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The prediction of heavy metal pollution in gemstone mines using contamination variables

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ABSTRACT

Although contamination variables have been widely employed in evaluating heavy metal pollution, the potential for their use to predict heavy metal pollution levels in the mining sector using regression analysis has been overlooked. Therefore, this study aimed to use contamination variables to monitor metal pollution level and its health hazards by adopting multiple regression analysis at the Ikinyinwa gemstone mine site. A systematic grid method of sampling was adopted to obtain soil samples from mining areas (MA) and non-mining areas (NMA). 15 soil samples were collected from MA and five samples from NMA at the depths of 0–20 cm. The samples were prepared and aggregated into rice bags, well labelled and sealed at the Central Laboratory, FUTA, before being sent to the LA-ICP-MS Laboratory at the Colorado School of Mines, Golden Colorado, USA for metal analysis. The results showed that there were higher concentrations of As (7.51±0.59) and Cd (0.65±0.26) in MA compared to NMA and the world average standard. On the contamination factor (Cf), the average value of Pb (0.44) was categorised as low (Cf < 1) contamination, while Ni (3.06) was classified as considerable (3 ≤ Cf < 6) contamination, and the others were categorized as moderate (1 < CF < 3) contamination. Based on the degree of contamination (Cdeg), Ni (46.05), As (36.02), and Cd (34.13) were classified as very high (32 ≥ Cdeg) contamination, while Pb (0.681) was classified as low (Cdeg < 8) contamination. The pollution load indices (PLI) for samples SS4, SS5, SS7, SS11, and SS13 were less than one (PLI<1) and classified as unpolluted. The regression analysis established that Cf and PLI variables are positive and significant predictors of heavy metal pollution at the investigated site. It was predicted that for every unit increase in Cf and PLI, the pollution level of heavy metals would increase by 0.138 and 0.578, respectively.

Keywords: *Gemstone mining, Contamination factor, Heavy metal, Pollution load index.*

1. Introduction

Gemstone mining has become one of the most important factors contributing to local and national economic development in many countries [1]. The developments in the use of explosives in gem mining have led to hundreds of low-tech processes where miners create hazardous environments for humans. In order to increase profits, miners engage in unskilled and unregulated practices that can pollute the soil and the environment [2]. Soil is known to be a natural body that contains three states of matter (solid, liquid, and gas) covering the earth's surface [3]. The influence of gemstone exploitation on the soil and its environment is highly dependent on the type of methods used and the geological setting of the minerals [1,4]. Gemstone mining is a source of income for people, especially in emerging nations, where economic hardship is prevalent and income from agricultural production is low [5,6]. As a result, individuals, politicians, and businessmen engaged in gem mining using unconventional methods that could lead to heavy metal pollution that results in health hazards [7-9]. Pollution during the exploration and development stages of gem mining leads to vegetation destruction, exposes the soil surface to varying climatic conditions and reduces surface soil fertility [1,10]. Toxic substances released during drilling and blasting contaminate both soil and water, adversely affecting host communities and their surroundings [11]. Equipment used in drilling emits measurable amounts of smoke and pollutant gases into the atmosphere [12]. Large amounts of waste generated are typically released into the surrounding soil and water [5,13]. One of the contaminants associated with generated waste that is of public health concern is heavy metals [6, 14].

Heavy metals are an essential group of pollutants that can cause serious damage to the environment and human health [15,16]. The main sources of heavy metals in soils in the study area are agricultural and mining activities [17,18] as well as human exposure to heavy metals through direct ingestion of plants, dermal contact, inhalation and ingestion [5,19]. Though, small quantities of heavy metals (zinc, copper, iron, manganese, and nickel) are essential for soil nutrients, while the non-essential heavy metals (mercury, Arsenic, cadmium, and lead) are toxic even in very small amounts [17,24]. The damage of high levels of heavy metals on people, animals, and plants includes reduced growth and development, impaired hematopoiesis and circulation, nervous system damage, kidney and reproductive system abnormalities, and, in severe circumstances, can lead to death [8,25,26].

