

Environmental Pollution by Heavy Metals in the Gold Mining Region of East Cameroon

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Abstract: The main objective of this study is to assess the level of heavy metals pollution in soil samples collected around gold mines located in Betaré Oya (BO) and Batouri (BA) in Eastern Cameroon. Elemental analysis of soil samples was carried out using Quant'X EDXRF spectrometer. Its sensitivity and accuracy were increased by using the Fundamental Parameter approach for quantification of the results. Analysis results showed high content of iron (39300±200 ppm), the presence of manganese (730±70 ppm), arsenic (4±2 ppm), zirconium (314±4 ppm) and lead (79±9 ppm) as compared to worldwide average upper continental crust (UCC). The mean Enrichment Factors (EF) decrease as Pb>As>Zr>Mn>Fe>Y>Ga>Zn>Cu>Ni> Rb>Sr>Sn>Ba in agreement with the Contamination Factors (CF). The values of pollution load index (PLI) were found to be low in all the studied samples and indicate that the studied sites in Bétaré Oya and Batouri are in low pollution status regarding the total of the studied metals. Results of this study pointed out that soils examined in the gold mining areas of Eastern Cameroon are polluted by heavy metals. It is therefore important that measures should be geared towards strengthening the monitoring of mining areas to stem down the level of contamination of soil. Phytoremediation of sites after closing open pits or the biogeosystem method would be a suitable way of regulation of the studies areas.

Keywords: Gold Mine, X-Ray Fluorescence, Heavy Metals, Enrichment Factor, Pollution Load Index

Introduction

Incorrect outdated industrialization is one of the main causes of worldwide environmental pollution. Among the various pollutants, heavy metals are the more serious in our natural environment (Caeiro *et al.*, 2015; Emmanuel *et al.*, 2014; Pawan, 2012). The study of heavy metals deposition and accumulation is a universal problem because of the negative consequences that heavy metals in soils may have on human health and on the environment (Likuku *et al.*, 2013). At low fodder supply for cattle-keeping, in pastures in the vicinity of the mining area the heavy metals from plants and soils could enter the digestive tract of grazing livestock

subsequently contaminating the humans food chain. Soils are usually regarded as the ultimate sink for heavy metals discharged into the environment (Banat *et al.*, 2005; Nazari and Razmara, 2014; Ezekiel and Ayinde, 2015; Adedeji *et al.*, 2014). In addition to the geochemical background, the amount of metals in soil may be increased by human activities and atmospheric deposition. The problem of soil pollution by heavy metals has received increasing attention in the last few decades in both developing and developed countries throughout the world.

Several gold mining sites are registered in Eastern Cameroon located at Bétaré Oya, Batouri, Kette, etc. including small, medium and large scale mining. In some

of these sites, gold is still collected from soils surface in rivers. The levels of heavy metals resulting from industrial activities such as mining and mineral processing have not been evaluated in almost all the mines in Cameroon.

In the present study, soil samples were collected from a depth of 0-5 cm in the areas of Bétaré Oya and Batouri, because of mining residues discharged in the immediate environment. These residues discharged without any treatment may lead to the contamination of surface and groundwater, sediments, soils and plants.

Materials and Methods

Description and Geology of Study Areas

Bétaré Oya and Batouri are two cities located at about 176 km and 135 km respectively from Bertoua, the regional capital of East-Cameroon. The study was conducted around the gold mines of Bétaré Oya

(latitudes 5°35'00" to 5°39'00"N; longitudes 14°04'00" to 14°07'00"E) and Batouri (latitudes 4°35'00" to 4°38'00"N; longitudes 14°24'00" to 14°26'00"E) as shown in Fig. 1.

The study areas are dominated by the remains of artisanal gold mining marked by the presence of several abandoned open pits scattered in areas of ancient or recent gold mining. These pits are of various sizes that can reach 200 m² and often contain stagnant water. This situation leads to a severe disturbance of the land surface and the disappearance of the cultivable land and gallery forest. In addition oil and fuel used for the engines, the vestiges of abandoned machines, rusty empty barrels and plastic tins, production of large amounts of fined-grained material due to the digging of numerous pits in river sediments tend to increase the risk of contamination (Penaye and Hell, 2013). The study areas are undergoing intense surface gold mining since 2006.

