ABSTRACT

The study mainly focuses on heavy metals’ quantification, investigation of physicochemical characteristics, source apportionments, human health risk assessment, and connectivity between the water of alpine glacial lakes and rivers. In this regard, twenty-six water samples are taken from sixteen lakes and two lakes-fed rivers. The physiochemical parameters, particularly heavy metals are quantified using atomic absorption spectrophotometer. The Pearson correlations are used to find out the relationship between various heavy metals and physicochemical parameters. The multivariate methods, Principal component analysis (PCA), and Hierarchal cluster analysis (HCA) are conducted for contamination source apportionment. In the findings of the current analysis, the mean concentration of Lead (Pb 0.51), Cadmium (Cd 0.04), Nickel (Ni 0.06), Chromium (Cr 0.08), Iron (Fe 0.06), and Manganese (Mn 0.09) is the most common heavy metal contaminants found in both lakes and river water. Copper (Cu 0.00) and Arsenic (As 0.001) concentrations are lower in both lakes and rivers. Other tested physicochemical characteristics, such as pH, Hardness, Chemical oxygen demands (COD), total dissolved solids (TDS), dissolved oxygen (DO) chloride (Cl), sulfate (SO4), nitrate (NO3), and bicarbonates (HCO3) are within the maximum permissible limits of World Health Organization’s (WHO).

HIGHLIGHTS

- The main contributors to the presence of heavy metals in both lakes and rivers are human activities and long-distance air transport.
- A significant ecological risk is posed by heavy metal contaminants to the glacial alpine lake and river system.
- In this research area, Pb and Cd posed a substantially greater potential ecological risk than other metals.
1. Introduction

The requirement of water for life is crucial as other foods and is one of the threatened resources globally. Various anthropogenic activities cause water quality to deteriorate (Şener et al. 2017; Wang and Zhang 2018; Varol 2020a). To regulate climate, regional hydrological cycle and aquatic biodiversity lakes water play a vital role (Wetzel 2001). Lakes are open systems and their water quality indicates the environmental disturbances that occurred in the surrounding area (Guo et al. 2020). Water contamination is currently one of the most important environmental concerns facing the world. Organic and inorganic chemicals, cations and anions, and pathogenic microorganisms are all common contaminants in water (Azizullah et al. 2011). Heavy metals are a severe problem among the numerous contaminants because they infiltrate and accumulate in the food chain. In urbanized regions, industrialization, domestic sewage, industrial effluents, atmospheric deposition, leaching from landfills/dumpsites, and storm runoff are the primary sources of potentially dangerous pollutants. On the other hand, the Glacier retreat, long-range air pollution transfer, and wildfire are all ongoing worldwide issues that have an impact on Alpine Glacial Lakes and River Hydrochemistry. In remote places, these characteristics play an important role as drivers of heavy metal contamination of freshwater systems.

The northern part of Pakistan is well-known for its vast number of freshwater lakes, rivers, and streams, spread across a wide range of heights and latitudes in Pakistan. Alpine lakes are distant and pristine ecosystems that are especially vulnerable to anthropogenic influences because they are used to monitor global water quality changes and anthropogenic repercussions on a large scale (Battarbee et al. 2009). As a result, high-altitude lakes are regarded as "early warning systems" for the entire alpine environment, and they are particularly vulnerable to climate change (Selene et al. 2020). Aside from lakes, river water is constantly contaminated by a variety of pollutants. These Alpine Glacial Lakes serve as catchment areas for two major rivers, the Swat and Panjkora, and play an important role in agriculture, drinking water, and other uses such as electricity and recreation for approximately 10 million population. A variety of biogeochemical activities occurred in lake ecosystem as lake are the hub of several biogeochemical activities that are necessary for the communities that rely on these ecosystems (Tranvik et al. 2009; Eiler et al. 2014). These ecosystems are very dynamics and lead to rapid changes in their environmental conditions. Lakes are freshwater bodies and are approximately 0.8% of the overall global water reservoir since lakes are important in the development of sustainable ecosystems (Harrison et al. 2016). A variety of biogeochemical activities occurs in these lakes which are vital for the communities living inside these lakes (Tranvik et al. 2009; Eiler et al. 2014). High altitude lakes are very susceptible to chemical, physical and biological such as microbial changes upon climate changes (Rofner et al. 2017), however eutrophication are less abundant or absent in low temperate environments such as high-altitude lakes (Stres et al. 2013). Heavy metals such as Fe, As, Zn, Pb, Cd, Hg, Ni, and Cu etc., have been found in various Pakistani water basins. Heavy metals are poisonous, non-biodegradable, persistent, have negative impacts on living organisms and harm for aquatic biota and accumulating in the food chain as well as detected in atmospheric deposition (Hussain et al. 2014; Sobhanardakani 2017; Sobhanardakani et al. 2018; Sobhanardakani 2019). High consumption of Pb, a
non-essential metal for the human body, can result in skeletal, neurological, circulatory, endocrine, enzymatic, and immune system diseases (Zhou et al. 2015). High Cu and Mn levels above permissible limit can cause mental illnesses including Alzheimer’s and Manganism (Cabral-Pinto et al. 2020). The High intake of Cd is linked to kidney and skeletal damage, as well as to itai-itai (ouch-ouch) illnesses (Abarikwu et al. 2017). Some of these heavy metals and carcinogens produce edoema of the eyelids, congestion of the nasal mucous membranes and throat, tumor, stuffiness of the brain, gastrointestinal, muscular, reproductive, neurological, and genetic abnormalities (Kar et al. 2008; Lin et al. 2020).