One of the best approaches to assess the distribution of heavy metal accumulation in the environment is the use of contamination variables to evaluate soil quality and predict the future sustainability of

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ecosystems that may have adverse effects on human health [27]. Studies have established unprecedented concentrations of heavy metals and their associated health hazards at mining sites. Alaba and Opafunso [28] reported over 500 child deaths from elevated lead concentrations in artisanal gold mines in Zamfara state. Adebayo et al. [29] found significantly higher concentrations of heavy metals in soils from active and abandoned mines in Itagunmodi, Osun State, while Wang et al. [30] identified high concentrations of heavy metals accumulation in contaminated soils from a typical mining city, Central China. Onisoya et al. [5] established a significant carcinogenic and non-carcinogenic risk for children and adults when they carried out a risk assessment of heavy metals in soil based in Anka, Nigeria. Onifade et al. [9] found concentrations of cadmium and iron in the Komu Mining Sites in Oyo state to be higher than the WHO permissible limits. The results of Rattikansukha et al. [31] show that the consumption of cadmium and mercury in the seafood from the Nai Thung coast poses moderate risk to human health.

Despite several studies on contamination variables to assess heavy metal pollution, only a little is known about its use to predict metal contamination levels in gemstone mining environments using multiple regression analysis. Therefore, this study facilitated its use to monitor metals pollution and their health hazards at the Ikinyinwa community.

2. Materials and methods

2.1. Description of the Study Area

Ikinyinwa is located close to the Obokun town in Obokun LGA of Osun State [32,33]. It is located at latitude 07°47" N, longitude 04°45" E and is 513 meters above sea level, as shown in Figure 1. The community is blessed with a large landmass that is good for agriculture. The area is sited in a tropical climate with a seasonal rainy season from April to early October and a dry season from November to March. The yearly temperatures vary from 28°C to 35°C, and the estimated yearly precipitation varies from 1,125 mm in the savannah to 1,75 mm in the rainforest zone. The region lies within the crystalline bedrock of Nigeria and is characterized by undulating topography [34-36]. The area has huge deposits of precious metals (gold) and precious stones (tourmaline), and the distance between the mining site and the town of Ikinyinwa is 1.5 to 2.0 km. Gemstone exploitation is a source of livelihood for the people of the Ikinyinwa community due to prevailing economic difficulties and low income from agricultural production. A visit to the site revealed that individuals, politicians, and businessmen were mining gold and tourmaline (gemstone) using unconventional local mining methods. The study area has more than 30 active and abandoned mines associated with various environmental problems arising from non-compliance with mining laws and regulations.

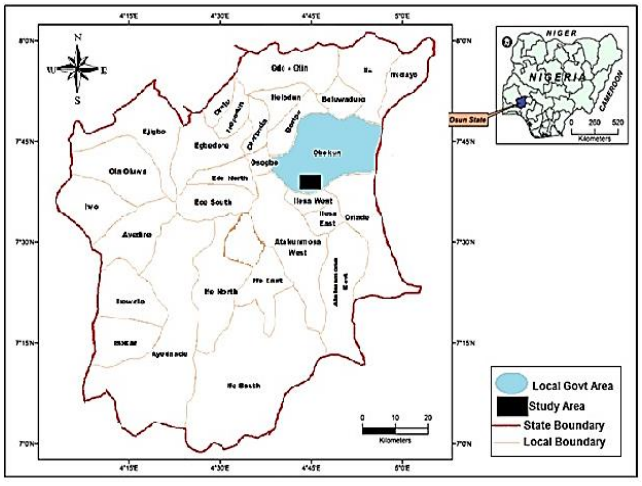

Figure 1. Modified Location of the Study Area (Source: Fashae et al. [37]).

2.2. Soil Sampling and Preparation

A systematic grid sampling method was adopted to collect soil samples from mining areas (MA) and non-mining areas (NMA). The MA and NMA were divided into different homogenous units using gridlines of 10 m intervals based on visual observation and samples were collected at the intersection of the gridlines between the depths of 0-20 cm with a shovel. The non-mining area where control samples were collected was approximately 2 km away from the mining site and had no record of mining activity. Soil samples were taken from 15 MA labeled SS_1-SS_{15}, and five soil samples were taken for control in NMA, labeled SSC_1-SSC_5, as shown in Figure 2. Five sub-samples were taken at each sampling site and mixed thoroughly to form a composite sample which was stored in a zip lock plastic bag and immediately taken to the Central Laboratory, Federal University of Technology Akure (CLFUTA) for preparation. Approximately 1.0 kg of the sample from each site was removed using the quarter method and air-dried at a temperature of 35±5°C. Samples were ground in an agate mortar and sieved through a 2 mm stainless steel sieve using standard methods. The samples were aggregated into rice bags, well labelled, sealed, and sent to the LA-ICP-MS Laboratory, Colorado School of Mines, Golden Colorado, USA for metal analysis.

