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# Effects Of Artisan Coal Mining On The Bioaccumulation Profile Of Heavy Metals In Medicinal Plants In Akwuke And Iva Valley Communities Enugu State, Nigeria

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**ABSTRACT**
The effects of artisan coal mining on the bioaccumulations profile of the heavy metals in medicinal plants of Akwuke and Iva valley mines has been investigated. The study focused on emphasizing the heavy metal status and bioaccumulation factors of potentially toxic metals (lead, mercury and arsenic) and essential metals (copper and manganese) in three medicinal plants (Veronia Amygdalina (Bitter leaves) Carica papaya (Pawpaw), and Azadirachta Indica (Neem Leaf) plants grown under two different areas of Akwuke and Iva valley near coal mines and those in fertile soils of Amorji Nike all in Enugu State of Nigeria. Also investigated are the physicochemical and phytochemical compositions of the soil and the leaves of the plants mentioned above. The results shown that the phytochemicals (Saponins, Terpenes, Tannins, Flavonoid, Glycoside and Alkaloids) were present at reduced values in the three medicinal plants grown near the two artisan coal mine areas. It also revealed that the mining activities reduced the medicinal values of these plants correlated with very lower values in the two mines areas. The bioaccumulation factors of the heavy metal were estimated to understand the soil to plant transfer pattern of the heavy metals. Results revealed significantly that metals investigated were founds in the medicinal plants in Akwuke than Iva valley mining areas indicating that majority of these metals are precipitated and were immobilized. The presence of these heavy metals in the medicinal plant is a health risk when the plants are used for treatment of diseases. It can be concluded that the artisan coal mining activities in the two communities in Enugu reduces the medicinal value of the three plants and resulted in heavy metals decompositions in the soil which bioaccumulated into the medicinal plants. These suggest that the soil near coal mine areas require proper treatment prior to cultivation in order to reduce the heavy metals concentration in the soil, land degradation and pollution.

**Keywords**: Artisan coal mining, Heavy metals, Bioaccumulation, Medicinal plants, Phytochemicals

## I. INTRODUCTION

Medicinal plants that have at least one of their parts (leaves, stem, barks, or roots) used for therapeutic purposes (Bruneton, 1993). The availability and relatively cheaper costs of medicinal plants in sub-saharan Africa, makes them more attractive as tonic agents when compared to modern medicines (Agbor and Ngogang, 2005; Agbor et al, 2005). The World Health Organization (WHO) has estimated that 65 to 80% of the world's populations depends on traditional medicine preparations as their primarily form of healthcare (WHO, 2002; Annan et al, 2013; Ezuruike and Prieto, 2014). Contamination of medicinal plants by heavy metals is of major concern because of the toxicity, persistence and bioaccumulative nature of such metals. Bioaccumulation of heavy metal in medicinal plants as a results of artisanal mining activities could disrupt functions of vital organs and glands in the human body such as brain, kidney and liver; leading to long term toxicities (Ray and Ray,2009). Illegal mining activities could thus lead to emission of heavy metal into environments, and consequently to heavy metal toxicity of medicinal plants used in traditional medicine preparations (Nagajyoti et al, 2001).

Heavy metal are non-biodegradable and persistent environmental contaminants, which may be adsorbed from contaminated soils via roots uptakes, or deposited on the surfaces and then adsorbed into tissue of most plants (Singh et al, 2011; Dzomba et al, 2011). Thus, medicinal plants can present health risks due to heavy metal contents of these plants (Nagajyoti et al, 2010; Annan et al, 2001).

Mining is an excavation made in the earth for the purpose of extracting ores, coal, precious stones, etc. a place where such minerals may be obtained, either by excavation or by washing the soil. The African continent had about 30% of the world's mineral resources, and possesses the largest known reserves of strategically important minerals, including gold. Nigeria is one of the countries in the sub-Saharan Africa where mining formed huge sources of export prior to oil boom period in 1970s (Ericsson, 2011). National interest in mining started to decline, since the oil boom period, causing increased sporadic, informal uncoordinated or unmonitored management of the existing and potential mines that resulted into intensified artisanal mining activities in the country (Edwards. 2014).

Artisanal mining is generally a small-scale practice where the basic tools and manual labour are generally used for excavation (Canavesio, 2019). Artisanal mining activities are also an informal procedure, which though have also been linked with economic benefits (including employment opportunities, tourism technology advancement, accessibility to both native and migrant population) Canavesio, 2014.

