

Monitoring the Anthropogenic and Geochemical Environment Surrounding the Butana Drinking Water Sources via the Determination of Heavy Metals Composition of the Soil, Streams Sediments and Gold Mining Tailings (II)

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Abstract:

Based on the results obtained in our previous research assessment regarding the quality status of the Butana drinking water, the present investigation is aimed at monitoring the anthropogenic and geochemical environment surrounding the sites of collection in the Dry Season Duration. To achieve these objectives the heavy metal composition of the soil, stream sediment and gold mining tailings be determined and some geochemical indices and factors must be calculated. Random sampling procedure was adopted in 11 sites inside and outside the mining domain covering an area of 16897.1 Km². The collection compilation of three categories of samples included 31 soil samples, 24 stream sediments and 14 gold mining tailings during the Dry Season of the year. Flame Atomic Absorption Spectrometer Savant AA 5th generation from GBC was adopted to determine the concentration levels of selected heavy metal: (Ni, Cr, Co, Zn, Cu, Pb, Cd, Mn, Fe and Hg) in the three sampled categories. The average concentration levels in the soil samples were compared with those average concentrations reported in most World Soils. Geochemical parameters such as Enrichment Factor (EF), modified degree of contamination, pollution load index and geoaccumulation index were calculated. Results obtained have shown remarkable variability in the concentrations of heavy metals during the Dry season: The highest concentrations of heavy metals were obtained at the center of mining area particularly inside the mining basins of gold extraction. It could be concluded that the distinctive high levels of mercury, lead and cadmium present an unambiguous risks for the contamination of the drinking water with these hazardous heavy metals in the future as the mining activities within this region would expected to intensified to more than two folds from its present level. The results also indicated that the soil samples which are collected from inside mining basins have a highest enrichment factor for most heavy metals, emphasizing that lead and mercury may surely be derived from anthropogenic source. It is recommended regular monitoring of heavy metals in the soils around the Artisanal gold Mining for conservation and protection from pollution. Gold mining tailings should need more attention and management from the local authorities as they are the sources of the hazardous heavy metals particularly mercury drinking water pollution.

Keywords: Butana Drinking Water, Geochemical Environment, Heavy Metals, Stream Sediment, Tailings, Gold Mining, Anthropogenic.

Introduction

Mining has been identified as one of the human activities which can have a negative impact on the quality of the environment [1] due to activities such as mineral excavation, ore transportation, smelting and refining, and disposal of the tailings and waste waters around mines [2,3]. It causes the destruction of natural ecosystems through removal soil and vegetation and burial beneath waste disposal sites funeral [4], and adverse environmental impacts from excessive heavy metals dispersed from mine and smelter sites include contamination of water and soil, phytotoxicity and potential risks to human health [5, 6, 7]. Mining waste can be divided into mine tailings, generated during processing of the ore, and mine waste rock produced when uncovering the ore body [8].

The metal content in soil is a sum of metals originating from natural processes and human activity. It is estimated that the contribution of metals from anthropogenic sources in soil is higher than the contribution from natural ones [9]. Significant increases in soil metal content are found in areas of high industrial activity where accumulation may be several times higher than the average content in

non contaminated areas. Additionally, areas distant from industrial centres also show increased metal concentrations due to long-range atmospheric transport. This fact has been observed by numerous authors [10, 11, 12]. Anthropogenic sources not only lead to increasing heavy metals concentrations in the environment, but also it can cause an unnatural enrichment, leading to metal pollution of the surface soils. The soil enriched with heavy metals can significantly cause an adverse impact on the population via inhalation, ingestion and dermal contact. The soil-accumulated heavy metals can also pose potential long-term hazards to plants and animals as well as humans that consume these plants [13].

Gold mining is responsible for most mercury (Hg) pollution in developing countries. (Hg) is considered to be one of the environmental pollutants with the greatest negative impact on the biosphere. The process by which many developing countries use elemental (Hg) to extract gold is becoming one of the primary sources of mercury pollution. The amalgamation method of artisanal gold mining causes mercury emissions to leak into the environment in several different ways. In Sudan studies on the impacts of artisanal small scale gold mining, known locally as public gold mining) have not been extensively carried out despite the fact that it employs more people (around 20 million).. Numerous environmental and social negative impacts aroused from artisanal small scale gold mining are gaining interest due to the serious impact and pressure that this sector is imposing on the environment as well as the serious health and safety impacts on those unskilled miners. Based on these foregoing justifications it was decided to set up the current research investigation on the objectives to monitor the anthropogenic and geochemical environment surrounding the Butana drinking water sources, which would also requires determination of heavy metals composition of the soil, streams sediments and gold mining tailings. The objectives of the present work would also include the determination of various geochemical indices such as index of geoaccumulation, degree of contamination, contamination factor and. normalized enrichment factor

