



Heavy metal pollution assessment in phosphate mining and processing sites, Hahotoé and Kpémé in Togo

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Abstract

The industrial activities related to phosphate mining and treatment constitute a threat to the population and its environment at Hahotoé and Kpémé (Togo) and released uncontrolled concentrations of toxic heavy metals that polluting soils, vegetation and water. The assessment of the pollution level would be useful for remediation, safeguard and prediction of risk to the environment and public. This study aims to assess the pollution level related to heavy metals in the soils, water and leaves at the phosphate mining (Hahotoé) and treatment (Kpémé) sites using spectroscopic techniques. Pollution indices such as contamination factor, degree of contamination, potential ecological risk index, pollution load index and heavy metal pollution index were used to achieve this goal. The average concentrations of Cd, Cr, Cu, Ni, Pb and Zn were investigated in the soils, leaves and water samples from the studied areas using the ZEEnit 700 with acetylene-air flame atomic spectrometer. High average concentrations of heavy metals were found in the samples exceeding the WHO limits. Heavy metal pollution index was used to assess the water quality with respect to heavy metals and very high value was found of 1168.84 at Kpémé and 733.58 at Hahotoé exceeding the critical value of 100. The results show unacceptable water quality for drinking on these sites. This study must help to design measures to reduce the pollutants released in the areas. The concerned issue poses a public health threat, so stakeholders would act in controlling the pollution level.

Keywords

Heavy metal, pollution Index, phosphate mining, Togo

Introduction

The pollution related to heavy metals constitute a great challenge for human and his environment. Human activities such as mining and manufacturing industries contribute significantly to the development of a country. They are the main sources of hazardous heavy metals (Sharma et al., 2019; Ma et al., 2019; Ashraf et al., 2019). The wastes produced from mining and mineral processing need a safe management in order to reduce the release of trace elements such as heavy metals and to protect the environment.

Heavy metals include Cadmium (Cd), Nickel (Ni), Lead (Pb), Iron (Fe), Zinc (Zn), Cobalt (Co),

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Arsenic (As), Chromium (Cr), Silver (Ag), and Platinum (Pt) can be naturally present in the soil. Their concentration in the matter determine the contamination level and the toxic effect on plants and living beings (Kolo et al., 2018; Ashraf et al., 2019). An excess of heavy metals in plants could lead to the absorption, accumulation, biochemical and physiological changes that causes the reduction and inhibition of the plant growth (Ameraoui et al., 2017; Majolagbe et al., 2018). Their toxic effects on plants can lead to lack of photosynthesis, degradation of nutrients and water balance to the plant death (Zhou et al., 2016; Wang et al., 2021). The inhalation of heavy metals and human expositions could lead to deaseses such as pulmonary alterations, lung disease, damage of nervous tissue, respiratory tract and lungs, dermatitis, rhinitis, ulceration of the skin, inflammatory lung disease, kidney damage as well as pneumonia and even gastritis or conjunctivitis (Ashraf et al., 2019; Wang et al., 2021; Fahimirad and Hatami, 2017; Verma and Sharma, 2017; Jaishankar et al., 2014). Many industrial and mining activities are developed in Togo. Open air activities concerned the Phosphate mining (Hahotoé) and treatement (Kpémé) industries of the "Société Nouvelle de Phosphate du Togo" (SNPT) and the cement industries such as Ciment of Togo (CIMTOGO), Diamond Cement Togo (DCT SA), West African Cement (WACEM SARL), etc., exposed the surrounding population to diseases and environmental pollution.

Past studies carried out in Togo have shown (i) a very high average values of heavy metals in the blood of workers and local residents of the phosphate mining area (Aduayi-Akue et al., 2015), (ii) the presence of heavy metals in agricultural products (corn, etc.) around the phosphate treatment area (Aduayi-Akue and Gnandi, 2014), (iii) the fluoride high content in the water with fluorosis of the teeth of the population (Tanouayi et al., 2016). Surface and groundwater in the mining area of Hahotoé-Kpogamé were contaminated by Cadmium and Lead (Tanouayi et al., 2015). High accumulation of heavy metals were found in the vegetables and in the soil along the Lomé-Aného (Togo) road near the industrial zone (Gnandi et al., 2008). The impact of the industrial activities on the health of the surrounding population and on its environment becomes crucial to know.

