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Ecological Risk Analysis of Stream Sediments Data and Impact on Aquatic Biota around Mining and Agricultural Regions

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Abstract

Potentially harmful elements enter into the environment through mining and agricultural activities, causing water and stream sediment pollution. Ecological risk analysis helps to determine sediment pollution, to recommend remediation measures for human health safety and the survival of aquatic species. The sediments were analysed for acidity and redox potential using a pH-meter and spectrophotometer, respectively. Nickel, cadmium, arsenic, chromium, lead, zinc, and iron were measured using atomic absorption spectrophotometer. The mean value of Cd exceeded the threshold effect limit guideline indicating its adverse effect to water dwelling organisms. Anthropogenic metal input identified cadmium, lead, arsenic, zinc and chromium contamination in locations 3, 6, and 7. Modified risk assessment code, toxic response index and comprehensive ecological risk values exhibited considerable to high ecological risks in locations 3, 6, and 7. The highest comprehensive ecological risk value recorded 653.2 in location 3, showing high ecological risk to water dwelling organisms. Durbin Watson ecological risk value (2.34) is between a critical value of $1.5 < d < 2.5$ showing auto correlation of the data. Potentially harmful elements obtained Durbin Watson value of 2.77, which exceeded the range showing lack of auto correlation. Strong correlation of arsenic, lead and zinc showed their affinity and common source of enrichment. Principal component analysis indicated that the sources of the elements were mostly geological weathering, sewage disposal, industrial wastes and agricultural fertilizers. The study integrated recent ecological risk indices with multivariate and regression statistics. This is helpful in interpreting related environmental problems by scientists in other parts of the world.

1. Introduction

Accumulation of soil and sediments with potentially harmful elements (PHEs) leads to ecological hazards [1]. Environmental pollution with PHEs is associated with human health risks [2]. Environmental geochemical studies provides data on geochemical characteristics, distribution and dispersion of PHEs in the environment [2]. Mineral exploration is associated with environmental pollution [3]. Mineral exploration has its economic benefits but it also causes environmental consequences [4]. Both underground and open cast mining methods are associated with levels of environmental pollution [5]. Soil acidity (pH) and oxidation potential (eH) control the ease of movement and bioavailability of PHEs [6]. Analysis of stream sediments reveal the impacts of separate human activities on aquatic species depending on stream sediment for their nutritional requirements. Sediments are the final sink of contaminants in an aquatic ecosystem and control groundwater, surface water, plants and animal species [7, 8]. Concentrations of PHEs in water above their threshold can become hazardous to sediment dwelling species causing death,

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retarded growth and decrease in their reproductive potential [9, 10]. Lead (Pb), cadmium (Cd), and arsenic (As) are toxic, have no biological benefit and deleterious to organisms even at low concentrations [11]. On the contrary, zinc (Zn), iron (Fe), copper (Cu), chromium (Cr), and manganese (Mn) are useful to living species within their threshold standards that are not toxic [11]. PHEs enter into the environment through chemicals in agricultural practices, mining, ceramic etc. Cr enters into the environment through paints, pigments, fungicides, preparation of catalyst, and alloys. Zn is mostly from mining, bronze is an alloy of brass and die-casting while iron oxides are utilised in pigments and water treatment coagulants [11]. Mining and farming activities contaminate the environment with heavy metals [12]. Mining activities have both negative ecological effects and positive economic and social gains [13]. Heavy metals can enter into the food chain and constitute human health threats. Open pit mining is environmentally sustainable through compliance with regulatory guidelines.

This study focussed on PHEs entry into the environment via cultivation of farmlands, and mining activities that are common in the study area. The residents have no source of potable water and recourse to water sources drained by runoffs from Pb-Zn mine ponds around cultivated farmlands. These farmlands contain fertilizers and pesticides, which are sources of PHEs. Stream sediments serve as major sinks where contaminants can be stored and as a sink of water pollutants and water dwelling organisms [14]. A thorough water quality study requires assessing the bottom sediments quality. Some water dwelling organisms are dependent on minor species for nutrients. The death of these minor species lead to the extinction of major water dwelling organisms causing a loss of biodiversity in the aquatic environment. Agricultural practices are likely sources releasing PHEs into water channels, but this area of research has not received considerable attention. This study seeks to address how cultivation activities contribute to stream sediment contamination with PHEs. Stream sediments contamination with PHEs degrade water quality and renders it unsuitable for domestic, industrial, irrigation and other uses.

Different studies, considered the environmental and human health consequences of mining activities [15-17]. The study of freshwater using sediment quality guidelines (SQGs) helps to identify PHEs suspected to be harmful to organisms that dwell in sediments [18]. Probable effect concentration (PEC) enables the assessment of sediments containing diverse chemical contaminants [19]. background values of some elements were estimated to exceed the probable effect level (PEL), severe effect level SEL) and toxic effect threshold (TET) [18]. The use of SQGs in assessing heavy metal contamination of stream sediments obtained metal concentrations lower than their proposed threshold effects [19]. This indicated no harmful effect from the elements. The concentrations of Pb was found tobe above the threshold probable effect level (PEL) in stream sediments. [20]. Heavy metal contamination study of stream sediments applied effect range low (ERL), effect range medium (ERM) and single pollution risk indices [21]. The study established high levels of Cd and identified geogenic, agricultural runoff, and atmospheric sources of heavy metal pollution in the stream sediments. A fresh water ecosystem study applying SQGs showed that threshold effect concentrations (TEC) and probable effect concentration (PEC) predicted the absence of sediments toxicity [22]. In addition, probable effect concentration (PEC) of the elements exceeded the standard compared and harmful to sediments dwelling organisms. River Basin stream sediments analysis recorded mean concentrations of Pb above SQGs, while Ni, Mn and Zn were lower than their average world shale values [23]. New indices for ecological risks analysis such as modified hazard quotient (mHQ) and ecological contamination provide a better approach in sediment contamination studies [24]. More recent ecological risk indices such as toxic risk index (TRI), mean ERM quotient (mERMQ) and contamination severity index (CSI) are reliable indicators of sediment pollution [25]. These indices can evaluate the impact of heavy metals in aquatic organisms. Ecological analysis of heavy metals in sediments using pollution indices and multivariate analysis reported different ranges of ecological risks [26, 27-31]. Contamination indices and risk assessment analysis identified geo-chemical and mineralisation processes, and mining activities as major sources of heavy metal contamination [32, 33]. Analysis of watershed stream discovered that streams receiving effluents from car wash and agricultural areas obtained higher metal values than the surrounding streams [34]. Application of minimum/maximum autocorrelation factor (MAF) and sequential Gaussian simulation in heavy metals pollution study was confirmed to be simple and accurate by various statistical checks [35].

A study on the sustainability of a mine using the modified Folchi method quantified the environmental impact caused by mining activities

[36]. The use of renewable energy in mining operations can reduce the cost and impact of mining on the environment [37]. The limiting factors with its usage are lack of knowledge of its advantages, inadequate trained personnel and high equipment maintenance cost. Diverse safety risks and management processes in underground mines help in reducing accidents among miners [38]. The main hazards identified in underground mines are required airflow, lack of proper scaling, post blasting scaling and proper ventilation of dust [38]. Studies on risk management applied game theory, multicriteria decision-making methods and decision matrix [39, 40]. Economic risks were the most important risks and social risk the least significant. These studies promote the health and safety of workers. Reduction risk can reduce the product grade and its seller price with an additional cost to generation of wastes, as obtained in a manganese deposit [41]. The study used Fuzzy Delphi Analytical Hierarchy Analysis method for weighting while Multi-Attributive Approximation Area Comparison (MABAC) estimated the dilution for each mine. Ranking in dilution risk of the different mines tested was by cavity monitoring system and Technique for Order of Preference by Similarity to Ideal Situation (TOPSIS). The study concluded that MABAC method is preferable to TOPSIS method in mine dilution ranking.