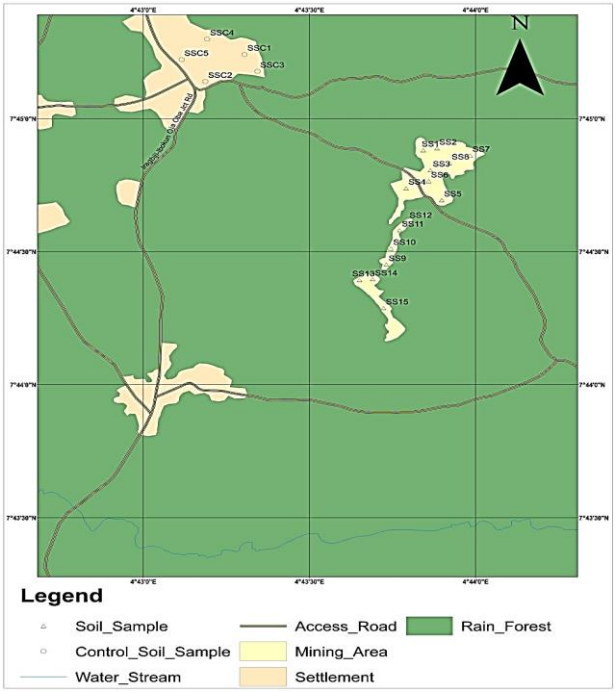

Figure 2. The Location of Soil Samples Collection.

2.3. Acid Digestion of Soil Sample

Digestion of soil samples was performed using a conventional acid digestion method recommended by the Environmental Protection Agency (EPA 3050B) [38,39]. One gram of soil sample was weighed into a 250 mL flask, mixed with approximately 10 mL of HNO_3 (1:1) and then covered with a watch glass in a digestion flask. The sample was refluxed at 95±5°C for 10-15 minutes without boiling and 5 mL of concentrated HNO_3 was added every 30 minutes after cooling until brown fumes escaped from the sample. Next, the sample was heated with a watch glass to the boiling point of 95 ± 5 °C to reduce the volume of the solution to approximately 5 mL. After cooling slowly and efficiently, 10 mL of 30% H_2O_2 was added and heated. The samples were then heated with 10 mL of 37% HCl at 95° C for 15 minutes. The resulting extract was filtered through a 0.45-µm membrane paper, diluted to 100 mL with deionized water, and stored at 4°C for analysis.

2.4. Heavy Metal Analysis and Quality Control

The concentrations of Fe, Zn, Cr, Cu, Pb, Ni, and As in the final digested solutions were measured using inductively coupled plasma optical emission spectrometry (Agilent 8900 ICP-MS QQQ). The quality assurance and quality control (QA/QC) were conducted following the Association of Analytical Community (AOAC) Peer-Verified Methods Program [40]. Caution was exercised to prevent any metal contamination throughout the entire investigation, from sampling to analysis. All apparatus and receptacles were immersed in a 10% solution of HNO_3 for 24 hours and thoroughly washed with deionized water before use. Soil samples were digested using AnalaR grade reagents that adhere to the standards set by the Analytical Reagents Committee of the American Chemical Society. The inductively coupled plasma optical emission spectrometry was auto calibrated when the coefficient of determination (R^2) was equal to or greater than 0.99. The limits of detection and quantification for Fe, Zn, Cr, Cu, Pb, Ni, and As were determined based on the standardized procedures of Baraud et al. [41]. Prior to analyzing each heavy metal, blank solutions were tested to ensure that no contamination occurred during the digestion process with the acids. Each sample was analyzed twice, and the relative standard deviations of the heavy metals were less than 5%.

2.5. Evaluation of Contamination Indices

The level of heavy metal contamination in the soil of the study area was evaluated using the following contamination indices: contamination factor (Cf), degree of contamination (C_{deg}) and pollution load index (PLI). These indices were computed using Equations (1-3) and classified as outlined in Table 1, in accordance with Omer and Mustafa [3].

(i) Contamination factor (Cf)

The contamination factor is a one-digit index used to describe the degree of soil contamination with toxic metals. It was estimated using Equation (1).

$$Cf = \frac{C_{soil}}{C_{ctr}} \quad (1)$$

Where Cf is contamination factor, C_{soil} is the value of metal accumulation in soil samples from MA, and C_{ctr} is the value of metal accumulation in soil samples from NMAor control.

