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Bioaccumulation and Health Risk Assessment of Heavy Metals in *Tectona grandis* LINN and *Gmelina arborea* Roxb around Ratcon Quarry site in Ibadan, Oyo State Nigeria.

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ABSTRACT

Global public health concerns associated with environmental contamination by Arsenic, Copper, Manganese, and Zinc among other metals are on the increase due to rising human exposure to these Heavy Metals through mining activities.The quarry mining operations cause the leaching of heavy metals into groundwater, and surface water,and further release into the ambient air.The increased activities of quarries in Oluyole due to the high demand for aggregates (sand, gravel, excavated rocks) can hurtan already stretched environment. This study was carried out to investigate heavy metals concentrations: copper (Cu), cobalt (Co), lead (Pb), cadmium (Cd), zinc (Zn), nickel (Ni), and manganese (Mn) in Tectona grandis, Gmelina arborea and soil samples from Ratcon quarry site in Ibadan. The plant leaf and soil samples were assessedfor heavy metal concentrations using atomic absorption spectrophotometer. The health risk index (HRI) of the heavy metals following the consumption of these plant materials by the populace for medicinal uses was determined using standardmethods. The concentrations of heavy metals in the leaf samples ranged from 0.030–15.300mg/kg, whereas that of the soil sample was within the range of 6.194– 40.001 mg/kg. Ni concentrations in the soil and leaf samples were above the maximum permissible level according to the World Health Organization (WHO). The bioaccumulation of the heavy metals followed the trend: Ni > Co > Zn > Mn > Cu > Cd (T. grandis); Zn > Ni > Co > Mn > Cu > Cd (G. arborea). The highest bioaccumulation of the heavy metals (Ni) occurred in T. grandis, lowest bioaccumulation of heavy metals (Cd) occurred in G. arborea. The estimated HRI of Ni in T. grandis and G. arborea were greater than 1. T. grandis-based herbs from the quarry contained Ni at a toxic level, whereas Cu, Cd, Zn, Pb, Co, and Mn concentrations were relatively within safe limits.

Keywords: Bioaccumulation, Tectona grandis, Heavy Metals, Gmelina arborea,

Introduction

Heavy metals and metalloids are the natural and structural parts of the earth's crust with a density greater than 5 g/cm3. They are environmentally persistent and non-degradable contaminants. They are deposited on the soil surface, then absorbed by the apoplast of plant roots and further distributed and accumulated into their edible and non-edible parts, with an imminent danger to man (Ahmad *et al.,* 2019; Alsafran *et al.,* 2021).Developing countries like Pakistan (Alam *et al.,* 2018), Bangladesh (Islam *et al.,* 2016), Ethiopia (Gebeyehu and Bayissa, 2020), Ghana (Ametepey *et al.,* 2018), South Africa (Fonge *et al.,* 2021), and India (Yadav *et al.,* 2015) have reported high levels of heavy metal in there urban areas due to rapid industrialization, wastewater irrigation, and other anthropogenic activities.

Tectona grandis Linn (teak) is a tropical plant with a wide range of applications. Teak is one of the best hardwoods on the planet; it can be harvested as early as the sixth year after they are planted. Teak species belonging to the family Verbenaceae, have a vital role in soil nutrients and properties. Exotic tree species have been found to have effects on soil properties differing from those of indigenous natural forests in terms of organic matter accumulation, soil conditions, and type of vegetation growing on the forest floor (Kai and Jiao-Jun, 2015). The soil system strongly influences the structure and function of ecosystems and acts as a buffer against global climate change (Pareek, 2017).

Gmelina arboreaRoxbis a deciduous tree, it belongs to the family Verbenaceae (Wang. 2004). Extracts from the leaves, fruits, and seeds of Gmelina arborea Roxb. Has been reported from various sources to contain bioactive chemicals called phytochemicals. These include alkaloids, steroids, anthrax quinones, glycosides, triterpenoids, saponins, phenolic compounds, flavonoids, proteins, and carbohydrates (Nayak *et al*, 2012).