The cause of acid mine drainage risk was determined using statistical regression analysis in an abandoned pile coalmine [42]. Sensitivity analysis done by the tornado diagram shows that the pile depth was by far the most critical factor affecting the remaining pyrite. Risk assessment of long walls, fly rock, dimension stone cutting and rock fall are for the safety of miners and sustainable mining operations [43-46]. The studies applied matrix priority, fuzzy fault tree analysis; multicriteria decision-making, failure mode, effects analysis and semi quantitative techniques in assessing risks. These risks analyses are important because they can cause loss, injuries, fatalities and destruction of mine equipment [44, 46].

This study considered the possibility of stream sediments pollution from both farmlands and mining activities. It will establish the extent of environmental pollution caused by mining for economic minerals, cultivation of farmlands and highlight the most suitable method for computing ecological risks in stream sediments. It will reveal whether mining activities contribute to environmental pollution more than agricultural activities and recommend remediation measures. The ultimate goal of this study is to assess the environmental risks of PHEs in stream sediments and their impact on water dwelling organisms. This aim is achievable through analysis of stream sediments for Cd, Pb, Ni, As, Zn, Cr, and Fe. Comparison of the analysed data with SQGs such as PEC, TEL, and ERM to ascertain the chance of survival of aquatic species in the water sources. Analysis of data using pollution indices such as anthropogenic metal input, mCd, SPI, and NCPI. Compute recent ecological indices like mRAC, TRI, RI, mPELQ, CSI, and mERMQ. Applying multivariate and regression statistical analysis to determine sources and relationships of chemical pollutants in stream sediments. This study will provide a basis of monitoring sediment and water quality within mining and agricultural regions on a regional scale. It will further elucidate the usefulness of sediment quality guidelines, recent ecological risk indices, multivariate statistics and regression statistical analysis in sediment pollution studies. This is an integrated approach with a reliable data in the analysis of sediments. The study will provide a guide for planning a mine and agricultural sites for the overall water management in the studied area.

2. Description of Studied Area

The studied area is situated between latitudes 6^0 14’ 0’’ to 6^0 9’ 30’’N and longitude 8^0 0’ 30’’ to 8^0 10’ 30’’ within the Abakaliki mining district in southeastern Nigeria (Figure 1). The inhabitants of the study area are agrarian, living in dispersed and scattered settlements. The terrain is characterised by low and high reliefs with outcrops of shale mineralised with zinc and lead ores [47]. The study area falls under the Abakaliki shale in the Lower Benue Trough in the rainforest zone [48]. The area is characterised by the rainy season, which last from April to October and the dry season caused by winds from the Sahara desert starts in November and end in March. The wet season is caused by ocean wind sweeping across the Atlantic Ocean [49]. The average annual rainfall is estimated at 1500 mm. The temperatures varies between 20 ^{0}C to 30 ^{0}C in the dry season and 16 ^{0}C to 28 ^{0}C in the wet season [50, 49]. The vegetation is dominantly grassy, and characterised by scrubs and tall trees [50]. The drainage is dendritic, the rivers flow from mines sites, and the runoffs enter into stream channels. There are Ishiagu, Eyingba, Ameri and Ameka mines in the area (Figure 1). The study area consist of brown to dark grey coloured and jointed shale with evidence of fissility. The shale and mud rocks are the host rocks for Pb-Zn

mineralisation intruded by baked shale [48]. The area is part of the Abakaliki Anticlinorium underlain by shale of the Asu River Group. The Asu River Group comprises a sequence of shale, mudstone, and siltstone alternations including sandstone, and lenses of limestone in few locations [51]. The sediments in the area are from poorly bedded sandstone and limestone lenses [52]. The shale is pink blue, pale grey due to weathering and Fe content [53].

3. Materials and Methods

3.1. Sample collection

Active sediments were collected from the bottom of the middle stream channels. The study covered an estimated area of 342.8 km^2. Ten stream sediments were collected from farmlands and mine sites areas while 1 sample was collected as a control sample distant from farming and mining areas (Figure 1). The samples were collected along stream and river courses and their tributaries around mine sites and cultivated farmlands. Stream sediments were sampled using a hand-held trowel made of plastic material not a metal hand trowel to avoid sample contamination. The samples were collected into clean unused polyethylene bags. The plastic hand trowel was washed using detergent and dried to avoid compromising the quality of samples. Duplicate samples were collected in each location. The samples were later mixed to form a composite and representative sample during sample preparation.

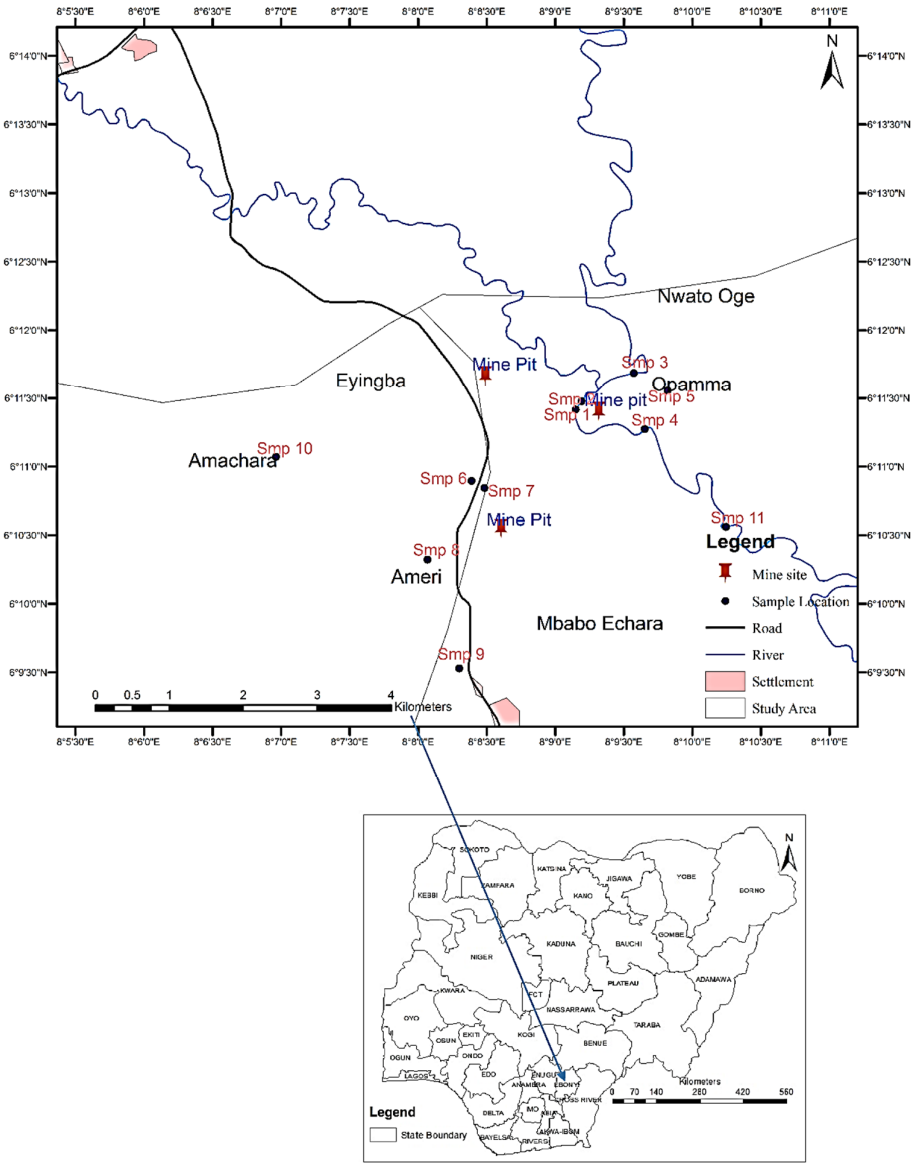

Figure 1. Sample location map of the studied area.

3.2. Preparation of samples and laboratory analysis

The stream sediment samples were air dried for three days and then later pulverised and sieved using a 2 mm mesh nylon sieve to separate large grains from fine grains. The two sub samples were mixed to form a composite sample. The composite samples were ground using an agate mortar and pestle. The ground samples were carefully put into sealed envelopes. The samples were well labelled for easy identification during chemical analysis.