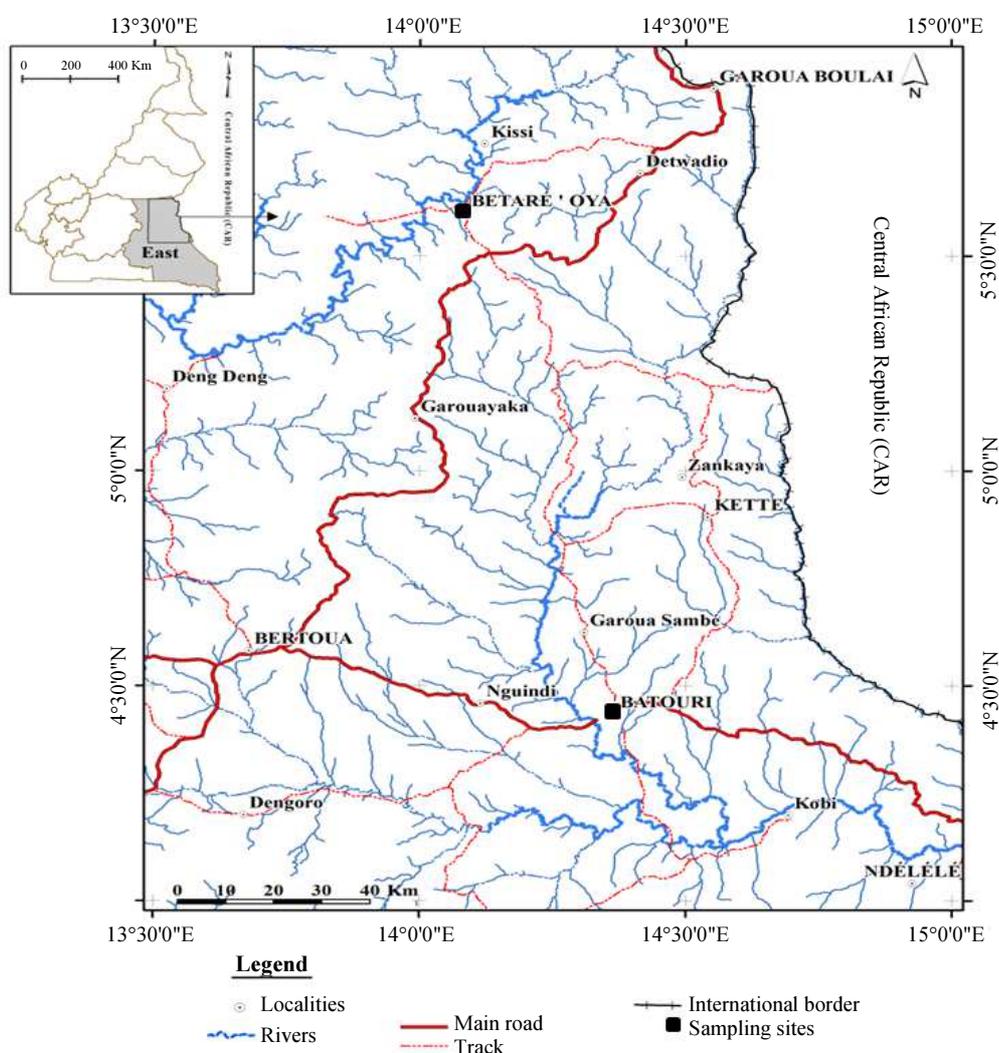


Fig. 1: Location of the gold mining areas of Batouri (Kambélé) and Bétaré Oya in Eastern Cameroon

Batouri and Bétaré Oya are respectively subject to the influence of hot and humid equatorial climate of classical Guinean type and tropical climate of sahelian Guinean type. There is a transition between the equatorial forest to the south and the savannah to the north. The climatic conditions of the study areas are (i) a rainy season from April to October, (ii) dry season from November to March and (iii) absolute low water in March. The annual average rainfall at Bétaré Oya is 1590 mm and approximately 1509 mm at Batouri (Penaye and Hell, 2013; CTFC, 2012). Most of the land is covered with sparse grass, shrubs and trees. According to the villagers, agricultural activities such as crop cultivation are in decline, while artisanal gold mining is still practiced.

The regional geology is dominated by the Neoproterozoic volcano-sedimentary rocks, metamorphosed under low-to medium-grade conditions and cross-cut by the Pan-African granitoids (Suh *et al.*, 2006; Tehna *et al.*; 2015; Yakeu *et al.*, 2016). Gold occurs as small particles disseminated within the quartz matrix and along microfractures within the veins. These veins have as much as 54 ppm gold. The quartz veins show variable textures. This pervasive variation in quartz vein textures is associated to the different generations and recrystallization events commonly exhibited by quartz crystals. Hand specimen samples for some of these quartz veins show visible gold associated with disseminated sulphides and oxides (Suh *et al.*, 2006). The quartzite show well preserved primary sedimentary structures such as current marks, indicating the flow direction, together with oblique cross stratification and load casts (Albert *et al.*, 2012). The soil in the area is predominantly acrisols, alisols, plinthosols, acid soil with clay-enriched lower horizon and low saturation of bases.

Sampling and Conditioning

Sampling was based on the proximity to potential pollution sources of mine (mining residues) due to processing, the presence of water flows and other activities (agricultural, livestock...). Thirty two (32) soil samples were collected within selected areas of the gold mining sites of Bétaré Oya and Batouri using a systematic random sampling basis because of the irregular form of the study areas. Twenty four (24) soil samples (0-5 cm) were collected in Bétaré Oya within 10 km radius from the perimeter of gold mining sites and eight (8) soil samples (0-5 cm) collected within 2 km radius from the perimeter of gold mining site of Kambele in Batouri. Each sample, with a total fresh mass of about 2 kg, was made up of a composite of material from 5 holes distributed at the

4 corners and centre of a 1 m² surface area with a maximum depth sampled of 5 cm (Dallou *et al.*, 2017). The soil samples were collected using a reusable stainless steel shovel and trowel. Each sample were stored in sealed plastic containers and carefully labeled. To avoid contamination between samples, the samplers were cleaned after each sample drawing. The soil samples were taken to the Institute of Geological and Mining Research (IRGM), Yaoundé for elemental analysis.

In the laboratory, heavy metal concentrations were determined in accordance with protocols at the Nuclear Technology Section of IRGM. Large particles were removed from the samples and the remaining sample dried in a UK Gallenkamp Hotbox Oven with fan size 3, for 2 days at 70°C. The samples were then pulverized using a German Fritsch Pulverisette with speed up to 60 rpm, sieved at about 2 mm diameter. A mixture of two grams (soil+10% of Licowax C Micropowder, PM, Hoechstwax) for each sample was weighed on a Sartorius analytical balance (TE 153S) with a precision of 0.001g, homogenized and ground in an agate mortar to obtain micron-sized diameters that were used to press pellets of 32 mm diameter under 20T using a Manual VANEON 25t Press. More details on sample preparation are given in Abdourahimi *et al.* (2016).

Analysis

Analyses of soil pellets were carried out by EDXRF Quant'X spectrometer. Spectra acquisition and quantification were performed using Wintrace 4.1. The Fundamental Parameter (FP) method was used for quantification. Combined analytical uncertainty included the calibration of EDXRF Quant'X, instability of the electronics, counting statistics and sample preparation. The unknown elements were estimated as SiO₂: 96%. The Certified Reference Materials (CRMs) NIST 2704, NIST 2709, NIST 2710, PACS 1 were used to make calibration for soil analysis (Abdourahimi *et al.*, 2016). The Z-score values recorded are given in Table 1. The heavy metals concentrations in the gold mining soil samples were subjected to Spearman's significant correlation analysis using statistical software R version 3.3.1 to determine the relationship and characteristic between the different heavy metals.

Data Processing

The results obtained were used to determine the Contamination Factor (CF), degree of contamination (Cdeg), Enrichment Factor (EF) and geoaccumulation Index (Igeo) of the metals in the studied environment.

Table 1: IAEA Worldwide Open Proficiency Test for X-Ray Fluorescence Laboratories PTXRFIAEA10, Abdourahimi *et al.* (2016)

Element	Analyte concentration (ppm)	Standard dev.	Assigned value of the analyte (ppm)	Standard dev.	Z score
Fe(g/kg)	6.064	0.254	6.100	0.0327	-0.09
Br	11.780	0.380	10.400	1.3500	0.79
Ca	0.340	0.200	0.283	0.0554	0.69
Cu	16.600	2.500	18.000	2.6300	-0.50
Mn	279.000	23.940	226.000	16.7000	2.21
Ni	5.250	0.520	8.760	1.5290	-2.31
Pb	14.600	2.240	26.300	3.4100	-3.03
Rb	52.810	0.390	41.800	2.2200	1.93
Sb	0.730	0.510	0.638	0.0858	0.56
Sr	65.980	1.610	53.700	3.8700	2.60
Y	11.330	0.510	10.800	2.3000	0.44
Zn	45.450	2.640	47.800	5.3100	-0.55
Zr	384.850	11.080	387.000	65.8000	-0.06