The present study aims to assess heavy metal pollution in soil, leaves and water in the surrounding area of the mining and industrial zone (phosphate) in Togo using spectroscopic techniques. The findings showed high average concentrations of heavy metals that exceeded the WHO limits for Zinc, Nickel, Chromium and Cadmium in soils and for Chromium, Cadmium and Lead in leaves and for Chromium and Cadmium in water.

High pollution indices were found indicating a considerable contamination. These results could aware the opinion on the industrial pollution in Togo and could help policy and decision makers in term of limitation and strategies of remediation to save human beings and the environment.

Materials and Methods

Study areas

The mining of the phosphate has been proceeded at Hahotoé and Kpogamé (6°12'47.99" N 1°30'42.03" E) sites and the processing is carried out at Kpémé (6°21'52.44" N 1°24'47.79" E), respectively by the "Société Nouvelle de Phosphate du Togo" (SNPT). The phosphate extraction area lays stretches from Avéta (south-west) to Dagbati (north-east) over a length of 36 km with a width of 2.5 km (Figure 1). Figure 1 shows the phosphate extraction area of Hahotoé with the different collected samples positions (soil, leaves and water). The phosphate processing area at Kpémé with the various collected sample positions (soil, leaves and water) is presented on figure 2. Hahotoé and Kpémé areas are part of "Bas-Togo" where the climate is Guinean subequatorial. This climate is subject to cold sea currents with the atmospheric circulation of the boreal continental trade winds (harmattan, November - February) and the southern maritime trade winds (monsoon, June-September). It is marked by two rainy seasons interspersed by two dry seasons as (i) the rainy seasons run from March



Figure 1. Google Earth map of the Hahotoé phosphate mining area showing the different samples collected



Figure 2. Google Earth map of the Kpémé phosphate processing area showing the different samples collected

to July and from September to October and (ii) the dry seasons are from July to September and from November to February. The average annual rainfall reaches 1000 mm of water and the average annual temperature is around 27 ° C. It is an area where the altitude is sometimes below the sea level at around 15m with an average humidity of 75% (Houedakor, 1997; Amoussou, 2002; Aduayi-Akue et al., 2015).

Sampling and instrumentation

The investigated area belongs to the coastal sedimentary basin of Togo which is includes three geological sets (Da Costa, 2005; Da Costa et al. 2006; Johnson, 1987; Johnson et al., 2000) such as (1) the Tabligbo group (sands, limestone, claystone) of Campanian to basal Eocene, (2) the Hahotoé-Kpogamè phosphatic complex (lower to middle Eocene) made of phosphatic marls (15 to 20 m), phospharenite layer (2 m to 8 m) and phosphatic clay (0.5 to 10 m) and (3) the upper detrital series (or Continental terminal sensu lato) of post Eocene age, is a reddish sandy-argillaceous complex made of conglomeratic clayey sands and clayey silty sands ("Terre de barre"). The various soils encountered in the studied area are (i) ferralitic red soils corresponding to the "Terre de barre" (ii) semi peaty soil on various alluvium and (iii) flooded, evolved and hydromorphic soils from alluvial valleys.