The samples were digested to destroy the matrix, which could inhibit atomization and possibly change in a single oxidation state [49]. After shaking to mix the sample thoroughly, 50 mL of each sample was put into a glass beaker and 20 mL of nitric and perclhoric acid mixture was added. The beaker with its contents were placed on an electric hot plate and evaporated down to about 20 mL. The cooled samples were filtered using Whatman 42 filter paper to remove some of the insoluble materials to avoid clogging of the atomizer. The volume was adjusted to 50 mL using distilled water free of metallic elements.

The samples were analysed using atomic absorption spectrophotometer (AAS). AAS can analyse the concentration of an element in the sample. The instrument requires a standard solution to establish the relationship between measured absorbance and the analytical and the analyte concentration relying on Beer Lamberts Law. In its elemental form, metals will absorb ultraviolet light and get excited. Each metal element possess a characteristic wavelength that will be absorbed. The AAS instrument identifies a particular metallic element concentration by concentrating a beam of ultraviolet light at a specific wavelength through a flame and into a detector. The metal of interest is aspirated into the flame. If the metal is present and the intensity is reduced, the instrument measures the variation in intensity of the flame [54].

3.3. Quality assurance and quality control

A control sample was collected from the field. In addition, blanks and duplicates were run as some of the samples. A thousand milligrams per litre standard solutions of each metal was used for calibrating the instruments. Mixed working standard solutions containing all the metals were prepared by dilution in appropriate procedures using doubled distilled deionised water. The measurement for each resolution was done in triplicate and the average was recorded [55]. The accuracy of the analytical method was determined by drawing calibration curves and the simultaneous performance of analytical blanks [56, 57].

3.4. Statistical treatment of data

The SPSS version 20 software was used to perform descriptive statistics, correlation analysis (CA), principal components analysis (PCA), and regression statistics. CA and PCA aided in evaluating sources and relationships among geochemical variables [57]. Varimax rotation was done to classify the loading variables and preclude ambiguities in the component matrix. Mat Lab 2013 version software was used in the construction of boxplots. PCA generates correlation matrix, extracted factors based on correlation coefficient of variables and maximised the relationship between some factors and variables [58].

3.5. Stream sediments pollution with heavy metals

Anthropogenic metal input was computed using the equation stated as follows:

$$AMI = \frac{X - X1}{X1} \text{ X } 100 \quad (1)$$

where X represent average concentration of the metal in sediments,

X_1 is the average background concentration of the metal [59].

Single pollution index was determined using the relation:

$$Pi = \frac{Ci}{Si} \quad (2)$$

where Pi represents single pollution index,

Ci stands for concentration of heavy metal i in stream sediment sample.

Si denotes the threshold value of heavy metal [60].

Numerov composite pollution index (NCPI) is given by the expression:

$$\text{NCPI} = \frac{\sqrt{(Pimax)^2 + (\text{P})^2}}{2} \quad (3)$$

where NCPI represents Numerov composite pollution index.

Pi_{max} is the maximum value of single pollution index.

P represents the mean value of SPI. SPI and NCPI classification are presented in Suppl. Data 1 and 2.

Contamination factor (Cf) is given as [61, 62]:

$$Cf = \frac{\text{Conentration of metal in sample}}{Background\ concnetration\ of\ metal}$$

Degree of contamination (Cd) is computed using the expression:

$$Cd = \sum_{ki=1}^{n} Cf$$

Modified degree of contamination is expressed as:

$$mCd = \frac{\Sigma Cf}{n} \quad (4)$$

where cf = contamination factor.

Cf and mCd was classification are presented in Suppl. Data 3 [63].

3.6. Environmental risk assessment

Potential ecological risk is given in the relation:

$$E_i ir = Tr^{i} \times CF^{i}$$

where Tr^{i} = toxic response factor of a metal Pb = 5, Zn = 1, Cd = 30, Cr = 2, Ni = 5, and As = 10.

where CF^{i} is the contamination factor [63].

The potential comprehensive ecological risk index was calculated using the expression:

$$RI = E_i ir_1 + E_i ir_2 \text{---}\ldots\ldots\ldots\ldots E_i ir_{ṇ} \quad (5)$$

$E_i ir$ is the ecological risk factor, and ṇ is the number of elements studied.

$E_i ir$ and RI classifications are presented in Suppl. Data 4

Toxic Risk Index (TRI) is expressed as follows:

$$TRIi = \frac{\sqrt{(C_i/TEL_i)^2 + (C_i/PEL_i)^2}}{2}$$

$$TRI = \sum_{I=1}^{n} TRI_i \quad (6)$$

where C_i represents the measured concentration of heavy metal I and ṇ the number of target heavy metals, TEL_i and PEL_i is the TEL value and PEL value of the target metals respectively [25].

The pollution values of TRI are classified as presented in supplemental data 5.

Ecological contamination index (ECI) is given by the expression:

$$ECI = \sum_{i=1}^{n} mHQ \quad (7)$$

where Bn is the reciprocal of derived Eigen value of heavy element concentration only. The proposed ranking risks posed by heavy metals to the ecological systems is computed based on the proposed formula as presented in:

$$mHQ = \sqrt{C_i / TEL_i} + \sqrt{C_i / PEL_i} + \sqrt{C_i / SEL_i}$$

where C_i represents the measured concentration of heavy metal I, TEL_i is the TEL value of the target heavy metal, PEL_i is the PEL value of the target heavy metal, and SEL_i is the SEL value of the target heavy metal i [24]. ECI classification is presented in Suppl. Data 5.

The Mean Effect Range Medium Quotient (mERMQ) was calculated using the relation:

$$ERMQ = \frac{Ci}{ERMi}$$

$$mERMQ = \frac{\sum_{i=1}^{n} ERMQ}{n} \quad (8)$$

where mERMQ is the mean effect range medium quotient of multiple metal contamination. ERMQ is the effect range medium quotient of heavy metal i, C_i is the measured content of the target heavy metal C_i, ERM is the ERM value of the target heavy metal i, and n are the metal and number of metals respectively [64, 65]. The mERMQ classification is as presented in supplemental data 6.

Mean Probable Effect level quotient (mPELQ) as presented in expression given as:

$$mPELQ = \frac{\sum_{i=1}^{n} \left(\frac{Ci}{PELi}\right)}{n} \quad (9)$$

where Ci is the concentration of metal i PELi is the probable effect level value for metal i and n is the sum of metals considered [66, 67]. The classification of mPELQ is given in Suppl. Data 6.

Modified risk assessment code (mRAC) was calculated was calculated as follows:

$$mRAC = \frac{\sum_{i=1}^{n} TriRACi}{\sum_{i=1}^{n} Tri} \quad (10)$$

where Tri stands for toxic response factor for a single metal i.

RAC is the risk assessment code of the i^{th} metal derived from summation percentage concentration of metal, n is the total number of heavy metals [68]. The mRAC was classified as given in Suppl. Data 7 [69].

Contamination Severity Index (CSI) was calculated using the equation:

$$CSI = \sum_{i=1}^{n} w_i[\sqrt{(C_i / ERL_i) + (C_i / ERM_i)^2}] \quad (11)$$

Where W_i is the weight of the heavy metal i, C_i is the measured content of the target heavy metal i, ERL_i is the ERL value of the target heavy metal i, ERM_i is the value and n is the number of selected metals [24].

The ratio $\frac{PCA}{FA}$ is used to obtain the weight (W_i) of each heavy metal. This method only considered the factors with human influence to calculate the weight. The weight (W_i) of each trace element is calculated as follows:

$$wi = \frac{loading\ value\ i\ _{x}\ eigen\ value}{\sum_i^n loading\ value\ _{x}\ eigen\ value}$$

CSI classification is presented in Suppl. Data 7.

4. Results and Discussion

4.1. Concentrations of trace elements in stream sediments

The statistical summary showing the concentrations of the physical parameters and PHEs in stream sediment samples are presented in Table 1. The mean values of PHEs were below SQGs limits with the exception of Cd exceeding TEL limit. PHEs concentrations and their spatial distributions are given in Figures 2a and 2b. The mean concentrations of elements decreased in the order Fe > Zn > Pb > As > Ni > Cd > Cr. Spatial variation of PHEs is presented in Figure 2.

Table 1. Summary of descriptive statistics of physical parameters and trace elements in stream sediments.

