.1	≤0.02	0.2
	Mean	<b>0.029</b>	<b>0.057</b>	<b>0.075</b>	<b>0.03</b>		
	Std Error	±0.0073	±0.0176	±0.0200	±0.018		
Fe	Range	0-0.4	0-0.3	0-0.06	0-0.9	---	--
	Mean	0.055	0.012	0.011	0.128		
	Std Error	±0.0199	±0.0139	±0.0064	±0.0704		
Al	Range	0-0.3			0-0.1	≤0.2	0.2
	Mean	0.045	ND <sup>d</sup>	ND	0.018		
	Std Error	±0.0167			±0.0138		
B	Range		0-0.4			0.3	0.3
	Mean	ND	0.042	ND	ND		
	Std Error		±0.0244				
Mn	Range	0-4.5	0-3.6	0-0.8	0-0.84	≤0.5	0.5
	Mean	<b>1.76</b>	<b>0.87</b>	0.177	0.187		
	Std Error	±0.3438	±0.3139	±0.0408	±0.0643		
Cu	Range	0-2.2	0-0.4	0-1.9	0-1.5	2	2
	Mean	0.45	0.1	0.358	0.412		
	Std Error	±0.1244	±0.0305	±0.0943	±0.1173		

Note. <sup>a</sup>number of samples; <sup>b</sup> source: Pakistan National Drinking Water Quality Standards, 2008; <sup>c</sup> World Health Organization (WHO, 2008); <sup>d</sup> not detected

**Human Health Risk Assessment**

**Chronic Daily Intake Indices.** According to the results, obtained from the human health risk analysis the Chronic daily intake indices (CDIs) for adults and children were calculated in water samples from Skardu were ordered as, Mn < Cu < Fe < Zn < Al < Cr < As and Ni < Hg , in Gilgit as, Mn < Cu < Zn < Ni < B < Cr < Fe < As < Hg , in Ghizer as, Cu < Mn < Zn < Ni < Cr < Fe < As < Hg, and in Nagar as, Cu < Al < Mn < Fe < Cr < Zn < Ni < As < Hg. The CDIs of B in water samples from Gilgit, Ghizer, and Nagar districts and Al in all water samples collected from four districts were not calculated for adults and children because their concentrations were found below the detection limit. Overall the CDIs in the study area were within the permissible limits recommended by USEPA (USEPA, 2005).

The high concentration of TTEs in the springs’ water is generally controlled by drainage basin, lithology, and hydrodynamic features of aquifers (Hussain, 2019). However, the availability of high concentrations in drinking water may cause various health problems. The chronic daily intake through drinking water is presented in Table 2.

Table 2  
Chronic daily intake (CDIs mg/lkg-1day-1) through drinking water (n<sup>a</sup> = 36×3)

Parameter	Individuals	Skardu n=10	Gilgit n=7	Ghizer n=12	Nagar n=7
Hg	Adults	0.00027	1.39E-05	5.56E-05	1.94E-05
	Children	0.0003	1.53E-05	6.12E-05	2.14E-05
As	Adults	0.0008	0.0003	0.00028	0.00027
	Children	0.00088	0.00033	0.0003	0.0003
Zn	Adults	0.0013	0.0021	0.00035	0.00038
	Children	0.0014	0.0023	0.0039	0.00039
Cr	Adults	0.00083	0.00055	0.00088	0.00039
	Children	0.00091	0.00061	0.00097	0.00042
Ni	Adults	0.0008	0.0015	0.002	0.00038
	Children	0.00088	0.0017	0.0022	0.00091
Fe	Adults	0.0015	0.00033	0.00032	0.0035
	Children	0.0016	0.00036	0.00033	0.0039
Al	Adults	0.0012	ND	ND	0.00051
	Children	0.0013	ND	ND	0.00055
B	Adults	ND	0.0011	ND	ND
	Children	ND	0.0012	ND	ND
Mn	Adults	0.048	0.024	0.0049	0.0051
	Children	0.053	0.026	0.0054	0.0057
Cu	Adults	0.012	0.0027	0.0099	0.011
	Children	0.013	0.003	0.01	0.012