Thus, ten (10) soil samples, five (05) plant leaf samples and ten (10) water samples from wells, rivers and runoffs, were collected from the phosphate mining (Hahotoé) and treatment (Kpémé) areas with a clean stainless steel shovel and were placed in labeled, well-sealed polyethylene bags for the laboratory. A Memmert UNB 400 oven was used to dry soil samples at 105°C for 24 hours. Soil samples were ground using a mortar and 100 g were sieved through a 150 μ m sieve, according to ISO 11464 (ISO 11464, 2006) before used to investigate the content of Cu, Cr, Cd, Zn, Ni and Pb. Thus, 3 g of dried and ground soil were transferred to a digestion vessel and then 21 ml of hydrochloric acid (37%) and 7 ml of nitric acid (HNO₃, 65%) from Merck (Darmstadt. Germany) were added successively according to ISO 11466 protocols (ISO 11466, 1995). The mixture was left to stand for 16 hours at room temperature for slow oxidation of the organic matter in the sample. The temperature of the mixture was slowly increased to reflux conditions and then maintained for 2 hours. The digested samples were then filtered and the filtrate was collected in a volumetric flask and the volume was increased to 100 ml with HNO₃ (0.5%). The supernatant was analyzed by flame atomic absorption spectrometry. For each set of measurements, a blank was prepared using the same procedure. Leaves samples were cleaned and washed with distilled water, dried at 80-90°C for 15-30 minutes and then at 65°C for 12-24 hours. Leaves were crushed and sieved through a 0.5 mm sieve. All samples were weighed before and after drying. For each analysis, 0.5 g of the leaf sample was digested with 4 ml of HNO₃ (65%) until a clear solution was obtained. The digested samples were then diluted to 50 ml and filtered. 100 ml of water samples were used without any treatment for the detection of heavy metals.

The heavy metals amount was investigated using a ZEEnit 700 atomic absorption spectrometer with acetylene-air flame and cathode lamps at Babeş-Bolyai University, Faculty of Environmental Science and Engineering, ISUMADECIP, Cluj-Napoca, Romania.

Heavy Metal Pollution Assessment

The pollution related to heavy metals in soil, leaves and water from the industrial areas in Togo was few studied (Aduayi-Akue et al., 2015; Tanouayi et al., 2016; Tanouayi et al., 2015; Gnandi et al., 2008). The present study performed the assessment of the contamination of heavy metals in soil, leaves and water on the phosphate mining (Hahotoé) and treatment sites (Kpémé) in Togo using contamination factor (C_{f}), degree of contamination (C_{deg}), potential ecological risk index (RI) and pollution load index (PLI).

The degree of contamination (C_{deg}) corresponded to the sum of contamination factors of each heavy metal, equation [1] and [2] (Kowalska et al., 2018; Hakanson, 1980; Newaz et al., 2021):

$$C_f = \frac{C_i}{C_n}$$
[1]

$$C_{deg} = \sum C_f$$
^[2]

Where: C_i is the mean concentration of individual metal, and C_n is the concentration of a reference value for individual metal. In this study, C_n for soil, leaves and water were the limits values from WHO (WHO, 1998; WHO, 2017) and international sources (European Parliament and Council of the European Union, 2019; FAO/WHO, 2018). The values of these indices with their associated interpretations were presented in the Table 1.

The degree of heavy metal pollution risk on ecology was investigated using the potential ecological risk index (RI) that related to the toxicity of metals and the response of the environment according to the equation [3] (Kowalska et al., 2018; Haridson, 1980; Newaz et al., 2021):

$$RI = \sum T_r C_f$$
[3]

Where: T_r is the metal toxic factor and C_f is the metal contamination factor. The toxic factor of heavy metal concerned in this study are Cd = 30, Ni=5, Cu = 5, Pb = 5, Cr = 2 and Zn = 1 (Fiori et al., 2013). The potential ecological risk can be qualified by the terminology defined for RI in table 1.

Table 1. Contamination factor, degree of contamination and potential ecological risk index, categories and interpretation (Newaz et al., 2021; Hakanson, 1980; Calmuc et al., 2021)

Indexes	Values	Interpretation				
	$C_{f} < 1$	Low contamination				
C	$1 < C_f < 3$	Moderate contamination				
C _/	$1 < C_f < 3$ $3 < C_f < 6$	Considerable contamination				
	$C_f > 6$	Very high contamination				
	<i>C_{deg}</i> < 8	Low degree of contamination				
C	8< C _{deg} <16	Moderate degree of contamination				
C_{deg}	$16 < C_{deg} < 32$	Considerable degree of contamination				
	$C_{der} > 32$	Very high degree of contamination				
	RI < 150	Low potential ecological risk.				
DI	150 < RI < 300	Moderate potential ecological risk.				
RI	300 < RI < 600	Considerable potential risk ecological risk.				
	RI > 600	Very high ecological risk				

The pollution load index (PLI) of a single site is the root number (n) of multiplied together contamination factor ($C_{\rm f}$) values (equation [4]) (Kowalska et al, 2018; Hakanson, 1980; Newaz et al., 2021).

$$PLI = \sqrt[n]{C_f^1 \times C_f^2 \times \dots \times C_f^n}$$
[4]

Where: n is the number of metals considered and C_r is the contamination factor.