Materials and Methods:

Site Description and Soil, Stream Sediment and Tailing Sampling:

The study area is located in central Butana area within the administrative boundaries of rural subagh locality of Gedarif State, Eastern Sudan, It comprises vast clay plains extending from the Gedarif area in the south up to latitude -15°N and from the banks of the Blue Nile River in the west up to banks of the Atbara River in the east. Climatically, the area is set within semi arid to savannah region [14] with high temperature in summer (March –June), and cold winter (December-February). The vegetation is spares, confined to the valleys and is made up of acacia type (Acacia Sayal and Acacia mallifera). The rainy season in central Butana is between July and September, raining although intermittent increases from north to south ranging from 150 to 400mm, and the average annual temperature in summer is around 40°C (March - October) and 25°C in winter (November-February). Soil cover of central Butana plain is formed mainly of sticky dark clays of montomorillonitic composition, which readily swell when wet, and shrink forming wide and deep cracks when they dry up.

The study area contains gold and minerals mines. Heaps of metal ores including precious, strategic, base metals and industrial minerals were scattered all over the area. The most important: Precious metals such as gold, silver and platinum, Base metals such as copper zinc and lead, Strategic minerals such as iron, manganese, chromium and **pentanium**, Industrial metals such as calcium carbonate, salt, talc and kaolin, silicon, and zircon and Building stones

Butana area contains many locally named mines like: Um Sarha, Fuwail, Khiari, Wad Bushara, Shawor, Gilaitah, Ead AL Wahash, and Birasi[Fig1].

