



## Research article

# Multivariate studies and heavy metal pollution in soil from gold mining area

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## ARTICLE INFO

## Keywords:

Environment  
Heavy metals  
Pollution  
Topsoil

## ABSTRACT

Mining generates large volumes of waste, which if not regulated can release toxic metals, causing widespread environmental contamination. This study focused on heavy metal contamination in topsoil within a mining area at Nangodi in the Northern Region of Ghana. A total of 24 soil samples were collected from the study area and control samples were analyzed for Hg, Pb, Cd, As, Cr, and Fe using atomic absorption spectroscopy. Results of Pollution Index estimations and Geo-accumulation index (Igeo) classified the soil samples as moderately contaminated to heavily contaminated. Soil samples were severely enriched with As and moderately enriched with b, and Hg. Multivariate analyses such as factor analysis and cluster analysis were employed to examine the relationship between the metals and also differentiate the influence of the natural background content of metals from that due to human activities. Factor analysis identified three polluted soil factor associations. Cadmium, Fe, As, and Pb associated with factor 1, were due to anthropogenic activities. The high intercorrelation revealed by As and Pb shows similarity in their sources. Factor two dominated by Hg is considered an anthropogenic component. Factor 3 correlated with Cr and can be considered a natural component. Correlation analysis and cluster analysis supported each other. Results from the bi-plots showed that sites S1, S8, S11, and S18, have similar metal composition as the control site. Heavy metal contents in soils sampled from Zones A and B have been influenced by the mining activities as seen from the associations of these sites in the bi-plots. The results are useful for metal source identification, and can contribute to monitoring and regulatory programs.

## 1. Introduction

Artisanal or small-scale gold mining is essential in many developing countries such as Ghana. Because it provides jobs for the indigenous people, particularly in rural communities where economic activities are critically limited and also make significant contributions to foreign exchange earnings [1]. Ghana's artisanal or small-scale mining sector is growing in size and significance. Its

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<https://doi.org/10.1016/j.heliyon.2022.e12661>

Received 18 September 2022; Received in revised form 19 December 2022; Accepted 20 December 2022

Available online 6 January 2023

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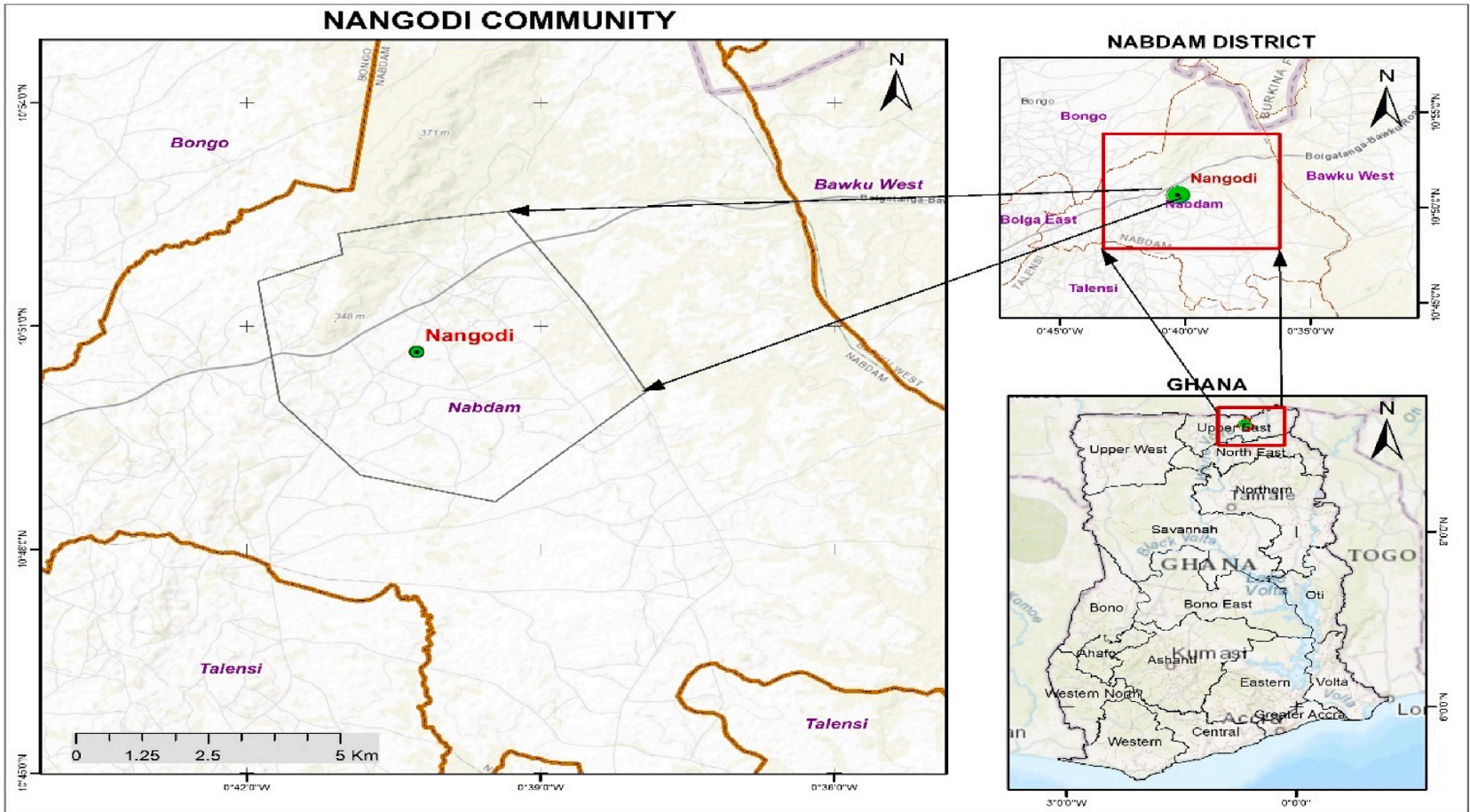


Fig. 1. Map of the study area.

contribution to wealth creation, employment and the economy make it one of the nation's most important livelihood activities, directly employing an estimated one million people and supporting approximately 4.5 million more [2].