The informal mining activities are characterized by low productivity, a lack of capital, poor technology, hazardous working conditions, land degradation, and pollution (EMEL, 2011).

Artisanal mining practices are common in Africa, and many researchers have investigated their attributes. For example, Hilson (2001) provides information about the working of the small-scale mining industry in the Ghana, and argued that initiatives have recently been taken to regularize and formalize the activities of the industry, with the intention of reducing the associated environmental impacts and land-use-conflicts. Except for the recent efforts by the Federal Ministry of Mine and Steel Development, gold mining in Nigeria is largely uncontrolled, and the majority of the operators are unlicensed (Oramah, 2015). Bartem, (2014) indicated that mine sites are around farmlands where chemicals may accumulate in fruits and leaves of arable and cash crops, and that soil contamination in mine sites can cause severe heavy metal contamination of water sources and poisoning by materials associated with mining has been associated with increased cases of kidney pain, respiratory problems, dizziness, and miscarriage in women, and deaths in many residents of communities where artisan mining activities are carried out.

## II. MATERIALS AND METHODS

**Materials**

The soil samples were collected from different artisan coal mining areas (Iva Valley and Akwuke ) in Enugu state of Nigeria.
Also collected were three different medicinal plants (Veronia amygdalina (Bitter leaves) Carica papaya (Pawpaw) and Azadirachta indica (Neem leaf) at the coal mining areas.

**Methods for Heavy Metal Analysis**

Heavy metal analysis was conducted using Varian AA240 Atomic Absorption Spectrophotometer according to the method of APHA 1995 (American Public Health Association)

Atomic absorption spectrometer's working principle was based on the sample being aspirated into the flame and atomized when the AAS's light beam is directed through the flame into the monochromator, onto the detector that measures the amount of light absorbed by the atomized element in the flame. Since metals have their own characteristics absorption wavelength, a source lamp composed of that elements is used, making the method relatively free from spectral or radiation interferences. The amount of energy of the characteristics wavelength absorbed in the flame is proportional to the concentration of the element in the sample.

**Dry Preparation of Minerals**

2g of the samples were put in a muffle furnace and heated for 2hrs at $550^0$C. The heated samples were the diluted with 20ml, 20% $H_2SO_4$, and filter with filter paper and make it up to 50ml mark with distilled water..

**Preparation of References Solution:**

A series of standard metal solution in the optimum concentration range was prepared; the references solutions were prepared daily by diluting the single stock element solutions with water containing 1.5ml consecrated nitric acid\liter. A calibration blank was prepared using the entire reagent except for the metal stock solutions. Calibration curve for each metal was prepared by plotting the absorbance of standards against their concentrations.

**Nitrate Determination**

Nitrate was determined using PD303UV spectrometer (APHA, 1998): A known volume

(50ml) of the samples was pipette into a porcelain dish and evaporated to dryness on a hot water. 2ml of phenol disulphonic acid was added to dissolve the residue by constant stirring with a glass rod. Concentrated solution of sodium hydroxide and distilled water were added with stirring to make it alkaline. This was filtered into a nesslers tube and made up to 50ml with distilled water, the absorbance was read at 410nm using a spectrophotometer after the development of color. The standard graph was plotted by taking concentration along X-axis and the spectrophotometer readings (absorbance) along Y-axis, the value of nitrate was found by comparing absorbance of sample with the standard curve and expressed in mg/l.

Conc. of sample= abs of sample x Conc. of std (1)

Abs of std

**Nitre** = 1ml of zinc Sulphate was added and made up to 50ml with distilled water

**Measurement of Acidity**

The acidity was measure by placing soil samples about ¾ full in sample jar and distilled water was added to cover the soil. The jar was cap and shake for about 20 minutes. The mixture was allowed to stand for 10 minutes to dissolve the salts in the soil and the solution was calibrated with a pH 7 and a pH 10 buffer solution. At the end the cap was removed and the wet soil slurry was tested by placing the pH tester and the pH were recorded.

**Measurement of Alkalinity:**

The 50ml burret was rinsed several times with 0.2N HCL and 100ml solution of soil samples were analyzed into a 250 ml Erlenmeyer flask and titrated to a bromocresol green (pH = 4.5) as end point. Alkalinity is expressed in terms of milligram of calcium carbonate per liter.