A quarry isthe exploitation of various lithologic materials given by nature to mankind. It is a place from which dimension stones, rocks, construction aggregates, sand, gravel, or slate have been excavated from the ground.

The Periodic table consists of heavy metals to a notable portion with high density and atomic weight. Among them, the majority in the biosphere, such as in water, soils, and rocks, are also released into the surroundings from anthropogenic resources, mostly commercial and industrial. (Azeh Engwa, 2019).The toxic principles of heavy metals have been known for decades. However, recent experimental investigations show that some, including nickel, copper, and zinc, are vital for humans and are widespread in nature(Azeh Engwa, 2019).

Dermal, respiratory, reproductive, immunological, neurological, liver cancer, genotoxic, and mutagenic effects are some of the human health problems linked with heavy metals poisoning (Ahmad *et.al*, 2019).

Quarrying has been the major source of construction materials globally. These activities are accompanied by the release of large quantities of heavy metals such as lead (Pb), cobalt (Co), cadmium (Cd), zinc (Zn), nickel (Ni), copper (Cu), manganese (Mn), and so on. Iroye and Igbozurike,(2018), reported that quarry activities caused the release of certain heavy metals such as Cd Cr, Ni, and As into the river Ona which are above WHO permissible limit.

This study was carried out to investigate heavy metal concentrations: Pb, Co, Cd, Zn, Ni, Cu, and Mn in Tectona grandis LINN and Gmelina arborea and soil samples from the Ratcon quarry site. Furthermore, the present study ascertained the bioaccumulation factor (BF) and related toxicological indices to establish the levels of human exposure to heavy metals within the residential locations of the quarry with health risk indicators associated with the consumption of these medicinal plants.

Materials and Methods

Description of the study area

Oluyole Local Government was established in 1976 and the Council occupies about 629 square kilometers. Based on the 2006 population Census its population is 202,725, however, the projected populationbased on the 2006 census is 290,800. The Agricultural products in the area include cocoa, citrus, cassava, maize and so on. The area is located between Latitude 7°3' 0" Nto 7° 21' 0" N and Longitude 3°42' 0" to 3°42' 0" E (Fig. 1). Ratconquarry site was chosen for this study because of its proximity to the residential area, which is associated with the release of a large number of pollutants into the environment.

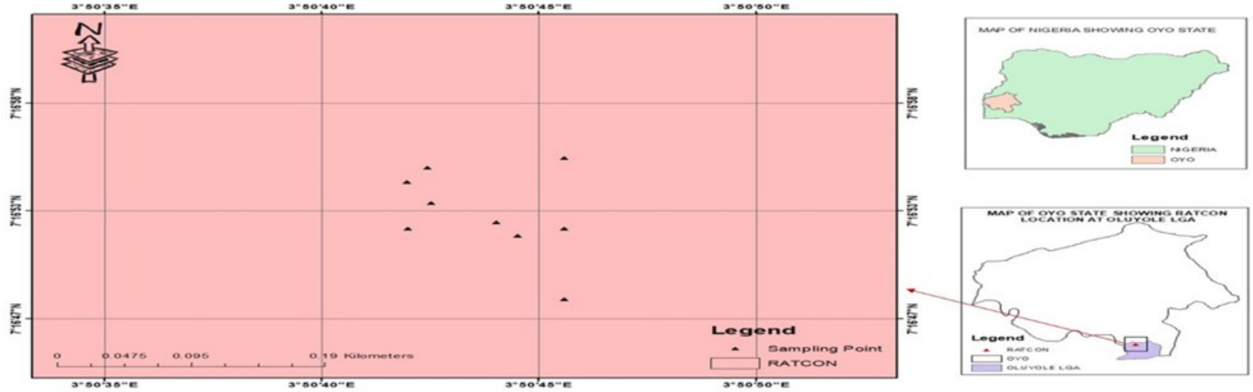

Fig 1: Map of Oluyole LGA, Ibadan, Oyo state showing the study area.