However, mining activities negatively impact the environment, and the impact's severity depends on the miners' methods and technical skills [3,4,5].

In Ghana, using rudimentary methods by small-scale gold miners has caused significant environmental damage, especially to landscape and ecosystem, impact on the quality of ground and surface water, impact on flora and fauna as well as on humans. Mining wastes contain minerals, including silicates, sulphides, oxides, hydroxides, and carbonates. When the sulphides come into contact with oxygen and water (humidity), they generate acid water which dissolves and mobilize heavy metals and releases them into the environment [6,7].

The potential effects of heavy metals on human health and the environment have become a major concern worldwide [6,8–10]. Because of their high toxicity and persistence, heavy metals such as, As, Pb, Cd and Hg are among the priority chemicals of public health concern. Long-term exposure to these heavy metals can lead to cancers and other diseases like lung and kidney damage, neurological disorders, DNA damage and infertility [11,12,13,14]. Several landscapes worldwide which, Ghana is not an exception, have been heavily damaged due to transient small-scale gold mining activity and this is occurring because of intensive levels of exploration activity [15].

Gold deposits discovered in Nangodi in the Nandam traditional area of the Northern Region of the Republic of Ghana have attracted a lot of small-scale mining ventures to the area. This has led to illegal small-scale gold mining activities such as improper tailings disposal, which may contain sulfide-bearing material such as pyrite. When pyrite is exposed to air in the presence of water, it produces acid mine drainage (AMD), one of the most significant environmental issues associated with mining and mine wastes [16,17]. Without treatment, the harmful effects of AMD lead to the destruction of surrounding ecosystems, including serious health impacts on affected communities [18]. Low pH acid mine drainage enhances the dissolution of heavy metals in water; hence this causes the metals to quickly move in the environmental media, adversely impacting the ecosystem's flora and fauna.

High levels of toxic metals and other contaminants are therefore released into the environment, which can pollute surface and groundwater. Unauthorized mining activities could also result in environmental havoc and destruction. Vast lands that could be used for other economic activities, such as agriculture, can lose their properties [19]. The activities have caused the systematic destruction of farmlands without compensation to the affected farmers and landowners [20].

Traditionally, Nangodi is a farming community where cereals and legumes are mostly grown. Activities of illegal small-scale gold mining can potentially increase metal pollution, threaten the quality of water, soil and plant growth, and deprive the indigenes of their livelihood. The present study seeks to estimate the extent of heavy metal pollution and identify the possible sources of these metals in soil within the Nangodi community using a multivariate statistical approach. The outcome of this study will highlight potential threats of mining activities to soil and human health and determine mitigation measures that would be needed.

## 2. Materials and method

### 2.1. Study area

Nangodi is located on latitude 10° 15' and 10° 60' north and longitude 0° 31' and 1° 05' west (Tom-Dery et al., 2012). Its total land area is 244.94 km<sup>2</sup> [21] Fig. 1. The topography is naturally undulating lowlands with gentle slopes ranging from 1% to 5% gradient with few hilly terrains mainly of quartzite and gneiss formations [22]. The underline rocks fall within Ghana's Tarkwaian, Birimian and Voltarian rocks [23].

The area experiences irregular rainfall patterns with annual mean rainfall between 800 mm and 1100 mm. The rainy season is usually from May to June and between September and October. The dry season, characterized by the dry northeast trade winds, extends from November to April. The area experiences a maximum temperature of 45 °C and a minimum of 12 °C in December with a relative humidity of 69%.

### 2.2. Sampling

Nangodi was selected for the study because of the extensive destruction of land by illegal small-scale gold mining activities within the community. In order to study the influence of illegal small-scale mining on the extent of heavy metal pollution in the soil, the area was divided into four (4) zones; Zone A represented areas where gold-bearing rocks are extracted; Zone B represented areas where mine rocks are washed and Hg added to extract gold; Zone C represented an area where there is no activity concerning gold mining (control) and Zone D represented an area where mining tailings are dumped. Ten (10) top-soil samples each were collected from different locations within Zone A and Zone B. Two (2) soil samples were also taken from different locations within Zone C, were used as control samples and two (2) other different soil samples were taken from Zone D. The soil samples were collected at a depth of 0–25 cm from the surface using a stainless-steel spatula. Samples collected were kept in plastic bags, labelled and transported to the laboratories of the Chemistry Department at Kwame Nkrumah University of Science and Technology (KNUST).

### 2.3. Sample treatment

Soil samples were air-dried at room temperature to constant weight. The dried samples were ground using a mortar and pestle and sieved through a 1 mm sieve to obtain a homogenous sample. The sieved soil samples were stored in labelled plastic containers until

digestion.