Contamination Factor and Degree of Contamination

To assess the level of heavy metals contamination in gold mining sites and surroundings, CF and Cdeg were calculated using the method presented by Rastmanesh *et al.* (2010). This method is commonly used by numerous researchers as an indicator of contamination in sediments and soils (Ghadimi, 2014; Banu *et al.*, 2013; Iqbal *et al.*, 2013; Abhijit and Ramkrishna, 2017). CF is determined by using the following Equation 1:

$$C^x F = \frac{C_o^x}{C_n^x} \quad (1)$$

where, $C^x F$ represent the contamination factor of the element of interest, C_o^x is the concentration of the element in the sample, C_n^x is the background concentration. In the present study the average Upper Continental Crust (UCC) was used (Taylor and McLennan, 1985; Esser and Turekian, 1993; Jochum *et al.*, 1993). $C^x F$ is defined according to four categories given as follows: <1 = low contamination factor, 1-3 = moderate contamination factors, 3-6 = considerable contamination factors and > 6 = very high contamination factor (Rastmanesh *et al.*, 2010; Likuku *et al.*, 2013; Sadhu *et al.*, 2012).

The degree of contamination (Cdeg) is the sum of CFs. It allows the assessment of the polymetallic contamination for each sampling site (Halim *et al.*, 2014; Tariq *et al.*, 2018). It is calculated according to the following Equation 2:

$$C_{deg} = \sum_{x=1}^N C^x F \quad (2)$$

Four categories have been defined for Cdeg as follows: <8 = low degree of contamination, 8-16 = moderate degree of contamination, 16-32 = considerable

degree of contamination and >32 = very high degree of contamination (Rastmanesh *et al.*, 2010).

Pollution Load Index

The Pollution Load Index (PLI) is used to comprehensively evaluate the pollution effect of study metals and for each site, it is defined as follows (Caeiro *et al.*, 2015; Feifei *et al.*, 2015):

$$PLI = \sqrt[n]{CF_1 \times CF_2 \times CF_3 \dots \times CF_n} \quad (3)$$

where, n is the number of metals studied and CF is the contamination factor calculated as described in an earlier equation. The PLI value greater than 1 is polluted, while less than 1 indicates no pollution (Tariq *et al.*, 2018; Khan *et al.*, 2017; Krika and Krika, 2017).

Enrichment Factor

EF of an element is based on normalizing a measured element against a reference element. It enables to differentiate heavy metals originating from anthropogenic sources and those of natural sources (Emmanuel *et al.*, 2014; Salati and Moore, 2010). The enrichment factor was calculated using the formula used by number of authors (Abhijit and Ramkrishna, 2017; Tariq *et al.*, 2018; Smuc *et al.*, 2015; Zahran *et al.*, 2015; Jiao *et al.*, 2015) and given by the following Equation 4:

$$EF = \frac{(C_n / C_{ref})_{sample}}{(B_n / B_{ref})} \quad (4)$$

where, C_n (sample) is the concentration of the examined element in the environment, C_{ref} (sample) is the concentration of the examined element in the reference environment, B_n is the concentration of the reference element in the environment and B_{ref} is the concentration of the reference element in the reference environment. Iron (Fe) was used as the reference element in the crust

(Abhijit and Ramkrishna, 2017; Taylor and McLennan, 1985). Five contamination categories were recognized on the basis of the enrichment factor. They are given by: $EF < 2$ = minimal enrichment, $EF = 2-5$ moderate enrichment, $EF = 5-20$ significant enrichment, $EF = 20-40$ very high enrichment and $EF > 40$ extremely high enrichment (Emmanuel *et al.*, 2014; Yongming *et al.*, 2016).

Geoaccumulation Index

The Geoaccumulation Index (Igeo), was introduced by Müller (1969) to assess the level of metal accumulation in the sediments and has been used by several researchers for various studies (Abhijit and Ramkrishna, 2017; Zahra *et al.*, 2014; Nowrouzi and Pourkhabbaz, 2014). It is mathematically expressed as follows:

$$I_{geo} = \log_2 \left(\frac{C_n}{1.5 \times B_n} \right) \quad (5)$$

where, C_n represents the measured concentration of heavy metal in the soil, B_n is the geochemical background concentration of the heavy metal. The factor 1.5 is incorporated in the relationship to account for possible variation in background data due to lithogenic effect. The geoaccumulation index (Igeo) scale consists of seven grades ranging from unpolluted to highly polluted as given below (Emmanuel *et al.*, 2014; Müller, 1969; Kabir *et al.*, 2011):

< 0 = practically unpolluted, $0-1$ = unpolluted to moderately polluted, $1-2$ = moderately polluted, $2-3$ = moderately to strongly polluted, $3-4$ = strongly polluted, $4-5$ = strongly to extremely polluted and > 5 = extremely polluted.

Results and Discussion

Heavy Metals Concentrations

The mean concentrations of elements in soil samples collected in gold mining sites of Bétaré Oya and Batouri decreases as: Fe > Mn > Zr > Pb > Rb > Zn > Sr > Ba > Y > Cu > Ga > Ni > As > Br > Sn.

High concentrations of iron (Fe) have been observed in soil samples collected from sites of Bétaré Oya. They range from 28131 to 55780 ppm with a mean of 43179 ± 315 ppm at Bétaré Oya (Table 2) and from 28328 to 30777 ppm with a mean 29552 ± 204 ppm at Batouri (Table 3). Toxic effects related to iron can appear if concentrations in the human body are higher than the critical value (50000 ppm) indicated by the WHO (World Health Organization), affecting negatively several organs particularly in children (Baby *et al.*, 2010; Chiroma *et al.*, 2014). The highest concentration (55780 ppm) of Fe recorded in BO3 site located behind the