Note. n = number of water samples; ND = not detected

**Health Risk Index.** The calculated health risk indices (HRIs) for adults and children in Skardu were ordered as, Mn < Cu < Ni < Zn < Cr, in Gilgit as, Mn < Ni Cu < Zn < Cr, in Ghizer as, Cu < Ni < Mn < Zn < Cr and in Nagar as Cu < Mn < Ni < Zn < Cr. HRIs of Zn for adults in Skardu, Gilgit Ghizer, and Nagar were calculated as 0.0043, 0.007, 0.0011, and 0.0012 for children of Skardu, Gilgit, and Ghizer, respectively and Nagar were calculated as, 0.0046, 0.0076, 0.0013, and 0.0013. The HRIs of Cr were 0.00055, 0.00036, 0.00058, and 0.00026 for adults and 0.0006, 0.0004, 0.00064 and 0.00028 for children in Skardu, Gilgit, Ghizer, and Nagar, respectively. The HRIs of Ni ranged from 0.01 to 0.075 for adults and from 0.011 to 0.085 for children. The HRIs of Mn were 0.34, 0.171, 0.035 and 0.036 for adults and were 0.37, 0.18, 0.038 and 0.04 for children, whereas the HRIs of Cu were 0.32, 0.072, 0.26 and 0.29 for adults and 0.35, 0.081, 0.27 and 0.32 for children in Skardu, Gilgit, Ghizer, and Nagar. The HRIs of Hg, As, Al, B, and Fe were not calculated because there is no reference dose for these elements. The HRI values ranged between safe limits (HRI < 1) in Gilgit-Baltistan, as shown in Table 3, suggesting that, at present, there is no risk associated with TTEs to the health of adults and children.

Table 3  
Health risk indexes (HRIs) through drinking water (n= 36×3)

Parameter	Individuals	Skardu n=10	Gilgit n=7	Ghizer n=12	Nagar n=7
Hg	Adults	NC	NC	NC	NC
	Children				
As	Adults	NC	NC	NC	NC
	Children				
Zn	Adults	0.0043	0.007	0.0011	0.0012
	Children	0.0046	0.0076	0.0013	0.0013
Cr	Adults	0.00055	0.00036	0.00058	0.00026
	Children	0.0006	0.0004	0.00064	0.00028
Ni	Adults	0.04	0.075	0.01	0.019
	Children	0.044	0.085	0.011	0.045
Fe	Adults	NC	NC	NC	NC
	Children				
Al	Adults	NC	ND	ND	NC
	Children				
B	Adults	ND	NC	ND	ND
	Children				
Mn	Adults	0.34	0.171	0.035	0.036
	Children	0.37	0.18	0.038	0.04
Cu	Adults	0.32	0.072	0.26	0.29
	Children	0.35	0.081	0.27	0.32

Note. n = number of water samples: NC = not calculated: ND = not detected

### Spatial Distribution of TTEs

Spatial variations based on the concentration of different elements were found in the study area, as presented in Figure 2. The post hoc or Tukey test highlighted that the concentration of Hg was higher ( $P < 0.05$ ) in Skardu compared to Gilgit and Nagar, as shown in Figure 2. It was reported that the high concentration of Hg in water mostly evolved from mining wastes (Hussain et al., 2019). The mean concentration of Zn was significantly higher ( $P < 0.05$ ) in Ghizer compared to Skardu and Nagar. Fe concentration was found to be