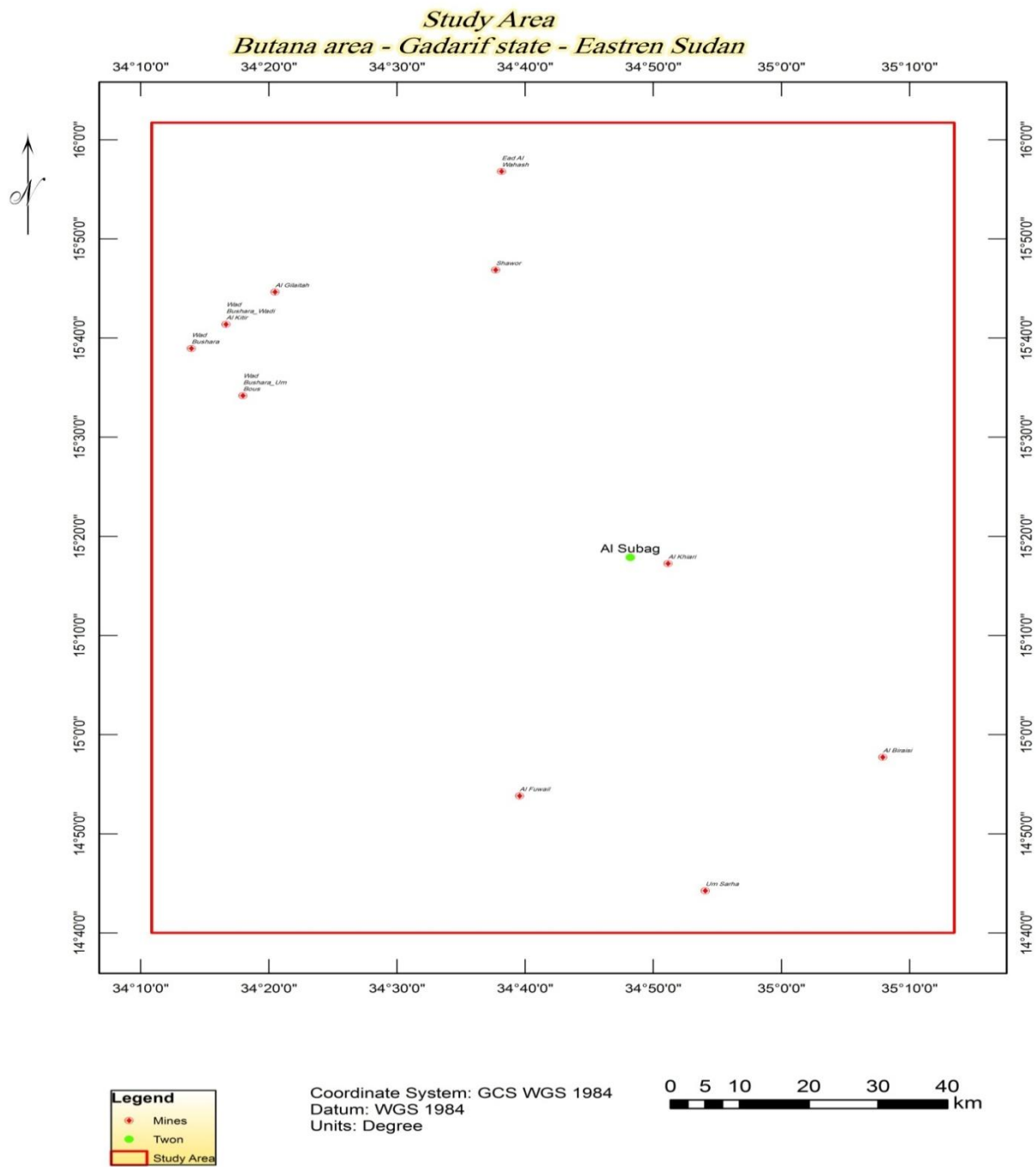


Fig 1: Distribution of Gold Mining Sites within Butana Region

Sampling Procedure:

Soil and Stream Sediments Sampling

A grand total of 87 samples were collected from 11 sites in Butana region, of which 31 soil samples, 42 stream sediments samples and 14 tailings samples. Random sampling was adopted. Sampling has been done in the May (summer) 2015. At selected sampling points, samples of the soil were taken from a depth of 80 to 90 cm using a soil probe. Sampling of Stream sediments

involved digging pits to bedrocks, Then the pits were channel-sampled along the entire profile from surface to bed rock to give uniform bullock sample weighing about 30Kg, The pits were generally 40-50cm deep. All samples were packed in plastic bags and transferred to the laboratory as soon as possible for analysis

Preparation of samples and standards

The samples were air-dried ($25 \pm 2^\circ\text{C}$) and primary crushed with Jaw Crusher to a size of -10 mm, secondary crushing by pulverize to -2 mm, splitting up to 200 gm and finally Grinding by Tema Mill to -125 mesh to obtain a very fine powder. Standard stock 1000 mg/L solutions were prepared for each of the heavy metals from which serial dilutions were made from known standard stock solutions of each. Calibration line method was used for the quantification of selected metals. The digests were appropriately diluted whenever required as reported in reference [15]. All measurements were made in triplicate. Similarly prepared blank solutions and standard reference materials were also analyzed alongside with the samples.

Determination of the concentration levels of heavy metals

The soil, stream sediment and tailing samples after air drying, were grounded and sieved to obtain a fine powder. Conventional aqua regia digestion was performed in 250 ml Teflon beakers. A well-mixed sample of 0.5 g of the sieved sediment soil or tailing) was digested in 12 ml of freshly prepared aqua-regia solution (1:3 HNO_3 -HCl, v/v) on a hotplate for 3 hours at 110°C . The solution was then evaporated to near dryness. The sample was diluted with aqueous nitric acid (20 ml, 2%) and filtered through Whatman No. 42 paper into a 100 ml volumetric flask and then diluted to 100 ml with deionized distilled water [16]. The determinations of heavy metals (Ni, Cr, Co, Zn, Cu, Pb, Cd, Mn, Fe, Hg) in the samples were performed using Savant AA 5th generation AAS from GBC. The air-acetylene flame was adjusted according to the manufacturer's recommendations.

Determination of the geochemical indices (Assessment of heavy metal pollution levels)

The Geo-accumulation index (*I-geo*) as defined by Muller [17]. The enrichment factor (*EF*) was calculated adopting the equation designed by Sutherland [18]. The contamination factor (*Cf*) of heavy metals, and contamination degree (*Cd*) of sampling sites were calculated as defined by Håkanson [19]. All of these soil contamination indicators are calculated with respect to local background, i.e. median values of metal concentrations in control soil samples.