Collection and preparation of samples

Twenty fresh leaf samples of T.grandis and G.arborea were harvested from the quarry site. The coordinates (latitude and longitude) of the collection points of the leaf samples were referenced using GPS map® 76 (Garmin Ltd.): (T. grandis) Latitude 7°3' 26"NLongitude 3° 84' 25.14" E(G. arborea) Latitude 7°2' 0.66" N Longitude 3°82' 25.14" E(Figure 1). The leaf samples were washed under a flowing current of deionized water to remove soil and dust particles and dried in an oven at 50℃ until they became crispy. The separate dried leaf samples were crushed intofine powder using a ceramic mortar and pestle and stored in well-labeled air-tight containers pending analysis. The method described by (Ibe *et al*., 2017) was adopted. The Analysis was conducted at the Laboratory of Soil Science Dept. University of Ibadan.

Soil samples were collected from the sub-areas of the quarry using a soil auger at 10 cm depth (topsoil) (Enyoh *et al.,* 2017). Ten soil samples from each sub-area were pooled and sieved to remove unwanted soil particles and debris. The prepared soil samples were dried in an oven at 50℃ to constant weight and represented the composite soil samples from the quarry. The separate composite dried soil samples were stored in corresponding air-tight containers.

Digestion and analyses of leaf and soil samples

One gram (1.0 g) of the powdered leaf and soil samples were weighed into separate corresponding test tubes and digested in 24 mL of aqua regia ($HCl: HNO3 = 3:1$ v/v) for three days as previously described. The concentrations of Pb, Co, Cd, Zn, Ni, Cu, and Mn in the leaf and soil samples were measured in triplicate using atomic absorption spectrophotometer (Buck Model 210) according to the methods reported by (Isiukuand Enyoh.,2020).

Bioaccumulation of heavy metals

From the results of Pb, Co, Cd, Zn, Ni, Cu, and Mn concentrations in T. grandis and G. arboreal leaf and soil samples, the ratio of plant leaf to soil bioaccumulation of heavy metal BF was calculated.

$BFHM = HMCPS / HMCSS$ …………… eqn. 1

Where HMCPS is the heavy metal concentration of a leaf sample and HMCSS is the heavy metal concentration of a soil sample.

Daily intake of heavy metals

The daily intake of heavy metals (DIHM, mg/kg per person/day) was estimated about the body weights of consumers. Because DIHM by individuals is directly proportional to the average daily consumption of the plant leaf, DIHM was calculated (Okereke *et al.,* 2016).

Thus:

$DIHM = CHM \times DFI / BAW$ ………………..eqn .(2)

Where CHM is heavy metal concentration in plant leaf (mg/kg), DFI is the daily consumption of T. grandis and G.arborea by adults and children based on related studies (Okereke *et al.*,2016), which was put at estimated averages of 0.345 and 0.232 kg-person/day, respectively, and BAW is the average body weights of the consumers based on field estimations using a mobile weighing scale (Hana instrument; China), which was put at 55.9 kg and 32.7 kg for adults and children, respectively according to (Okereke *et al.,* 2016).

Health risk index

The health risk index (HRI) was estimated as described in previous literature.

$HRI = DIHM / ORD$ ……………. Eqn. (3)

Where ORD: Oral reference dose (mg/kg person/day). The ORD is an estimated exposure of metal to the human body per day associated with no potential hazardous effect during a lifetime. The ORD (mg/kg person/day) for Pb, Co, Cd, Zn, Cu, Ni and Mn used were 0.004, 0.02, 0.001, 0.3, 0.02, 0.04, 0.02, and 0.033 mg/kg per person/day, respectivelyaccording to(Gebreyohannes and Gebrekidan, 2018). The consumer population is considered safe when HRI is less than 1 (Okereke *et al*., 2016).

Results and Discussion

The heavy metals concentrations in T. grandis and G. arborea leaf samples from the Ratcon quarry site are presented in Table 1. The heavy metal concentrations in the leaf samples were in the following order: Ni > Zn > Mn > Co > Cu > Cd > Pb for T. grandis; Ni > Zn > Co > Mn > Cu > Cd > Pb for G. arborea.

