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A Particle Induced X-Ray Emission (PIXE) Analysis of Heavy Metals in Soil and Plantain (*Musa paradisiaca*) Leaves at an Artisanal Gold Mining Settlement in Southwestern Nigeria

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Abstract

The study examined the contamination levels of the soil and plantain leaves in three communities in Atakunmosa west local government area of Osun State in southwest Nigeria, where gold mining activities have recently become intensive. Plantain is a major food crop, whose leaves are used to wrap food items for household consumption. The objectives were to examine the heavy metal concentrations in the soil and plantain leaves around the gold mine site and compare them with standard recommended safe limits for the environment. Soil and plantain leaves were sampled at different locations around the sites and at a control site from a neighboring local government area with no history of gold mining activities between March 2015 and February 2016. Both soil and leave samples were processed and analyzed for selected heavy metals (Cd, As, Cu, Zn, Cr, Mn and Fe) using the Particle Induced X-ray Emission (PIXE) in the laboratory. The study showed higher concentrations than the World Health Organization's recommended safe limits of the heavy metals in the soils and plantain leaves. Dry season concentrations of the variables were also higher than the wet season and the heavy metal concentrations at the control station were significantly ($p < 0.05$) lower than those of the mining environment. The study concluded that the ecosystems in the artisanal gold mining region are vulnerable to bioaccumulation of heavy metals and the leaves from the sites are sources of heavy metal contamination if consumed or used to wrap food items.

Keywords

Artisanal Mining, Soil and Food Contamination, Particle Induced X-Ray Emission, Bioaccumulation

1. Introduction

Heavy metals are so classified based on their high atomic weight and relative density which is about five times higher than that of water [1]. They are present in various levels in soil, sediment, water, food and even air from various sources such as domestic, agricultural, medical, pharmaceutical, commercial and industrial. Heavy metals are toxic with various serious health implications when exposed even at low concentrations. Artisanal mining, especially of gold metal, has been regarded as one of the most prominent sources of heavy metal contamination of soil, sediment, plant and water [2] [3]. It has been known to release metals such as Pb, Cd, As, Cu, Fe, Cr, Ni, Mn, Zn into the environment where they constitute hazards. Metal toxicity to man is particularly heightened from human exposure through food chain as the soil is a major sink for these metals [4] [5]. Human exposure can also be through direct contact with such contaminated soil or plant during farming, fishing and other activities [6].

Plants have been known to absorb these metals from soil in various amounts [7]; they are accumulated through the root from the soil and can also be taken through other organs above the ground. While the essential ones are needed for plant growth, the latter may be required by some aquatic organisms for their survival [8]. Copper is an active center in many proteins and enzymes [9]; its deficiency symptoms in plant usually lead to loss of biomass. Nickel is equally an active centre for urease [10]; its deficiency, which is rarely observed, is hyperaccumulation of urea to toxic level. While Zn is equally essential in plant growth, it is an indispensable constituent of many proteins and enzymes [11]; its deficiency includes stunted growth and wilting. In the study area, plantain is second only to Cocoa as the Agricultural products.

Plantain (*Musa paradisiaca*) is an important biennial food crop in Nigeria. It is a popular staple food that is eaten boiled, baked, fried or mashed. Different parts of the plant are used for different purposes. Edible parts of the plants include the pulp, peel [12] and the stem, while the leafy part is found usage as platters for serving food, for packing and wrapping food materials, especially when dried. It is also a cheap source of food for some domestic animals. The leave and peel have been also recently found ready use as adsorbents [13] for heavy metal remediation in the environment which makes the determination of heavy metal content important as it will also serve to indicate the degree of pollution/contamination of the soils on which it is grown [14] [15].

2. Study Objectives

The main objectives were to examine the heavy metal concentrations in the soil and plantain leaves around the gold mine site and compare them with standard recommended safe limits for the environment.