#### 2.4. Digestion of soil samples

Approximately 1 g of each sieved soil sample was weighed and transferred into conical flasks. 10 mL of a mixture of HNO<sub>3</sub> - HClO<sub>4</sub> (9:4) was added [24]. Each sample was placed on a hot plate and heated until the production of red NO<sub>2</sub> fumes ceased. The contents were further heated until the volume reduced to about 4 mL and became yellowish. Digested samples were allowed to cool and filtered through Whatman No.1 acid-washed filter paper into 100 mL volumetric flask and filled to the mark with distilled water. The resulting solution was preserved at 4 °C in pre-cleaned bottles until analysis [24].

Concentrations of Cr, Cd, Fe and Pb were measured using Atomic Absorption Spectrophotometry (AAS 220; PerkinElmer, AAS). Mercury was analyzed by Cold Vapour Atomic Absorption Spectrophotometry using an Automatic Mercury Analyser model HG-5000 developed at the National Institute for Minamata Disease (NIMD). A Hydride Generator Atomic Absorption Spectrophotometer was used in determining As in all digests [25].

Soil pH and electrical conductivity (EC) were measured in soil/distilled water mixture of ratio 1:10 (w/v). The solution was allowed to settle for 30 min. Electrical conductivity and pH were measured using a calibrated conductivity meter and a pH meter (HANNA HI 98150) with the ion-selectivity hydrogen electrode respectively. Soil organic matter content was determined using the Loss on Ignition method while soil texture was determined by the pipette method described by Kroetsch [23].

#### 2.5. Enrichment factor

Enrichment factor (EF) is used to determine the presence and intensity of heavy metals pollution by anthropogenic deposition in soil compared to their natural abundance. The EF calculation described by Ref. [25] as defined in equation (1) was adopted for this study.

$$EF = \frac{(X/Fe)_{soil}}{(X/Fe)_{background}} \quad (1)$$

where (X/Fe)<sub>soil</sub> is the ratio of heavy metal (X) to Fe in the soil from the mining site.

(X/Fe)<sub>background</sub> is the natural background value of the metal to Fe ratio.

Iron (Fe) was used as a reference element because Fe is naturally present in soils and its concentration is not altered anthropogenically [26].

The higher the EF value of a metal the more severe its contribution from anthropogenic sources.

#### 2.6. Geoaccumulation Index (I<sub>geo</sub>)

Another factor used to determine the intensity of metal pollution in the soil is the geoaccumulation index (I<sub>geo</sub>) introduced by Ref. [27]. The geoaccumulation index (I<sub>geo</sub>) was calculated using equation (2)

$$I_{geo} = \log_2 \frac{C_m}{1.5B_n} \quad (2)$$

C<sub>m</sub> = measured concentration of metal in soil.

B<sub>n</sub> = geoaccumulation background values of metals

1.5the background matrix correction factor due to the lithogenic effects

The I<sub>geo</sub> scale consists of seven grades (0–6) ranging from uncontaminated to extremely contaminated.

#### 2.7. Pollution index

Pollution index (PI) is used to evaluate the pollution status of heavy metals in soil. The PI was calculated from equation (3).

$$P_i = C_i/S_i \quad (3)$$

where P<sub>i</sub> the pollution index of heavy metal (i).

C<sub>i</sub> (mg/kg) is the concentration of heavy metal in the soil.

S<sub>i</sub> (mg/kg) is the concentration of standard heavy metal value for soil quality.

#### 2.8. Statistical analysis

The mean and standard deviation of the studied physicochemical parameters and heavy metals were calculated with Microsoft Office Excel (2013) spreadsheet. All statistical analyses were done using JMP 10 Statistical Software (SAS Institute). Pearson's correlation was performed to identify the important parameters affecting soil chemistry (at a significant level *p* less than 0.05). Factor analysis and Cluster analysis were carried out to describe the degree of association and identify possible sources of metals in the soil samples. The principal components were extracted with eigenvalues >1 through a varimax rotation. Cluster analysis was also



performed based on Euclidean distance using Ward's clustering method.

### 3. Results and discussion

#### 3.1. Physicochemical parameters of the soil samples

The results of the mean values of physicochemical parameters (pH, EC, TOM and Texture) measured in the soil samples collected from different sampling sites from the Nangodi illegal gold mining area are represented in Table 1.

The pH of soil samples from the sampling zones at Nangodi are represented in Table 1. The pH varied from  $5.6 \pm 1.2$  at Zone A, where the gold-bearing rocks were extracted to  $7.7 \pm 0.6$  at Zone D where the mine tailings are kept, indicating that the soil is slightly acidic to near neutral. pH is considered a parameter that controls the bioavailability of metals in soil [27]. For most heavy metals, the acidity of the soil results in a higher solubility than adsorption. Thus, the mobility of metals is promoted by an acidic pH. The mean soil pH values recorded from the study area may not significantly influence the solubility and mobility of the heavy metals in the soil because the acidic pH is high and near neutral [28]. Soil EC is defined as the ability of soil water to carry electrical current [29]. In general, the greater the concentration of the ions in the soil, the higher the conductivity. EC of soil samples from the study area varied from  $142.5 \pm 7.6$   $\mu\text{S}/\text{cm}$  in Zone A to  $206.7 \pm 2.3$   $\mu\text{S}/\text{cm}$  in Zone C. Soil samples from the control site (Zone C) recorded the highest EC values. This may be because the mining activities have compromised the soil compatibility in the mining areas and the soil has become loose and more porous.