To evident spatial and temporal variations in water quality and to identify the major parameters and influences responsible for these variations, multivariate statistical approaches were used (Achieng et al. 2017; Chounlamany et al. 2017; Siepak and Sojka 2017; Ustaoglu et al. 2020; Varol 2020b). In its application, cluster analysis (CA) aims to separate a big set of items into small groups (clusters) based on their similarities. As a result, the resulting clusters must have a high degree of internal homogeneity and a high degree of external heterogeneity (Muangthong and Shrestha 2015). It can assist in the interpretation of data and the identification of patterns (Hajigholizadeh and Melesse 2017) by establishing hypotheses (exploratory analysis) rather than making data confirmations (confirmatory analysis). Another multivariate technique commonly used to assess water quality is (PCA). PCA minimize the size of the data set by forming factors that represent the original data’s whole structure (Ustaoglu et al. 2020). The study was focused to analyses the water quality and determine the spatial distribution of hazardous heavy metals in surface water of the various Glacial Alpine Lakes and rivers of Khyber Pakhtunkhwa Pakistan. Human health risks were assessed by using the hazard quotients of individual heavy metals and the hazard index of mixed heavy metals. The pathways of ingestion and dermal absorption, the carcinogenic risks from the heavy metals were also assessed and identified the natural and/or anthropogenic sources of these heavy metals using multivariate statistical techniques and Hierarchical Cluster analysis. The multivariate statistical technique i.e., PCA assessed the spatial variability of monitoring locations and to better understand the relationship between the key characteristics and the lentic and lotic environments. To protect the public health from the adverse effects of heavy metals contaminations the present study is conducted and will be a valuable reference and guidelines for water resource and water quality management in the Eastern Hindukush regions, Pakistan. According to our best knowledge this study is first time reported in terms of research area.

2. Materials and methods

2.1. Study area profile, climatology, and hydrology

The present research area represents three districts (Swat, Lower Dir and Upper Dir) and is large area of Khyber-Pakhtunkhwa Pakistan (Figure 1). District Swat also called Switzerland of Pakistan due to its greenish and beautiful nature. Swat is located in the northern mountain ranges which are temperate zone having an altitudinal range from 500 to 6500 m above sea level (Qasim et al. 2011). District Swat covering approximately 5337 square kilometers and runs from 34 to 36 degrees north latitude.
and 71 to 73 degrees east longitude. Swat is located on the north by Chitral, on the northeast by Gilgit Agency, on the west by Dir Valley, on the south by Malakand and Buner Districts, on the east by Kohistan and Shangla Districts, and on the west by Hazara (Qasim et al. 2011). Swat climate is Mediterranean in the northern region however subtropical at southern region. The annual rainfall in the area ranges from 750 to 1350 mm and the average temperature ranges between \(-10^\circ\text{C}\) and \(25^\circ\text{C}\) (Shah et al. 2010a; 2010b). The maximum rainfall in the studied area occurs in a monsoon season from June to September. April is least humid month takes 40% humidity to a high of 85% in July (Nafees et al. 2008). The geology of the studied area is part of the Indian plate that have undergone high pressure and low temperature metamorphism and are made up of a volcano-sedimentary, discontinuous belt of volcanic, and sedimentary rocks, as well as a chaotic assemblage of mafic and ultramafic rocks like met basalts serpentinite, talc-carbonate schist and green schist (Shah et al. 2010b; Arif et al. 2011). The Swat River rises in Kalam at the junction of the Ushu and Utror rivers and flows for 160 kilometers through the valley to Chakdara. From Kalam to nearCharsada, the river stretches for 250 kilometers. Along the river’s path, many large and small tributaries join it. River Panjkora is 220 Km long and flows south through Dir Upper and Dir Lower districts and joins River Swat at Bosaq at the junction of district Dir Lower, district Malakand and Bajaur Agency and from where the water body flows downstream as River Swat. River panjkora finally joins River Kabul in district Charsadda (Ali 2018).

2.2. Sampling collection and physiochemical analysis of the water samples

The samples were collected in triplicates and temporarily stored in ice and were analyzed (Physiochemical parameters) within 6-h after transferring to PCRWR laboratory.
Peshawar. For quantification of heavy metals samples were transferred to centralized resource laboratory University of Peshawar Pakistan, and samples were kept at 4°C for further analysis. GPS was used to take the geographical position of the Each Lake and River. The minerology and lithology of nearby mountains and Lakes was identified taking help from geologist. Samples were collected in different bottles for different analysis such as for heavy metals 250 mL precleaned polythene bottles which were previously soaked and washed with 10% HNO₃ and for COD, DO, Turbidity and Hardness etc. 1 L of water were collected. Chemical Oxygen Demand (COD), nitrate, phosphate, Dissolved oxygen etc. of the water samples were determined according to Standard Methods (APHA, 2005). All samples were filtered by using 0.45 µm membrane filter. One part of each sample was acidified with 5% HNO₃ (3 to 5 drops) for the quantitative analysis of heavy metals. The other part of the sample was not acidified but examined for the Chloride, sulphate, and Nitrate etc. analysis. During sampling collection, pH, electrical conductivity (EC), and total dissolved solids (TDS) were determined in situ with the help of portable instruments, to confirm the results, pH, electrical conductivity (EC), and total dissolved solids (TDS) were countercheck in laboratory. The concentrations of Heavy Metals were analyzed by Acetylene flame atomic absorption spectrophotometer (Perkin Elmer AAS 700, USA) and the quality assurance and control were assessed using duplicates, certified reference materials (CRM 700), with each batch of water samples and reagent blanks.