Public School of Bétaré Oya is slightly higher than the critical value indicated above. High concentrations of Manganese (Mn) compared to the worldwide average Upper Continental Crust (UCC) in some sites were measured. These concentrations range from 498.8 to 1169.3 ppm with a mean of 840 ± 44 ppm at Bétaré Oya and from 402.3 to 476.1 ppm with a mean 439 ± 25 at Batouri. However, these concentrations are lower than critical value (2000 ppm) authorized by the WHO (1996) in all sites (Table 2). ATSDR (2012) and, more recently, Lucchini *et al.* (2015) reviewed the toxicity of manganese. These studies showed that exposure to concentrations higher than this value can cause toxic effects primarily on the brain and central nervous system. The mean concentrations of Fe and Mn in this study area exceeded concentrations reported in similar studies on heavy metals contamination associated with gold mining in Sudan (Mushtaha *et al.*, 2017) and Ghana (Crentsil and Anthony, 2016). The concentrations of lead (Pb) ranged from 28.8 to 293.4 ppm with a mean of 105 ± 17 ppm at Bétaré Oya and from 0.0 to 26.7 ppm with a mean value of 13.4 ± 5 at Batouri. In most sites they exceeded worldwide average value, but are lower than the critical value (100 ppm) authorized by the WHO (1996), excepted mining site located behind the Government High School of Bétaré Oya (BO4) (Table 2). Pb presents a variety of dangers because the Pb accumulated in the human body affects negatively several organs: bones, liver, kidney, brain, lung and central nervous system (Castro-Gonzalez and Mendez-Armenta, 2008). Although toxicity of lead from industrial settings has been relatively controlled, it remains a pervasive toxicant worldwide (Arif *et al.*, 2015; Azizi and Azizi, 2010). In comparison with other similar studies, the mean concentration of Pb in soil of study area was significantly higher than the concentrations reported in Sudan, Ghana and South Africa, respectively by Mushtaha *et al.* (2017), Crentsil and Anthony (2016) and Caspah *et al.* (2016), in addition, this concentration was also higher than obtained in the studies carried out by Feifei *et al.* (2015) and Sijin *et al.* (2015), respectively around a gold mine and Zinc-Lead mining area in China (Table 4). Concentrations of Arsenic (As) range from 0.0 to 16.2 ppm with a mean value of 4.6 ± 4 ppm (Table 2) at Bétaré Oya and from 0.0 to 5.6 ppm with a mean value of 3 ± 2.4 at Batouri (Table 3). They exceeded the worldwide average value in some sites. The highest concentration of As was recorded in a mining site abandoned of Bétaré Oya (BO5) while in sites BO3, BO4 and BA2 (Narké crossroads, Batouri), they were below the detection limit. This high value in BO5 site could be explained by the presence of vestiges abandoned machines and mineral residues discharged in the pits scattered. Despite being the 20th most abundant element in the earth's crust, As ranks highest on the list of hazardous substances toxic to

public health (Arif *et al.*, 2015). Inorganic arsenic is considered as a human carcinogen (cancer-causing agent) by the U.S. and exposure to arsenic has been linked to increased incidence of irritation of mucous membranes and lung cancer (WHO, 1996; Qian *et al.*, 2014; Mo *et al.*, 2016). In general, As concentrations in all the sites are lower than normal value (20 ppm) authorized by the WHO. Concentrations of copper (Cu) ranged from 13.1 to 28.5 ppm with a mean of 21 ± 7 ppm (Table 2) at Bétaré Oya while in sites in sites BA1 et BA2 of Batouri, they were below the detection limit. In humans Cu is necessary for their development, but may be toxic when its concentration exceeds the reference value (100 ppm) given by the WHO for soils (Chiroma *et al.*, 2014; Ran *et al.*, 2017; Sabah *et al.*, 2012)). The observed concentration of Cu did exceed the earth crust threshold value prescribed in soil for BO3 site, but not for others. The concentrations of zinc (Zn) ranged from 20.3 to 87.3 ppm with a mean of 45 ± 10 ppm (Table 2) at Bétaré Oya and from 24.6 to 29.3 ppm with a mean value of 27 ± 6 at Batouri (Table 3). The observed concentrations of Zn did not exceed the worldwide average value prescribed in soil except for BO5 site of Bétaré Oya. Copper (Cu) and zinc (Zn) provide clear examples, both being essential for normal metabolism and both can be toxic in high concentrations (Verdejo *et al.*, 2016; USEPA, 2013). When the quantities of Cu and Zn in the body increase they become toxic, which can result in damaged and malfunctioning human organs (USEPA, 2013; Bost *et al.*, 2016; Fraga, 2005; Tobias *et al.*, 2013). The level of Zn in this study area was higher than the concentrations found in studies on soils around gold mine in China (Feifei *et al.*, 2015) and Sudan (Mushtaha *et al.*, 2017) but lower than Zn concentration (112.3 ppm) reported in Zinc-Lead mining area in China (Sijin *et al.*, 2015).

Concentrations of Zr ranged from 128.5 to 182.1 ppm with a mean of 149 ± 18 ppm (Table 2) at Bétaré Oya, below the average value in the upper continental crust (190 ppm) while at Batouri, they were ranged from 616.7 to 832.7 ppm with a mean of 724.7 ± 32 ppm (Table 3) above the average value in the upper continental crust. The highest concentrations of Zr were recorded in Batouri (BA1 and BA2) sites. In other sites concentrations did not exceed the average in the earth crust. In soil, Zr is more than twice as abundant as copper and zinc and has ten times more abundance than lead (Shahid *et al.*, 2013). Its geochemistry is dominated by its lithophilic nature. Although data are scarce, it appears that Zr have low toxicity to organisms (Shahid *et al.*, 2013). Concentrations of gallium (Ga) ranged from 5.8 to 18.1 ppm with a mean of 10.8 ± 5 ppm at Bétaré Oya and from 8.1 to 17.5 ppm with a mean value of 12.8 ± 4.3 ppm at Batouri. Observed concentrations of Ga did not exceed the worldwide average value prescribed in soil in most sites except for BO5 and BA1 (Kambélé, METALICOM Company) sites. Gallium causes increased unbound Fe owing to its ability to substitute for Fe in numerous moieties involved in vital cellular functioning (Auger *et al.*, 2013; Bignucolo *et al.*, 2013). It is known to be toxic at high concentrations (Sujeenthara *et al.*, 2017). The concentrations of yttrium (Y) ranged from 15.8 to 26.3 ppm with a mean of 20 ± 7 ppm at Bétaré Oya and from 11.2 to 15.5 ppm with a mean value of 13.4 ± 4 ppm at Batouri. The highest concentration of Y was recorded in Ali-Bachir mining site located at Bétaré Oya (BO1). In other sites, concentrations did not exceed the reference value considered in the soil. As a heavy metal, yttrium is not entirely free of toxicity. It can be a threat to the liver when its concentration exceeds the safe limits in the human body (He, 2013).

Table 2: Summary statistics of heavy metals (ppm) concentrations in gold mining sites of Bétaré Oya (n = 24)

Elements	Minimum	Maximum	Mean	Std. Deviation	Worldwide average*	Maximum permissible Level in soil
Mn	498.9	1169.3	840.0	44.0	600.0	2000 ^a
Fe	28131.0	55780.0	43179.0	315.0	35000.0	50000 ^a
Ni	6.8	10.1	9.0	4.5	20.0	50 ^a , 40 ^b
Ba	-	45.2	17.1	7.3	550.0	N.A
Cu	13.1	28.5	21.0	7.0	25.0	100 ^a , 30 ^b
Zn	20.3	87.3	45.0	10.0	71.0	300 ^a
Ga	5.8	18.1	10.8	5.0	17.0	N.A
As	-	16.2	4.6	4.0	1.5	20
Br	-	2.2	1.3	1.1	N.A	N.A
Rb	34.2	61.4	46.8	10.0	112.0	N.A
Sr	23.1	54.5	33.3	9.0	350.0	N.A
Y	15.8	26.3	20.0	7.0	22.0	N.A
Zr	128.5	182.1	149.0	18.0	190.0	N.A
Pb	28.8	293.4	105.0	17.0	20.0	100 ^{ab}
Sn	-	0.5	0.1	0.1	5.5	N.A

N.A = not available, * worldwide average upper continental crust (Taylor and McLennan, 1985; Esser and Turekian, 1993; Jochum *et al.*, 1993), ^a Chiroma *et al.* (2014) and ^b Tobias *et al.* (2013): The maximum acceptable limits of heavy metals in soils established by standard regulatory bodies such as World Health Organization (WHO), Food and Agricultural Organization (FAO) and the European Union.