3. Materials and Methods

3.1. Study Area

The area selected for the study are Sabo, Itagunmodi and Igun mining villages in Atakumosa West Local Government area of Osun State in Southwestern Nigeria within latitudes 7.51˚N and 7.65˚N and longitudes 4.61˚E and 4.85˚E (**Figure 1**) and covers an area of 577 km^2 and a population of 68,643 (NPC, 2006). Three mining locations each was sampled in the selected villages. The control site selected for the study is Tonkere village in Ife Central Local government area of Osun State, about 13 km from the study area.

3.2. Sample Collection

One hundred and eight (108) samples of each soil and plantain leaves were collected from Sabo (SB), Itagunmodi (IT) and Igun (IG) gold mining villages of Atakumosa-west Local Government area of Osun State from March 2015-February 2016. One (1) kg of soil sample was collected with a hand trowel at a depth of 15cm from surface soil around the pits dug for mining. The leafy part of the plantain tree were cut, the soil and leave samples were packed differently in polythene bags sealed properly and tagged.

3.3. Laboratory Analysis

In the laboratory, the soil samples were air dried ground and pelletized for Particle Induced X-ray Emission (PIXE) analysis for metal concentration determination. The PIXE is a nuclear analytical technique employed in the multi-elemental determination of metals in samples. Leafy part of the plant was selected for the study. The leaves were collected and kept in labeled polythene bags; effort was made to avoid soil contamination of the samples. Samples collected were dried at 103˚C for 24 hrs, ground and sieved with a 2 mm mesh, pelletized and kept for heavy metal analysis.

3.4. Data Treatment

Data obtained from PIXE analysis of the samples were subjected to statistical analysis using an up-to-date statistical package (SPSS V24). Descriptive and inferential analyses were equally carried out and the results compared with environmental standard limits recommended by world regulatory bodies.

4. Results and Discussion

4.1. General Distribution of Selected Heavy Metals in Soils and Plantain Leaves

The mean concentrations of eight (8) heavy metals obtained from PIXE analysis of soil and plantain leaves collected from mining locations in three villages of the

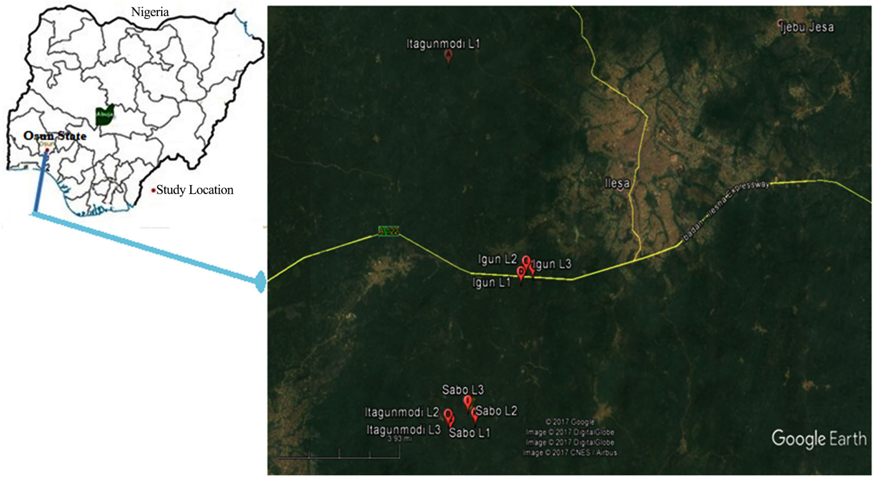

Figure 1. Location of the sampling stations in Atakumosa local government area of Osun State, Nigeria.