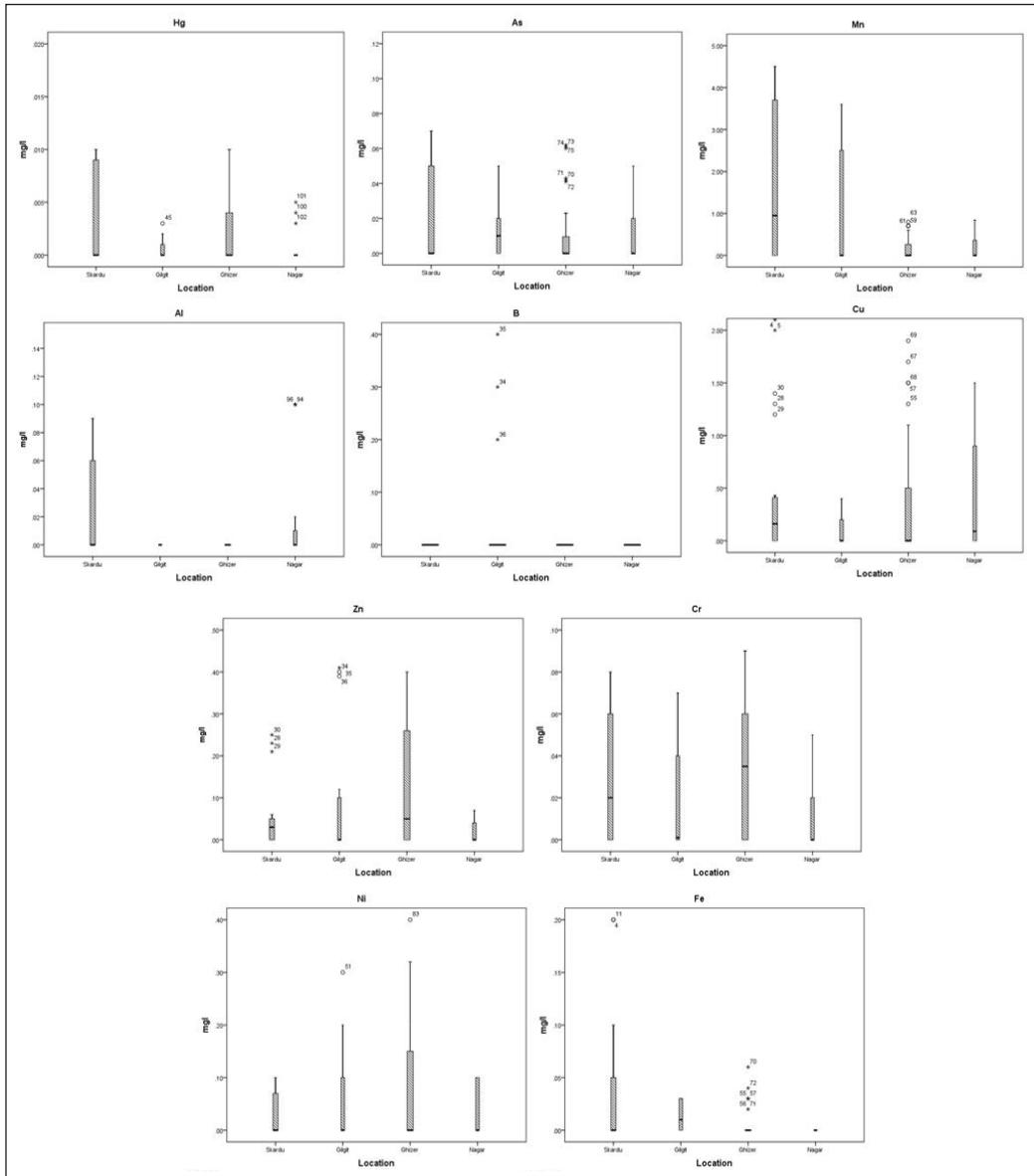


Figure 2. One-way ANOVA box plot comparison of selected TTEs with sampling locations

significantly higher in water from the Nagar than in Ghizer. The concentration of Al in water samples collected from the Skardu was considerably higher than in Gilgit and Ghizer. Similarly, the concentration of B in water samples collected from Gilgit was significantly higher than the concentrations measured in water samples collected from Ghizer Nagar and Skardu. The concentration of Mn in Skardu was higher than in Gilgit, Ghizer, and Nagar. An insignificant difference ( $P > 0.05$ ) was observed among As, Cr, Ni, and Cu in the analyzed water samples. Geologically, these elements can be evolved by reaction with passing through various rock strata and can behave similarly/differently from one another (Kavcar et al., 2009).

### Correlation Analysis

One-way ANOVA analysis for correlations among elements was performed using SPSS software V17. The correlations among different elements provide very useful evidence of the concentrations and pathways of TTEs (Huma-Khan et al., 2016). Based on the results obtained from our study, very strong correlation was found among different elements, such as Hg-As ( $r = 0.983$ ,  $P > 0.05$ ), Hg-Al ( $r = 0.884$ ,  $P > 0.05$ ), Hg-Mn ( $r = 0.852$ ,  $P > 0.05$ ), As-Al ( $r = 0.905$ ,  $P > 0.05$ ), As-Mn ( $r = 0.921$ ,  $P > 0.05$ ), Cr-Zn ( $r = 0.669$ ,  $P > 0.05$ ), and Zn-Ni ( $r = 0.945$ ,  $P > 0.05$ ). However, in some water samples, strong negative correlations were observed, such as Zn-Fe ( $r = -0.878$ ), Ni-Al ( $r = -0.816$ ), and Cu-B ( $r = -0.971$ ). The positive correlation between Mn and Zn represents the intimate source of domestic and agricultural activities (Hussain et al., 2019). The correlation matrix has pointed to two elements with the same sources of agricultural activities, industrial and sewage effluents, and geogenic origin from weathering of sulfide-bearing minerals (Huma-Khan et al., 2016). Correlation matrixes of selected TTEs in water samples are shown in Table 4.