Total organic matter (OM) recorded in the soil samples ranged from  $0.9 \pm 0.4\%$  at Zone A to  $1.8 \pm 1.2$  at Zone B. The extreme removal of humus horizon (topsoil) from the activity zones and the poor vegetation cover coupled with annual bushfires have resulted in low levels of OM within the study area. This low OM content of the soil samples will not influence the cation-holding capacity of the soil, resulting in low levels of EC in the soil samples.

#### 3.2. Levels of heavy metals in soil

The results of the mean concentrations of heavy metals (Hg, Cr, Cd, As, Pb, Fe) in the soil samples from the four selected Zones of illegal mining around Nangodi community are represented in Table 2.

Mean Hg levels in the soil samples ranged from  $2.20 \pm 0.14$  to  $7.46 \pm 2.96$  mg/kg. Zone C and Zone B recorded the lowest and highest mean Hg concentrations, respectively, as presented in Table 2. The illegal gold mining activity has increased the mean Hg levels in the soil samples from Zone B above the control samples. This is a result of the Hg added to amalgamate the gold after washing. The observed level of Hg at Zone C (control site) may be because Hg is volatile and can easily be transferred through the atmosphere to remote areas. In a similar study in the Amansie West District of the Ashanti Region of Ghana [17], recorded Hg levels as high as 11.20 mg/kg, which was equivalent to an increase of 300% of Hg over control sample, suggesting a higher use of Hg for gold mining by small-scale gold miners in Ghana. Mercury in soil is an indicator of an industrial activity such as mining and has a unique property of propagation over long distances [30].

Mean Pb concentration in the soil samples was highest in Zone D ( $21.65 \pm 0.21$  mg/kg) and lowest in Zone C ( $1.45 \pm 0.21$  mg/kg). The highest mean Pb concentration was observed at the site where the tailings are dumped after the gold-bearing rocks are crushed and gold extracted. It was, however, lower than the 1200 ppm [31] recommended for non-residential areas. Leopold [30] observed 19.02 mg/kg Pb in a traditional gold mining site in Kombo-laka, Cameroun. Bempah and Ewusi [32] recorded 35 mg/kg Pb around Obuasi gold mine in Ghana. The present study tends to support the observation made by Ref. [33] that anthropogenic activities contribute to Pb concentrations in soil. The high concentration of Pb in the active mining areas compared to Zone C could be due to the nature of the gold-bearing rocks in the study area, which may contain high levels of Pb, as observed at Zone D where the tailing was dumped after gold extraction.

Average Cd concentrations recorded from the different zones during the study ranged from  $2.0 \pm 0.28$  mg/kg at Zone C to  $14.60 \pm 0.28$  mg/kg at Zone D. differences in Cd concentrations between Zones A, B and D were significant, indicating possible anthropogenic influence except that of Zone C. According to WHO, the standard requirement for Cd in the soil is 1.4 mg/kg [34], suggesting that the Cd recorded from Zones A, B and D, as presented in Table 2, were extremely high. This indicates Cd pollution in the soils within the study area. However [35], recorded 0.5–3.8 mg/kg Cd in the artisanal mining site in Dakoto-Shega in the Upper East Region of Ghana.

Levels of arsenic As in the soil samples from Zone D were extremely high ( $21.7 \pm 0.57$  mg/kg) compared to values recorded in soils from the other Zones. Soil samples from Zone C recorded the least As the concentration of  $0.35 \pm 0.07$  mg/kg (Table 2). Mean As

**Table 1**  
Mean  $\pm$  SD of physicochemical parameters of soil samples from the zones in Nangodi.

Parameters		Zone A	Zone B	Zone C	Zone D
pH	Range	4.8–6.4	5.1–8.0	6.5–6.6	7.5–8.1
	Mean	$5.6 \pm 1.2$	$6.8 \pm 1.6$	$6.5 \pm 0.1$	$7.7 \pm 0.6$
EC( $\mu\text{S}/\text{cm}$ )	Range	133.8–158.1	162.9–204.2	205.1–208.4	165.6–167.4
	Mean	$142.5 \pm 7.6$	$179.2 \pm 13.8$	$206.7 \pm 2.3$	$166.5 \pm 1.3$
TOM (%)	Range	0.2–1.2	0.1–3.5	0.1–2.2	0.3–1.7
	Mean	$0.9 \pm 0.4$	$1.8 \pm 1.2$	$1.7 \pm 0.3$	$1.1 \pm 0.9$
Texture		Sandy/Loamy	Clay	Sandy/Loamy	Clay

**Table 2**

Range and mean concentrations of heavy metals (mg/kg) in soil samples from the selected zones in Nangodi.

Heavy Metals		Zone A	Zone B	Zone C	Zone D
Hg	Range	3.9–7.5	3.4–12.4	2.1–2.3	3.3–3.4
	Mean	5.97 ± 0.94	7.46 ± 2.96	2.2 ± 0.14	3.35 ± 0.07
Pb	Range	7.43–12.60	1.93–11.43	1.30–1.68	21.51–21.82
	Mean	11.71 ± 3.86	7.31 ± 2.52	1.45 ± 0.21	21.65 ± 0.21
Cd	Range	1.92–11.44	4.76–11.31	1.80–2.25	14.44–14.81
	Mean	8.78 ± 4.55	8.45 ± 2.03	2.0 ± 0.28	14.60 ± 0.28
As	Range	0.82–11.14	0.53–9.25	0.34–0.48	21.38–22.16
	Mean	5.30 ± 3.63	4.67 ± 2.94	0.35 ± 0.07	21.70 ± 0.57
Cr	Range	2.87–13.35	5.438–10.61	2.77–2.99	8.55–9.34
	Mean	7.87 ± 3.53	7.75 ± 14.97	2.80 ± 0.14	8.90 ± 0.57
Fe	Range	463.61–634.76	412.92–585.13	248.73–296.21	824.60–843.28
	Mean	571.40 ± 69.80	484.53 ± 70.10	272.35 ± 33.50	833.80 ± 13.10