The relative total water quality and the impact of each individual heavy metal on the overall water quality were investigated on the sites using the heavy metal pollution index (HPI) (Li et al., 2020; Venkata Mohan et al., 1996) using the equations [5] and [6].

$$HPI = \frac{\sum_{1}^{n} Q_i W_i}{\sum_{1}^{n} W_i}$$
[5]

$$Q_i = \frac{C_i}{S_i} \times 100; \quad W_i = \frac{k}{S_i} \quad [6]$$

Where: Q_i is a sub-indicator of the heavy metal pollution index, W_i is the unit weight of the heavy metal parameter with the proportional constant (k) set to 1 (Pejman et al., 2015), C_i is the mean concentration value of the specific heavy metal i parameter (mgL⁻¹), and S_i corresponded to the highest standard permissible value of the i^{-th} parameter.

The values of these indices with their associated interpretations were presented in the Table 2.

Table 2. Pollution loa	d index. heavy	metal pollution	index. categories and
<u>interpretation (Newaz e</u>	t al., 2021)		

1		,				
Indexes	Values	Interpretation				
	PLI < 1	Denote perfection				
PLI	PLI =1	Only baseline levels of pollution				
	PLI > 1	Deterioration of soil quality				
HPI	0 < HPI < 25	Very good				
	26 <hpi <50<="" td=""><td colspan="3">Good</td></hpi>	Good				
	51 <hpi <75<="" td=""><td>Poor</td></hpi>	Poor				
	HPI > 75	Very poor (unsuitable for drinking)				

Results and discussion

Heavy Metal Concentrations in soils, leaves and water

Concentrations of heavy metals such as Zn, Cr, Cd, Ni, Pb, Cu were investigated in soil, leaf and water samples from the phosphate mining and processing areas. The obtained toxic element average concentrations from these areas are listed in the Table 3. The results were compared to the standard permissible limits (WHO, 1998; WHO, 2017; European Parliament and Council of the European Union, 2019; FAO/WHO, 2018). For the soil samples, heavy metal concentrations followed an order at Hahotoé as: Zn > Cr > Ni > Cu > Cd > Pb and at Kpémé, as: Zn > Cr > Cu >Ni > Pb > Cd, respectively. The comparison of the proportions of heavy metals in leaves followed the arrangements at Hahotoé as: Zn > Cr > Ni > Cu > Pb > Cd and at Kpémé as: Zn > Cu > Ni > Pb > Cr> Cd, respectively. The average concentration of heavy metals measured in soil samples at phosphate

processing site (Kpémé) is lower than the value measured at phosphate mining (Hahotoé) for all the elements. The concentration of Cr and Cd were sharply higher than the permissible limits. The concentration of Zn and Ni were very higher than the permissible limits in the soil from Hahotoé. This could constitute danger for the pollution in the two zones. For leaf samples, the concentration of Cr, Cd and Pd were very higher than the permissible limits. For water samples, the heavy metal proportions at Hahotoé followed the order Cd > Ni> Zn > Cu > Pb > Cr and at Kpémé, as: Zn > Cr > Ni > Cu > Pb > Cd, respectively. In the water samples, the average concentration of Cd and Pb were higher than the permissible limits.