Variable	Minimum	Maximum	Mean	Standard Dev.	Skewness	Kurtosis	Average shale	TEL	PEL	SEL	ERM
pH	5.3	6.9	6.4	0.5	-1.0	0.4	NA	NA	NA	NA	NA
eH	189	390	304.3	60	-0.3	-0.1	NA	NA	NA	NA	NA
Ni	0.0	21.4	5.2	8.2	1.6	1.2	68	15.9	42.8	75	50
Cd	0.0	6.8	1.7	2.3	1.5	1.1	0.3	0.7	4.2	10	9
As	0.0	17.6	6.1	6.8	0.8	-1.0	13	7.2	41.6	33	85
Pb	0.0	95.4	19.6	36.8	1.7	1.5	20	30.2	112.2	250	110
Zn	0.0	191.1	36.4	73.2	1.9	2.1	95	124	271	820	270
Cr	0.0	0.9	0.4	0.4	0.8	-1.2	90	52.3	160.4	110	145
Fe	20	1980	376.3	552.6	2.9	8.9	47200	NA	NA	NA	NA

NA- not applicable TEL-threshold effect level [70]; SEL-severe effect level [71]; PEL-probable effect level [71]; ERM- effect range median [72].

The pH is weakly acidic and the redox potential (eH) is of medium range. The pH and eH control the mobility and dispersion of elements in the environment. High eH values reflect oxidising environment and low pH reflects reducing conditions. The pH controls the bioavailability and speciation of heavy metals in water dwelling plants and animal species. At a pH value less than 4, the toxicity of trace elements increases. The values of eH obtained in this study are a function of the acidity of the environment of deposition of stream sediments. The elements Cd, As, Pb, and Zn obtained higher concentrations than those for Ni, Cr, and Fe. This is traceable to their relative mobility in the secondary geochemical environment, caused by mining and farming activities in the study area. Higher concentrations of Zn, As, Cd, and Pb indicated that they are from anthropogenic sources like domestic sewage and agricultural runoffs. The mean values of Cd in different sample locations exceeded background average shale value implying anthropogenic inputs. The spatial distribution (Figure 2) of As, Cd, Cr, and Zn show their relative mobility in the environment more than other elements. Samples 5, 6, 7, and 10 has more spatial variation of trace elements concentrations than other locations. This is attributable to their proximity to the mine sites and farmlands (Figure 1). The elements As, Cd, Cr, and Zn are chalcophilic elements common in sediment samples around sphalerite and galena mining areas. The mean value of Cd is above the background average value of shale (0.3 mg/kg). Cd is a carcinogen, and it is toxic even at minimal concentrations. In stream sediments, the source of Cd is mostly from geological weathering, agricultural runoffs and wastes disposal sites. Excessive Cd intake has harmful effects to animal species. Cd is transferable into the food chain through vegetation. In this study, the TEL for Cd exceeded the range at which adverse effects are likely to occur to water dwelling organisms.

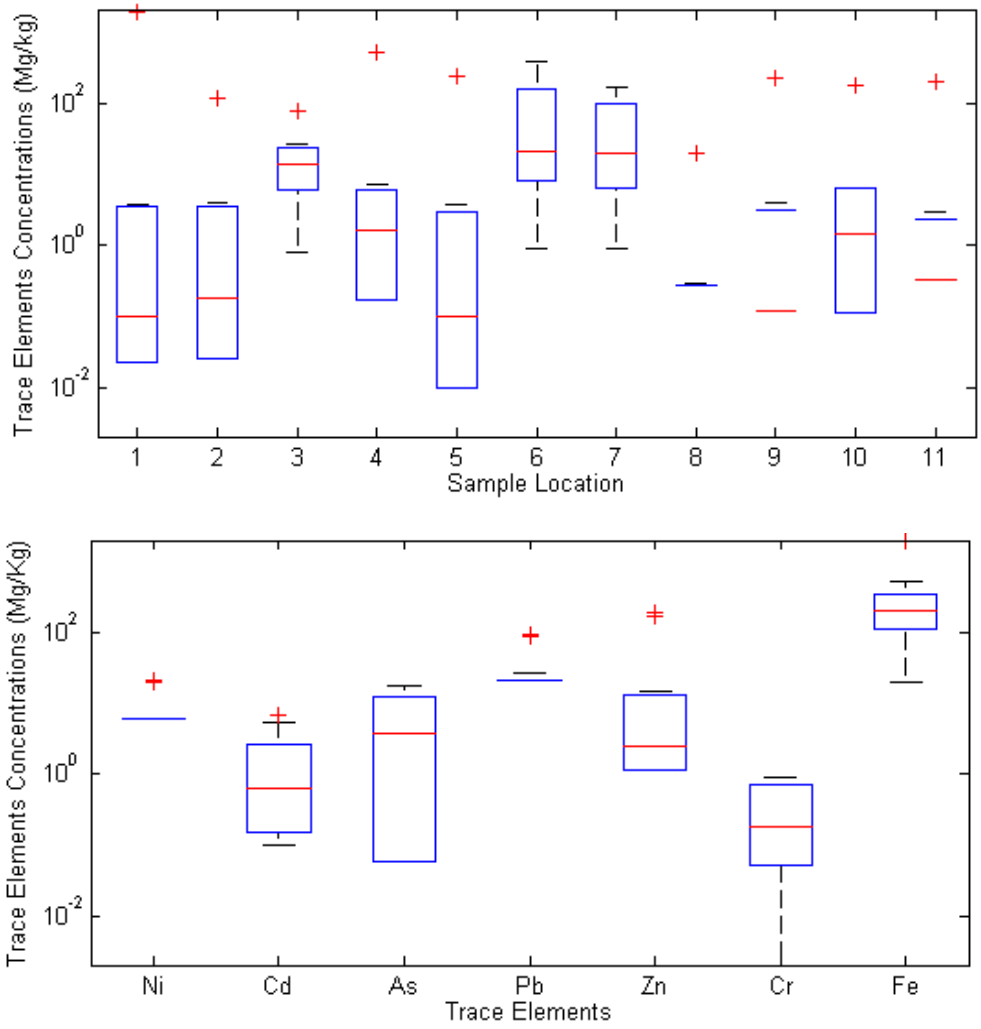

Figure 2. Trace elements concentrations (a) and spatial distribution in sample locations (b).

4.2. Pollution indices

Anthropogenic metal input

Anthropogenic metal input displayed high concentrations of As, Cd, Pb, and Zn mostly in locations 3, 6, and 7. The elements Ni and Zn obtained high anthropogenic inputs in locations 6 and 7. Cr and Fe possess the least anthropogenic inputs in the study area (Figure 3).

High anthropogenic metal input in some locations may be because of mining activities for Pb-Zn ore. Fertilizer applications during farming activities may be the source of anthropogenic inputs in these locations. High anthropogenic metal input in control site, which is location 11, shows that Cd may not be from mining and farming activities alone. Anthropogenic metal input demonstrated high concentrations of Cd in locations 3, 6, and 7. Most of the indices also reflected high ecological risks in these locations. This can be due to Cd occurring as an associative metal at Pb-Zn mine sites. The source of Cd can also be from automobile emissions.

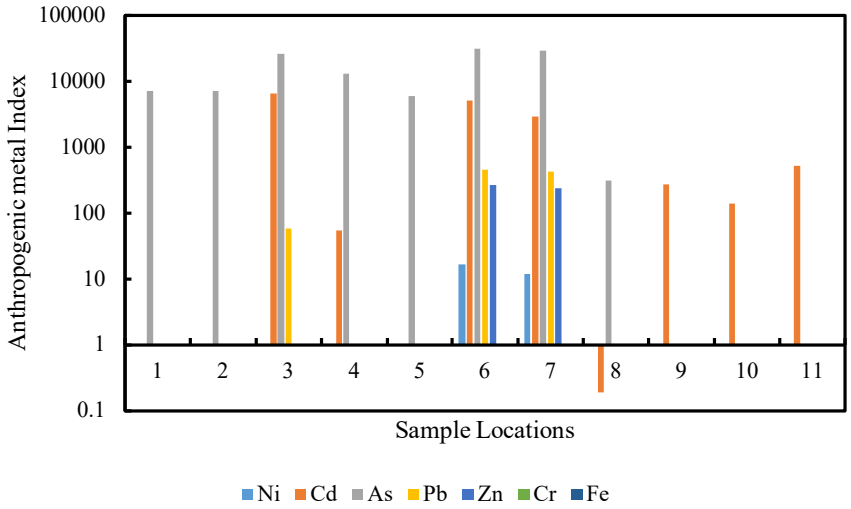

Figure 3. Distribution of anthropogenic metal index in the studied area.