Table 4  
Correlation matrixes of selected TTEs in water samples ( $n = 36 \times 3$ )

	Hg	As	Zn	Cr	Ni	Fe	Al	B	Mn	Cu
Hg	1									
As	<b>0.983</b>	1								
Zn	-0.152	-0.259	1							
Cr	0.582	0.462	0.699	1						
Ni	-0.465	-0.554	<b>0.945</b>	0.432	1					
Fe	-0.0163	0.017	<b>-0.878</b>	-0.65	-0.785	1				
Al	<b>0.884</b>	<b>0.905</b>	-0.586	0.166	<b>-0.816</b>	0.437	1			
B	-0.413	-0.285	0.146	-0.314	0.276	-0.479	-0.493	1		
Mn	<b>0.852</b>	<b>0.921</b>	-0.215	0.347	-0.467	-0.17	0.741	0.108	1	
Cu	0.56	0.463	-0.321	0.259	-0.479	0.562	0.682	<b>-0.971</b>	0.088	1

Note. n = number of water samples

## CONCLUSION

The present study investigated the contamination and human health risks in four districts, including Gilgit, Skardu, Ghzer and Nagar of Gilgit-Baltistan, Pakistan. This study's results revealed that the present Hg, As, Cr, Ni, Al, and Mn levels in springs' water from Gilgit-Baltistan are higher than the permissible limits of WHO, and Pak-NDWQS. Meanwhile Fe and Cu concentrations were under the safe limits. Overall, no risks to human health were detected ( $HRI < 1$ ), but in the near future, the increasing concentration may reach to HRI level recommended by US-EPA. One-way ANOVA showed significant positive/negative correlations among different elements. The positive correlations revealed that the mode of occurrence of TTEs is lithological and anthropogenic. The results of this study provide useful information for policymakers and local administration to control TTE<sub>s</sub> pollution to provide safe drinking water to the residents of Gilgit-Baltistan, Pakistan.

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## REFERENCES

- Ahmed, N., Bodrud-Doza, M., Islam, A. R. M. T., Hossain, S., Moniruzzaman, M., Deb, N., & Bhuiyan, M. A. Q. (2019). Appraising spatial variations of As, Fe, Mn and NO<sub>3</sub> contaminations associated health risks of drinking water from Surma basin, Bangladesh. *Chemosphere*, *218*, 726-740. <https://doi.org/10.1016/j.chemosphere.2018.11.104>
- Ali, S., & Rubina, S. H. (2018). Assessment of freshwater springs, associated diseases and indigenous perception in Ghizer, Gilgit-Baltistan, Pakistan. *Pakistan Journal of Medical Sciences*, *34*(1), 121-124.
- Ali, U., Batool, A., Ghufran, M., Asad-Ghufran, M., Sabahat-Kazmi, S., & Hina-Fatimah, S. (2019). Assessment of heavy metal contamination in the drinking water of muzaffarabad, Azad Jammu and Kashmir, Pakistan. *International Journal of Hydrology*, *3*(5), 331-337. <https://doi.org/10.15406/ijh.2019.03.00196>
- Alidadi, H., Tavakoly Sany, S. B., Zarif Garaati Oftadeh, B., Mohamad, T., Shamszade, H., & Fakhari, M. (2019). Health risk assessments of arsenic and toxic heavy metal exposure in drinking water in northeast Iran. *Environmental Health and Preventive Medicine*, *24*, Article 59. <https://doi.org/10.1186/s12199-019-0812-x>
- Alina, M., Azrina, A., Yunus, A. M., Zakiuddin, S. M., Effendi, H. M. I., & Rizal, R. M. (2012). Heavy metals (mercury, arsenic, cadmium, plumbum) in selected marine fish and shellfish along the Straits of Malacca. *International Food Research Journal*, *19*(1), 135-140.
- Arif, M., Henry, D., & Moon, C. (2011). Host rock characteristics and source of chromium and beryllium for emerald mineralization in the ophiolitic rocks of the Indus Suture Zone in Swat, NW Pakistan. *Ore Geology Reviews*, *39*(1-2), 1-20.