concentrations in soil from Zone D exceeded the recommended level by WHO which is 12 mg/kg [34]. In other studies, Gyasi et al. [36] recorded relatively lower mean As concentration (0.16 mg/kg) in soil from Buruli Ulcer endemic communities in the Ashanti Region of Ghana, where activities of small-scale gold mining are high. The extremely high levels of As recorded in soils from Zone D is mainly from arsenopyrite gold-bearing rocks tailings dumped haphazardly at that site. Thus As levels in the soil have been enriched by the activities of the mining operations and the mine tailings dumped at that site.

Chromium in the soil is predominantly affected by rock weathering and erosion and only slightly by anthropogenic activities [37]. Chromium concentrations in the soil samples varied from  $2.8 \pm 0.14$  mg/kg at Zone C to  $8.9 \pm 0.57$  mg/kg at Zone D. Another study by Ref. [38] recorded much higher Cr concentrations in the range 104.3–938.7 mg/kg in Dakoto-Shega another illegal gold mining community Upper East Region of Ghana. The miners' activities have not significantly impacted the levels of Cr in the soils from the study area.

Iron is one of the most abundant metals in soil [39]. It was the most predominant of all the metals considered in this study because it recorded a relatively higher concentration than the other metals with concentrations varying from  $272.35 \pm 33.45$  mg/kg to  $833.8 \pm 13.01$  mg/kg (Table 3). The presence of iron-rich laterite sub-soils which FAO classifies as ferralsols and oxisols accounted for high Fe concentrations observed in this study [40]. made a similar observation in his study in Tarkwa gold mining community in Ghana.

### 3.3. Enrichment factor (EF)

Enrichment factor values are presented in Table 3. All the metals had EF values equal or exceeding 1.0, except for Cd (0.99) in Zone C and Hg (0.50) in Zone D. The highest EF value 20.26, was recorded for As in Zone D.

Enrichment factor (EF) estimations for heavy metal concentrations in soils are necessary for geochemical studies in differentiating heavy metal sources that originated from human activities and lithogenic processes. EF can be used to determine the source and intensity of metal contamination from human activities in soil [41,42]. EF values calculated for Cd and Hg in the soil samples from Zones C and D were less than 1, indicating that their main sources are mainly from lithogenic processes. EF for all the studied metals in the soil sampled from Zone A showed minor to moderate enrichment except for As, which showed moderately severe enrichment (7.55). EF values recorded for all the studied metals (Hg, Pb, Cd, As and Cr) showed moderate enrichment in Zone B, with Hg recording the highest value. This can be attributed to the amalgamation that occurred at Zone B as the main source of Hg. Soils in Zone D showed moderate to minor enrichment for Pb, Cd and Cr (Table 3). Arsenic in Zone D showed the highest EF value of 20.26. This can be attributed to the fact that associated minerals in the gold-bearing rocks are principally arsenopyrite and the soil around where the mine tailings are dumped recorded high concentrations of As. This As could leach into surface and groundwater and increase their As concentrations. From the study, As and Pb in the soil, from Zones A, B and D showed high EF values. These metals are highly toxic, have no known biological importance, and can potentially enter the food chain and cause adverse health impacts to humans [43,44]. The enrichment of the metals (As, Pb, Cd, Cr and Hg) in the soil samples was influenced by mining which is the main human activity in the study area.

**Table 3**

Mean EF values for the soil samples collected from selected zones in the study area.

Metals	Zone A	Zone B	Zone C	Zone D
Hg	1.30	1.91	1.01	0.50
Pb	3.94	2.95	1.02	4.88
Cd	2.06	2.44	0.99	2.39
As	7.55	4.46	1.02	20.26
Cr	1.32	1.61	1.01	1.04

### 3.4. Geoaccumulation Index ( $I_{geo}$ )

In this study,  $I_{geo}$  was used to determine and describe the extent of heavy metal contamination in the soils by comparing recent concentrations with background levels.  $I_{geo}$  class includes seven grades (0–6) varying from uncontaminated to highly contaminated. Calculated means  $I_{geo}$  for heavy metal concentrations in all the zones varied from 0 to 5.37 (Table 4), which suggests contamination in some Zones. The  $I_{geo}$  values obtained for the metals in Zone A indicate that Hg and Cr were uncontaminated to moderately contaminated; however, As and Pb were moderately to heavily contaminated in the soil, as presented in Table 4. In Zone B, all metals except Cr were in class 2, representing moderate contamination for the metals in the soil. Chromium and Fe were in Class 1 with  $I_{geo}$  values of 0.86. However, Zone D was uncontaminated for Hg (0.02), Cr was moderately contaminated with  $I_{geo}$  values between 1 and 2. Cadmium (2.28) was put in Class 3. Pb was heavily contaminated in Zone D. Results revealed that  $I_{geo}$  value for As was >5, which puts As in class 6, implying that the soil is extremely contaminated with As. The average  $I_{geo}$  value of the studied metals calculated for all the zones was in the order Hg < Cr < Cd < Pb < As. This shows that Hg and Cr were the least polluted metals in the study, while Pb and As showed significant pollution in all the activity areas within the study area. The result of  $I_{geo}$  suggests that gold mining, the major anthropogenic activity in the area, has significantly affected concentrations of As, Pb and Cd in the soils.