Single pollution index (SPI) and Numerov composite pollution Index (NCPI)

Single pollution index (SPI) displayed high values in locations 3, 4, 6, and 7 (Figure 4). These are locations of Pb-Zn mining activities. Numerov composite pollution index (NCPI) showed moderate pollution in locations 3, 6, and 7 (Figure 5).

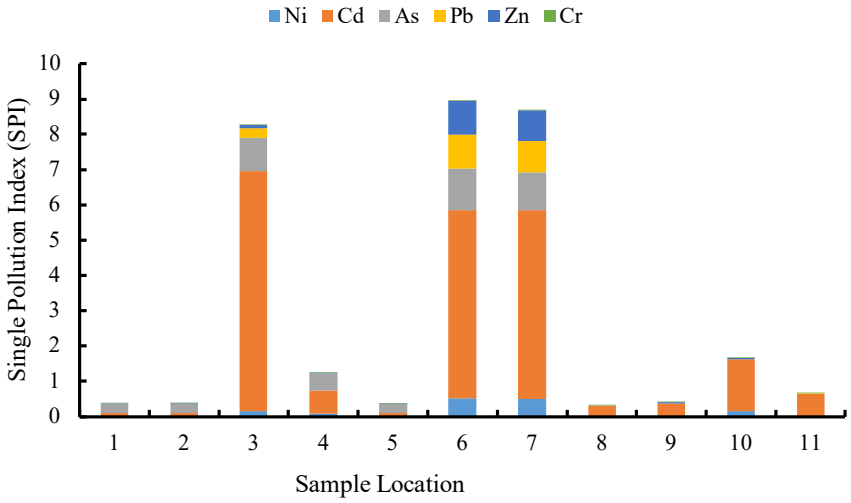

Figure 4. Spatial distribution of SPI values in the studied locations.

The SPI of all the PHEs excerpt Cd fall in the range of SPI $\leq$ 0.1, reflecting clean and safe sands. However, Cd in locations 3, 6, and 7 fall in the range of 3.0 < SPI < 5.0, depicting moderate pollution of the sediments with PHEs. Location 3 obtained Cd SPI > 5, indicating severe pollution of sediments. These locations (3, 6, and 7) are locations of intense mining operations. The NCPI also represent clear sands (NCPI $\leq$ 0.7). Similarly, locations 3, 6 and 7 obtained NCPI in the range of 2 < NCPI $\leq$ 2.0. There is moderate pollution in location 3 (1 < NCPI $\leq$ 2.0). This location shows slight pollution exceeding the background level (Figures 4 and 5).

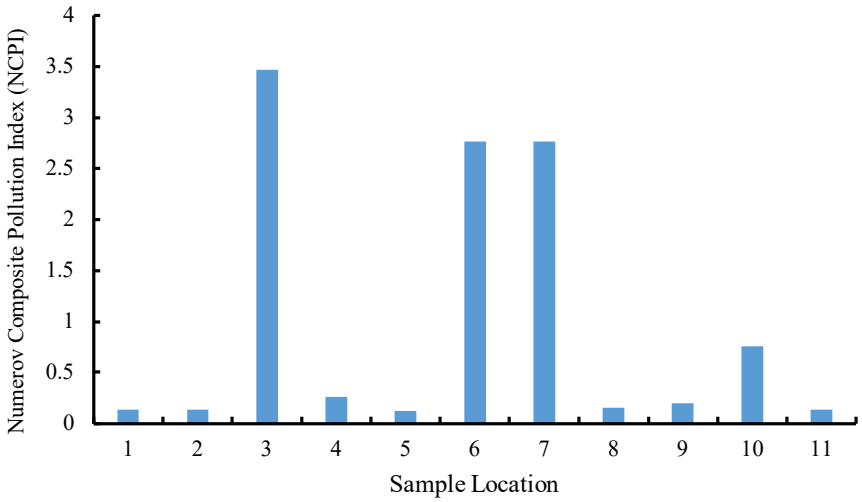

Figure 5. Spatial distribution of NCPI values in sample locations.

Contamination factor (Cf) and modified degree of contamination (mCd)

Levels of Cf are presented in Table 2 show that Cf values fall mostly under low contamination (Cf < 2). However, the Cf of As in location 3, 6 and 7 are of moderate contamination levels. The Cf of Pb in locations 3, 6, and 7 including Cd in locations 4, 5, 8, 9, 10, and 11 are within moderate contamination (1 < Cf < 3). The Cf of Zn in locations 6 and 7 are also of moderate contamination range. Cd Cf in locations 4 and 7 fall under high contamination (Cf > 6). Table 2 shows that mCd is between moderate and high in locations 4, 6, and 7. Other locations fall under nil to low mCd (Suppl. data 3). Locations with high mCd implies more anthropogenic input of heavy metals into stream sediments [48]. The mCd in stream sediments increase in the order: location 4 > location 7 > location 6 > location 10 > location3 > location 5 > location 11 > location 9 > location 8 > location 1 > location 2.

Table 2. Contamination factor (Cf) values of trace elements

	Ni	Cd	As	Pb	Zn	Cr	Fe	C_d	mC_d
1	0.0	0.33	0.30	0.0	0.026	0.001	0.041	0.7	0.933
2	0.0	0.33	0.310	0.0	0.023	0.002	0.003	0.668	0.926
3	0.088	0.33	1.101	1.39	0.163	0.001	0.002	3.075	1.461
4	0.052	22.67	0.556	0.0	0.017	0.0	0.011	23.306	5.956
5	0.0	2.2	0.287	0.0	0.011	0.0	0.005	2.503	1.334
6	0.305	0.33	1.35	4.77	2.011	0.010	0.008	8.784	2.729
7	0.31	10.27	1.248	4.44	1.867	0.009	0.002	18.146	4.810
8	0.0	1.0	0.02	0.0	0.0	0.0	0.0004	1.0204	1.004
9	0.0	1.27	0.0	0.0	0.043	0.001	0.005	1.319	1.071
10	0.088	4.97	0.0	0.0	0.071	0.005	0.004	5.138	1.919
11	0.0	2.13	0.0	0.152	0.0	0.004	0.004	2.29	1.287

Locations 4 and 7 are locations of mining and farming areas and recorded high and considerable contamination (Cf > 6) of Cd. The likely sources of Cd is pesticides and fertilizer application and weathering of geological materials. The highest value of Cf in locations 7 (Figure 2) is due to its proximity to a mine site (Figure 1). Contamination index (C_d) value greater than 1.5 is evidence of anthropogenic pollution. C_d values in only locations 4 and 7 recorded moderate and considerable degrees of contamination while other locations fall under low degree of contamination. This is corroborative of other pollution indices applied in this study. The modified degree of contamination is of moderate range in location 7 (2 < mCd < 4) and high (4 < mCd < 8) in location 4. Other locations fall under nil to low modified degree of contamination (Suppl. data 3). This study area is predominantly nil to low contamination (mCd < 1.5) of PHEs.

4.3. Environmental risk indices

4.3.1. Comprehensive potential ecological risk

The potential ecological risk index (RI) values are higher in locations 3, 6, and 7 (Figure 6) than other locations. Location 3 is under very high ecological risk while locations 6 and 7 are of considerable ecological risks and other locations are of low ecological risks. The RI ranged from low ecological risk index (RI < 150) to very high ecological risk index (RI > 600).

The mCd and RI are indices used to evaluate the comprehensive risk of heavy metal enrichment in an area. RI values in location 6 and 7 fall under considerable ecological risk, while location 3 is of very high ecological risk. There is intense mining in these locations confirming that mining activities can release PHEs into the environment. Location 3 recorded the highest ecological risk of 698.6. The EirF of Cd in these locations is evidence of the adverse effect it has on benthonic fauna ecosystem. Moderate ecological risk in stream sediments, can have unfavourable effect to benthonic fauna species. RI value exceeded 150 in locations 3, 6, and 7, which show unfavourable conditions to benthonic organisms.