Other works were carried out on phosphate mining sites to assess heavy metals in the soils of the phosphate mining area of Kpogamé and Hahotoé (Togo) and the finding showed high concentrations for Cd, Pb, Cr, Cu, Ni, V, Zn, Zr and Sr (Gnandi and Tobschall, 2002). The finding in the present study on the water quality related to the heavy metal pollution on surface and groundwater in the Hahotoé-Kpogamé mining area (Togo) showed high contamination by Cadmium and Lead (Tanouayi et al., 2015). The measurements carried out on the soil samples, confirmed the pollution of

the sites by different heavy metals such as Cadmium and Lead.

Table 3. Mean heavy metals concentration in the soil, leaf and water samples from the phosphate mining (Habotoé) and treatment (Kpémé) sites

<u> </u>		-	-	6.1	N 71		0
Samples	Locations	Zn	Cr	Cd	Ni	Pb	Cu
Soil (mg/kg)	Kpémé	193.7	103.7	19.3	39.5	23.4	37.2
	Hahotoe	459.9	262.1	35.7	103.6	34.6	58.5
	Limits *	300	2	1.5	50	120	300
Leaf (mg/kg)	Kpémé	51.7	6.9	3.4	8.6	7.2	16.4
	Hahotoe	69.4	6.8	6.3	5.6	6.5	12.7
	Limits***	100	0.19	0.2	50	0.3	100
Water (mg/L)	Kpémé	0.1136	ND**	0.0468	0.0511	0.0265	0.0769
	Hahotoe	0.0139	0.0045	0.0297	0.0376	0.0127	0.0144
	Limits*	3.0	0.05	0.003	0.07	0.01	2.0

* (WHO, 1998; WHO, 2017; European Parliament and Council of the European Union, 2019) - ** ND = Not detectable - *** source (WHO, 1998; FAO/WHO, 2018)

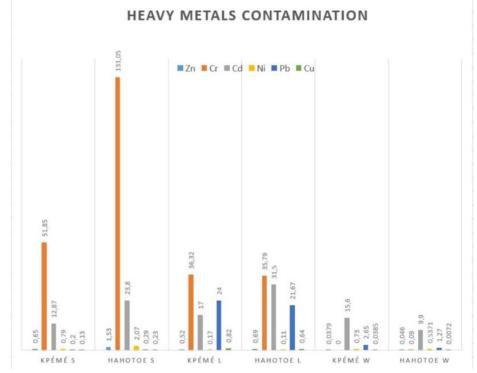


Figure 3. Metal contamination factor at the phosphate mining (Hahotoé) and treatment (Kpémé) sites. L= leaf, S=Soil, W= water

Contamination assessment of soils, plants and water

Contamination factor (C_f) . The heavy metals contamination was assessed by using contaminant factor (C_f) . Figure 3 shows the average values of

the contamination factor of the measured elements in the studied soils, plants and water from the phosphate mining (Hahotoé) and treatment (Kpémé) sites. The C_f of Zn, Ni and Cu was found to be smaller than one and corresponding to low contamination of these heavy metals in all leaf and water samples. The C_f value for Zn and Ni was less than 3 in soil samples from Hahotoé indicating a moderate contamination. The C_f value for Cr was considerabily higher than 6 indicating a very high contamination in soil and leaf samples. The C_f value for Cadmium was very high in soils, plants and water from the phosphate mining (Hahotoé) and treatment (Kpémé) sites and means very high contamination. The C_f value for Pb in leaf and water on the sites was considerable and indicates very high contamination.

many heavy metals had their concentrations higher than the permissible limit, demonstrating their anthropogenic origin. Contamination of heavy metals on these sites would due to the industrial activities contributions (Shankar, 2019).

Contamination Degree (C_{deg}). The C_{deg} in the phosphate mining (Hahotoé) and treatment (Kpémé) soils presented very high degree of contamination (table 3). In the leaf samples, the C_{deg} varied between considerable (Kpémé, 24.00) to very high degree (Hahotoé, 158.98). In water sample, the C_{deg} varied from moderate (Hahotoé, 11.94) to very high degree (Kpémé, 90.40).