study area presented in **Table 1** and **Table 2**. In the soil, Pb occurred mostly at Itagunmodi, between 20.65 ± 2.72 and 24.31 ± 2.88 mg/kg but was lowest at Sabo with a range of 11.05 ± 2.20 to 21.26 ± 3.30 mg/kg. The mean concentration at Igun mining site ranged between 14.42 ± 1.67 and 19.12 ± 2.39 mg/kg, Although the mean value at Itagunmodi was within the safe limit, being lower than the 100 mg/kg maximum permissible limit set for soil [16] [17], the mean concentrations of Cd (158.50 ± 9.71 - 346.03 ± 10.12 mg/kg) and Cu (149.68 ± 2.74 - 295.63 ± 4.88 mg/kg) were greater than the recommended allowable limits of 3 mg/kg and 100 mg/kg, respectively at all the mining settlements (**Table 1**). Zn occurred within the safe limit at most of the sampled areas being lower than 300 mg/kg recommendation of the WHO (2008), except at Itagunmodi. Fe occurred at above normal concentrations (4016.96 ± 120.24 - 8765.45 ± 210.11 mg/kg) at all the sampling region, but the concentrations of Cr (Igun, 91.03 ± 7.91 - 121.37 ± 10.31; Itagunmodi, 68.87 ± 6.89 - 78.31 ± 3.15 and Sabo, 42.28 ± 10.62 - 47.45 ± 8.59 mg/kg) exceeded the recommended safe limits.

In the plantain leave samples, most of the investigated metals (except As, Cr and Mn) occurred in abnormally high concentrations (Pb, 3.97 ± 0.29 - 7.69 ± 0.63 mg/kg; Cd, 6.48 ± 0.58 - 22.05 ± 1.90 mg/kg, Cu, 25.13 ± 2.18 - 59.34 ± 5.21 mg/kg, Zn, 12.36 ± 1.06 - 30.34 ± 2.31 mg/kg and Fe, 48.51 ± 4.29 - 82.51 ± 7.34 mg/kg) greater than the recommended safe limit, WHO, [18] in the leaves at all the selected locations (**Table 2**). The Cu concentration was higher than recommended safe limit for in the plantain leaves at the control station, and also suggested the vulnerability of the non-mining site to bioaccumulation from other Cu sources. Studies showed that Cu sources include the use of $CuSO_4$ as herbicide in farmlands.

The lower concentrations of these metals recorded at the control sites for all the metals in both soil and plantain leave samples implicated the mining activities as sources of these metals. Heavy metals have been reported to be present in parent materials (such as rock, and minerals) in particularly high concentrations

Table 1. Mean concentrations (mg/kg) of selected heavy metals in soils around mining and non-mining (control) sites in southwest Nigeria.

Settlements	Sample locations	Pb	Cd	Cu	Zn	As	Cr	Mn	Fe
IGUN	IG1	16.21 ± 1.80	223.25 ± 9.60	162.43 ± 4.36	128.46 ± 11.25	19.72 ± .81	121.37 ± 10.31	245.44 ± 9.54	4959.07 ± 131.41
	IG2	14.42 ± 1.67	225.47 ± 7.20	167.30 ± 3.66	129.74 ± 10.25	20.7 ± 32.75	91.03 ± 7.91	198.87 ± 8.22	5008.97 ± 139.17
	IG3	19.12 ± 2.39	272.35 ± 8.62	191.67 ± 9.12	116.96 ± 8.26	18.92 ± 1.65	106.87 ± 3.25	289.61 ± 10.15	4016.96 ± 120.24
ITAGUNMODI	IT1	24.31 ± 2.88	346.03 ± 10.12	284.26 ± 4.36	244.08 ± 13.25	25.04 ± 1.82	78.31 ± 3.15	196.34 ± 8.25	8678.62 ± 206.09
	IT2	21.87 ± 2.65	304.50 ± 7.85	295.63 ± 4.88	222.10 ± 11.29	23.78 ± 2.62	68.87 ± 6.89	192.42 ± 10.10	8765.45 ± 210.11
	IT3	20.65 ± 2.72	328.73 ± 10.02	264.36 ± 2.77	427.13 ± 13.26	27.38 ± 3.94	71.27 ± 9.55	204.20 ± 8.29	6161.66 ± 140.25
SABO	SB1	12.15 ± 2.25	158.50 ± 9.71	157.56 ± 1.51	211.51 ± 7.65	14.08 ± 1.42	46.98 ± 5.16	134.98 ± 5.00	4711.57 ± 129.74
	SB2	11.05 ± 2.20	166.42 ± 7.54	173.35 ± 1.85	192.15 ± 5.36	12.59 ± .81	42.28 ± 10.62	118.78 ± 5.54	4475.04 ± 122.51
	SB3	21.26 ± 3.30	245.68 ± 8.22	149.68 ± 2.74	236.67 ± 5.96	12.32 ± 2.19	47.45 ± 8.59	103.75 ± 5.21	5888.21 ± 139.12
CONTROL		9.39 ± 3.60	147.46 ± 4.04	125.72 ± 1.15	116.01 ± 3.69	10.84 ± 1.55	21.64 ± 10.11	84.94 ± 4.30	5352.06 ± 141.32
MAL		100	3	100	300	20	50	2000	2000