4.3.2. Toxic Risk Index (TRI)

TRI values classified as low toxic risk were obtained in locations 3 and 6 (Figure 6). The rest of the locations are of nil to low toxic risks as presented in Suppl. data 5. All the locations obtained TRI values ranging from no toxic risk (TRI $\leq$ 5) to low TRI (5 < TRI $\leq$ 10). TRI values in sample locations trend as 6 > 3 > 7 > 10 > 4 > 11 > 1 > 5 > 2 > 9 > 8. TRI recorded values reflecting low toxic risk. Other locations are of nil to no toxic risk values. Locations 3, 6, and 7 obtained low toxic risks, while other locations were of nil to no toxic risks.

In Suppl. data 5, TRI values in locations 3, 6, and 7 fall under 5 < TRI $\leq$ 10 of low toxic risk. Only Cd display low toxic risk. Other PHEs are classified under no toxic risk. Sample 3 is contiguous to a mine pit. Very high ecological risk shows that pollution is from human activities. Integrated toxic risk of sample locations 3, 6, and 7 fall under low toxic risk. When the toxic unit sum is more than four (4), it suggest moderate toxicity to the ecosystem. TRI is an acceptable method for the accurate evaluation of ecological toxicity. Cd obtained high TRI values in this study reflecting high Cd pollution risk. From this study, only aquatic species in location 6 are at risk of high severity of contamination. Pb is at low risk of ecological hazard to aquatic species in locations 6 and 7. The mRAC displayed medium potential adverse effect to aquatic species in locations 3, 6, and 7. These are mine sites where there is active mining of Pb-Zn ore leading to the release of PHEs into the environment.

4.3.3. Effect Range Median Quotient (ERMQ)

The effect range medium quotient (mERMQ) (Figure 6) show values in the range of mERMQ $\leq$ 0.1 and 0.1 $\leq$ mERMQ $\leq$ 0.5 (Suppl. Data 6). This implies that the study area is of low priority and medium priority risks of 9% and 21%, respectively. All the locations possess mERMQ in the range of mERMQ $\leq$ 0.1. These shows that the sites are of low priority risk level and the probability of being toxic is 9%.

In all locations, values of mPELQ, mERMQ, CSI, and ECI show low degree of contamination, low priority, very low severity of contamination and uncontaminated, respectively. The RI in location 3 fall under high ecological risk reflecting anthropogenic input in this location while other locations indicate geogenic source of pollution. Location 6 is of considerable ecological risk depicting anthropogenic influence.

4.3.4. Contamination Severity Index (CSI)

Contamination severity risk (CSI) was in the range of CSI < 0.5 and 1 < CSI < 1.5 (Suppl. data 7). This implies that the locations were uncontaminated with harmful elements while locations 6 and 7 are of very low severity contamination.

4.3.5. Modified risk assessment code (mRAC)

Modified risk assessment code (mRAC) were less than 1 in locations 1, 2, 3, 5, 8, 9, and 11 indicating no potential adverse effect to aquatic biota (Suppl. data 7). In locations 3, 6, and 7, mRAC values fall between 10%-29% showing evidence of medium potential adverse effect. Locations 4 and 10 recorded values between 1-9 % implying low potential adverse effect to aquatic species.

4.3.5. Ecological contamination index (ECI) and mean probable effect level quotient (mPELQ)

ECI obtained values less than 2 in all the locations depicting uncontaminated sediments (Suppl. data 5).

The mPELQ values were all less than mPELQ $\leq$ 0.1 in all the locations indicating low

contamination. Ecological indices in different sample locations (Figure 6) show higher levels of RI, mRAC, and mCd than other indices. In this study, mRAC, RI, and TRI proved to be more reliable ecological risk assessment indices. Ecological risk evaluation is crucial in managing heavy metals in the environment. Very high ecological risk represents pollution by human activities.

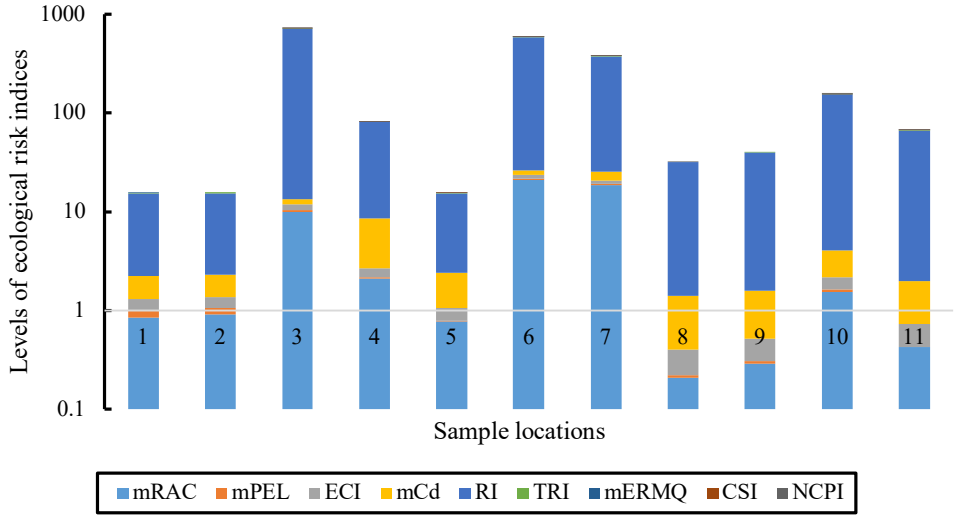

Figure 6. Pollution and ecological indices in sample locations.

4.4. Multivariate statistical analysis

Pearson correlation analysis (CA) of physical and trace elements show the relationship between chemical species (Table 3). Significant correlation at $p \leq 0.5$ confidence limit exist between Cd and Pb. Significant correlation between elements at $P \leq 0.1$ exist between Ni and As, Pb, Zn and Cr. Cd also correlated with As and Cr. As correlated with Pb, Zn, and Cr. Pb correlated with Zn and Cr. Zn correlated with Cr.

Table 3. Pearson correlation matrix of trace elements.

	pH	eH	Ni	Cd	As	Pb	Zn	Cr	Fe
pH	1								
eH	0.034	1							
Ni	0.218	0.072	1						
Cd	0.041	0.310	0.681	1					
As	0.432	0.197	0.845**	0.822**	1				
Pb	0.359	0.166	0.969**	0.698*	0.877**	1			
Zn	0.360	0.082	0.968**	0.578	0.809**	0.985**	1		
Cr	0.011	0.426	0.858**	0.880**	0.806**	0.860**	0.788**	1	
Fe	0.268	-0.335	-0.161	-0.234	-0.058	-0.148	-0.113	-0.259	1

Pearson correlation matrix of ecological risk indices

In Table 4, Pearson correlation show significant correlation between ecological risks and pollution indices as mCd did not correlate with NCPI. The mCd did not correlate with RI, TRI, mERMQ, CSI, but correlated with NCPI. All other ecological indices correlated significantly excluding the above listed indices.

Principal component analysis (PCA) obtained a total percentage cumulative of 76.4% and generated two principal components (PCs). PC 1 has Eigen values of 5.27 and PC 2 has Eigen value of 1.64 (Figure 7). Components with Eigen values more than or equal to 1 were considered in the analysis as source of variation in the data. Variables with components of 0.5 were considered as significant correlation [73]. Rotated component matrix was applied in the analysis of data. Significant variables of PC1 are Ni, Cd, As, Pb, Zn, and Cr, while PC2 has significant loadings of pH, Fe and eH. Principal component plots in rotated space is presented in Figure 7.