Alkalinity = (ml HCL titrant) x (normality of HCL) x (50,000) (2)

(ml of water samples)

**Phosphate Determination**

Phosphate was measured using standard method 4500-P B.5 and 4500-PE (APHA; 1998)

Exactly 100ml of the homogenized and filtered sample was pipetted into a conical flask. The same volume of distilled water (serving as control) was also pipette into another conical flask. 1ml of 18M $H_2SO_4$ and 0.89g of ammonium per sulphate were added to both conical flasks and gently boiled for $1_{1/2}$ hours, keeping the volume of 25-50cm$^3$ with distilled water.

It was then cooled, one drop of phenolphthalein indicator was added and after neutralized to a faint pink color with the 2M Na0H solutions. The pink color was discharged by drop wise addition of 2M HCL, and the solutions made up of 100ml with distilled water. For the colorimetric analysis, 20ml of the samples was pipette into test tubes, 10ml of the combined reagent added, shaken and left to stand for 10 min before reading the absorbance are 690nm on a spectrophotometer, using 20ml of distilled water plus 1ml of the reagent as references. Standard phosphate solution: 219.5mg of dried AR potassium hydrogen phosphate was dissolved in distilled water and made up to 1000ml, where 1ml= 50.0ug.of phosphate. 10ml of the stock solutions was made up to 1000ml to give 1ml = 0.05mg. Standard of strength ranging from 0 (blanks) to 0.05mg/L at intervals of 0.01mg is prepared by diluting the stock with distilled water.

Conc. of sample= abs of sample x Conc. of std (3)

Abs of std

**Sulphate Determination**

Sulphate analyzed according to ALPHA standard method (APHA; 1998)

A 20cm$^3$ of the sample was added with 10mlof 10% barium chloride and 10ml of conditioned reagents. It was allowed to stand for 30 min for color developments and the absorbance was read at 880nm at the UV spectrophotometer.

**NB**: The soil sample was diluted in1:10 dilutions.

**Plants and Soil heavy metal analysis**

Plants and soil heavy metal analysis were carried out using the methods of Kalagbor et al.(2014) and Masvodza et al.(2014) respectively. Wet digestion is used in the preparation of the plant samples to avoid loss of some of the metal by high temperature (dry) ashing. The heavy metal load (Pb, Zn ,Fe, Cu, Hg, As, and Mn ) in the medicinal plants samples (leaves and root) and the soils will be determined using atomic absorption spectrometer.

**Bioaccumulation factor (BAF) and translocation factor (TF)**

Bioaccumulation factor (BAF) is the ability of the plants to accumulate the heavy metals with respects to the metal concentration in the ecosystem. Translocation factor (TF) is the plant's ability to translocate heavy metal from root to harvestable aerials part.(Oti, 2015a). Assessments of bioaccumulation factor (TF) of the metals in the plats will be determined using the methods of Masvoda et al. (2013) and Oti (2015a) respectively.

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The index of the plant's ability to accumulate metals from soils is calculated as follows:

$$BAF = \frac{CPLANT}{CSOIL}$$

Where $C_{plants}$ and $C_{soil}$ represents the heavy metal concentration in plants parts and soils respectively. Translocations factors are expressed as:

$$TF = \frac{Cshoot}{Croot}$$

Where $C_{shoot}$ and $C_{root}$ is the concentration of metal in short and root respectively.

**Characterization of the Extracts**

The phytochemical analysis of the leaf extracts obtained was carried out using American Society for Testing Methods, ASTM (D4903). Both qualitative and quantities analysis of the leaf extracts were carried out using methods described in the literature to detect and determine the chemical compositions such as Saponins, Terpenes, Tannins, Flavonoid, Phlobatannis, Anthraquinones, Glycoside and Alkaloids.