All the heavy metal concentrations in the leaf samples were below the maximum permissible level of the WHO, whereas the concentrations of Zn, Cd, and Ni in both plants `were higher than the WHO permissible limits (Table 1). The Mn concentrations in the leaf samples were very low when compared with the WHO permissible level for Mn (200 mg/kg). The low Mn concentration was probably due to the leaching of the metal to the deeper strata of the soil.

Apoptosis, necrosis, and genotoxicity are associated with Co poisoning (Simonsen *et al.,*2012). Lung cancer, anemia, kidney failure, and bone disorder are peculiar diseases of Cd toxicity (Edward *et al.,*2013). Zn is known to play a major role in body development and function, but excessive exposure to Zn can provoke respiratory disorder, epigastric pain, diarrhea, the elevated risk of prostate cancer, focal neuronal deficits, and nausea (Plum *et al*., 2010).

The concentrations of Cd, Zn, Cu, Co, and Mn in leaf samples of both T.grandis and G. arborea from the study area were similar to the report of (Franklyn *et al,* 2020) except for higher Nickel. Additionally, the reports of Erakhrumen and Inaede, (2018)also show that lead Pb was below the detection limit in the leaf of T.grandis analyzed this is in agreement with the present study.

Heavy metal concentrations in the soil samples

The concentrations of Cu, Co, Zn, Mn, Pb, Cd, and Ni in the soil samples collected from the quarry site are shown in Table 1. The mean concentrations of the heavy metals followed the order: of Zn > Mn > Cu > Ni > Cd > Co > Pb.

All heavy metal concentrations in the soil samples were within the permissible level of WHO (WHO, 1996), except Cd, which gave a concentration of 8.640 mg/kg as against the WHO's maximum permissible level of 0.8 mg/kg. The concentration of Cd in the soil samples from this area was higher than that reported for Shao North Central Nigeria, Nigeria by Ogundele *et al*, (2015)which was 0.033 mg/kg, and Nashwa andManal, (2022) which was 0.14mg/kg.

These are indications that the soil samples from the Ratcon quarry were highly polluted with Cd. By implication,Cd exposure can result in a variety of adverse effects, such as renal and hepatic dysfunction, pulmonary edema, testicular damage, osteomalacia, and damage to the adrenals and hemopoietic system.The high concentration of Cd in the soil samples from the area of study can be attributed to the quarry activities in the area.

The concentration of Zn from soil samples from the present area of study was lower compared to the WHO Maximum Permissible limit which is 50mg/kg. The mean Ni concentration (40.001mg/kg) which is higher than the WHO MPL also conformsto the findings of (Iroye and Igbozurike,2018) which reported higher Ni concentration in the river Ona which is very close to the Ratcon quarry site. A lower concentration of Cu (15.782mg/kg) was noted in soil samples from the present study sites. Pd (3.543mg/kg) has the lowest mean concentration for the present study.

Table 1. Maximum permissible limits (MPL) of heavy metals in soils and plants according to the Food and agriculture organization and world health organization (WHO/FAO) and heavy metal concentrations (HMC) in leaf and soil samples from Ratcon Quarry site.

Heavy metals	MPL (mg/kg)		Leaf Samples HMC (mg/kg)		Soil Samples HMC
	Soil	Plant	T.grandis	G.arborea	
Copper (Cu)	36	10	0.22± 0.03	0.43 ±0.02	15.782±1.10
Cobalt (Co)	ND	1.5	0.34±0.02	0.574±0.00	6.194 ±1.36
Zinc (Zn)	50	0.6	0.93±0.06	6.657±0.04	18.531±2.84

Manganese (Mn)	ND	200	0.40±0.02	0.560±0.11	15.929±3.67
Lead (Pd)	85	2	ND	ND	ND
Cadmium (Cd)	0.8	0.02	0.04 ± 0.02	0.03± 0.05	8.640± 5.30
Nickel (Ni)	35	10	15.3 ± 0.30	11.2± 0.05	13.001± 1.25

Not Detected -ND

Bioaccumulation of heavy metals (BHM)

Among the analyzed leaf samples, the T. grandis exhibited the highest bioaccumulation of Ni, Co, and Zn, whereas G. arborea gave the highest concentration of Zn.