Table 4. Pearson correlation of ecological risk indices.

	mRAC	mPELQ	ECI	mCd	RI	TRI	mERMQ	CSI	NCPI
mRAC	1	0.963**	0.980**	0.449	0.812**	0.952**	0.999**	0.998**	0.296
mPELQ		1	0.963**	0.338	0.842**	0.949**	0.958**	0.963**	0.884
ECI			1	0.442	0.900**	0.985**	0.981**	0.989**	0.897
mCd				1	0.208	0.345	0.434	0.462	0.951
RI					1	0.949**	0.820**	0.834**	0.970
TRI						1	0.956**	0.962**	0.962
mERMQ							1	0.998**	0.885
CSI								1	0.901
NCPI									1

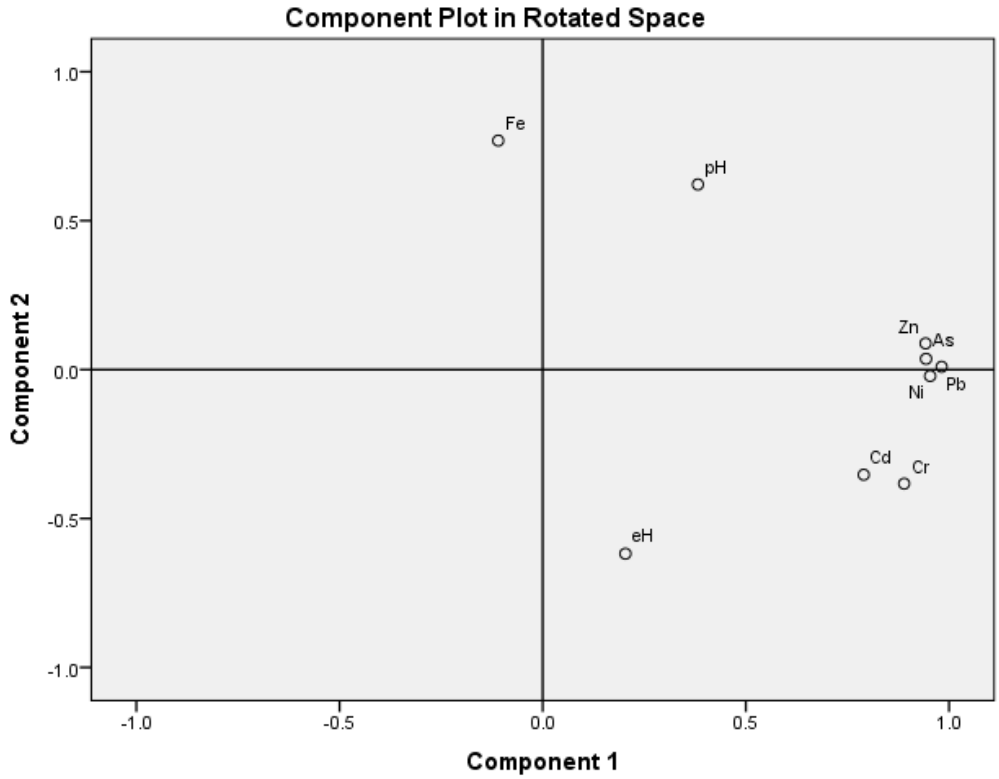

Figure 7. Principal components plot in rotated space.

Pearson correlation shows that mCd and NCPI have significant correlation but mCd has no correlation with ecological risk indices (mRAC, mPELQ, ECI, TRI, mERMQ, and CSI). NCPI and mRAC exhibited weak correlation showing no similar interpretation. NCPI and mCd correlated showing similar interpretation as pollution indices. Strong positive correlation among mRAC, mPELQ, RI, TRI, and CSI show ecological indices with similar interpretation. Biplots (Fig. 8) showing regression expressions indicate no strong correlation between mCd and ECI, which is in agreement with Pearson correlation (Table 4) of pollution and ecological risk indices. Regression expression shows R^2 values having strong correlation between mPELQ *vs* mRAC, TRI *vs* RI and CSI vs mERMQ. Biplots of PC1 *vs* PC2 (Fig. 7) shows PC1 has more contribution to data interpretation than PC2. Low $R^2 = 0.161$ reflects different levels of contribution by the variables PC1 and PC2. Regression analysis is to test the linear dependence between two variables while correlation seeks to test the degree of variability between two variables. Correlation also measures the closeness of fit of the regression lines.

4.5. Regression statistical analysis

Regression model of heavy metals (Suppl. data 8) shows $R^2 = 0.939$, which is significant. The Durbin Watson value is 2.77. The significant factor change is 0.02. The regression sum of squares is

51.4 and the degree of freedom is 6 and a residual of 3.32. The mean square of regression is 8.56 and the mean square of residual is 4. The regression model of ecological and pollution indices shows R^2 = 0.835 and Durbin Watson value of 2.34. The regression sum of squares is 24.5 and the residual is 4.83. The degree of freedom 2.17 is statistically significant at a value of 0.28, which is greater than 0.05 (Suppl. Data 8).

Biplots of PC2 versus PC1 show uneven distribution of scattered points (Figure 8). Biplots of ecological indices (Figure 9) show that R^2 of mPELQ versus mRAC, TRI versus RI, CSI versus mERMQ were higher showing more correlation than mCd versus ECI.

In this study, the line of closeness of fit of the scatter diagram (Figure 8) shows CSI versus mERMQ have more closeness. The regression model of heavy metals (Suppl. data 8) shows Durbin-Watson value of 2.77 exceeded the critical values of 1.5 < d < 2.5. Durbin Watson model summary for ecological risk indices was 2.34 which is within the critical value of 1.5 < d < 2.5. That means there is linear auto correlation in the data for ecological risk indices than heavy metals data. These findings are in agreement with biplots of ecological risk indices in Figure 8.

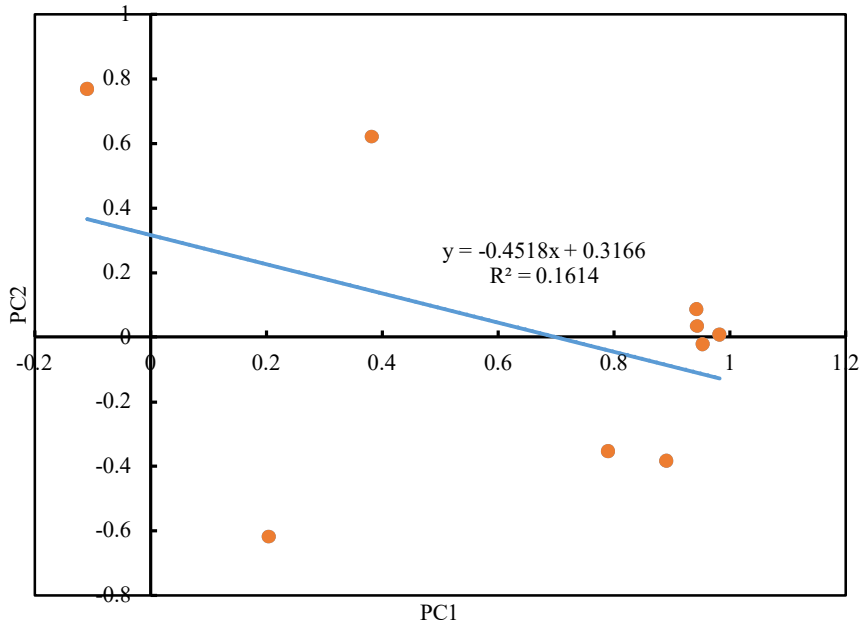

Figure 8. Biplots of principal component analysis.

Recent ecological indices such as ECI, mPELQ, and ECI did not obtained values showing ecological risk of aquatic species in the study area. The use of multiple recent pollution and ecological indices has unfolded the ecological risk status of the studied area. On the other hand, mRAC, RI, and TRI reported few locations of ecological risk in the area. Related studies considered few ecological risk indices compared to this study. For instance if mPELQ, mERMQ, and CSI only were considered in this study, the result will show no evidence of pollution in all the locations. Considering additional indices such as RI, TRI, and mRAC made it obvious that there are some locations of ecological risks to aquatic species. This underscores the need for an integrated risk analysis of this type for a better interpretation. Regression and multivariate analysis supported the usefulness of mRAC, TRI, and RI in understanding the ecological risk status of this study area compared to other indices considered.