(ii) Degree of contamination (C_{deg})

The degree of contamination is the sum of the contamination factors (Cf) determined for all heavy metals in the sample and was estimated using Equation (2).

$$C_{deg} = \sum Cf \quad (2)$$

Where C_{deg} is the degree of contamination, and $\sum Cf$ is the summation of contamination factors for all the investigated heavy metals.

(iii) Pollution load index (PLI)

The pollution load index is the most commonly used method to estimate the pollution of several heavy metals in an area and was estimated using Equation (3).

The pollution load index is the most commonly used method for estimating multi-heavy metals pollution present in an area and it was estimated using Equation (3).

$$PLI = \sqrt[n]{Cf_1 \times Cf_2 \times Cf_3 \dots \dots \dots \dots . Cf_n} \quad (3)$$

Where PLI is the pollution load index; $Cf_1 \times Cf_2 \times Cf_3 \dots \dots \dots \dots . Cf_n$ is the multiplication of each contamination factor, and n is the number of investigated heavy metals. The evaluation of the physicochemical characteristics and pollution indices of the soil was carried out through the utilization of SPSS version 6, which involved the utilization of descriptive statistical methods, such as charts, percentiles, mean, standard deviation, minimum, and maximum.

3. Results and discussion

3.1. Heavy Metal Concentration

Table 2 presents the descriptive results of heavy metal concentrations in MA and NMA. The results revealed that the mean value of Fe, Zn, Cr, Cu, Pb, and Ni in MA was higher than in NMA but below global average standards. Comparable to prior research conducted at other mining sites in Osun State, the results obtained in this study were similar to those of Abiya et al. [8] and Adebayo et al. [29] but contradicted those obtained from mining sites in Ghana and Zambia [42,43,44]. Additionally, the study revealed that the soil samples from MA exhibited elevated levels of Cd and As in comparison to those from NMA. The comparison of the mean value of Cd (0.65 mg/kg) was found to be higher than those in the soil of both Denmark and Finland (0.3 mg/kg), Canada (0.5 mg/kg) [45], and the world average standard (0.41 mg/kg) [27]. According to Alshahri and Taher [25], while low levels of Cd intake are acceptable, its accumulation in the human body can be extremely toxic. Therefore, the buildup of Cd intake in the study area can cause various health problems, including damage to the respiratory, renal, skeletal, and

Table 1. Classification of Cf, Cdeg, and PLI.

Contamination Factor (Cf)	Degree of Contamination (Cdeg)	Contamination Classification	Pollution Load Index (PLI)	Pollution Classification
Cf < 1	Cdeg ≤ 8	Low contamination	PLI < 1	No Metal Pollution
1 < Cf < 3	8 ≤ Cdeg ≤ 16	Moderate Contamination	PLI = 1	Baseline Pollution
3 < Cf < 6	16 ≤ Cdeg ≤ 32	Considerable contamination	PLI > 1	Metal Pollution
6 > Cf	32 ≥ Cedg	Very high contamination	-	-

(Source: Omer and Mustafa [3]).

Table 2. Descriptive statistics of heavy metals concentrations (ppm) in the study areas.

Parameter	Fe	Zn	Cr	Cu	Pb	Ni	As	Cd
Mean (mg/kg)	20.11	9.13	8.39	10.09	0.69	2.93	7.51	0.65
Std. Dev.	0.31	0.08	1.01	0.10	0.37	0.01	0.59	0.26
Minimum	10.08	3.66	3.36	3.29	0.12	0.38	3.53	0.14
Maximum	33.11	14.01	12.36	15.81	1.23	6.90	13.77	1.35
Q_1	12.31	5.78	6.17	4.97	0.31	0.76	4.21	0.43
Q_3	25.53	12.23	11.51	13.91	1.04	4.59	11.43	0.91
CTR (NMA)	10.91	6.58	5.61	5.88	0.53	1.22	2.34	0.25
WSA[a] (mg/kg)	-	70.00	59.50	38.90	27.00	29.00	0.67	0.41