Table 3: Summary statistics of heavy metals (ppm) concentrations in gold mining sites of Batouri (n = 8)

Elements	Minimum	Maximum	Mean	Std. Deviation	Worldwide average*	Maximum permissible Level in soil
Mn	402.3	476.1	439	25	600	2000 ^a
Fe	28328	30777	29552	204	35000	50000 ^a
Ni	6	7.2	6.6	3	20	50 ^a , 40 ^b
Ba	5.0	64.3	34.7	8	550	N.A
Cu	-	-	-	-	25	100 ^a , 30 ^b
Zn	24.6	29.3	27	6	71	300 ^a
Ga	8.1	17.5	12.8	4.3	17	N.A
As	-	5.9	3	2.4	1.5	20
Br	2.3	3.3	2.8	2	N.A	N.A
Rb	23.4	27.2	25.3	6	112	N.A
Sr	36.8	56.8	46.8	8	350	N.A
Y	11.2	15.5	13.4	4	22	N.A
Zr	616.7	832.7	724.7	32	190	N.A
Pb	-	26.7	13.4	5	20	100 ^{a,b}
Sn	1.1	1.9	1.5	1.4	5.5	N.A

Table 4: Comparison of average heavy metal concentrations with those of similar studies carried out in other countries

Country	Mean concentration (ppm)											Reference
	Hg	As	Cd	Pb	Zn	Cu	Ni	Co	Cr	Fe	Mn	
China	0.17± 0.064	7.11± 1.32	0.33± 0.023	28.3± 4.6	39.7± 3.5	12.9± 2.1	-	-	-	-	-	Feifei <i>et al.</i> (2015)
China	0.12± 0.09	10.6± 6.50	0.24± 0.29	39.2± 19.5	112.3± 70.6	30±22	28.4± 13.8	-	59.8± 25.6	-	-	Sijin <i>et al.</i> (2015)
China	1.62± 2.24	6.90± 3.10	0.65± 0.71	109± 145.1	155± 63.7	58.30± 21.20	54.20± 15.20	20.17± 5.60	189.9± 73.7	-	-	Qian <i>et al.</i> (2014)
China	0.12± 0.12	8.57± 6.07	0.15± 0.06	819.7± 1126	104.2± 44.8	46.92± 21.03	28.40± 9.43	-	88.61± 47	-	-	Mo <i>et al.</i> (2016)
South Africa	0.09	79.40	0.05	4.79	51.30	42.51	112.06	25.56	278.8	-	-	Caspah <i>et al.</i> (2016)
Ghana	1.06± 0.65	6.83± 2.42	0.19± 0.13	19.96± 3.11	61.87± 16.5	16.03± 3.22	41.77± 13.60	-	16.88± 2.47	531.5± 97	445.6± 108	Crentsil and Anthony (2016)
China	2.91	16	2.45	252	286	46.4	-	-	-	-	-	Ran <i>et al.</i> (2017)
Sudan	-	-	-	12.46	20.85	19	15.33	5.2	-	8473	238.1	Mushtaha <i>et al.</i> (2017)
Oman	-	-	5.33	96.82	964	3240	282	101.2	-	14108	2865	Sabah <i>et al.</i> (2012)
Cameroon	-	4±2	-	79±9	40±8	15±7	8±4	-	-	39300± 200	730± 70	Present study

Concentrations of nickel (Ni), Tin (Sn), rubidium (Rb), strontium (Sr), barium (Ba) and bromine (Br) are low in the study area (Table 2 and 3) and do not exceed the earth crust threshold values prescribed in soil. The mean concentrations of arsenic (As) and nickel (Ni) in soil in this study area were slightly lower than the concentrations in the other countries (Table 4). Mercury (Hg) and cadmium (Cd) were below the detection limit in the all samples.

Contamination Factor, Degree of Contamination and Pollution Load Index of Heavy Metals in Soils

Results presented in Table 5 show that mean contamination factors of heavy metals follow a decreasing order: Pb>As>Zr>Mn>Fe>Y>Ga>Cu>Zn>Ni>Rb>Sr>Sn>Ba. Overall, the present study shows that the highest mean

value of contamination factor observed is 3.95 for Pb, i.e., a considerable contamination while contamination factors of heavy metals such as As, Zr, Mn and Fe were considered as moderates. Contamination factor for elements such as Y, Ga, Cu, Zn, Ni, Rb, Sr, Sn and Ba is less than 1 indicating a low contamination from anthropogenic sources (Likuku *et al.*, 2013).

The degrees of contamination of soil samples from various sites are presented in Fig. 2. Generally, values obtained for the degrees of contamination were higher than 8, meaning that samples from the sites have moderate degrees of contamination and considerable degrees of contamination for the BO5 and BO4 sites except the BA2 site of Batouri which presents a low degree of contamination as shown in Table 5. Mean degrees of contamination of all heavy metals compared to their respective degree of contamination reveal a

moderate degree of contamination, indicating a moderate anthropogenic pollution (Abhijit and Ramkrishna, 2017). Comparison between degrees of contamination of gold mining sites shows a decreasing order as follows: BO5>BO4>BA1>BO3>BO1>BO2>BA2.

A similar result can also be obtained by examining the spatial variations in the PLI, by calculating the values of soils in the study area, in particular for both sites BO4 (n = 11) and BO5 (n = 14) that are 0.78 and 0.67, respectively. These values were found to be low in all the studied samples and varied between 0.43 and 0.78, indicating that the studied sites in Bétaré Oya and Batouri are in low pollution status regarding the total of the studied metals (Mmolawa *et al.*, 2011). The soil of

Bétaré Oya (BO) indicate the higher degrees of contamination values, which mean that pollution caused by heavy metals is higher than that of Batouri (BA). The conclusion is consistent with the PLI.