The Geo-accumulation index (*I-geo*) was calculated as follows [20].

$$I\text{-geo} = \log_2 [C_n / 1.5B_n] \quad (1)$$

Enrichment factor *EF* of heavy metals was calculated as the ratio of elemental concentration of sediment normalized to *Fe* [17]:

$$EF = (C_n / Fe)_{\text{sediment}} / (C_n / Fe)_{\text{reference}} \quad (2)$$

Contamination factor *Cf* was determined as [19]:

$$Cf = C_n / B_n \quad (3)$$

The contamination degree (*Cd*) was defined as Aksu [21]:

$$\text{The sum of all contamination factors (Cf)}$$

In the above formulas, *C_n* is the concentration of the examined element 'n' in the surface sediments, and *B_n* is the geochemical background concentration of metal 'n'. The world surface rock average [22] is used as background concentration reference for this study. The factor 1.5 in the

I-geo formula (1) is incorporated to account for possible variation in the background data due to lithologic effect [20]. The *EF* geochemical normalization 2, was obtained using *Fe* as the reference element and as a conservative tracer to differentiate natural from anthropogenic components, following the hypothesis that its content in the earth crust has not been troubled by anthropogenic activity and because natural sources (98%) greatly dominate its contribution [23].

Statistical Analysis

SPSS statistics 24 was used for data analysis.

Results and Discussion:

It is noteworthy to mention that, the current research investigation is a continuation of our previous reported research outcomes [24], in which it was concluded that the impermissible levels of some hazardous minerals to international specification values were, presumably due to washings that were discharged from the mining sites and then drained to the drinking water sources (Haffirs reservoirs). These washings were originally constituted of gold mining wastes (tailings) and also to other unorganized activities such as mineral exploitation, crude gold ore transportation, smelting, refining and the inconspicuous public disposal of mine wastes activities. Based on these justifications it was then recommended and decided to carry out this current study to monitor the anthropogenic factors prevailing within the environment surrounding the drinking water sources. The research investigation requires implementation of the following phases: determination of the concentration levels of 10 most expected heavy metals prevailing within the surroundings soils, sediments and tailings. The current study is also intended to evaluate some geochemical parameters that would assist in justifying acceptable conclusions and recommendations.

Table 1, presents the maximum, minimum and mean concentration levels of the 10 heavy metals in the surrounding soil, stream sediment and gold mining tailings of the study area during the Dry Season: It could be noticed from this Table that iron Fe has shown the highest mean concentration level of in the surrounding soil. The heaps of tailings (gold mining wastes) have shown the highest concentration levels of mercury Hg 20.54 mg kg⁻¹, lead Pb 1015.90 mg kg⁻¹ and Zinc Zn 266.29 mg kg⁻¹, as expected contaminants of the drinking water in Butana Region. The concentration level of mercury in the Tailings has been found to be two folds of its concentration level in either the Soil or Stream Sediments. The soil surroundings have shown the highest concentration levels of iron Fe 42,649 mg kg⁻¹, which is approximately twice its concentration in either the stream sediments or tailings. It has also been shown that high concentrations of cadmium in the soil and sediment Cd 24.88 mg kg⁻¹ and copper Cu 769.25 mg kg⁻¹. On the other hand the streams sediments have shown the highest concentration levels of nickel Ni 81.188 mg kg⁻¹, chromium Cr 204.21 mg kg⁻¹, cobalt Co 45.64 mg kg⁻¹ and manganese Mn 2861.6 mg kg⁻¹. It could also be noticed from Table that the order of increasing concentration level of Ni, Cr, Co and Mn could be in the sequence: stream sediment > soil > tailing. Zinc Zn and Lead Pb were rated as Tailing > Soil > Stream Sediment samples, Copper Cu and Iron Fe are rated as: soil > tailing > stream sediment samples. Cadmium Cd is rated in the sequence: soil > stream sediment > tailings. The Mercury Hg concentration level is rated in the sequence: Tailings > Stream sediment > Soil samples.

Table 1: Heavy Metals Levels in the Soil, Stream Sediment in and Gold Mines Tailings in Comparison with reported levels in Most World Soil MWS [25].