- Bortey-Sam, N., Nakayama, S. M., Ikenaka, Y., Akoto, O., Baidoo, E., Mizukawa, H., & Ishizuka, M. (2015). Health risk assessment of heavy metals and metalloids in drinking water from communities near gold mines in Tarkwa, Ghana. *Environmental Monitoring and Assessment*, 187, Article 397. <https://doi.org/10.1007/s10661-015-4630-3>
- Dessie, B. K., Gari, S. R., Mihret, A., Desta, A. F., & Mehari, B. (2021). Determination and health risk assessment of trace elements in the tap water of two Sub-Cities of Addis Ababa, Ethiopia. *Heliyon*, 7(5), Article e06988. <https://doi.org/10.1016/j.heliyon.2021.e06988>
- Dieter, H. H., Bayer, T. A., & Multhaup, G. (2005). Environmental copper and manganese in the pathophysiology of neurologic diseases (Alzheimer's disease and Manganism). *Acta hydrochimica et hydrobiologica*, 33(1), 72-78. <https://doi.org/10.1002/ahch.200400556>
- Habib, M. A., Islam, A. R. M. T., Bodrud-Doza, M., Mukta, F. A., Khan, R., Siddique, M. A. B., Phoungthong, K., & Techato, K. (2020). Simultaneous appraisals of pathway and probable health risk associated with trace metals contamination in groundwater from Barapukuria coal basin, Bangladesh. *Chemosphere*, 242, Article 125183. <https://doi.org/10.1016/j.chemosphere.2019.125183>
- Huma-Khan, N., Nafees, M., & Bashir, A. (2016). Study of heavy metals in soil and wheat crop and their transfer to food chain. *Sarhad Journal of Agriculture*, 32(2), 70-79. <http://dx.doi.org/10.17582/journal.sja/2016/32.2.70.79>
- Hussain, R., Wei, C., & Luo, K. (2019). Hydrogeochemical characteristics, source identification and health risks of surface water and groundwater in mining and non-mining areas of Handan, China. *Environmental Earth Sciences*, 78, Article 402. <https://doi.org/10.1007/s12665-019-8350-9>
- Islam, A. R. M. T., Islam, H. T., Mia, M. U., Khan, R., Habib, M. A., Bodrud-Doza, M., Siddique, M. A. B., & Chu, R. (2020). Co-distribution, possible origins, status and potential health risk of trace elements in surface water sources from six major river basins, Bangladesh. *Chemosphere*, 249, Article 126180. <https://doi.org/10.1016/j.chemosphere.2020.126180>
- Ji, Y., Wu, J., Wang, Y., Elumalai, V., & Subramani, T. (2020). Seasonal variation of drinking water quality and human health risk assessment in Hancheng City of Guanzhong Plain, China. *Exposure and Health*, 12, 469-485. <https://doi.org/10.1007/s12403-020-00357-6>
- Jiang, Z., Xu, N., Liu, B., Zhou, L., Wang, J., Wang, C., Dai, B., & Xiong, W. (2018). Metal concentrations and risk assessment in water, sediment and economic fish species with various habitat preferences and trophic guilds from Lake Caizi, Southeast China. *Ecotoxicology and Environmental Safety*, 157, 1-8. <https://doi.org/10.1016/j.ecoenv.2018.03.078>
- Kavcar, P., Sofuoglu, A., & Sofuoglu, S. C. (2009). A health risk assessment for exposure to trace metals via drinking water ingestion pathway. *International Journal of Hygiene And Environmental Health*, 212(2), 216-227. <https://doi.org/10.1016/j.ijheh.2008.05.002>
- Khan, S., Rauf, R., Muhammad, S., Qasim, M., & Din, I. (2016). Arsenic and heavy metals health risk assessment through drinking water consumption in the Peshawar District, Pakistan. *Human and Ecological Risk Assessment: An International Journal*, 22(3), 581-596. <https://doi.org/10.1080/10807039.2015.1083845>