MAL: Maximum Allowable Limit (WHO, 2004).

Table 2. Mean concentrations (mg/kg) of selected heavy metals in plantain leaves around mining and non-mining (control) sites in southwest Nigeria.

Heavy metals	IGUN			ITAGUNMODI			SABO			Control	LIMITS	
	IG1	IG2	IG3	IT1	IT2	IT3	SB1	SB2	SB3		NMC	WHO
Pb	6.02 ± 0.55	5.41 ± 0.44	5.23 ± 0.42	7.56 ± 0.63	6.44 ± 0.56	7.69 ± 0.63	4.51 ± 0.35	3.97 ± 0.29	4.73 ± 0.36	3.54 ± 0.27	5 - 10	2
Cd	12.84 ± 1.34	13.48 ± 1.13	11.42 ± 0.96	21.82 ± 1.83	22.05 ± 1.90	18.54 ± 1.56	9.12 ± 0.79	8.02 ± 0.70	6.48 ± 0.58	5.18 ± 0.45	0.1 - 2.4	--
Cu	54.95 ± 4.88	48.36 ± 4.22	53.29 ± 4.65	59.34 ± 5.21	44.51 ± 3.90	51.63 ± 4.34	41.21 ± 3.48	25.13 ± 2.18	36.26 ± 3.13	30.82 ± 2.66	5 - 20	10
Zn	20.09 ± 1.73	20.29 ± 1.74	16.27 ± 1.39	30.34 ± 2.31	22.75 ± 1.90	27.91 ± 2.51	16.27 ± 1.47	12.36 ± 1.06	14.32 ± 1.21	10.51 ± 0.86	1 - 40	5
As	3.03 ± 0.26	2.75 ± 0.19	1.99 ± 0.17	3.78 ± 0.31	2.91 ± 0.21	3.48 ± 0.25	2.26 ± 0.19	2.50 ± 0.18	2.07 ± 0.16	1.96 ± 0.15	0.02 - 5	2
Cr	4.37 ± 0.37	4.06 ± 0.36	3.53 ± 0.26	5.47 ± 0.48	4.2 ± 0.36	5.02 ± 0.45	3.53 ± 0.26	2.72 ± 0.19	3.61 ± 0.27	2.29 ± 0.17	0.03 - 14	1 - 30
Mn	7.23 ± 0.63	6.36 ± 058	5.71 ± 0.53	10.12 ± 0.87	8.9 ± 0.76	10.63 ± 0.87	8.68 ± 0.78	8.42 ± 0.76	7.72 ± 0.61	7.02 ± 0.59	--	--
Fe	78.53 ± 6.57	69.1 ± 6.07	73.03 ± 6.13	66.75 ± 5.68	82.51 ± 7.34	73.42 ± 6.14	68.32 ± 6.02	48.51 ± 4.29	62.17 ± 5.44	40.39 ± 3.47	--	20

NMC (Normal Metal Concentration), [19].