cardiovascular systems, as well as the development of lung, kidney, prostate, and stomach cancers [26,46,47]. At the same time, the mean arsenic (13.8 mg/kg) concentration in this study was in line with the results obtained in Thailand (13.60 mg/kg) but was lower than those reported in soils from Italy (41.0 mg/kg), Portugal (17.0 mg/kg), and the UK (16.0 mg/kg), and higher than those observed in soils from the USA (7.0 mg/kg) and Japan (11.0 mg/kg) [48]. The high concentration of As in the study area may result in damage to the immune system [49,50], skin lesions during arsenicosis [51], cardiovascular disorders [52,53], diabetes mellitus [54,55], neurological disorders [56,57], and pulmonary diseases [58, 59]. The significant disparity between the Q1 and Q3 interquartile ranges for heavy metal concentration at all sampling sites indicates that several factors, such as the period of mining, the volume of work done, the equipment used, and the method of waste disposal at each sample site influence their concentrations and associated health risks.

3.2. Heavy metal contamination variables

3.2.1. Contamination factor (Cf)

The average value of the contamination factor in MA was highest for Ni (3.06 mg/kg) and lowest for Pb (0.44 mg/kg), as shown in Figure 3. The concentrations of Fe ranged from 0.63-1.99 mg/kg, Zn from 0.65-3.70 mg/kg, Cr from 0.36-2.35 mg/kg, Cu from 0.57-2.09 mg/kg, Pb from 0.14-0.85 mg/kg, Ni from 0.38-6.96 mg/kg, As from 0.52-4.71 mg/kg, and Cd from 0.29-6.05 mg/kg, respectively. In comparison with Table 2, the contamination factor (Cf) average value of Pb (0.44) was categorized as low (Cf < 1) contamination, while Ni (3.06) was classified as considerable (3 ≤ Cf < 6) contamination, and others were moderate (1 < CF < 3) contamination. Meanwhile, the minimum values of the metals were categorized as low contamination. Similarly, only the maximum value of Pb was classified as low contamination, while those of Fe, Cr, and Cu were classified as moderate contamination; As and Zn as considerable contamination; and Ni and Cd as very high contamination. The low contamination range value of Pb concentration indicates that it does not pose a health risk to the study environment, unlike other metals whose contamination ranges from moderate to very high contamination. Comparing with the results obtained in previous studies, similar results were reported in the surface soils of Kumasi, Ghana [60] and in an automobile repair workshop in Ikare-Akoko, Ondo State, Nigeria.

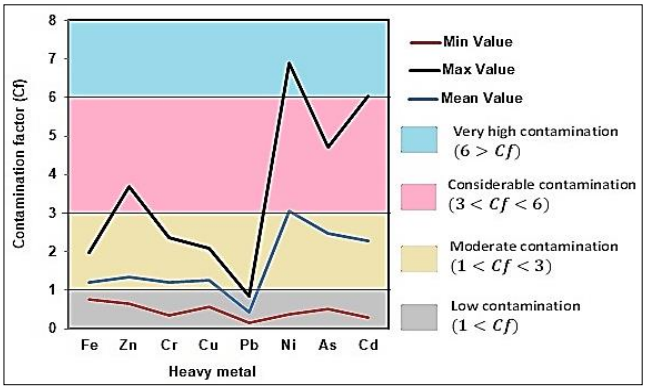

Figure 3: Classification of the Contamination Factor of Heavy Metals in Soil Samples.

3.2.2. Degree of Contamination ($Cdeg$)

Figure 4 presents the values of Cdeg for the investigated heavy metal in the study area. The results revealed that Pb (5.83) was classified as low contamination (Cdeg≤8). Meanwhile, Fe (17.83), Zn (20.22), Cr (17.76), and Cu (19.8) were classified as having considerable contamination (16≤Cdeg≤32). Also, Ni (46.08), As (36.83), and Cd (35.12) were classified as having very high contamination (32≥Cdeg). The results indicated that the study area may be contaminated with heavy metals except for Pb, as the contamination levels were low. The very high contamination values of Ni, As, and Cd indicate serious health risks in the study environment.

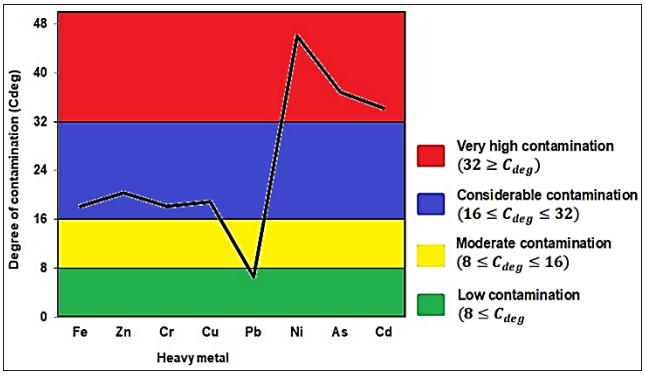

Figure 4. Classification of the Degree of Contamination of Heavy Metals in the Soil Samples.