### 3.5. Pollution index (PI)

PI was calculated to evaluate the pollution status of heavy metals in the soil from the study area. PI of the metal in soil from Zone A revealed that Hg, Cd and Cr were moderately polluted. Lead and As recorded PI values of 8.08 and 15.14, respectively, indicating that the pollution levels of these metals are high. PI values for Hg and Cd in Zone B were >3 but Pb and As once again recorded high PI values of 5.04 and 7.63, respectively, at Zone D. Hg had the least PI value at Zone D, indicating moderate pollution (Table 5). However, all other metals in soils sampled from zone D recorded PI > 3, indicating high pollution, with As showing the highest PI value of 62, Pb (14.93), Cr (3.18).

The intense illegal gold mining activities characterized by the arsenopyrite nature of the gold-bearing rocks, haphazard dumping of mine residues and tailings, which are potential sources of As and Pb and by-products of mining which releases other contaminants into the soil have contributed to the buildup of the heavy metals in soils within the study area.

### 3.6. Correlation analysis

Pearson's correlation coefficients of heavy metals in soil samples and physicochemical parameters of the soil are summarised in Table 6. A significant positive correlation was observed between Pb–As (0.81) and Cd–Cr (0.72), indicating that the same anthropogenic activity might have influenced these metals at a 99% confidence level. Again, Fe has a significant correlation with Pb (0.68), Cd (0.64) and As (0.64) at 95% confidence level, which suggests that these metals may have the same source or sink in some locations. There was no significant correlation between Hg and the other metals, signifying a degree of uniqueness, indicating that different anthropogenic activities influence Hg. Cd and Pb also exhibited a positive correlation at p (0.05). There was also a negative but significant correlation between EC and Pb and EC and Fe.

### 3.7. Factor analysis

To be able to properly identify the possible sources (lithogenic or anthropogenic) and better understand the relationship among the metals in the soil, factor analysis was applied using Varimax rotation to the experimental data [40,45]. The results in Table 7 show that only three eigenvalues are >1.00 and explain 87.6% of the total variance of the system.

The first factor (F1) accounted for 49.5% of the total variation and was dominated by Pb, Cd, As and Fe (Table 8). Lead and As are strongly associated, while Cd and Fe are highly affiliated with these metals. This association could be tentatively named as ore specific since these metals' common source of occurrence is the basic rock and the gold-bearing rock, which contain As and Pb [46]. This represents the effect of pollution caused by gold mining activity. The second factor (F2) accounted for 20.4% of the total variance and was highly dominated by Hg, indicating the main metal contribution from the influence of the amalgamation process, which involves the use of Hg. Factor 3 accounted for 17.62% of the total variance and was dominated by Cr. The mean concentration of Cr in the topsoil samples from the control area was not significantly different from the active mining areas. Again EF,  $I_{geo}$ , and PI values calculated for Cr, showed that Cr was not significantly affected in the active mining areas. Therefore, Cr levels in the soil are controlled mainly by natural factors of the lithogenic process during the weathering process of parent rocks.

**Table 4**  
Mean  $I_{geo}$  values of soil samples collected from selected zones in the study area.

Metals	Zone A	Zone B	Zone C	Zone D
Hg	0.84	1.07	0	0.02
Pb	2.37	1.63	0	3.32
Cd	1.29	1.45	0	2.28
As	2.91	1.60	0	5.37
Cr	0.70	0.86	0	1.08

**Table 5**  
Pollution Index values of soil from in the study area.

Metals	Zone A	Zone B	Zone C	Zone D
Hg	2.71	3.39	1.00	1.52
Pb	8.08	5.04	1.00	14.93
Cd	4.39	4.18	1.00	7.30
As	15.14	7.63	1.00	62.00
Cr	2.81	2.86	1.00	3.18

**Table 6**  
Correlation matrix for heavy metal concentrations and Physico-chemical Parameters.

Variables	Hg	Pb	Cd	As	Cr	Fe	pH	EC	TOM
Hg	1								
Pb	-0.25	1							
Cd	0.10	<b>0.66*</b>	1						
As	-0.35	<b>0.81**</b>	0.45	1					
Cr	0.25	0.37	<b>0.72**</b>	0.26	1				
Fe	0.09	<b>0.68*</b>	<b>0.64*</b>	<b>0.64*</b>	0.45	1			
pH	0.00	0.017	0.15	0.17	0.00	-0.03	1		
EC	-0.17	<b>-0.51</b>	-0.32	-0.19	-0.26	<b>-0.50</b>	<b>0.52</b>	1	
TOM	<b>0.54</b>	-0.27	-0.03	-0.23	0.09	-0.06	0.20	0.19	1

\*\*Correlation is significant at 0.01 level.

\*Correlation is significant at 0.05 level.

**Table 7**  
Cumulative variance of factor Coefficients.

	F1	F2	F3	F4	F5	F6
Eigenvalue	2.97	1.22	1.05	0.34	0.25	0.14
Variability (%)	49.59	20.44	17.62	5.73	4.24	2.38
Cumulative (%)	49.59	70.03	87.65	93.38	97.62	100.00

**Table 8**  
Varimax-rotated factor loadings of metals in the soil samples.

	F1	F2	F3	F4	F5	F6
Hg	0.04	<b>0.73</b>	0.17	0.01	0.05	0.00
Pb	<b>0.86</b>	0.014	0.00	0.00	0.04	0.07
Cd	<b>0.61</b>	0.18	0.01	0.17	0.00	0.02
As	<b>0.75</b>	0.11	0.00	0.04	0.04	0.05
Cr	0.01	0.14	<b>0.81</b>	0.04	0.00	0.00
Fe	<b>0.71</b>	0.05	0.05	0.08	0.11	0.00

\*high positive factor loading is shown in bold.