Season	Type of samples	Range	Average concentration of elements mg kg ⁻¹										
			Ni	Cr	Co	Zn	Cu	Pb	Cd	Mn	Fe	Hg	
Dry	Soils	Max	68.097	172.1	71.52	131.41	1532.8	105.67	48.55	2137.7	83,863.	10.00	
		Min	0.9778	13.00	07.52	02.47	05.71	19.62	01.22	12.05	1,436	08.90	
		Mean	34.53	92.55	39.52	66.94	769.25	62.64	24.88	1092.8	42,649	09.45	
	Streams sediment	Max	106.136	366.33	59.59	70.62	84.69	352.08	25.39	4894.7	39,467.	11.34	
		Min	56.2414	42.105	31.70	15.27	23.22	136.19	02.80	828.65	11,814	10.42	
		Mean	81.188	204.21	45.64	42.94	53.955	244.13	14.09	2861.6	25,640	10.88	
	Tailings	Max	60.793	150.4	30.11	526.66	474.9	1996.0	09.07	721.79	43,416	24.17	
		Min	7.5905	13.80	08.30	05.92	16.92	35.83	01.80	63.26	8,417	16.92	
		Mean	34.191	82.10	19.20	266.29	245.91	1015.9	5.435	392.52	25,916	20.54	
	Common range in MWS	Soil	Max	500.00	1,000.	40.00	300.00	100.00	200.00	00.70	3000,00	55000.00	-
			Min	05.00	01.00	01.00	10.00	02.00	02.00	00.01	20.00	7000.00	-
			Mean	40.00	100.0	08.00	50.00	30.00	10.00	00.06	600.00	38000.00	-

Compared to the Most World Soils (Table 1) the concentration levels obtained in the present study could, confidently, be affirmed that the nickel and chromium concentration levels in stream sediment samples in all sites of Butana region is twice higher than Ni and Cr in Most concentrations in Most World Soils, Table 1.

The Cobalt, Copper, lead and Cadmium concentrations in all sites of the Butana region are extremely higher than it is reported in the World Common Soil. The Lead concentration levels in soil and stream sediment of the Butana region are extremely above the Most World Soil. Another sign of the involvement of anthropogenic factor is the concentration level of lead obtained in the present study, which is above the concentration levels reported by [26, 27 and 28].

The concentration of Zn in tailings and the concentration of manganese in stream sediment samples are in levels extremely higher than concentration in Most World Common Soil. The mean concentration levels of zinc, iron and manganese in soil are higher than their reported concentration in Most World Common Soil.

On the other hand, it was observed that the concentration of chromium and cadmium in soil, stream sediment and tailing samples of Butana region is extremely higher than that reported in the Canadian soil Quality Guidelines (Table 2). It was also being observed that the Nickel, Cobalt and Lead concentrations in stream sediment samples in all sites were above its concentration in Canadian Soil Quality Guidelines. Similarly, the concentrations levels of Zinc, lead and Copper in tailing samples is extremely higher than their concentration levels in Canadian Quality Guidelines.

Also the Cu concentrations in soil samples are extremely higher than its concentrations in Canadian soil Quality Guidelines.

Table 2: Canadian Soil Quality Guidelines for analyzed elements expressed in mg/kg

Element	Canadian Soil Quality
Cd	1.4
Cr	64
Co	40
Cu	63
Ni	50
Pb	70
Zn	200

It is important to mention that the concentration of Ni that occurred in all sites with different values of concentrations in such micro-sites such as: Um Sarha, Wad Bushara, Wadi Alkitar, Fuwail and Ed Al Wahash. The differences in the ratios of the Ni concentration in the various samples and sites are rather small. It is then evident that the whole region could be considered as rich source in Ni metal.

It could briefly, be mentioned that the highest concentration level of Cr in Butana region have been detected in the micro sites such as Um Sarha, Braisi, Fuwail and also beside the Subagh boarder. Whilst the highest concentrations of cobalt in stream sediments were found in the micro-sites Shawor, Ed Al Wahash and also beside Wad Bushara. Low concentrations of this heavy metal were found in Birasi, Gilaitah, Subagh and Khiari micro-sites.

Tailing samples have shown the highest concentration of zinc in Fuwail micro-sites and low concentration found beside Subagh twon. The concentration of copper in soils have shown the highest concentration of copper in Ed Al Wahash micro-site and law concentration was found in Shawor and Wad Bushara micro-sites. Tailing samples have shown the highest concentration of lead and was found in Fuwail micro-site and low concentration found in Shawor, Gilaitah and Wad Bushara micro-sites. The highest concentration of Cd in soil samples was found in Subagh and Khiari micro-sites and low concentration found in Birasi micro-site. The highest concentration of Mn in stream sediment was found in Shawor micro-site and low concentration in Wad Bushara-Um Bous micro-sites and beside Subagh micro-sites. The highest concentration of Fe was found in soil samples was found in Um Sarha, Gilaitah micro-sites, Wad Bushara-Wadi Alkitar, Wad Bushara-Um Bous micro-sites. Low concentration of this heavy metal was detected beside Shawor and Subagh micro-sites. Tailing samples have shown the highest concentration of Hg in Fuwail, Gilaitah micro-sites, Wad Bushara-Wadi Alkitar, Wad Bushara-Um Bous macro-sites. Low concentration was detected for Hg in Shawor micro-site.