## III. RESULTS AND DISCUSSIONS

**Table 1: Physicochemical analysis of Iva Valley and Akwuke soils**

| Parameter | Sample A (Iva Valley artisan coal mine area) | Sample B (Akwuke artisan coal mine area) | EPA Standard (WHO) |
|---|---|---|---|
| Acidity | 50mg/kg | 35mg/kg | 40mg/kg |
| Alkalinity | 25mg/kg | 75mg/kg | 50mg/kg |
| Nitrate | 2.79mg/kg | 8.22mg/kg | 50mg/kg |
| Nitrite | 3.14mg/kg | 7.49mg/kg | 4mg/kg |
| Phosphate | 1.31mg/kg | 2.01mg/kg | 6.5mg/kg |
| Sulphate | 91.22mg/kg | 103.72mg/kg | 80mg/kg |
| pH | 4.6 | 4.8 | 4.5-6.5 |

**Key**: EPA-Environmental Protection Agency, WHO -World Health Organization

Soils are considered safe and not contaminated if the concentrations of undesired substances do not exceed the WHO safe limit. In Iva Valley artisan coal mine area (sample A), parameters such as acidity and sulphate exceeded the permissible limit with others such as alkalinity, nitrate, nitrite and phosphate fall below permissible limit. Whereas, in Akwuke artisan coal mine area (sample B), parameters such as alkalinity, nitrite and sulphate exceeded the permissible limit with others such as acidity, nitrate and phosphate fall below permissible limit.

The differences are attributed to various bottom sediments of organic matters and localized coal mining activities in the area. However, the pH level indicated normal for the both artisan coal mine areas studied.

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**Table 2: Phytochemical analysis of leave extracts of control area (Amorji Nike Enugu)**

| S/N | Phytochemical | Paw-paw leave Extracts (% $\frac{w}{w}$) | Neem leave Extracts (% $\frac{w}{w}$) | Bitter leaf Extracts (% $\frac{w}{w}$) |
|---|---|---|---|---|
| 1 | Saponins (% $\frac{w}{w}$) | 5.983 | 7.087 | 6.170 |
| 2 | Terpenes (% $\frac{w}{w}$) | 7.762 | 9.762 | 8.211 |
| 3 | Tannins ($\frac{mg}{100g}$) | 7.432 | 9.969 | 8.311 |
| 4 | Flavonoid (% $\frac{w}{w}$) | 2.362 | 5.821 | 2.992 |
| 5 | Phlobatannis (% $\frac{w}{w}$) | - | - | - |
| 6 | Anthraquinones (% $\frac{w}{w}$) | - | - | - |
| 7 | Glycoside ($\frac{mg}{100g}$) | 0.143 | 1.582 | 0.876 |
| 8 | Alkaloids (% $\frac{w}{w}$) | 3.328 | 6.333 | 3.758 |

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Table 2 revealed the phytochemical analysis of leave extracts of the control area. The results indicate that saponins, terpenes, tannins, flavonoid, glycoside and alkaloids were present while phlobatannis and anthraquinones are absent in the three leaves studied. The chemical structures of most of these phytochemical contained electron rich bond or hetero atoms that facilitated their electron donating ability.

**Table 3: Phytochemical analysis leave extracts from coal mining area (Iva Valley)**

| S/N | Phytochemical | Paw-paw leave Extracts (% $\frac{w}{w}$) | Neem leave Extracts (% $\frac{w}{w}$) | Bitter leaf Extracts (% $\frac{w}{w}$) |
|---|---|---|---|---|
| 1 | Saponins (% $\frac{w}{w}$) | 4.583 | 5.647 | 3.976 |
| 2 | Terpenes (% $\frac{w}{w}$) | 5.887 | 7.364 | 6.631 |
| 3 | Tannis ($\frac{mg}{100g}$) | 6.113 | 6.766 | 5.754 |
| 4 | Flavonoid (% $\frac{w}{w}$) | 2.007 | 4.432 | 1.096 |
| 5 | Phlobatannis(% $\frac{w}{w}$) | - | - | - |
| 6 | Anthraquinones (% $\frac{w}{w}$) | - | - | - |
| 7 | Glycoside($\frac{mg}{100g}$) | - | 0.998 | 0.453 |
| 8 | Alkaloids (% $\frac{w}{w}$) | 2.098 | 4.421 | 1.9788 |