In soils, plants and water at Hahotoé and Kpémé,

Items —		Kpémé			Hahotoé	
	Soil	Leaf	Water	Soil	Leaf	Water
Cdeg	66.49	78.83	19.06	158.97	90.40	11.85
RI	496.05	708.11	485.13	990.58	1129.37	306.30
PLI	1.44	3.20	0.59	2.95	3.25	0.24
HPI			1168.84			733.58

Table 4. Pollution indices at the phosphate mining (Hahotoé) and treatment (Kpémé) sites

Potential ecological risk index (RI). The potential ecological risk for soil, leaf and water samples from the phosphate mining (Hahotoé) and treatment (Kpémé) sites was investigated to assess the hazard of heavy metals contamination and related risks. From Table 3, a very high ecological risk index (RI, 708.11-1129.34) were found exceeding the maximum index of 600 and it's corresponding to very high ecological risk sites excepted the water samples from Hahotoé and Kpémé where the ecological risk index ranged from 309.43 to 503.34, indicating a considerable potential ecological risk.

Pollution Load Index (PLI). In this study, the overall toxicity status was investigated for soil, leaf and water samples from the phosphate mining (Hahotoé) and treatment (Kpémé) sites using the pollution load index (PLI) value. This index is a tool used to compare the hazard of heavy metal contamination level on the sites. The PLI values for all metals in Table 3 show low value less than 1 for the water samples from the two sites indicating that the water may be uncontaminated by heavy metals. For soil and leaf samples, the PLI values were greater than 1 and showing a deterioration of their quality. And the continuously industrial activities may increase the pollution level in the areas (Laniyan et al., 2020).

Heavy metal pollution index (HPI). The quality of water samples from the phosphate mining (Hahotoé) and treatment (Kpémé) sites was assessed using the HPI values. The mean heavy metal pollution indices (HPI) at Hahotoé and Kpémé were 741.45 and 1261.93, respectively (Table 3) and were found above the critical index value (100) indicating that the water quality was very poor and was unsuitable for drinking. These high index values confirmed that heavy metal pollution may be due to the spreading and the leaching of heavy metals from the mining and the processing industries, respectively. This technique takes into account the overall quality of water and is suitable for this duty (Tiwari et al., 2015). It was used to assess surface water quality related to heavy metal content in many countries such as: Turkey (Kutlu and Mutlu,

2021), India (Tiwari et al., 2015; Shankar, 2019), Sudan (Eldaw et al., 2020), Syria (Abou Zakhem and Hafez, 2015) and in groundwater for drinking in the Oti community and in the Tarkwa mining area in Ghana (Boateng et al., 2019; Yankey et al., 2013). The HPI values classified the water samples in the two sites very poor and unsuitable quality for drinking by heavy metal. The results of this study show the poor quality of local water and the danger to the health of the population and confirm the results of similar work that has been done on the impact of the discharge of phosphate waste at Kpémé by the "Société Nouvelle des Phosphates du Togo" (SNPT) on the marine ecosystem and the population of the area (Gnandi, 2005).

Conclusions

The present study investigated on the heavy metal pollution assessment in soil, leaf and water samples in the industrial areas of SNPT and the impact on the living beings. This was achieved using pollution indices such as contamination factor (C_{ℓ}) , degree of contamination (C_{deg}), potential ecological risk index (RI), pollution load index (PLI) and heavy metal pollution index. The results showed that the concentrations of heavy metals in the soil samples from the phosphate mining (Hahotoé) exceeded the measured values in the phosphate processing (Kpémé) areas. This state of the pollution of the soil is related to the phosphate mining in the area which contributed to the level of the pollution in leaf samples and all the water samples regarding to Cadmium and Lead. The high HPI at the processing area (Kpémé) correspond to an additionnal amount of heavy metals due to the leaching of these elements through the mineral treatment. The concentrations of heavy metals are high compared to the international standard limit values. The finding showed the poor quality of local water and its related hazards to the health of the population and the environment comprising the marine ecosystem. This present environmental pollution assessment confirm that anthropogenic sources could increase the risk of toxic metal contamination in soil and environment around industrial areas. Then, continuously m onitoring of heavy metals contents should be recommended in different areas of industrial sites while the socio economic activities is increasing in the studied area.

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