It was noticed that the concentration levels of Cd, Pb, Cu, Zn, Cr and Hg were highest near the mining sites and then decreases gradually further from the center of mining and washing basins. Conclusively, based on the foregoing survey of the results obtained in this study, it could be inferred that the anthropogenic factors are predominantly affecting the mineral composition of the environment surrounding the water sources.

Moreover, the above analytical results were verified and supported by the determination of some geochemical parameters such as Geo-accumulation index (*I_{geo}*), enrichment factor EF, and

contamination factor CF and contamination degree CD. For example, the Geo-accumulation values are widely used to assess the degree of metal accumulation and contamination in terrestrial, aquatic and marine environments [30]. The Geoaccumulation index scale, as defined by Muller [17], consists of seven grades (0 – 6) ranging from unpolluted to extremely polluted (Table 3).

Table 3: Muller's Classification for Geoaccumulation index (*I_{geo}*) scale

<i>I_{geo}</i> value	Class	Degree of contamination
0	0	Unpolluted
0-1	1	From unpolluted to moderately polluted
1-2	2	Moderately polluted
2-3	3	From moderately polluted to strongly polluted
3-4	4	Strongly polluted
4-5	5	From strongly polluted to extremely polluted
> 5	6	Extremely polluted

Based on the Muller scale (Table 3), the calculated *I_{geo}* for the sediment samples (Table 4) indicated that the sediment in Butana mining area was unpolluted, with values for Ni, Zn, Cu, Pb and Fe metals *I_{geo}* ≤ 0, unpolluted to moderately polluted' or enriched for Cr (0.445), Co (0.605) and Mn (0.861), all having *I_{geo}* = 0-1 and strongly polluted by Cd (3.232) have *I_{geo}* = 3-4. On the basis of the mean values of *I_{geo}*, the Butana sediment is greatly enriched with the following heavy metals in the order: Cd > Mn > Co > Cr.

Table 4: Geoaccumulation index (*I_{geo}*) values of heavy metals in sediments of Butana mining area

Elements	Ni	Cr	Co	Zn	Cu	Pb	Cd	Mn	Fe
<i>I-geo</i>	-0.734	0.445	0.605	-3.612	-1.475	-1.713	3.232	0.861	-1.49

The Enrichment Factor (EF) is calculated by a comparison of each tested metal concentration with that of a reference metal, and the normally used reference metals are Mn, Al and Fe [30], in this study iron was used as a conservative tracer to differentiate natural from anthropogenic components. Enrichment factor *EF* of heavy metals (Table 5) was calculated as the ratio of elemental concentration of sediment normalized to *Fe* [17]:

Table 5: Muller's classification for Enrichment Factor

EF Value	Degree of contamination
< 2	Slight
2-5	Moderate
5-20	Severe
20-40	Very severe
> 40	Excessive

The results of the calculated *EF* values of heavy metals in the sediment samples of Butana mining area (Table 6) enriched for metals in the following order, are Cd (26.37) > Mn (5.10) > Co(4.27) > Cr (3.82) > Pb (3.04) > Zn (0.229) > Ni (1.68) > Cu (1.01) > Fe (1.0) respectively.

Table 6: Enrichment Factor values of the heavy metals in sediments of Butana mining area

Elements	Ni	Cr	Co	Zn	Cu	Pb	Cd	Mn	Fe
<i>EF</i>	1.68	3.82	4.27	0.229	1.01	3.04	26.37	5.10	01.00

The calculated *EF* values for the heavy metals in the sediments of Butana mining area (Figure 2), indicate that the stream sediment were slight contaminated by Ni, Zn, Cu, and Fe, Moderate contaminated by Cr, Co and Pb and severe contaminated by Mn and very severe by Cd.