Enrichment Factor of Heavy Metals in Soil

Enrichment factors of heavy metals presented in Fig. 3 decrease as follows: Pb>As>Zr>Mn>Fe>Y>Ga>Zn>Cu>Ni>Rb>Sr>Sn>Ba. Determining enrichment factor (EF) is an essential part of geochemical studies and is generally used to differentiate between metals originating from anthropogenic and geogenic sources and to assess the degree of metal contamination (Moore *et al.*, 2009).

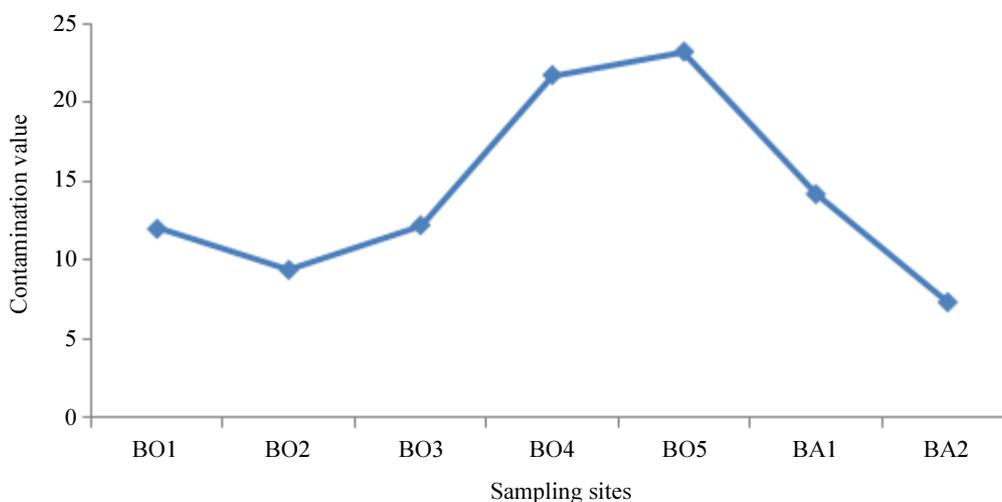


Fig. 2: Degree of Contamination of soil sampled from gold mining areas of Bétaré Oya and Batouri (BO = Bétaré Oya, BA = Batouri)

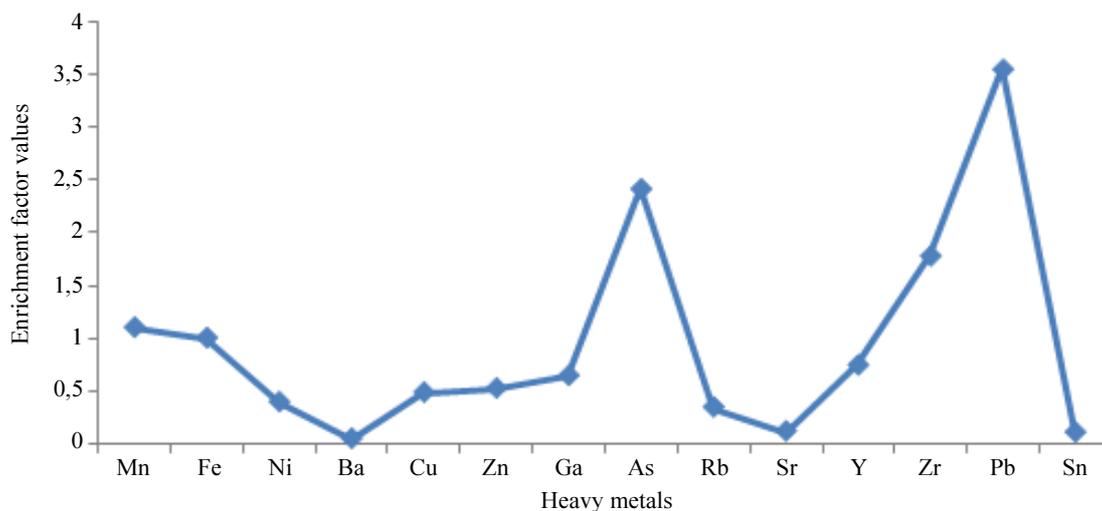


Fig. 3: Enrichment factor of soil sampled from gold mining areas of Bétaré Oya and Batouri

Table 5: Contamination Factors (CF) and degree of contamination (Cdeg) of soil from gold mining areas of Bétaré Oya (BO) and Batouri (BA)

Sampling site	Contamination Factor (CF)														Contamination Degree
	Mn	Fe	Ni	Ba	Cu	Zn	Ga	As	Rb	Sr	Y	Zr	Pb	Sn	
BO1	0.83	1.42	0.46	0.04	0.81	0.58	0.34	3.40	0.46	0.07	1.20	0.96	1.44	0.00	12.02
BO2	1.12	0.80	0.51	0.03	0.53	0.55	0.60	1.12	0.37	0.16	0.74	0.83	2.08	0.00	9.42
BO3	1.95	1.59	0.45	0.00	1.14	0.29	0.73	0.00	0.31	0.08	0.88	0.70	4.13	0.00	12.25
BO4	1.83	1.04	0.50	0.00	0.71	0.54	0.44	0.00	0.55	0.11	0.72	0.68	14.67	0.00	21.79
BO5	1.26	1.31	0.34	0.08	0.97	1.23	1.06	10.80	0.40	0.07	0.95	0.76	3.97	0.08	23.28
Mean BO	1.40	1.23	0.45	0.03	0.83	0.64	0.63	3.06	0.42	0.10	0.90	0.79	5.26	0.02	15.76
BA1	0.79	0.81	0.30	0.01	0.00	0.41	1.03	3.94	0.21	0.11	0.70	4.38	1.33	0.20	14.23
BA2	0.67	0.88	0.36	0.12	0.00	0.35	0.48	0.00	0.24	0.16	0.51	3.25	0.00	0.35	7.36
Mean BA	0.73	0.85	0.33	0.07	0.00	0.38	0.76	1.97	0.23	0.14	0.61	3.82	0.67	0.28	10.84
Mean values	1.21	1.12	0.42	0.04	0.59	0.56	0.67	2.75	0.36	0.11	0.81	1.65	3.95	0.09	14.34

BO2 site is located in town centre of Bétaré Oya

Table 6: Enrichment factor of heavy metals from gold mining areas of Bétaré Oya (BO) and Batouri (BA)