**Table 4: Phytochemical analysis leave extracts from coal mining area (Akwuke)**

| S/N | Phytochemical | Paw-paw leave Extract(% $\frac{w}{w}$) | Neem leave Extract(% $\frac{w}{w}$) | Bitter leaf Extracts (% $\frac{w}{w}$) |
|---|---|---|---|---|
| 1 | Saponins (% $\frac{w}{w}$) | 3.875 | 5.009 | 3.875 |
| 2 | Terpenes(% $\frac{w}{w}$) | 5.665 | 6.876 | 5.654 |
| 3 | Tannis ($\frac{mg}{100g}$) | 5.878 | 5.456 | 5.223 |
| 4 | Flavonoid (% $\frac{w}{w}$) | 2.547 | 3.765 | 2.342 |
| 5 | Phlobatannis(% $\frac{w}{w}$) | - | - | - |
| 6 | Anthraquinones (% $\frac{w}{w}$) | - | - | - |
| 7 | Glycoside($\frac{mg}{100g}$) | - | 1.008 | 0.675 |
| 8 | Alkaloids (% $\frac{w}{w}$) | 1.965 | 3.542 | 1.675 |

Table 3 and 4 demonstrated the phytochemical analysis of leave extracts of the Iva Valley coal mining area The results indicate that saponins, terpenes, tannins, flavonoid and alkaloids were present at a reduced values while phlobatannis and anthraquinones are absent in the three leaves studied. Glycoside indicated absent in paw-paw leave extracts but present at reduced values in neem leave and bitter leaf **e**xtracts. This indicated that the deposition of free radicals due to artisan coal mining activities in those areas reduced the formation of these phytochemical in the medicinal plants grown in those areas. This is in agreement with other studies.

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**Heavy Metal Analysis**

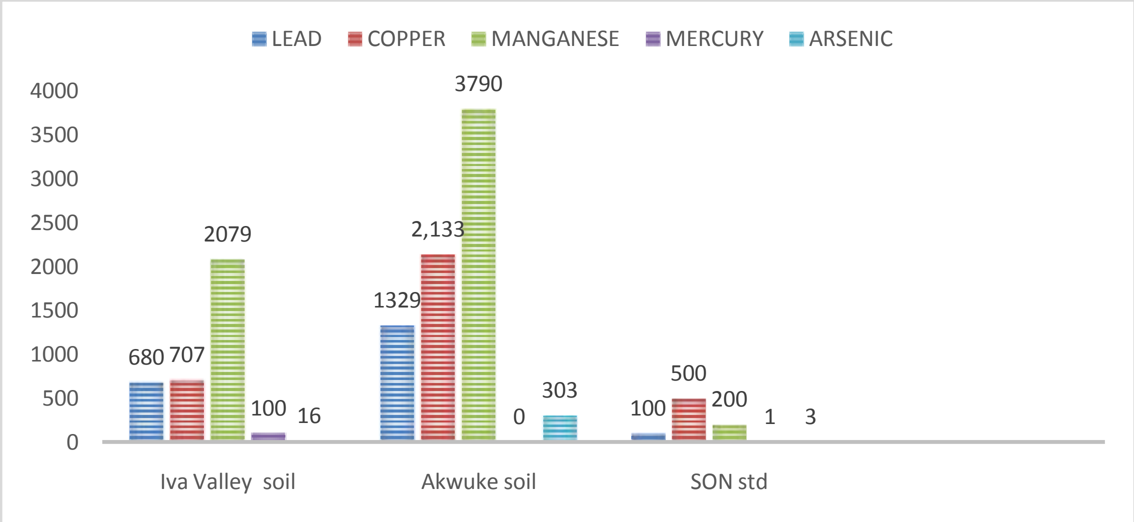


**Figure 1**: Comparative studies of coal mine areas of Iva Valley and Akwuke soil

All the heavy metals investigations showed higher values as presented in figure 1 above. The metals analyzed are lead, copper, manganese, mercury and arsenic. These metals, due to their toxic effects, impose danger to the environments which when absorbed into the plants grown in the area could disrupt functions of vital organs and glands in the human body such as brain, kidney and liver; leading to long term toxicities (Ray and Ray,2009). The results revealed that most of the heavy metals determined exceeded the permissible limits of sets standards except manganese which indicated absent in Akwke mine area.