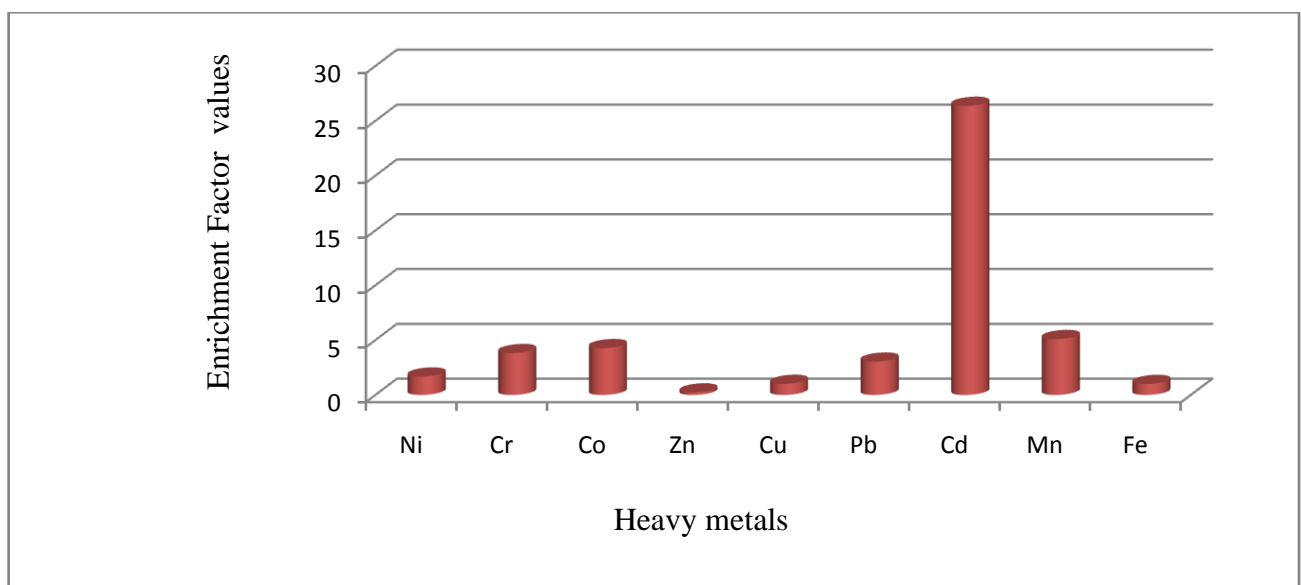


Figure 2: Enrichment Factor values of the heavy metals in sediments of Butana mining area

The Contamination *factor* (*CF*) is described as a ratio of the concentration of the element in samples to the pre-industrial reference value for the element [19], the world surface rock average values [22] are used for the calculation of *CF* as reference baselines. The degree of contamination (*Cd*) used in this study is also defined as the sum of all contamination factor values of the heavy metals [21]. Both contamination factor (*CF*) and contamination degree (*Cd*) are classified in five categories (Table 7). According to the definition by (19).

Table 7: Contamination factor (CF) and contamination degree (Cd) categories

CF classes	CF and Cd terminologies	Cd classes
CF < 1	Low CF indicating low contamination / Low Cd	Cd < 6
1 ≤ CF < 3	Moderate CF / Cd	6 ≤ Cd < 12
3 ≤ CF < 6	Considerable CF / Cd	12 ≤ Cd < 24
CF ≥ 6	Very high CF / Cd	Cd ≥ 24

The results of Contamination Factor and Contamination degree of Butana mining area in (Table 7) and (Figure 3) indicates that the sediments were low contaminated for Ni (0.90), Zn (0.122), (0.641), Cu (0.54) and Fe (0.53), Moderate contaminated for Cr (2.04), Co (2.28), Pb (1.63) and Mn (2.72), considerable contaminated for Co(5.705) and very high contaminated for Cd(14.09).

Table 8: (*CF*) and (*CD*) of the heavy metals in sediments of Butana mining area

Element	Ni	Cr	Co	Zn	Cu	Pb	Cd	Mn	Fe	CD
CF	0.90	2.04	2.28	0.122	0.54	1.63	14.09	2.72	0.53	24.852

The results of contamination degree (*Cd*) showed that sediments of Butana mining area was very high contaminated ($Cd \geq 24$) by anthropogenic pollution input. It is noteworthy to mention that Mercury pollution was not included in the Muller's classification and that is why its effects were not measured and hence its evaluation not being attempted.