2.3. Risk assessment of heavy metals

Two ways are considered for the human health risk assessment of heavy metals in the water of the study area such as ingestion and skin absorption (Drinking and dermal contact) (Zeng et al. 2015). The following two equations (Eq 1 and 2) are used to quantify dietary average daily dose (ADD) exposures.

\[
\text{ADD}_{\text{ingestion}} = \frac{C_w \times IR \times EF \times ED}{BW} \times AT
\]

\[
\text{ADD}_{\text{dermal}} = \frac{C_w \times SA \times Kp \times EF \times ET}{BW} \times AT
\]

The details of the above two equations are presented below:

SA = Surface skin area (cm²), 1800 cm² = Adults and 6660 cm² = Children

EF = Exposed frequency (days/year) and are 350 days/year in the present study (Zeng et al. 2015; Xiao et al. 2019)

Cw = Average value (µg/L) of heavy metals in the water sample

IR = Daily intake (ingestion) rate (L/day) and are 2.0 = Adults and 0.64 = Children (Xiao et al. 2019)

ET = Exposure time (h/day) 0.58 h/day = Adults and 1.0 h/day = Children (Wang et al. 2011)

AT = Average time for noncarcinogenic (days) and are 25,550 days = Adults and 2190 days = Children (USEPA 2011).
Kp = Skin adherence time in the samples (cm/h) and are 0.0001 cm/h = Pb, 0.001 cm/h = Cu, 0.004 cm/h = Ni and Co, and 0.0006 cm/h = Zn as taken in the present study (USEPA 2011)

ED = Exposure duration in years and are 70 years = Adults and 6 years = Children (USEPA 2011)

BW = Average body weight (kg), 65 kg = Adults 20 kg = Children (Xiao et al. 2019).

The hazard quotient value (HQ) was used to calculate noncarcinogenic risk (NCR) which are presented in Eq. 3 (USEPA 2011) and HQ value > 1, predicts potential noncarcinogenic effects.

\[
\text{Hazard Quotient (HQ)} = \frac{ADD}{\text{RfD}}
\]  

(3)

Where RfD = Reference dose (mg/kg/day) for various analytes

\[
\text{RfDdermal} = \frac{\text{RfD}}{\text{ABSg}}
\]

ABSg = Gastrointestinal absorption factor and are unitless.

The RfDdermal values of the studied heavy metals in the surface water variables such as As, Cd, Cr, Fe, Mn, Ni, Pb, Zn, and Cu were 0.0008, 0.001, 0.00006, 0.001, 0.14, 0.0054, 0.00053, 0.06, 0.012 mg/kg/day, and RfDingestion values were 0.0003, 0.001, 0.003, 0.7, 0.14, 0.02, 0.0035, 0.3, and 0.04 mg/kg/day, respectively. The cancer slope factor values for surface water variables such as Cd, Pb, Cr, Ni, and as were 15, 8.5, 0.5, 0.91, and 1.5 mg/kg/day, respectively.

The total potential human NCR caused by different pathways was quantified by hazardous index HI (Eq. 4):

\[
\text{Hazard Index (HI)} = \sum (\text{HQing} + \text{HQderm})
\]  

(4)

HI values > 1 indicates toxic human health effect and should be taken into consideration (Rashid et al. 2020; Noor et al. 2022; Rashid et al. 2022a; 2022b) (USEPA 2011).

2.4. Statistical analysis

The ARC.GIS 10.3 software is used for creating a map of geographical locations of the study area. Further, for the correlation plots and other statistical analysis such as Pearson correlation matrix, hierarchal cluster analysis and principal component analysis XLSTAT 2019.2.2 and SPSS-20 are utilized. Principal component analysis (PCA) extracts from the covariance matrix of original data set the eigenvectors and eigenvalues (Chabukdhara and Nema 2012). PCA is used to reduce the large data sets into small data sets from original data utilizing linear combination and produce new orthogonal non-correlated variables but still have maximum information in the large data set and to evaluate possible sources of heavy metals in surface water. To measure the significance of the association among pairs of variables the Pearson correlation matrix is used and to measures how well the variance of each constituent can be explained by relationship with each other (Liu et al. 2003). A dendrogram and cluster
analysis was produced based on the level of similarity with the help of wards method and to identify the groups that have similar sources. The most similar observations were prioritized first following consequent observations using the hierarchical clustering technique.

3. Results and discussion

3.1. Physicochemical analysis of lakes and river water

This study tested and analyzed the water quality of alpine glacial lakes and these lakes fed rivers. Table 1 represents the average, standard deviation, as well as minimum and maximum values of water quality tested of eighteen lakes and two rivers and compared with WHO (2017). All the parameters tested are represented in mg/L except for EC in μS/cm, pH in number, the temperature in °C as well as color, odor, and taste have no units. The water has no objectionable taste and odor, taste and is clear. Water shows a slightly alkaline nature with an average pH value of 7.72. The finding reveals that the pH values in the study areas ranged from 7.5 to 8.1 and the result showed that, pH values are within the permissible limit in almost all samples. However, the highest pH (8.1) value was recorded at the Katora Lake and the lowest pH (7.5) value was observed at Pari Lake, Seri Kalam, and Nasir Ullah Lake. Fluctuations of the pH values are due to altitudinal differences in the tested lakes and river contamination sources and location. A similar result in terms of pH values was reported for Hemkund lake (High altitudinal snow fed Lake) (Deep et al. 2020), neutral pH was observed in lake Satopanth (High altitudinal glacial Lake) (Sharma and Kumar 2017)

Table 1. Describes descriptive statistics and maximum permissible limits of Lakes and River Water (n = 26) for physicochemical parameters and heavy metals.