[19] and will be released into the environment by anthropogenic activities such as mining. High value of heavy metal, particularly Cu, Fe and Zn found in the soil samples compares well with those reported in Bagana mining area, Zamfara state, Nigeria [20]. Similar study carried out in one of the studied areas also reported heavy pollution with these heavy metals [21]. Cu is known to be particularly adsorbed by soil and plant [22] while the high Fe content in the soil samples can be attributed to the high percentage of the metal in the earth crust. The required low organic matter mineralization for effective plant growth will be disturbed as a result of negative effects of high concentrations of heavy metals on microbial activities in the soil and this is undesirable. The high concentration of

these metals recorded in plantain leaves is also a confirmation of their accumulation by plants. High values of heavy metals recorded in this study for plantain leaves did not however compare well with those reported for plant materials in a neighbouring mining site in the same local Government area [23]; this might not be unconnected with the relatively low rate of artisanal mining compared to the study site.

4.2. Analysis of Variance

The results of the analysis of variance that was used to investigate spatial variations in the heavy metal concentrations across the selected locations indicate significant variations in soils (**Table 3**) and plantain leaves (**Table 4**). Except for Fe which indicated a non-significant difference between Igun and Itagunmodi on one hand and between Igun and Sabo on the other hand, As (between Sabo and the control), Zn (between Igun and the control) and Cu (between Igun and Sabo), the overall variance showed significant differences in the heavy metal concentrations in soils at the locations.

4.3. Temporal Variations

Figure 2 and **Figure 3** shows the monthly distributions of selected variables in soils and plantain leaves. The metals generally occurred in higher concentrations in the dry months (November - March) than the wet months (April-July), probably due to the influence of rainfall that may be linked with a wash-off or dilution effects. Lower values of the metals observed in samples from control sites is an indication of contribution of mining activities to the heavy metal level in the study areas since other anthropogenic sources that might be responsible for the presence of high values of these metals such as presence of waste dump sites [24], leaching from incineration points, discharge from industries, leaching from smelting and fabrication [25] etc. are obviously absent in the areas. The

Table 3. Results of the Analysis of Variance of selected heavy metals in soils around mining and non-mining sites across the different locations.

Selected heavy metals	Multiple comparison						Overall ANOVA	
	Igun			Itagunmodi		Sabo		
	Itagunmodi	Sabo	Control	Sabo	Control	Control	F-Value	F-Probability
Pb	0.004*	0.724*	0.014	0.001*	0.001*	0.104*	14.82	0.001*
Cd	0.001*	0.001*	0.001*	0.001*	0.001*	0.005*	125.70	0.001*
As	0.001*	0.001*	0.001*	0.001*	0.001*	0.566	59.22	0.001*
Cu	0.001*	0.406	0.001*	0.001*	0.001*	0.028*	112.87	0.001*
Zn	0.001*	0.062	0.998	0.082	0.082	0.211*	11.39	0.001*
Fe	0.896	0.940	0.001*	0.578	0.001*	0.001*	10.77	0.001*
Cr	0.001*	0.001*	0.001*	0.001*	0.001*	0.007*	79.76	0.001*
Mn	0.002*	0.001*	0.001*	0.001*	0.001*	0.003*	40.43	0.001*

Table 4. Results of the Analysis of Variance of selected heavy metals in plantain leaves across the different locations.