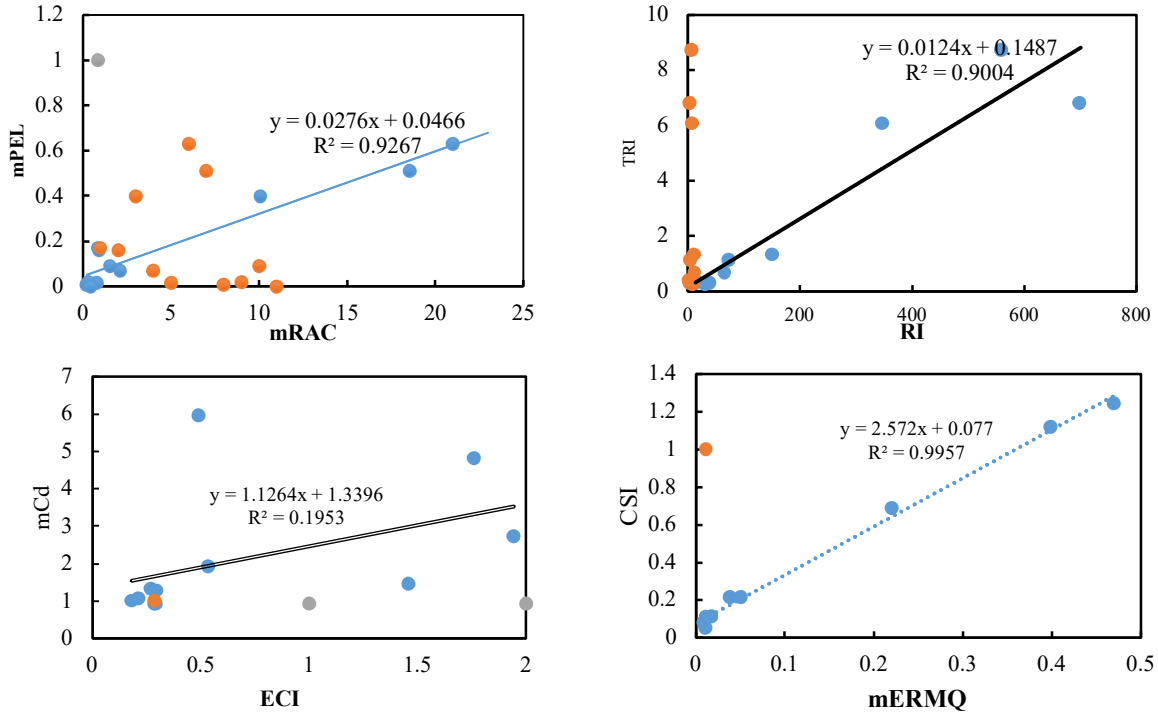

Figure 9. Biplots of ecological risk indices.

5. Conclusions

Comparison of SQGs shows Cd mean value is above TEL guideline indicating its adverse effect to water dwelling organisms. Other trace elements are within acceptable ranges to sediment dwelling organisms. The mRAC, RI, and TRI revealed locations of high to considerable ecological risks more than other ecological risk indices. These findings were further supported by multivariate and regression statistical analysis. The mCd and ECI obtained low correlation compared to other indices. The mCd and NCPI are pollution indices while other indices are ecological risk indices that evaluate the risk of aquatic species in the ecosystem. The sources of the trace elements were mostly geological weathering, industrial wastes, and sewage, disposal and fertilizer applications to farmlands. The elements Cd, As, and Pb are the principal pollutants in the stream sediments in mining and farming locations of the studied area.

Pollution of some sites with Cd, As, and Pb is a cause for concern, because these elements are identified by WHO as priority carcinogens. The elements are also capable of causing food and soil poisoning. A geochemical map showing Cd, As, and Pb distribution is important for easy monitoring of these elements to avert water and soil poisoning, which can be very lethal to the inhabitants. Further intensive research on the mode of water and soil remediation of these pollutants is necessary to forestall the outbreak of any cancer epidemic. Pollution and ecological indices identified possible areas of stream sediment pollution with PHEs by anthropogenic sources in locations of intense mining and agricultural activities. This is an integrated method of stream sediments pollution reproducible by scientists in other climes to solve similar environmental problems.

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List of Abbreviations

pH	Acidity	SQG	Sediment Quality Guidelines
eH	Oxidation potential	PEL	Threshold Probable Effect Level
AAS	Atomic Absorption Spectrophotometer	PEC	Probable Effect Concentration
PHEs	Potentially Harmful Elements	PEL	Probable Effect Level
Ni	Nickel	SEL)	Severe Effect Level
Cd	Cadmium	TET	Toxic Effect Threshold
As	Arsenic	ERL	Effect Range Low
Cr	Chromium	ERMQ	Effect Range Medium Quotient
Pb	Lead	TEC	Threshold Effect Concentrations
Zn	Zinc	SPI	Single Pollution Index
Fe	Iron	NCPI	Numerov Composite Pollution Index
AMI	Anthropogenic Metal Input	US EPA	United States Environmental Protection Agency
TEL	Threshold Effect Limit	mHQ	Modified Hazard Quotient
mRAC	Modified Risk Assessment Code	mPELQ	Mean Probable Effect Level Quotient
TRI	Toxic Response Index	WHO	World Health Organization
RI	Comprehensive Ecological Risk	PC	Principal Component
PCA	Principal Component Analysis		

تجزیه و تحلیل ریسک اکولوژیکی داده‌های رسوبات جریان و تأثیر آن بر موجودات آبزی در اطراف مناطق معدنی و کشاورزی

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چکیده:

عناصر بالقوه مضر از طریق فعالیت‌های معدنی و کشاورزی وارد محیط زیست می‌شوند و باعث آلودگی آب و رسوب نهرها می‌شوند. تجزیه و تحلیل خطر اکولوژیکی به تعیین آلودگی رسوبی، توصیه اقدامات اصلاحی برای ایمنی سلامت انسان و بقای گونه‌های آبزی کمک می‌کند. رسوبات از نظر اسیدیته و پتانسیل ردوکس به ترتیب با استفاده از pH متر و اسپکتروفتومتر مورد تجزیه و تحلیل قرار گرفتند. نیکل، کادمیوم، آرسنیک، کروم، سرب، روی و آهن با استفاده از اسپکتروفتومتر جذب اتمی اندازه گیری شد. مقدار میانگین کادمیوم از دستورالعمل حد اثر آستانه فراتر رفت که نشان دهنده تأثیر نامطلوب آن بر ارگانیسم های ساکن آب است. ورودی فلزات انسانی آلودگی کادمیوم، سرب، آرسنیک، روی و کروم را در مکان‌های ۳، ۶، و ۷ شناسایی کرد. کد ارزیابی ریسک اصلاح‌شده، شاخص پاسخ سمی و ارزش‌های خطر زیست محیطی جامع خطرات زیست محیطی قابل توجهی تا بالا را در مکان‌های ۳، ۶ و ۷ نشان دادند. بالاترین مقدار ریسک جامع اکولوژیکی با ۶۵۳.۲ در مکان ۳ ثبت شد که نشان دهنده خطر بالای اکولوژیکی برای موجودات ساکن آب است. ارزش خطر زیست محیطی دوربین واتسون (۲.۳۴) بین یک مقدار بحرانی $1.5 < d < 2.5$ است که همبستگی خودکار داده‌ها را نشان می‌دهد. عناصر بالقوه مضر ارزش دوربین واتسون ۲.۷۷ را به دست آوردند که از محدوده نشان دهنده عدم همبستگی خودکار فراتر رفت. همبستگی قوی آرسنیک، سرب و روی نشان دهنده میل ترکیبی و منبع غنی سازی مشترک آنها بود. تجزیه و تحلیل مؤلفه‌های اصلی نشان داد که منابع عناصر عمدتاً هوازدگی زمین شناسی، دفع فاضلاب، پسماندهای صنعتی و کودهای کشاورزی بوده است. این مطالعه شاخص‌های خطر زیست محیطی اخیر را با آمارهای چند متغیره و رگرسیون ادغام کرد. این در تفسیر مشکلات زیست محیطی مرتبط توسط دانشمندان در سایر نقاط جهان مفید است.

کلمات کلیدی: آلودگی رسوبی، شاخص‌های اکولوژیکی، گونه‌های آبزی، فاکتور پاسخ سمی، اسپکتروفتومتر جذب اتمی.