| Parameters       | Minimum | Maximum | Mean   | Standard Deviation | WHO standard values |
|------------------|---------|---------|--------|--------------------|---------------------|
| Color            | Colorless |        |        |                    |                     |
| Odor             | Unobjectionable |    |        |                    |                     |
| Taste            | Unobjectionable |    |        |                    |                     |
| Turbidity (NTU)  | 2       | 4.1     | 2.9    | 0.62               | < 5                 |
| pH               | 7.5     | 8.1     | 7.72   | 0.17               | 6.5-8.5             |
| EC (μS/cm)       | 20      | 85      | 41.96  | 16.39              | 200                 |
| Hardness (mg/L)  | 48      | 112     | 74.31  | 15.68              | 500                 |
| Cl (mg/L)        | 9.4     | 68      | 16.20  | 10.96              | 250                 |
| TDS (mg/L)       | 9.5     | 40      | 19.52  | 7.78               | 200                 |
| SO$_4$(mg/L)     | 2       | 17      | 6.19   | 4.13               | 45                  |
| NO$_3$(mg/L)     | 1.1     | 9.1     | 3.75   | 1.97               | 45                  |
| COD (mg/L)       | 56      | 200     | 108.88 | 29.36              |                     |
| HCO$_3$ (mg/L)   | 24      | 58      | 40.69  | 9.24               | 0.5                 |
| DO               | 2.12    | 5.8     | 3.76   | 0.69               | 5.0                 |
| Zn (mg/L)        | 0.003   | 0.182   | 0.03   | 0.05               | 3.0                 |
| Cu (mg/L)        | 0.0012  | 0.0053  | 0.00   | 0.00               | 2.0                 |
| Fe (mg/L)        | 0.02    | 0.126   | 0.06   | 0.04               | 0.3                 |
| Cd (mg/L)        | 0.019   | 0.052   | 0.04   | 0.01               | 0.02                |
| Cr (mg/L)        | 0.005   | 0.178   | 0.08   | 0.05               | 0.5                 |
| Ni (mg/L)        | 0       | 0.17    | 0.06   | 0.04               | 0.02                |
| Mn (mg/L)        | 0.035   | 0.146   | 0.09   | 0.03               | 0.01                |
| Pb (mg/L)        | 0.027   | 0.91    | 0.51   | 0.32               | 0.003               |
| As (mg/L)        | 0.001   | 0.002   | 0.001  | 0.001              | 0.01                |
The values of total dissolved solids (TDS) also vary and their values range from 9.5 to 40 mg/L. The value of TDS was maximum at Kalam and minimum at Chokyatan. Higher TDS values hinder photosynthesis, reduce water clarity and elevate temperature (Rahman and Gagnon 2014). The observed TDS values were found lower than the WHO standards value of 1000 mg/L. The lower values of TDS are the indication of freshwater and remote areas. Similar results were observed by Das et al. (2021). Higher values were observed in lakes water by Bhat et al. (2011) in Pangong Lake and by Rawat et al. (2021) in Lake Tarakund. The observed EC values were almost similar with a mean value of 41.96 µS/cm. The EC values were lower than the WHO standard value of 80–1000 µS/cm indicating lower mineral dissolution. The mean values of anions and other water quality parameters were observed according to the following decreasing order: bicarbonate > chloride > sulfate > nitrate. The hardness of the water was observed lower than the WHO standard values (48 and 112 mg/L respectively) and lower values of hardness the indication of soft water. The hardness of the water is due to the presence of the ionic species, anions and cations. The values of bicarbonate HCO₃ were observed to range from 24 mg/L to 58 mg/L. The responsible source of HCO₃ in surface water could be the rock weathering process and dissolution of silicate and carbonates minerals by carbonic acids as well as from gaseous CO₂ (Pandey et al. 2001; Chabukdhara and Nema 2012). The main source of sulphate in water is weathering of pyrite and marcasite and its range varying from 2 to 17 mg/L which is similar to the result reported by Das et al. (2021) in the lake Satopanth Tal. However lower values of sulphate were observed by Ghimire et al. (2013) in the lakes of Nepal and Rawat et al. (2021) in lake Tarakund. Eutrophic nature of the water is determined through nitrate values in the water body and the range of nitrates in this study was from 1.1 to 9.1 mg/L the nitrates concentration was observed lower than the (45 mg/L) WHO standard values for human consume water. These results were somewhat similar to the study reported by Rawat et al. (2021) in lake Tarakund and Bhat and Pandit (2014) for nitrates in Wular Lake. The chloride ions do not form insoluble precipitates as it cannot be absorbed into inorganic surfaces or mineral (Embaby et al. 2016). Chloride is utilized for the of water quality determination. The maximum and minimum values for chloride ions observed in the present study were from 9.4 to 68 mg/L. Analogous finding were observed by Das et al. (2021) in the lake Satopanth Tal. However lower values of chloride were observed by Rawat et al. (2021) in lake Tarakund.

Chemical oxygen demand is commonly used to measure the amount of organic compounds present in water which makes COD as an indicator of organic pollution in surface water (Kumar et al. 2011). COD points to a deterioration of the water quality caused by the discharge of industrial effluent (Mamais et al. 1993). The maximum and minimum concentrations of COD observed in the present study were 200 and 56 and 200 mg/L. The average value of the COD was 108.88 mg/L, and the highest COD was reported in sample Chota kalam and lowest in Chakdara. All of these values were far below the safe limit suggested by WHO and are supported by Pan et al. (2016). Dissolved Oxygen (DO) is vital for the health of aquatic life. The DO values varied between 2.12 and 5.80 mg/L. The median value of DO (3.762 mg/L) was lower from the limit developed in the national legislation (>5 mg/L). However, only
one sample (5.80 mg/L) exceeds the standard value. Similar results were reported by Thakur et al. (2013) for the Rewalsar Lake; Sharma and Tiwari (2018) for Nachiketa Tal and Rawat et al. (2021) in lake Tarakund. Turbidity of water indicated water clarity and is vital to analyse water quality. The turbidity of water is due to suspended particles inhibiting the transmission of light through the water, and it leads to damage to local fora and fauna (Ustaoğlu et al. 2020; 2020) The mean, maximum and minimum values of the turbidity value (>5 NTU) was lower than the established limit (100 NTU). Lower turbidity values in lentic environments were also observed by (Wu et al. 2016; Li et al. 2019). However higher turbidity was observed by Rawat et al. (2021) in lake Tarakund. It was concluded from the physicochemical parameters that the water of the Lake and River is satisfactory in terms of all the physical parameters tested in the present study and have no deleterious effects on aquatic biota and human health.