For further assessment of the extent of heavy metal contamination in the study, source identification and metal distribution Figs (2) 1a, 1b and 1c were used. A loading plot 1a is used to identify the sources of metals in the samples. Three (3) components similar to that of Table 4 were extracted. These three components constitute the metals and their association with samples. These 3 components significantly explain metal contamination in the study area, which constitutes the metals and their associations with the soil samples, as seen in plot1b. For instance, mercury is highly associated with samples from sites S12, S13, S15, S16, S17, S19 and S20, all in Zone B (Fig. 3). Chromium is highly associated with samples obtained from S4, S5, S6, S10 and S14. Arsenic and Pb are highly associated with sites R1 and R2, where mining waste (tailings) are dumped.

No association was observed between metals and the control samples (C1 and C2) as the concentration of all metals within the control zones was low. Similarly, low metal concentrations were detected in S1, S3, S8, and S18 in the back box as they had no associations with the metals (Fig. 2), which suggests these areas had low metal concentrations or the presence of the metal in the samples were from natural sources.

### 3.8. Cluster analysis (CA)

CA organizes variables into homogenous groups where variables within the same cluster exhibit associations and are dissimilar to variables in other clusters. Clusters are formed based on existing similarities, which become useful for data interpretation and pattern



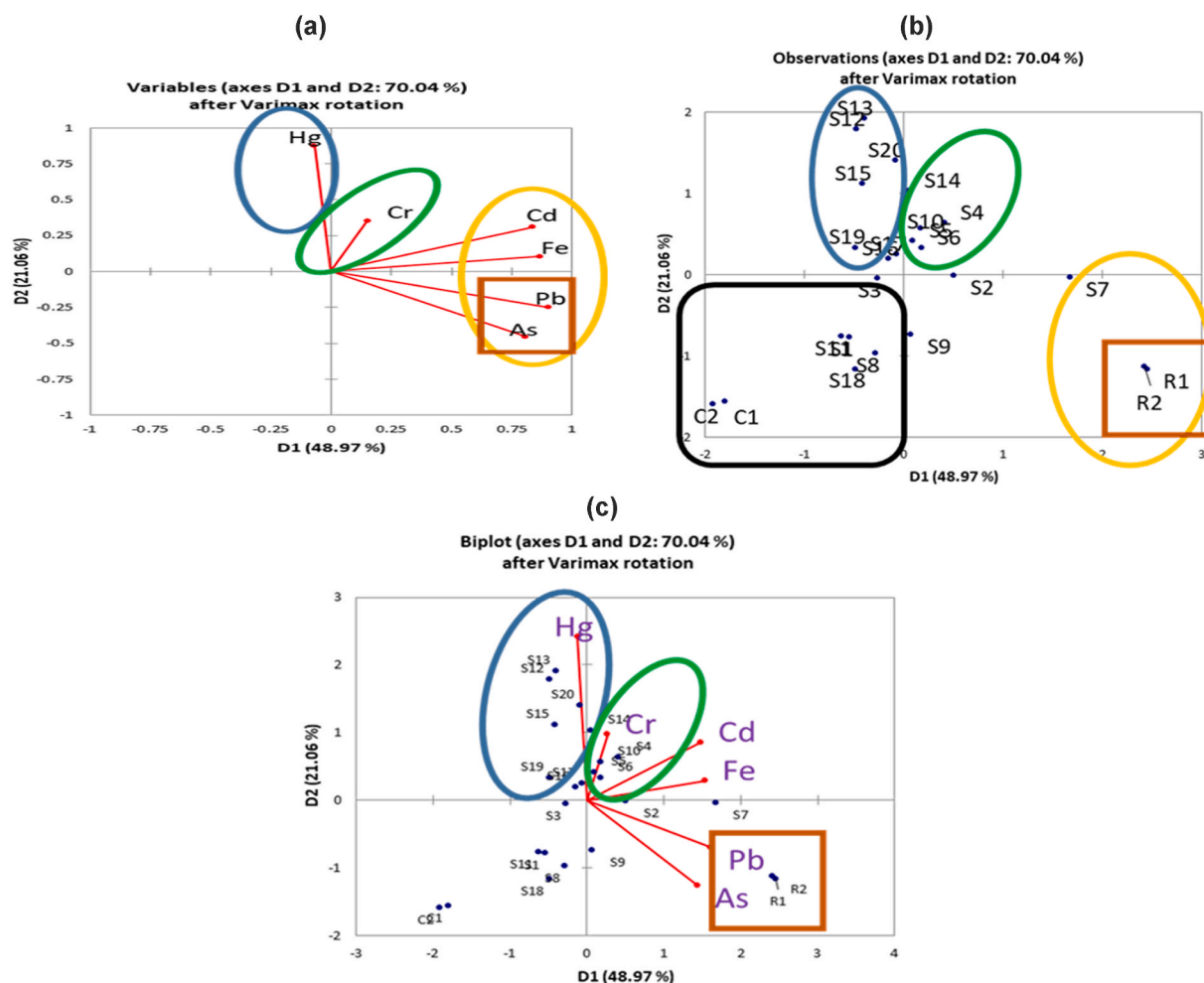


Fig. 2. Biplot of metal concentration (1a) sampling sites (1b) and a combination of the plots A (loading plot) and B (score plot) (1c).

recognition. This study grouped sampling locations into clusters based on Euclidean distance using Ward’s method. The results of the CA are presented in a dendrogram as shown in Fig. 3. In this dendrogram, all sampling sites were grouped into four statistically significant clusters.