**Bioaccumulation factors (BAFs)**

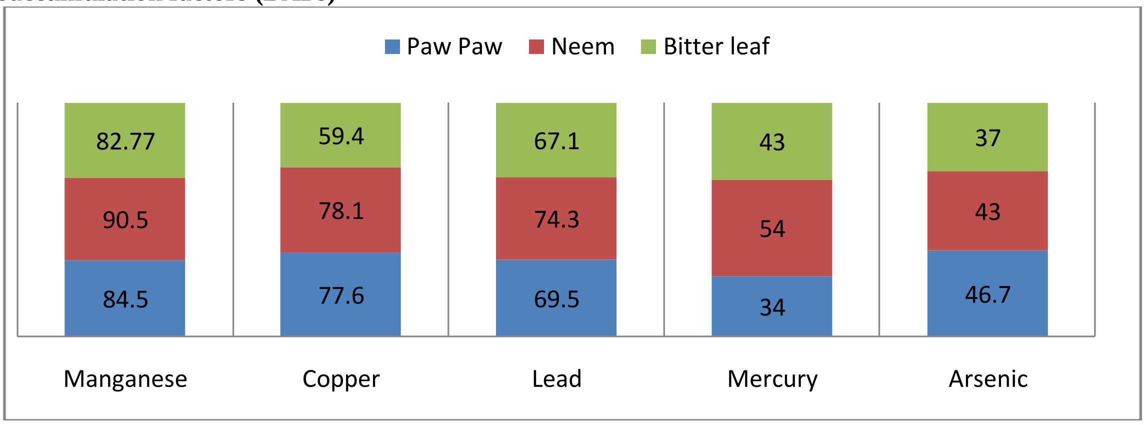


**Figure 2**: Bioaccumulation factor of heavy metals in Iva Valley coal mine area in Enugu

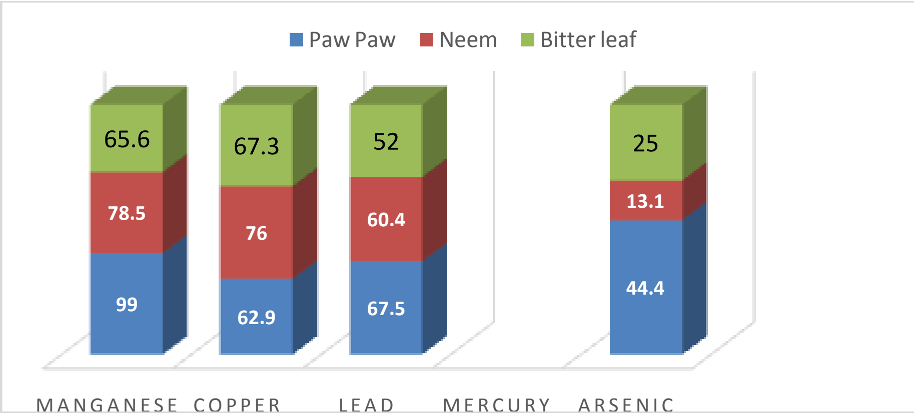


**Figure 3**: Bioaccumulation factors of heavy metals in Akwuke coal mine area in Enugu

Accumulation of the heavy metals in the plants was assessed by bioaccumulation factor (BAF) which helps in comparing this ability of different plants in taking up metals from soil (kovaeik et al, 2013). In this present study most of the medicinal plants revealed relatively moderate BAF values while some lower BAF (on the average which showed limited ability of the plants to accumulate the metals).

In the Iva Valley artisan coal mine area, manganese accumulation was highest, followed by copper, lead and mercury with arsenic the lowest in the three medicinal plants investigated. While in Akwuke artisan coal mine area, the result revealed manganese accumulation was highest, followed by copper, lead and arsenic with mercury the absent in the three medicinal plants investigated. This indicated that some of the heavy metal were precipitated and immobilized into the soil.

## IV. CONCLUSION

The study had revealed that artisan coal mining activities in Iva Valley and Akwuke communities contaminate the soil near the area investigated.

The heavy metals bioaccumulated into the plants grown on those areas. It is an evident the medicinal plants in the area contain some metals like manganese, copper, lead, mercury and arsenic which confirm that the plants are not clinically suitable to be used for treatment unless the metals concentration are reduced to acceptable limits. Due to the presence of heavy metals and acidity level in the soil of Iva Valley and Akwuke communities, the soil need to be ameliorated before usage for any cultivation.

### Recommendation

Based on the outcome of the study the following are recommended;

1) It is recommended that regular monitoring of artisan mining activities by appropriate authorities be conducted in the study area this will ensure that incidences of contamination are noticed earlier for remedial action to be taken.
2) The communities should be educated on the dangers of this artisan coal mining activities.
3) Recommend that more research work should be carried out to determine the translocation factor and occurrence of oxidative stress in the medicinal plants in those areas.

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