3.2. Heavy metals contamination in the lake and river water

The mean concentration of tested heavy metals in both the Lakes and River water is presented in and variates due to geographical locations. The concentration level of Pb (0.051 mg/L) was the highest, followed by that of Mn (0.09 mg/L), Cr (0.08 mg/L), Ni (0.06 mg/L), Fe (0.06 mg/L), Cd (0.04 mg/L) and Zn (0.03 mg/L). However, the lowest average level was found for as (0.001 mg/L) and Cu (0.00 mg/L). Lower concentration of Cu is previously observed in Mangla dam reservoir (Saleem et al. 2015). Significant fluctuations in the water samples were observed from different sites in terms of heavy metals especially Pb which has greater standard deviation as compared to other heavy metals. The mean concentration of all heavy metals was found higher from permissible limit in 34.61%, 100%, 100% and 88.46% samples of Mn, Cd, Pb and Ni respectively, except Cu, Fe, Cr, Zn. Iqbal and Shah (2013) reported highest concentrations of heavy metals such as Cd, Ni and Mn in Khanpur and Simly Lakes (Iqbal and Shah 2013). Muhammad Saleem et al. (2015) observed the elevated level of heavy metals such as Cd, Ni, Cr and Pb in Mangla reservoir Pakistan (Saleem et al. 2015).

Among heavy metals, Pb enter into surface water from industries, paint, vehicles and household emissions and have toxic and carcinogenic effects (Arain et al. 2009), as well as from geogenic mafic and ultramafic sources (Khan et al. 2018; Rashid et al. 2018; Liu et al. 2020). The average concentration of Pb was found higher than that in the studies by Shah et al. (2020) (Jehan et al. 2020) in water from the Swat River, Pakistan and lower concentrations of Pb were found in Lake Nasser by Rizk et al. (2022). Higher concentration of Pb was observed in various studies analyzing Lakes, Reservoir and River water (Ogoyi et al. 2011; Iqbal and Shah 2013; Saleem et al. 2015; Dey et al. 2021). Chronic exposure of Pb can cause nerves and brain disorders. Similarly, Ni has a significant toxic effect and carcinogenic effects on living organisms. The average concentration of Ni was found higher than those reported by Haq (2005) in both surface and groundwater in Pakistan, and Shah et al (2020) in surface water of River swat Pakistan (Jehan et al. 2020). Cr concentration is lower than WHO values. Lower concentration of Cr is detected in the Lake Bosomtwe Ghana
Cr is mutagenic and cause allergic reactions. Cd is very toxic for living cells and destroys both the DNA and membrane of living cells. Cd concentrations is higher than WHO standard values and are opposite to the results obtained by Rizk et al. (2022) in Lake Nasser. Higher concentration of Zn in water causes nausea, vomiting and appetite however lower concentration can cause sickle cell disease and chronic liver and renal diseases. The concentration of Zn is lower than the permissible limit of WHO and were analo-

gous to the previous study reported by Asare-Donkor et al. (2016) in Lake Bosomtwe Ghana, Shiretorova et al. (2020) in Baikal Lake Russia and by Rizk et al. (2022) in Lake Nasser. Anthropogenic activities and long-range air transport are the main cause of water contamination in Rivers and Lakes. Arsenic concentrations were lower than the detection limit and both the Copper and Arsenic concentrations were detected lower than the WHO standard values and are similar to results reported by Rizk et al. (2022) in Lake Nasser and Shiretorova et al. (2020) in Baikal Lake Russia. Higher concentration of Fe damage DNA, Lipids and proteins and cause coma and liver failure. Fe concentration were lower in this study and are opposite to the study reported by Asare-Donkor et al. (2016) in Lake Bosomtwe Ghana. The concentration of Mn in water causes cancer nervous and reproductive disorder. The concentration of Mn in this study is higher in river water as compared to lakes water.

Table 2. Describe health risk exposure of Lakes and River water consumed by children and adults of Swat and Dir Valley.

| Elements | ADD_{Children} | ADD_{Male} | ADD_{Female} |
|----------|----------------|------------|--------------|
| Cd       | Min Max average | Min Max average | Min Max average |
| Cu       | 0.0014 0.0039 0.0030 | 0.0010 0.0026 0.0020 | 0.0009 0.0024 0.0019 |
| Fe       | 0.0001 0.0004 0.0001 | 0.0001 0.0003 0.0001 | 0.0001 0.0002 0.0001 |
| Pb       | 0.0020 0.0683 0.0385 | 0.0014 0.0455 0.0253 | 0.0013 0.0427 0.0237 |
| Zn       | 0.0002 0.0137 0.0021 | 0.0002 0.0091 0.0014 | 0.0001 0.0085 0.0013 |
| Cr       | 0.0004 0.0134 0.0061 | 0.0003 0.0089 0.0041 | 0.0002 0.0083 0.0038 |
| Ni       | 0.0000 0.0128 0.0043 | 0.0005 0.0085 0.0029 | 0.0005 0.0080 0.0027 |
| Mn       | 0.0026 0.0110 0.0068 | 0.0018 0.0073 0.0045 | 0.0016 0.0068 0.0042 |
| As       | 0.0001 0.0002 0.0001 | 0.0001 0.0001 0.0001 | 0.0000 0.0000 0.0001 |