Cluster 1 is composed of S2, S3, S4, S5, S6, S9, S10, S12, S13, S14, S15, S16 and S17. The sampling sites grouped in this cluster are either in Zones A or B, indicating similarities in heavy metal content in topsoil samples. Thus, the metal content in the top soil from these sites has been influenced by similar anthropogenic factors. The associations of these sites can also be seen in loading plots in plot 1b. Cluster 2 is represented by site 20. Cluster 3 includes S1, S8, S11, S18, C1 and C2, indicating their heavy metal content similarity to that of the control samples. Thus, the heavy metal content of soil from these sites is not affected by the mining activity and could be attributed to lithogenic sources. This result is consistent with that of the score plot, plot 1b. The fourth cluster is made up of S7, R1 and R2 and are associated with As and Pb as observed in the Factor analysis Tables 7 and 8 loading plots (plot 1b and 1c). Correlation analysis, FA and CA highlight the sources of heavy metals in the top soil of the study site. The combination of these three (3) statistical analyses showed a strong correlation between Pb, As, Cd and Fe represented in PC1 (Table 8) indicating significant associations. Furthermore, these metals had high positive loads of 0.86, 0.75, 0.61 and 0.71, respectively, which explains that the level of pollution of these metals was not affected by their natural characteristics in the soil, but precisely by anthropogenic activities within the study area, which was mining. All the clusters formed were based on existing relationships between soil samples and metals.

### 5. Conclusion

Findings of the study indicated that heavy metal (Hg, Pb, As, Cd, Cr and Fe) levels in top soils from the mining areas were higher than that of the control site. EF, Igeo and PI values estimated for As, Pb, Cd, Hg showed widespread pollution with varying degrees. The soil samples’ sources of heavy metals were examined using multivariate statistical techniques. The results of correlation, FA and CA supported each other. The examined metals were classified into three main groups. Concentrations Fe, Cd, Pb and As in the soil samples

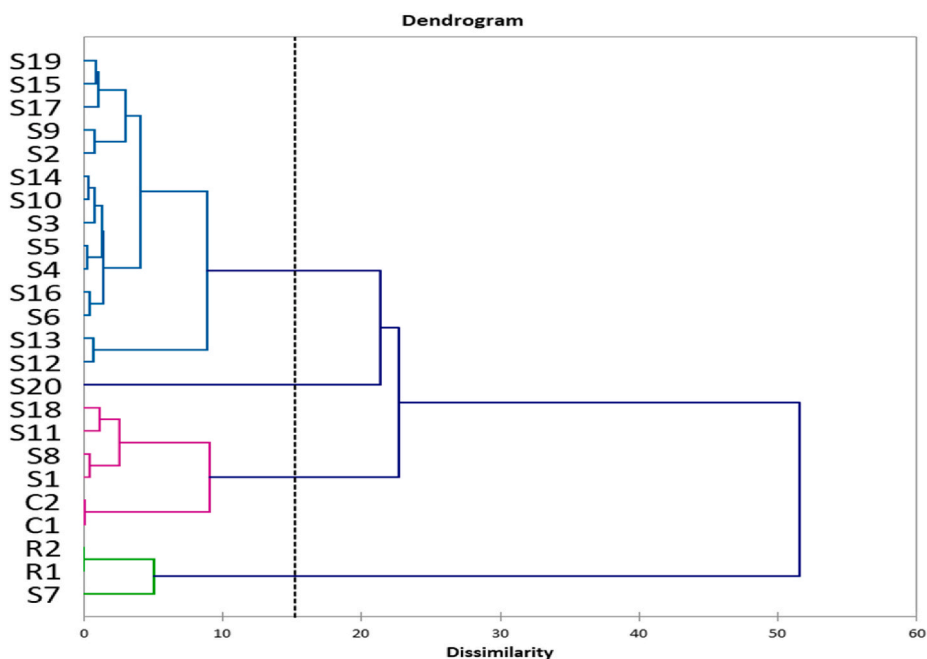


Fig. 3. Dendrogram of sampling sites.

significantly contribute significantly to the mining activity. The amalgamation process-controlled Hg distribution in the soil and can be considered an anthropogenic component since Hg levels in soil samples from Zone B were extremely high. Cr distribution in the soil is not affected much by the mining activities since the geoaccumulation and EF values showed that Cr has background concentration at most of the sampling sites. Results from CA and the loading plots showed that sites S1, S8, S11, S18, have a similar metal composition as the control, indicating that soils from these sites are not affected by the mining activity. The high intercorrelation revealed by As and Pb in this study shows similarity in their sources. The study showed preliminary yet relevant information concerning the pollution status of artisanal gold mining on soils. The results also provide useful information that can contribute to monitoring and regulatory programs at regional and national levels.

### Funding

The Ghana Government supported this work through the Postgraduate's Scholarship Programme.

### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

### Acknowledgement

The authors would like to thank the Technicians at the Department of Chemical Engineering in KNUST for the analysis.

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