3.2.3. Pollution Load Index (PLI)

PLI is utilized to assess the overall level of soil contamination which provides an easy way to demonstrate the degradation of soil quality due to the accumulation of heavy metals [62]. Figure 5 presents the PLI value of heavy metals in the study area with sample SS13 exhibiting the least value and sample SS6 exhibiting the highest value. The PLI values were greater than one (PLI > 1) in samples SS1, SS2, SS3, SS6, SS8, SS9, SS10, and SS15, indicating high heavy metal pollution. Similar results have been reported in the literature concerning soils from other regions worldwide. For instance, Jalali et al. [63] reported the high levels of trace element pollution in Iranian agricultural soils. Javaid et al. [64] published a severely heavy metal-polluted region in proximity to industrial and residential sites along the Hudiara drain. In contrast, samples SS4, SS5, SS7, SS11, and SS13 exhibited PLI values less than one (PLI<1), indicating low heavy metal pollution, and we classified them as unpolluted samples. Samples SS12 and SS14 were at a baseline value of one (PLI = 1), indicating that the samples were at the maximum permissible contamination limit. The variance in pollution levels among the samples may be attributed to factors, such as the period of mining, the volume of mining operations performed, the types of equipment used, and the method of waste management at each sample site.

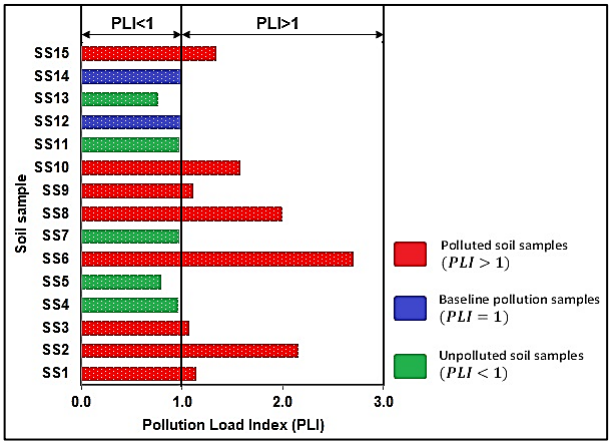

Figure 5. Classification of the Pollution Load Index (PLI) in Soil Samples.

By comparing the value of the contamination factor and degree of contamination of each of the heavy metals (Figures 3 and 4) with their pollution load indices (Figure 5) at each sampling site, it was observed that only lead (Pb) contamination levels were negligible at all sampling

sites. However, samples SS1, SS2, SS3, SS6, SS8, SS9, SS10, and SS15 displayed moderate contamination levels of iron (Fe), zinc (Zn), chromium (Cr), and copper (Cu), while nickel (Ni), arsenic (As), and cadmium (Cd) displayed elevated contamination levels in the same samples. Therefore, Fe, Zn, Cr, and Cu pollution should be closely monitored at these sites due to their moderate contamination levels, while Ni, As, and Cd pollution should be addressed through remediation measures. This will prevent additional decline in the nearby surroundings and shield the inhabitants of the study area from being exposed to Ni, As, and Cd health risks, such as skin damage, kidney failure, cancer risk, and circulatory system issues [24].

3.3. Modeling of contamination hazards

3.3.1. Correlation Analysis

Table 3 displays the correlation between heavy metal concentrations and contamination variables. The strong and significant correlation between HM-CF, HM-Cdeg, and HM-PLI indicates the sources of heavy metal concentration. The concentration of heavy metals in the soil at the sample sites determines the extent of contamination risks to both the environment and human health. CF and HM exhibit a positive and significant correlation which establishes the contamination level of every toxic metal in the soil. Cdeg-HM and Cdeg-CF also show a positive and significant correlation, determining the overall contamination of each heavy metal in the soil samples. PLI exhibits positive and significant correlation coefficients of 0.985, 0.902, and 0.991 with HM, CF, and Cdeg which ascertains the pollution load index of heavy metals in the soil samples.

Table 3: Pearson Product Correlations Coefficient Analysis.