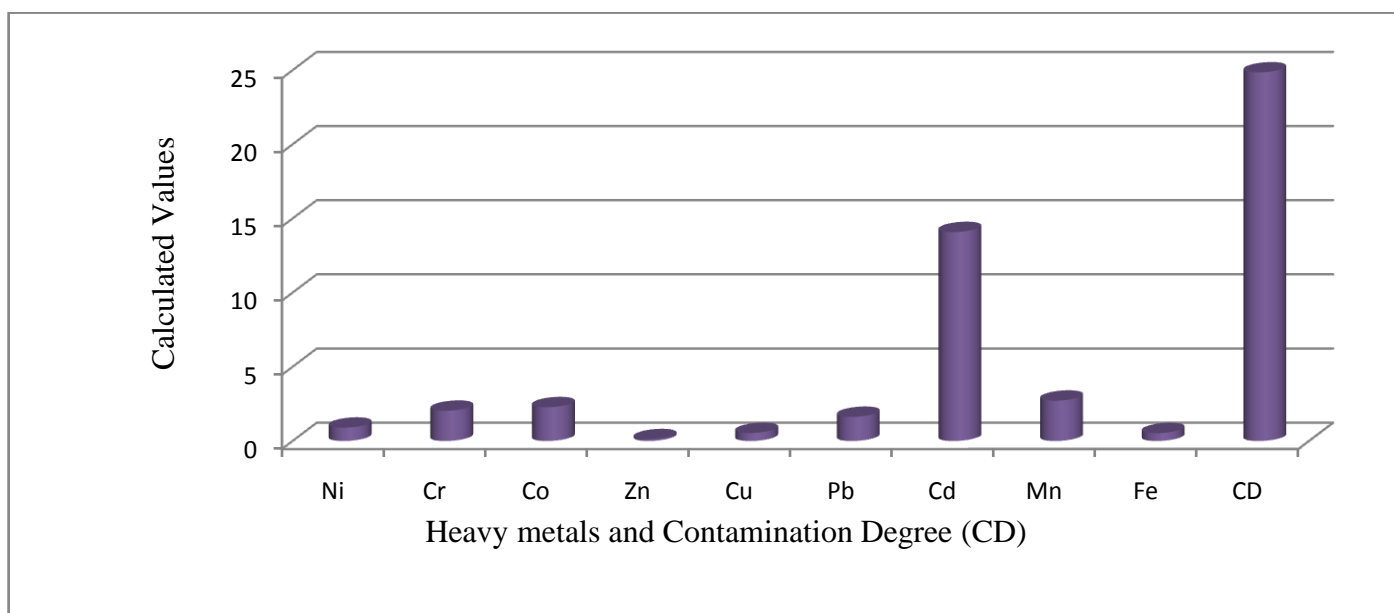


Figure 3: Histogram of the Contamination factor (*CF*) and contamination degree (*CD*) of the heavy metals in sediments of Butana mining area

Conclusions and recommendations:

It is concluded that this study, first of its kind, to confirm the influence of artisanal gold mining on heavy metals concentration at Butana mining area. It could also be, generally, concluded that the primary influencing factor of Butana drinking water pollution is actually anthropogenic related to the heavy and unorganized gold mining public activities leading to pollution of water with hazardous minerals such as mercury, cadmium, lead and chromium. Natural influencing factors, not to be ignored, are also prevailing due to the richness of the Butane region with various kinds of heavy metals such as gold, iron, cobalt, nickel, zinc and manganese. The former and latter factors were also being confirmed by the results obtained from the mineral composition of the surrounding soils stream sediments as well. Further confirmations were attained from the determination of the geochemical parameters: Geoaccumulation index *Igeo*, contamination factor *CF* enrichment factor *EF* and contamination degree *Cd*, whose values were pertinent with the analytical results obtained

in the present study. It is recommended that public gold mining should either be prohibited in the Butana region, particularly nearby the water sources and also workers in these gold mines be exposed to intensive training and educational programs on precautions, environmental protection awareness, safe-keeping measures including management of wastes in gold mining areas.

Acknowledgement

Our thanks are due to the Ministry of minerals –Geological Research Authority (GRAS) for funding the research project. Thanks are also due to The State of Gedarif local Government Office for facilitating the tours around the sites in which the work has been carried out. Our thanks and gratitude is due to the University of Medical Sciences and Technology and University of Khartoum for the supervision of this research project.

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