The ratio of plant leaf to soil bioaccumulation of Cd and Mn was lowest in T. grandis. Likewise, the ratio of plant leaf to soil bioaccumulation of Co was also lowest in T. grandis. The pattern of increase in bioaccumulation of heavy metals in the leaf samples is as follows: Ni > Co > Zn > Mn > Cu > Cd (T. grandis); Zn > Ni > Co > Mn > Cu > Cd (G. arborea) (Table2).

The transfer factor of heavy metals from soil to plants is the ratio of the concentration of heavy metals in a plant to the concentration of heavy metals in the soil-RBHM (Lago-Villa *et al*.,2015). The transfer factor signifies the extent to which heavy metals in the soil accumulated in the plants. The BHM was calculated to have a good knowledge of the extent of risk and hazard associated with the ingestion of plants from the study area. The soil-to-plant transfer factor is one of the key components of the measure of the level of human exposure to metals in the food chain. Okereke *et al.,*(2016), posited that the physicochemical properties of soil and plants could influence the transfer or mobility of metals from soil to plant. These physicochemical properties are on the other hand influenced by industrial activities such as quarrying or mining. Thus, the RBHM of the various heavy metals in this study area might have been influenced by quarry activities and other industrial processes going on in the area. The BHM of the heavy metals was noted to vary directly with the heavy metal concentration in the leaf samples. Therefore, it implies that the transfer factor of heavy metals from soil to plants was a determinant of the level of heavy metal pollution of the plants cultivated in the area.

The high BHM of the heavy metals for T. grandis was an indication that the teak around the study site was highly polluted and as such not safe for medicinal purposes

Table 2. Bioaccumulation of heavy metals from Ratcon quarry site in Ibadan.

Ratio Plant Samples	Cu	Co	Zn	Mn	Pb	Cd	Ni
Tectona grandis	0.014	0.054	0.050	0.025	ND	0.005	0.383
Gmelina arborea	0.028	0.093	0.359	0.035	ND	0.004	0.280

ND- Not detected

Daily intake of heavy metals

The DIHM in the study area is shown in Table 3. The estimated DIHM in both plants followed the same trend.

The DIHM was in the order: of Ni > Zn > Mn > Co > Cu > Cd > Pd (T. grandis); Ni > Zn > Mn > Co > Cu > Cd > Pd (G. arborea). Table 3 showed that Ni represented the highest DIHM in T.grandis and G. arborea for adults (0.0944 and 0.0691 mg/kg person/day, respectively). Additionally, Cd intake gave the lowest DIHM (0.0003 and 0.0002 mg/kg per person/day) following the consumption of T.grandis and G. arborea.

The Joint FAO/WHO Expert Committee on Food Additives (JECFA) stated that the tolerable daily intake of Pb, Co, Cd, Zn, Ni, Cu, and Mn was set at 0.0036, 0.0230, 0.0010, 0.4300, 0.0050, 0.5000, and 0.0110 mg/kg person/day, respectively(JEFCA,1999). The estimated DIHM was all below the provisional tolerable daily intake level specified by JECFA, except for high Ni. Pb was below the detection limit for both plants.

Maghrabi, (2014) reported similar results from different samples of tree plants in Saudi Arabia, with the levels of Pb below the WHO permissible limit (Maghrabi, 2014). However, contrary to this finding, the concentration of lead above the WHO maximum permissible limit was reported in previous studies (Samali *et al*., 2017). Lead is one of the most toxic heavy metals and progressive exposure may cause poor muscle coordination, gastrointestinal symptoms, brain and kidney damage, hearing and vision impairments, and reproductive defects(Weaver *et al*., 2014). Lead bioaccumulates in biological tissues; patients who use medicinal herbs with even low concentrations of Pb over a long period might be at risk of chronic Pb toxicity and should be monitored for any signs of lead poisoning (Zamir *et al*.,2020).

The concentration range of copper in tested samples was 0.0014 –0.0027 mg/kg as shown in Table 3. The maximum concentration was reported in G. arborea while the minimum amount was reported in T.grandis. This result revealed that the concentration of copper in both samples was found below the WHO permissible limit (10 mg/kg).