Non-cancer risk

| HQ_{Children} | HQ_{Male} | HQ_{Female} |
|---------------|-----------|-------------|
| Cd            | 2.850 7.800 6.040 | 1.900 5.200 3.996 | 1.781 4.875 3.746 |
| Cu            | 0.002 0.011 0.004 | 0.002 0.007 0.003 | 0.002 0.007 0.002 |
| Fe            | 0.000 0.000 0.000 | 0.000 0.000 0.000 | 0.000 0.000 0.000 |
| Pb            | 0.056 1.896 1.069 | 0.038 1.264 0.703 | 0.035 1.185 0.659 |
| Zn            | 0.001 0.046 0.007 | 0.001 0.030 0.005 | 0.000 0.028 0.004 |
| Cr            | 0.000 0.009 0.004 | 0.000 0.006 0.003 | 0.000 0.006 0.003 |
| Ni            | 0.000 0.638 0.215 | 0.025 0.425 0.144 | 0.023 0.398 0.135 |
| Mn            | 0.019 0.078 0.048 | 0.013 0.052 0.032 | 0.012 0.049 0.030 |
| As            | 0.250 0.500 0.365 | 0.167 0.333 0.244 | 0.156 0.313 0.228 |
| Hl            | 3.178 10.977 7.753 | 2.144 7.318 5.129 | 2.010 6.860 4.808 |

Cancer Risk

| Cancer risk Children | Cancer risk Male | Cancer risk Female |
|----------------------|------------------|-------------------|
| Cd                   | 0.021 0.059 0.045 | 0.014 0.039 0.030 | 0.013 0.037 0.028 |
| Pb                   | 0.017 0.580 0.327 | 0.011 0.387 0.215 | 0.011 0.363 0.202 |
| Cr                   | 0.000 0.007 0.003 | 0.000 0.004 0.002 | 0.000 0.004 0.002 |
| Ni                   | 0.000 0.012 0.004 | 0.000 0.008 0.003 | 0.000 0.007 0.002 |
| As                   | 0.000 0.000 0.000 | 0.000 0.000 0.000 | 0.000 0.000 0.000 |
| Thl                  | 0.039 0.657 0.379 | 0.026 0.438 0.250 | 0.025 0.411 0.234 |
3.3. Human health risk assessment

The outcome of health risk of both the Lakes and Rivers heavy metals present in water are presented in Table 2. The risk to health of the heavy metals in water was calculated according to USEPA the health risk assessment model and two population groups were considered (Adults and Children). The health risk related to water, HQingestion, HQdermal, and HI (HQingestion + HQdermal) were categorized as high to low level indicating significant to moderate hazardous risk. The values range from 0.00 to 1.90 for adults and 0.00 to 2.85 for children taking HQingestion is the exposure route and indicates potential noncarcinogenic risk to humans (Figures 2 and 3). The values of hazardous heavy metals for the HQdermal route were > 1 for both the adults and children (adults 0.00 to 5.20, children 0.00 to 7.80), indicates potential hazards to both the adults and children. The values of HQingestion, HQdermal, and HI for children were observed to be higher than those for adults and the order of HI in surface water was Cd > Pb > As > Ni > Mn > Zn > Cu > Fe in children and Cd > Pb > As > Ni > Mn > Zn > Cu > Fe in adults. The present study indicates that heavy metals posed risk to the local inhabitants causing diseases such as cardiac arrest, anemia, lung and stomach cancer and hypertension. The results in our study for HQdermal and HQingestion was observed to be higher than unity for both the Cd and Pb. Indicating higher risk of water samples through dermal and skin exposure.

3.4. Multistatistical analysis

3.4.1. Pearson correlation matrix analysis

To find out the inter-elemental correlation matrix of the physiochemical parameters and heavy metals (Variables) in the water of Lakes and River of Swat and Dir Khyber Pakhtunkhwa Pakistan the Pearson correlation matrix analysis was utilized. The Pearson correlation matrix analysis performed among the heavy metals and physiochemical parameters revealed moderate to strong correlation defining those heavy metals and physiochemical parameter (Variables) are closely associated in the water
of the study area (Table 3). The p value shows the significance level of correlation matrix and signifies the strength of correlations among (Variables) such as p value having 0.01 and 0.05 values indicate strong and significant correlations respectively. Further, significant positive correlation ($p < 0.05$) was found between: TDS and EC ($r = 1$), COD and EC ($r = 0.59$), Cu and EC ($r = 0.50$), Cu and TDS ($r = 0.50$), COD and TDS ($r = 0.59$), Cu and SO4 ($r = 0.57$), Mn and Cr ($r = 0.77$), Cr and Pb ($r = 0.76$), As and Mn ($r = 0.45$), As and Fe ($r = 0.80$). It is concluded from both the principal component analysis and person correlation matrix analysis that both the anthropogenic sources from the industrial waste and long-range air transport would be the main cause of toxic metals presence in the present study area.

### 3.4.2. Principal component analysis (PCA)

Heavy metals and physiochemical parameters correlation can predict the important information such as the emission pathways and the source of these pollutants. The principal component analysis was used to find out the origins of contamination and their aim is to reduce the multivariate dataset dimensionality into less components which are used to explains important information of the dataset. The Bartlett’s sphericity test was significant ($p < 0.0001$) and the Kaiser-Meyer-Olkin (KMO) test value for water was 0.470 which indicates that the PCA was nonidentity matrix and effective for the studies (Varol 2011). The whole dataset PCA contributes a maximum of five components and cover for 71.28% of the total variance having eigenvalue ($\lambda > 1.0$). Varimax rotation is used to maximize the sum of variances of the factor coefficients and to better describes the heavy metals influential possible factors. Figure 4a describes the scree plot of eigenvectors which is used to identify function of factor number and the prominent principal components number was takes using Kaiser criterion which is having eigenvalue greater than 1 (Kaiser 1960). The scree plot has obvious change of slope after the fifth eigenvalue in the present study. Majority of the physicochemical parameters indicates similar behavior such as major ions Cl, SO4, NO3, Fe and HCO3. These variables are due to ion exchange and water rock interaction. The origin of Fe in surface water is due to rock weathering of minerals