- Khan, T. A. (2011). Trace elements in the drinking water and their possible health effects in Aligarh City, India. *Journal of Water Resource and Protection*, 3(7), 522-530. <https://doi.org/doi:10.4236/jwarp.2011.37062>
- Khan, A., Ahmad, I., Waqarullah, ul Zia, I., & Zakirullah. (2015). *Field report of Gilgit-Baltistan*. <https://pdfcookie.com/documents/geological-field-report-of-gilgit-baltistan-by-arsalan-khan-51q3yxmqd8v7>
- Kisku, G. C., Pandey, P., Negi, M. P. S., & Misra, V. (2011). Uptake and accumulation of potentially toxic metals (Zn, Cu and Pb) in soils and plants of Durgapur industrial belt. *Journal of Environmental Biology*, 32(6), 831-838.
- Krishna, A. K., Satyanarayanan, M., & Govil, P. K. (2009). Assessment of heavy metal pollution in water using multivariate statistical techniques in an industrial area: A case study from Patancheru, Medak District, Andhra Pradesh, India. *Journal of Hazardous Materials*, 167(1-3), 366-373. <https://doi.org/10.1016/j.jhazmat.2008.12.131>
- Lambert, M., Leven, B., & Green, R. (2000). *New methods of cleaning up heavy metal in soils and water*. Hazardous Substance Research Centres. [https://cfpub.epa.gov/ncer\\_abstracts/index.cfm/fuseaction/display.files/fileID/14295](https://cfpub.epa.gov/ncer_abstracts/index.cfm/fuseaction/display.files/fileID/14295)
- Maigari, A., Ekanem, E., Garba, I., Harami, A., & Akan, J. (2016). Health risk assessment for exposure to some selected heavy metals via drinking water from Dadinkowa dam and river gombe abba in Gombe state, Northeast Nigeria. *World Journal Analytical Chemistry*, 4(1), 1-5.
- Malsiu, A., Shehu, I., Stafilov, T., & Faiku, F. (2020). Assessment of heavy metal concentrations with fractionation method in sediments and waters of the Badovci Lake (Kosovo). *Journal of Environmental and Public Health*, 2020, Article 3098594 <https://doi.org/10.1155/2020/3098594>
- Mohammadi, A. A., Zarei, A., Majidi, S., Ghaderpoury, A., Hashempour, Y., Saghi, M. H., Alinejad, A., Yousefi, M., Hosseingholizadeh, N., & Ghaderpoori, M. (2019). Carcinogenic and non-carcinogenic health risk assessment of heavy metals in drinking water of Khorramabad, Iran. *MethodsX*, 6, 1642-1651. <https://doi.org/10.1016/j.mex.2019.07.017>
- Mohmand, J., Eqani, S. A. M. A. S., Fasola, M., Alamdar, A., Mustafa, I., Ali, N., Liu, L., Peng, S., & Shen, H. (2015). Human exposure to toxic metals via contaminated dust: Bio-accumulation trends and their potential risk estimation. *Chemosphere*, 132, 142-151. <https://doi.org/10.1016/j.chemosphere.2015.03.004>
- Muhammad, S., Shah, M. T., & Khan, S. (2010). Arsenic health risk assessment in drinking water and source apportionment using multivariate statistical techniques in Kohistan region, northern Pakistan. *Food and Chemical Toxicology*, 48(10), 2855-2864. <https://doi.org/10.1016/j.fct.2010.07.018>
- Muhammad, S., Shah, M. T., & Khan, S. (2011). Health risk assessment of heavy metals and their source apportionment in drinking water of Kohistan region, northern Pakistan. *Microchemical Journal*, 98(2), 334-343. <https://doi.org/10.1016/j.microc.2011.03.003>
- Muhammad, S., Ullah, R., & Jadoon, I. A. (2019). Heavy metals contamination in soil and food and their evaluation for risk assessment in the Zhob and Loralai valleys, Baluchistan province, Pakistan. *Microchemical Journal*, 149, Article 103971. <https://doi.org/10.1016/j.microc.2019.103971>
- Muhammad Tahir, S., Suleman, M., Abdul Baqi, S., Sattar, A., & Khan, N. (2020). Determination of heavy metals in drinking water and their adverse effects on human health. A review. *Pure and Applied Biology (PAB)*, 9(1), 96-104. <http://dx.doi.org/10.19045/bspab.2020.90012>