Similar results were reported in Brazil (Tschinkel, *et al.,* 2020), and Nigeria (Ekeanyanwu, *et al*., 2013). Copper is an essential element for the human metabolic system. It regulates various biological processes like redox reactions, energy production, connective tissue formation, iron metabolism, and synthesis of neurotransmitters (Ekeanyanwu, *et al*., 2013). However, chronic exposure to a high concentration of copper irritates nasal mucosa, vomiting, nausea, diarrhea, kidney, and liver damage.

Table 3. Daily intake of heavy metals of adults (mg/kg person/day) in Ibadan

Leaf Samples	Daily intake (mg/kg person/day)						
	Cu	Co	Zn	Mn	Pb	Cd	Ni

Tectona grandis	0.0014	0.0021	0.0057	0.0025	-	0.0003	0.0944
Gmelina arborea	0.0027	0.0035	0.0411	0.0035	-	0.0002	0.0691

Below detection limit

Health risk assessment

Health risk assessment (HRA) defines the basis for establishing health risks posed by heavy metals in the consumption for medicinal purposes of T. grandis and G. arborea. Accordingly, HRA encapsulates the DIHM and HRI.

The HRI of the heavy metals by the utilization of leaves of plants under study for adults in the study area is shown in Table 4. The HRI of the heavy metals for adults was in the order: of Ni > Cd > Zn > Co > Mn > Cu

(T. grandis); Ni > Zn > Cd > Co > Mn > Cu (G. arborea). The range of values of HRI of the heavy metals were as follows: 4.720–0.035 and 3.455–0.068 for Tectona grandis and Gmelina arborea respectively.

The estimated Health risk index (HRI) of the heavy metals were all below one except Ni for both plants, which had an HRI of 4.720 and 3.455. HRI <1 indicates that the consumer population is safe from any potential health posed by heavy metals, whereas HRI >1 implies that the consumer population is not safe (Okereke *et al.,* 2016). Since it was observed that the HRI of Ni following the medicinal use of both plant leaves was >1, it implies that Ni could cause severe health risks to consumers of T.grandis and G. arborea in the study area. Depending on the dose and length of consumption, as an immunotoxicological and carcinogenic agent, Ni can cause a variety of health effects, such as contact dermatitis, cardiovascular disease, asthma, lung fibrosis, and respiratory tract cancer (Chen *et al.,* 2017). Inhalation exposure in occupational contexts is the main route for nickel-induced toxicity in the respiratory tract, the lung, and the immune system.

It has been reported that the food chain (soil-plant-man) is the most common and important exposure pathway to heavy metals such as Pb, Cd, and Ni, in humans (Okereke *et al.,* 2016). Thus, a reduction in the concentration of pollutants such as Ni in the soil would reduce its health risks to humans. Possible remediation techniques for Ni in soil include chemical immobilization/stabilization methods by adding some non-toxic materials into the soil to reduce the solubility of the heavy metal and mixing the polluted soils with clean unpolluted soils to reduce the heavy metal concentration in the soil. Other contemporary remediation methods electromigrationtion process, washing/leaching/flushing the soil with chemical agents such as cyclodextrins, surfactants, chelating agents, and organic acids either ex-situ or in-situ (Wuana *et al.,* 2010).

Table 4. Health risk index of heavy metals for adults in Ibadan

Health Risk Index

Leaf Samples	Cu	Co	Zn	Mn	Pb	Cd	Ni
T.grandis	0.035	0.105	0.285	0.076	-	0.300	4.720
G. arborea	0.068	0.175	2.055	0.106	-	0.200	3.455

Below detection limit

Conclusion

The present study noted high bioaccumulation of toxic heavy metals from soil samples in the Oluyole area of Ibadan. The HRI of Ni was reported to be greater than 1, indicating that the consumption of such plants will pose a significant health risk to the populace, especially. It is therefore recommended that the local populace of the Oluyole area in Ibadan should abstain from T.grandis and G. arboreal-prepared to reduce Ni bioaccumulation and concomitant health risks. However, the concentrations of Co, Cd, Zn, Pb, Cu, and Mn were relatively within safe limits.

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