![Figure 3. Shows cancer risk of PTEs used by three age groups in the study area.](image-url)
Table 3. Pearson correlation table shows correlating pairs in lakes and river water of Swat and Dir valley.

| Variables | pH | EC | Hardness | Cl | TDS | SO4 | NO3 | COD | HCO3 | DO | Zn | Cu | Fe | Cd | Cr | Ni | Mn | Pb | AS |
|-----------|----|----|----------|----|-----|-----|-----|-----|------|----|----|----|----|----|----|----|----|----|----|
| pH        | 1.00 |    |           |    |     |     |     |     |      |     |    |    |    |    |    |    |    |    |    |    |
| EC        | -0.14 | 1.00 |           |    |     |     |     |     |      |     |    |    |    |    |    |    |    |    |    |    |
| Hardness  | 0.09 | 0.05 | 1.00      |    |     |     |     |     |      |     |    |    |    |    |    |    |    |    |    |    |
| Cl        | 0.08 | 0.19 | 0.04      | 1.00 |     |     |     |     |      |     |    |    |    |    |    |    |    |    |    |    |
| TDS       | -0.14 | 1.00 | 0.20      | 1.00 |     |     |     |     |      |     |    |    |    |    |    |    |    |    |    |    |
| SO4       | -0.17 | 0.31 | -0.05     | 0.02 | 0.30 | 1.00 |     |     |      |     |    |    |    |    |    |    |    |    |    |    |
| NO3       | 0.01 | 0.41 | -0.13     | -0.18 | 0.42 | -0.21 | 1.00 |     |      |     |    |    |    |    |    |    |    |    |    |    |
| COD       | -0.20 | 0.59 | 0.09      | -0.04 | 0.59 | 0.38 | 0.04 | 1.00 |      |     |    |    |    |    |    |    |    |    |    |    |
| HCO3      | 0.36 | 0.12 | 0.22      | 0.27 | 0.13 | -0.31 | 0.17 | 0.08 | 1.00 |     |    |    |    |    |    |    |    |    |    |    |
| DO        | 0.28 | 0.11 | -0.02     | 0.07 | 0.10 | 0.19 | 0.19 | -0.05 | 0.24 | -0.02 | 0.24 | 0.11 | 1.00 |     |    |    |    |    |    |
| Zn        | 0.00 | 0.19 | -0.19     | -0.10 | 0.19 | -0.19 | 0.19 | -0.05 | 0.24 | -0.02 | 0.24 | 0.11 | 1.00 |     |    |    |    |    |    |
| Cu        | -0.48 | 0.50 | -0.18     | -0.05 | 0.50 | 0.57 | -0.08 | 0.39 | -0.38 | -0.24 | -0.04 | 1.00 |     |    |    |    |    |    |    |
| Fe        | 0.07 | -0.22 | 0.03     | 0.04 | -0.23 | -0.05 | -0.01 | -0.56 | -0.42 | -0.01 | -0.10 | 0.05 | 1.00 |     |    |    |    |    |    |
| Cd        | -0.22 | -0.30 | -0.24     | -0.08 | -0.31 | -0.12 | -0.10 | -0.14 | -0.28 | -0.07 | 0.01 | -0.02 | 0.02 | 1.00 |     |    |    |    |    |
| Cr        | 0.03 | -0.75 | -0.21     | -0.13 | -0.75 | -0.07 | -0.62 | -0.31 | -0.32 | 0.04 | -0.17 | -0.04 | 0.14 | 1.00 |     |    |    |    |    |
| Ni        | 0.19 | -0.47 | 0.04      | -0.30 | -0.48 | 0.04 | -0.14 | -0.17 | -0.09 | 0.04 | 0.01 | -0.26 | -0.04 | -0.04 | 0.42 | 1.00 |     |    |    |
| Mn        | -0.30 | -0.42 | -0.45     | -0.22 | -0.41 | 0.08 | -0.51 | 0.08 | -0.20 | -0.35 | 0.01 | 0.25 | -0.30 | 0.21 | 0.77 | 0.21 | 1.00 |     |    |
| Pb        | 0.35 | -0.79 | 0.15      | -0.03 | -0.79 | -0.37 | -0.48 | -0.54 | 0.29 | -0.12 | 0.07 | -0.56 | 0.08 | 0.13 | 0.76 | 0.38 | 0.31 | 1.00 |
| AS        | -0.13 | -0.02 | -0.18     | -0.20 | -0.02 | -0.12 | -0.04 | 0.18 | 0.30 | -0.01 | 0.11 | -0.14 | 0.80 | -0.14 | 0.25 | 0.22 | 0.45 | 0.13 | 1.00 |
such as muscovite, mica, fluorite and biotite. Bicarbonates are due to dissolution and weathering of minerals such as carbonate and albite and SO$_4$, NO$_3$ are due to the surrounding agriculture in the river side however in the Lake side these ions are leached from local animal grazing in large extent. Table 4 Explain the percentage of variance, cumulative percentages and factor loadings by each factor and Figure 4b. Describe the principal components analysis, components plot in circulatory form. The PCA first component varifactor (VF1) was responsible for 26.61% of the total correlated variance (loading > 0.90) with TDS and EC and indicating the strength of physiochemical parameters such as TDS and EC which have dominant role in factor loading contribution. The VF2 is responsible for 15.90% of the total variance and Cu and Mn are mainly responsible having high loadings (> 0.60). The possible source of these metals is correlated with anthropogenic activities (Gao et al. 2019). VF3 indicates that Fe is responsible factor for the contamination of water quality in the study area and
their loading value was observed to be greater than 0.80 and have linked with both the anthropogenic and natural sources. The VF4 and VF5 indicates significant loading for Zn and NO3 (> 0.40) and Ni (> 0.60), respectively and promotes the dissolution of Ni in the water aquifers in the study area.