- Murtaza, B., Amjad, M., Shahid, M., Imran, M., Shah, N. S., Abbas, G., & Naeem, M. A. (2020). Compositional and health risk assessment of drinking water from health facilities of District Vehari, Pakistan. *Environmental Geochemistry and Health*, *42*, 2425-2437. <https://doi.org/10.1007/s10653-019-00465-6>
- Pidwirny. (2006). *Characteristics of Metamorphic Rocks*. Physical Geography.net. <http://www.physicalgeography.net/fundamentals/10g.html>
- Rasool, A., Xiao, T., Farooqi, A., Shafeeque, M., Masood, S., Ali, S., Fahad, S., & Nasim, W. (2016). Arsenic and heavy metal contaminations in the tube well water of Punjab, Pakistan and risk assessment: A case study. *Ecological Engineering*, *95*, 90-100. <http://dx.doi.org/10.1016/j.ecoleng.2016.06.034>
- Sadeghi, P., Loghmani, M., & Frokhzad, S. (2020). Human health risk assessment of heavy metals via consumption of commercial marine fish (*Thunnus albacares*, *Euthynnus affinis*, and *Katsuwonus pelamis*) in Oman Sea. *Environmental Science and Pollution Research*, *27*, 14944-14952. <https://doi.org/10.1007/s11356-020-07907-0>
- Saleh, H. N., Panahande, M., Yousefi, M., Asghari, F. B., Oliveri Conti, G., Talaei, E., & Mohammadi, A. A. (2019). Carcinogenic and non-carcinogenic risk assessment of heavy metals in groundwater wells in Neyshabur Plain, Iran. *Biological Trace Element Research*, *190*, 251-261. <https://doi.org/10.1007/s12011-018-1516-6>
- Sharma, R. K., Agrawal, M., & Marshall, F. M. (2008). Heavy metal (Cu, Zn, Cd and Pb) contamination of vegetables in urban India: A case study in Varanasi. *Environmental Pollution*, *154*(2), 254-263.
- Stephanie, S. (2010). Trace elements. *Current Anaesthesia & Critical Care*, *21*(1), 44-48. <https://doi.org/10.1016/j.cacc.2009.08.004>
- USEPA. (2003). *Technical Standard Operating Procedure (SOP EH- 01)*. United States Environmental Protection Agency. <https://www.epa.gov/risk/guidelines-carcinogen-risk-assessment>
- USEPA. (2005, March). *Guidelines for carcinogen risk assessment (EPA/630/P-03/001F)*. United States Environmental Protection Agency. <https://www.epa.gov/risk/guidelines-carcinogen-risk-assessment>
- Vetrimurugan, E., Brindha, K., Elango, L., & Ndwandwe, O. M. (2017). Human exposure risk to heavy metals through groundwater used for drinking in an intensively irrigated river delta. *Applied Water Science*, *7*, 3267-3280. <https://doi.org/10.1007/s13201-016-0472-6>
- Wasserman, G. A., Liu, X., Parvez, F., Ahsan, H., Levy, D., Factor-Litvak, P., Kline, J., Geen, A. V., Slavkovich, V., Lolocono, N. J., Cheng, Z., Zheng, Yan., & Graziano, J. H. (2006). Water manganese exposure and children's intellectual function in Arai hazar, Bangladesh. *Environmental Health Perspectives*, *114*(1), 124-129. <https://doi.org/10.1289/ehp.8030>
- WWF. (2009). *Climate Change Resilience in Gilgit*. [https://www.wwfpak.org/our\\_work\\_/climate\\_and\\_energy\\_/climate\\_change\\_resilience\\_in\\_gilgit/](https://www.wwfpak.org/our_work_/climate_and_energy_/climate_change_resilience_in_gilgit/)
- Yang, F., & Massey, I. Y. (2019). Exposure routes and health effects of heavy metals on children. *Biometals*, *32*, 563-573. <https://doi.org/10.1007/s10534-019-00193-5>
- Zhang, Y., Sillanpää, M., Li, C., Guo, J., Qu, B., & Kang, S. (2015). River water quality across the Himalayan regions: Elemental concentrations in headwaters of Yarlung Tsangpo, Indus and Ganges River. *Environmental Earth Sciences*, *73*, 4151-4163.