Sampling site	Enrichment Factor (EF)														
	Mn	Fe	Ni	Ba	Cu	Zn	Ga	As	Rb	Sr	Y	Zr	Pb	Sn	
BO1	0.59	1.00	0.32	0.03	0.57	0.41	0.24	2.39	0.33	0.05	0.84	0.67	1.01	0.00	
BO2	1.40	1.00	0.63	0.04	0.65	0.68	0.74	1.39	0.46	0.19	0.92	1.03	2.58	0.00	
BO3	1.22	1.00	0.28	0.00	0.71	0.18	0.46	0.00	0.19	0.05	0.55	0.44	2.59	0.00	
BO4	1.76	1.00	0.48	0.00	0.68	0.52	0.42	0.00	0.53	0.11	0.69	0.65	14.05	0.00	
BO5	0.97	1.00	0.26	0.06	0.74	0.94	0.81	8.27	0.31	0.05	0.73	0.58	3.04	0.06	
Mean BO	1.19	1.00	0.39	0.03	0.67	0.55	0.53	2.41	0.36	0.09	0.75	0.67	4.65	0.01	
BA1	0.98	1.00	0.37	0.01	0.00	0.51	1.27	4.87	0.26	0.13	0.87	5.42	1.65	0.25	
BA2	0.76	1.00	0.41	0.13	0.00	0.39	0.54	0.00	0.28	0.18	0.58	3.69	0.00	0.40	
Mean BA	0.87	1.00	0.39	0.07	0.00	0.45	0.91	2.44	0.27	0.16	0.71	4.56	0.83	0.33	
Mean values	1.10	1.00	0.39	0.04	0.48	0.52	0.64	2.42	0.34	0.11	0.74	1.78	3.56	0.10	

Table 6 shows that enrichment factors for Mn, Fe, Y, Ga, Zn, Cu, Ni, Rb, Sr, Sn and Ba vary between 0 and 2, indicating the minimal enrichment in soil collected in Bétaré Oya and Batouri. For Pb and As, mean enrichment factor values vary between 2 and 5, indicating a moderate enrichment in soil (Zahran *et al.*, 2015). Sites BO4 and BO5 of Bétaré Oya present respectively Pb and As values ranging between 5 and 20, meaning that Pb and As are the main elements of anthropogenic loading into the soil of Bétaré Oya. In BA1 site, enrichment factors of Zr range between 5 and 20, testifying that Zr is the main element of anthropogenic loading into the soil of Batouri.

Geoaccumulation Index of Heavy Metals in Soils

The degree of soils pollution can be assessed by the determination of indices such as Geoaccumulation Index (Igeo) (Nowrouzi and Pourkhabbaz, 2014). The calculated Igeo values for different samples are summarized in Table 7.

As displayed in Fig. 4, Igeo values for Cu, Y, Ga, Zn, Ni, Rb, Sn, Sr and Ba fall mainly in unpolluted class (≤ 0). The Igeo values for Pb shows that 33.33% of the sites in the all study area belong to the unpolluted class (≤ 0), 16.67% in the unpolluted to moderately polluted class (0-1), 33% in the moderately polluted class (1-2), while the remaining 16.67% are strongly polluted class (3-4). For As, 25% of the sites are in the unpolluted

class, 50% in the moderately polluted class and 25% in the moderate to strongly polluted class. For Zr, 71.43% of the sites are included in the unpolluted class and the remaining 28.57% find themselves in the moderately polluted class. For Fe, sites mainly fall in the unpolluted class (85.71% of total sites) and the remaining 14.29% in the unpolluted to moderately polluted class. Finally for Mn, 71.43% of the sites are included in the unpolluted class and the remaining 28.57% belong to the unpolluted to moderately polluted class. The results in Table 7 indicate that soil samples collected in Bétaré Oya locality can be categorized as follows: Unpolluted with Cu, Y, Ga, Zn, Ni, Rb, Sn, Sr, Ba, Fe, Mn and Zr (mean $I_{geo} < 0$) and moderately polluted with As and Pb ($1 < \text{mean } I_{geo} < 2$) (Moore *et al.*, 2009), while these of Batouri locality can be categorized as follows: unpolluted with Cu, Y, Ga, Zn, Ni, Rb, Sn, Sr, Ba, Fe, Mn and Pb (mean $I_{geo} < 0$) and moderately polluted with As and Zr ($1 < \text{mean } I_{geo} < 2$).

Elements Correlation Matrix

Spearman correlation analysis was performed between various elements (with a level of significance: $p \leq 0.01$ and $p \leq 0.05$) for soil samples from various sites of Bétaré Oya and Batouri. The obtained results are given in Table 8. The study pointed out strong positive correlations between some elements such as Cu and Fe (0.84), Pb and Mn (0.96) at 1% significant levels.

Table 7: Geoaccumulation index of heavy metals in soil from gold mining sites of Bétaré Oya (BO) and Batouri (BA)

Geoaccumulation index (Igeo)														
Sampling site	Mn	Fe	Ni	Ba	Cu	Zn	Ga	As	Rb	Sr	Y	Zr	Pb	Sn
BO1	-0.85	-0.08	-1.71	-5.07	-0.89	-1.36	-2.14	1.18	-1.69	-4.51	-0.33	-0.65	-0.06	n.d
BO2	-0.42	-0.90	-1.57	-5.72	-1.51	-1.45	-1.33	-0.43	-2.01	-3.27	-1.02	-0.85	0.47	n.d
BO3	0.38	0.09	-1.74	n.d	-0.40	-2.39	-1.04	n.d	-2.30	-4.27	-0.76	-1.09	1.46	n.d
BO4	0.29	-0.52	-1.57	n.d	-1.09	-1.47	-1.78	n.d	-1.45	-3.76	-1.06	-1.15	3.29	n.d
BO5	-0.25	-0.20	-2.15	-4.19	-0.64	-0.29	-0.50	2.85	-1.91	-4.51	-0.66	-0.98	1.40	-4.19
Mean BO	-0.17	-0.32	-1.71	-4.99	-0.91	-1.39	-1.36	1.20	-1.87	-4.06	-0.77	-0.94	1.31	-4.19
BA1	-0.92	-0.89	-2.33	-7.35	n.d	-1.86	-0.54	1.39	-2.84	-3.83	-1.09	1.55	-0.17	-2.89
BA2	-1.16	-0.77	-2.06	-3.68	n.d	-2.11	-1.65	n.d	-2.63	-3.21	-1.55	1.11	n.d	-2.09
Mean BA	-1.04	-0.83	-2.20	-5.52	n.d	-1.99	-1.10	1.39	-2.74	-3.52	-1.32	1.33	-0.17	-2.49
Mean values	-0.42	-0.47	-1.88	-5.20	-0.91	-1.56	-1.28	1.25	-2.12	-3.91	-0.92	-0.29	1.07	-3.06

n.d = mean not defined

Table 8: Spearman correlation matrix of heavy metals in soils of Bétaré Oya and Batouri