### 3.4.3. Hierarchical cluster analysis

For the reduction of the clustering error, hierarchical agglomerative clustering was used and categorized the freshwater data into three main classes of (1) severely polluted, (2) polluted and, (3) less polluted cluster. Within the class variability is 61.01% and between the classes’ variability is 36.99%. In cluster C1 the distance between the class centroid were (0, 25.489 and 51.71) for less polluted class. The distance between the class centroid for moderate polluted cluster C2 were (25.489, 0 and 56.70). Similarly for cluster C3 severely polluted were (51.71, 56.70 and 0) respectively. Cluster C1, C2 and C3 represent 8, 11 and 7 lake and river water samples. C1 and C2 contain 8 and 11 lake water samples. Class C3 represents 7 samples from both the river swat and river panjkora. The centralized objects for the lake and river water data of three cluster analysis C1, C2 and C3 were Observation 3, 26 and 17 respectively. The mean concentrations of pH, EC, hardness, Cl, TDS, COD, HCO3, DO, Zn, Cu, Fe, Cd, Cr, Ni, Mn, Pb, As were calculated to be (7.850, 45.875, 82.000, 19.800, 21.250, 4.375, 4.913, 94.875, 46.250, 4.138, 0.051, 0.001, 0.079, 0.039, 0.046, 0.052, 0.052 and 0.324 mg/L, respectively) for class C1 in the study area. Similarly, for C2 were (7.700, 26.818, 72.000, 13.527, 12.409, 5.636, 2.427, 99.000, 39.636, 3.467, 0.014, 0.002, 0.055, 0.042, 0.135, 0.073, 0.116, 0.329 and 0.002 mg/L, respectively) in the lakes and river water samples of the study area. Similarly, for C3 were (7.586, 61.286, 69.143, 16.286, 28.714, 9.143, 4.486, 140.429, 36.000, 3.797, 0.025, 0.003, 0.037, 0.039, 0.079, 0.046, 0.052, 0.052 and 0.324 mg/L, respectively) for class C3 in the study area.

#### Table 4. Principal component analysis results of lakes and river water of Swat and Dir valley.

| Component | F1  | F2  | F3  | F4  | F5  |
|-----------|-----|-----|-----|-----|-----|
| pH        | -0.20 | -0.62 | -0.15 | -0.17 | 0.19 |
| EC        | 0.96  | 0.03  | -0.14 | -0.03 | -0.04 |
| Hardness  | 0.07  | -0.38 | 0.01  | -0.59 | 0.24 |
| Cl        | 0.17  | -0.25 | 0.05  | -0.45 | -0.62 |
| TDS       | 0.96  | 0.03  | -0.16 | -0.04 | -0.05 |
| SO4       | 0.34  | 0.50  | 0.18  | -0.33 | 0.31 |
| NO3       | 0.50  | -0.37 | -0.09 | 0.60  | 0.13 |
| COD       | 0.59  | 0.41  | -0.36 | -0.25 | 0.11 |
| HCO3      | 0.03  | -0.48 | -0.69 | -0.19 | -0.27 |
| DO        | 0.14  | -0.46 | -0.09 | 0.21  | 0.19 |
| Zn        | 0.08  | -0.07 | -0.31 | 0.46  | -0.10 |
| Cu        | 0.50  | 0.67  | 0.28  | -0.08 | 0.00 |
| Fe        | -0.13 | -0.28 | 0.87  | 0.05  | 0.03 |
| Cd        | -0.26 | 0.21  | 0.27  | 0.39  | -0.35 |
| Cr        | -0.86 | 0.36  | -0.12 | -0.11 | -0.07 |
| Ni        | -0.52 | 0.04  | -0.14 | 0.02  | 0.65 |
| Mn        | -0.50 | 0.77  | -0.22 | 0.05  | -0.13 |
| Pb        | -0.89 | -0.21 | -0.15 | -0.17 | -0.11 |
| As        | -0.14 | 0.29  | -0.83 | 0.12  | 0.06 |
| Eigenvalue| 5.06  | 3.02  | 2.52  | 1.64  | 1.31 |
| Variability (%) | 26.61 | 15.90 | 13.24 | 8.61  | 6.91 |
| Cumulative %  | 26.61 | 42.51 | 55.75 | 64.37 | 71.28 |
0.037, 0.038, 0.092, 0.038 and 0.002 mg/L, respectively) in the present study area. The details of the HCA are presented in Figure 5.

4. Conclusion

The present study is based on water quality especially heavy metals evaluation of water from Lakes and Rivers. These Lakes and Rivers are the main source of water and utilized for various applications such as agricultural, aquaculture, hydroelectric power plants, recreational and drinking purposes for millions of people. Among the

Figure 5. Clustering analysis of water data in the study area.
investigated heavy metals, Ni, Cd, and Pb are found greater than the maximum permissible values. Further, the poor water quality in the present study area is mainly due to heavy metals. Lakes are observed to be more polluted than the rivers. The studied Physiochemical parameters are within the standard recommended values. Further, in future geochemical analysis of the selected lakes should be analyzed to link the contamination sources of the Lakes. Mercury is one of the great problems in high altitudes and should be considered in future for heavy metals analysis. Moreover, the seasonal analysis can be helpful to better understand about the contamination level and sources (Inaotombi and Gupta 2014; Kumar and Sharma 2018; Rana et al. 2018).

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Disclosure statement

No potential conflict of interest was reported by the author(s).

Consent to participate

All authors are voluntarily agreed to participate in this research study

Availability of data and codes

The data and codes used to prepare the manuscript are available with the corresponding author and can be provided upon request.

Ethical statement

All procedures followed were in accordance with the ethical standards of the Helsinki Declaration of 1975, as revised in 2000.

Consent to publish

All authors are agreed to publication; there is no legal constraint in publishing the data used in the manuscript.

Authors’ contribution

All authors contributed equally.

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