Metals	Multiple Comparison						Overall ANOVA	
	Igun			Itagunmodi		Sabo	F-Value	F-Ratio
	Itagunmodi	Sabo	Control	Sabo	Control	Control		
Pb	0.04*	0.273	0.001*	0.001*	0.001*	0.06	23.78	0.001*
Cd	0.001*	0.001*	0.001*	0.001*	0.001*	0.01*	30.76	0.001*
As	0.001*	0.001*	0.001*	0.001*	0.001*	0.66	98.25	0.001*
Cu	0.001*	0.406	0.001*	0.001*	0.001*	0.03*	14.63	0.001*
Zn	0.001*	0.062	0.998	0.082	0.002*	0.21	25.16	0.001*
Fe	0.896	0.940	0.001*	0.578	0.001*	0.001*	0.985	0.336
Cr	0.001*	0.001*	0.001*	0.001*	0.001*	0.01*	26.25	0.001*
Mn	0.002*	0.001*	0.001*	0.001*	0.001*	0.003*	73.76	0.001*

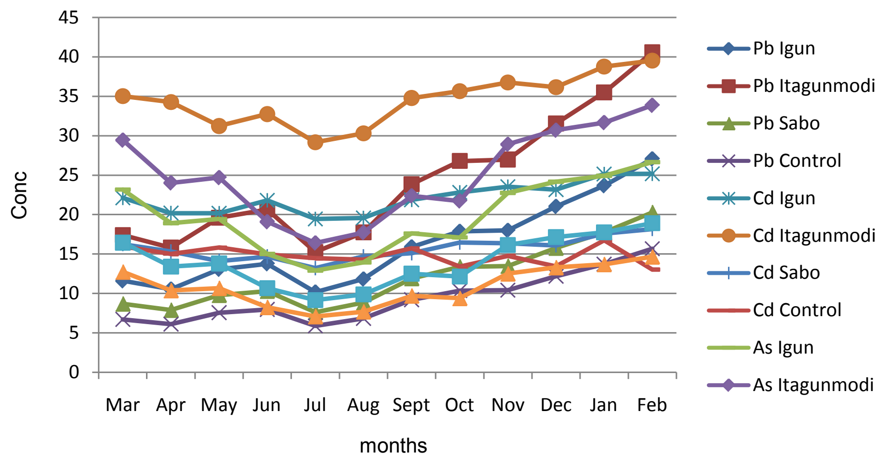

Figure 2. Monthly distribution of Pb, Cd and As in soil samples.

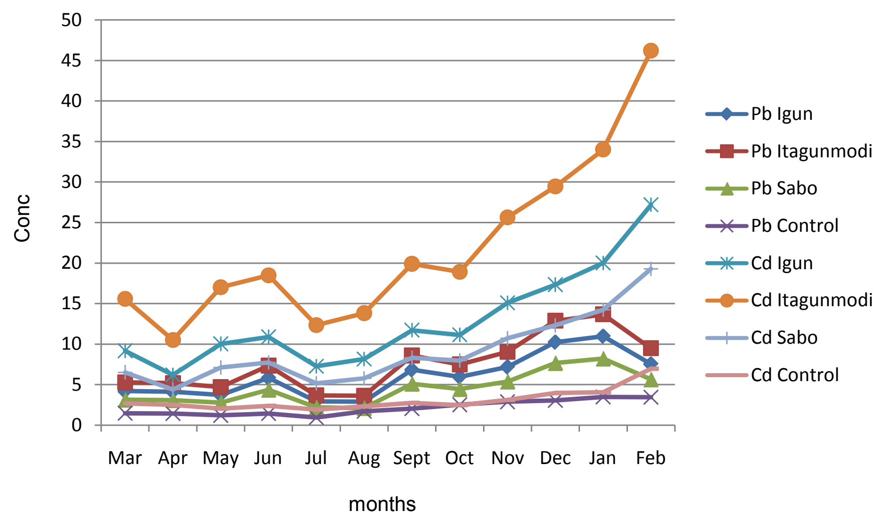

Figure 3. Monthly distribution of Pb and Cd in plantain leaves.

study concluded that high concentrations of the metals resulted from excessive digging for the gold metal which brings the core of the soil, and increases their concentrations.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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