	Mn	Fe	Ni	Ba	Cu	Zn	Ga	As	Br	Rb	Sr	Y	Zr	Pb	Sn
Mn	1.00														
Fe	0.53	1.00													
Ni	0.39	-0.01	1.00												
Ba	-0.66	-0.16	-0.33	1.00											
Cu	0.81*	0.84**	0.16	-0.27	1.00										
Zn	0.00	-0.03	0.12	0.37	0.19	1.00									
Ga	0.14	-0.10	-0.68	0.01	0.12	0.00	1.00								
As	-0.22	-0.11	-0.52	0.33	0.03	0.70*	0.51	1.00							
Br	-0.67	-0.53	-0.46	0.19	-0.66	-0.5	0.25	0.03	1.00						
Rb	0.5	0.35	0.66	-0.14	0.48	0.60	-0.50	-0.03	-0.92**	1.00					
Sr	-0.36	-0.70	0.25	0.05	-0.72*	-0.48	-0.21	-0.59	0.45	-0.34	1.00				
Y	0.46	0.64	0.19	0.00	0.81*	0.60	-0.03	0.40	-0.57	0.57	-0.84**	1.00			
Zr	-0.92**	-0.46	-0.55	0.52	-0.70*	-0.07	0.07	0.37	0.82*	-0.67	0.18	-0.35	1.00		
Pb	0.96**	0.42	0.48	-0.66	0.70*	0.10	0.03	-0.22	-0.78*	0.64	-0.28	0.39	-0.96**	1.00	
Sn	-0.70*	-0.35	-0.79*	0.59	-0.59	-0.19	0.39	0.22	0.57	-0.66	0.28	-0.61	0.66	-0.70*	1

**Correlation significant at the 0,01 level (bilateral); *correlation significant at the 0,05 level (bilateral)

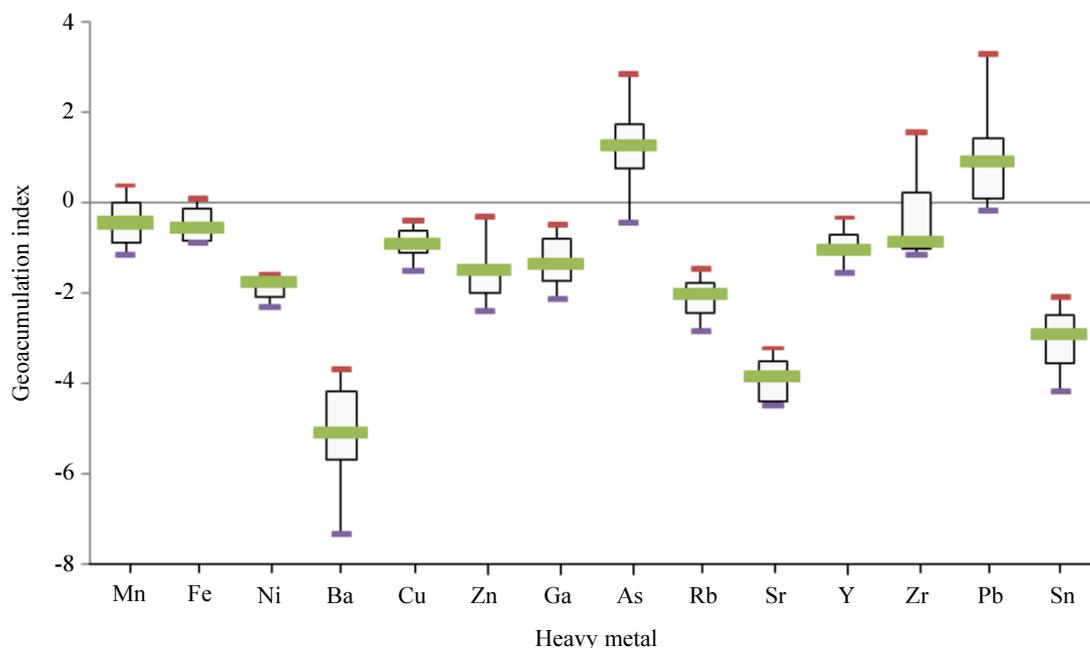


Fig. 4: Geoaccumulation index of heavy metals in soil samples of Bétaré Oya and Batouri

However, it shows a strong positive correlation between Cu/Mn (0.81), Y/Cu (0.81), Zr/Br (0.82), Pb/Cu (0.70) and As/Zn (0.70) at 5% significant levels. While Rb/Br, Sr/Cu, Y/Sr, Zr/Mn, Zr/Cu, Pb/Br, Pb/Zr, Sn/Ni and Sn/Pb (Table 8) show a strong negative correlation.

The strong positive correlations between some elements signify that these elements come from the same activities and are probably governed by the same physicochemical processes (Emmanuel *et al.*, 2014; Bai *et al.*, 2011; Otari and Dabiri, 2015). Among the common anthropogenic sources, there are oil and fuel used for engines, the vestiges of abandoned machines, rusty empty barrels and plastic tins since these sources are noted to be contributing one or the pair correlated metals to the natural environment. However, their contribution is moderate.

Conclusion

The level of pollution in soils by heavy metals in the gold mining sites of Bétaré Oya and Batouri was evaluated using enrichment factors, contamination factors, degrees of contamination, pollution load index and geoaccumulation index. The study revealed that there were high concentrations of certain metals (As, Pb, Mn, Zr and Fe) in terms of enrichment of the soil from some sites. For instance, the enrichment factor of lead (Pb) found for the site (BO4) in Betaré Oya indicates a significant enrichment and the geoaccumulation index for Pb in soil shows that this site is strongly polluted. With regard to the overall contamination of soils at the studied sites, the analysis indicates that contamination by the metals originated from anthropogenic sources. Based on the findings of this study, it is urgent to regulate mining activities in Cameroon to prevent environmental pollution, protecting people against harmful effects of toxic elements. For eliminating unacceptable health hazards from these metals and ensuring public and environmental safety; this study suggests the phytoremediation or the biogeosystem method (Kalinitchenko *et al.*, 2016) as technics of regulation. The advantage of the biogeosystem method is to provide a long-term stable high productive soil evolution and environmentally safe waste recycling.

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Author's Contributions

Dallou Guy Blanchard: Participated in all experiments, coordinated the data-analysis and contributed to the writing of the manuscript.

Ngoa Engola Louis: Participated in experimental planning and data-analysis interpretation.

Abdourahimi: Participated in all experiments, coordinated the data-analysis and helped in manuscript writing.

Bongue Daniel: Participated in experimental planning and data-analysis interpretation.

Saïdou: Coordinated all experimental planning and data interpretation and contributed to the writing of the manuscript.

Ndjana Nkoulou II Joseph Emmanuel: Participated in experimental planning and data-analysis interpretation.

Kankeu Boniface: Participated in experimental planning.

Kwato Njock Moïse Godfroy: Participated in experimental planning and critical data-analysis.

Ethics

Authors declare no conflict of interest.

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