	HM	CF	Cdeg	PLI
HM	1	.	.	
CF	.953**	1		
Cdeg	.970**	.892**	1	
PLI	.985**	.902**	.991**	1

**. Correlation is significant at 0.01 level (2-tailed).

3.3.2. Prediction of Heavy Metal Pollution Level

Contamination variables (Cf, Cdeg, and PLI) using regression analysis were used to predict heavy metal pollution in the study area, as shown in Tables 4–6. According to Tables 4 and 5, contamination variables accounted for 99.4% of heavy metal pollution in the study area [$F(3,4) = 233.305$, $p < 0.01$]. In predicting the level of heavy metal pollution, it was found that Cf ($p < 0.017$) and PLI ($p < 0.030$) are positive and significant predictors of heavy metal pollution in the study area, as shown in Table 6. The predictive equation (Equation 4) established that for every unit increase in Cf and PLI, the heavy metal pollution level in the study area increases by 0.138 and 0.578, respectively. Therefore, the heavy metal pollution level with its risk factor can be frequently estimated by adopting Equation 4.

$$HM = 0.064 + 0.138Cf + 0.578PLI \quad (4)$$

Table 4. Model Summary.

Model	R	R^2	Adj. R^2	RMSE
1	.997[a]	.994	.990	.085

Table 5. ANOVA.

	Model	SS	df	MS	F	Sig.
1	Regression	5.105	3	1.702	233.305	<.001
	Residual	.029	4	.007		
	Total	5.134	7			

Table 6. Coefficients.

Model		B	SE	β	t	Sig.
1	(Constant)	.604	.217		2.783	.050
	Cf	.138	.035	.346	3.968	.017
	Cdeg	-.025	.022	-.323	-1.118	.326
	PLI	.578	.176	.993	3.294	.030

4. Conclusions

The study used the Ikinyinwa gemstone mining site in Nigeria as a case study to predict heavy metal pollution using contamination variables and multiple regression analysis. The results showed that the mean values of Fe (20.11 ± 0.31), Zn (9.13 ± 0.08), Cr (8.39 ± 1.01), Cu (10.09 ± 0.10), Pb (0.69 ± 0.37), Ni (2.93 ± 0.01), As (7.51 ± 0.59), and Cd (0.65 ± 0.26) in MA were higher than in NMA. The average value of the contamination factor (Cf) of Pb (0.44) was classified as low ($Cf < 1$) contamination, while Ni (3.06) was classified as considerable ($3 \leq Cf < 6$) contamination, and others were moderate ($1 < CF < 3$) contamination. The average value of the degree of contamination (Cdeg) of Pb (5.83) was classified as low (Cdeg ≤8) contamination, while Fe (17.83), Zn (20.22), Cr (17.76), and Cu (19.8) were classified as having considerable (16≤Cdeg≤32) contamination, and Ni (46.08), As (36.83), and Cd (35.12) were classified as very high (32≥Cdeg) contamination. The pollution load index (PLI) revealed that samples SS4, SS5, SS7, SS11, and SS13 exhibited the PLI values less than one (PLI<1), indicating low heavy metal pollution, and we classified them as unpolluted samples. Samples SS12 and SS14 were at a baseline value of one (PLI = 1), indicating that the samples were at the maximum permissible contamination limit. Meanwhile, the PLI values were greater than one (PLI > 1) in samples SS1, SS2, SS3, SS6, SS8, SS9, SS10, and SS15, indicating high heavy metal pollution. The results revealed that Cf ($p < 0.017$) and PLI ($p < 0.030$) were positive and significant predictors of heavy metal pollution in the study area. The study, therefore, established that for every unit increase in Cf and PLI, the heavy metal pollution level in the study area increases by 0.138 and 0.578, respectively.

5. Strengths and Limitations

The strengths of this study are the use of primary data and the careful analysis and interpretation of the results. Another strength is the detailed discussion of the contamination variables and regression analysis in predicting the levels of heavy metal pollution in gemstone mining environments. The predictive equation obtained from this study is not limited to the extraction of gemstones and can be used for other minerals. However, the equation is restricted by the sole use of Cf, Cdeg, and PLI contamination variables.

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Conflict-of- interest statement

The authors declare that they have no conflict of interest.

Author contributions:

OC carried out fieldwork and was involved in the writing of the manuscript; AK conducted the experiments and was involved in the writing of the manuscript, while AG carried out results interpretations and was involved in the writing of the manuscript.

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