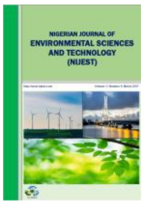
Nigerian Journal of Environmental Sciences and Technology (NIJEST)
www.nijest.com
ISSN (Print): 2616-051X | ISSN (electronic): 2616-0501
Vol 2, No. 1 March 2018, pp 89 - 95



Heavy metals in soil and accumulation in medicinal plants at an industrial area in Enyimba city, Abia State, Nigeria

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ABSTRACT

The study assessed heavy metals in the soil and subsequent accumulation in plants at an industrial site at Enyimba city, Abia State, Nigeria. Soil and medicinal plant samples were analyzed for zinc (Zn), lead (Pb) and cadmium (Cd). The highest concentration of Zn (142.06 ± 2.91 mg/kg), Pb (18.06 ± 1.30 mg/kg) and Cd (27.055 ± 2.468 mg/kg) were obtained at the sampling points of 2, 7 and 5, respectively. The highest concentrations of Zn (27.09 ± 1.44 mg/kg) and Cd (2.000 ± 0.156 mg/kg) were accumulated by *Azadiractha indica* while the highest concentration of Pb (4.58 ± 0.51 mg/kg) was accumulated by *Mangifera indica*. The levels of Zn and Cd in soil were 13.77 ± 1.35 to 142.06 ± 2.91 and 0.695 ± 0.106 to 27.055 ± 2.468, and their concentrations in *Azadiractha indica* were 5.06 ± 0.35 to 27.09 ± 1.44 and 0.002 ± 0.001 to 2.000 ± 0.156 mg/kg, respectively. The concentrations of Zn and Cd in soil and Cd in *Azadiractha indica* reflected a state of pollution relative to Dutch criteria for soil and the FAO/WHO Codex Alimentarius Commission for soil and herbal plants.

Keywords: Medicinal plants, heavy metals, industrial area, Enyimba city, Nigeria

1.0. Introduction

The use of medicinal plants for therapeutic purpose or as a dietary supplement dates back beyond records of history, but have increased substantially in the last decade (WHO, 2002). Medicinal plants are applied as single plants, which action is directed at individual ailments, as plant mixtures, syrups, plants and fruit-plant teas and as spices (Ozarowski and Jaroniewski, 1987). Thus, a large populace of people in developing countries relied heavily on medicinal plants for their primary health care because of the low price and safety of active ingredients in plant materials.

The atmosphere is an important pathway for transport of metals and the major external input of bio-available metals in the environment, which are potential threats to the health and survival of man. This is because urban atmosphere is submitted to large inputs of metals arising from stationary source such as industries. The metals are either deposited directly on plant surfaces such as leaves, flowers, branches, and stems or on soils, which are absorbed from the soil solution into plants via the roots. Therapeutic use of medicinal plants contaminated with heavy metals may lead to hazards of enriching human alimentary canal with toxic levels of metals. For instance, the prevalence of upper gastrointestinal cancer in the Van region of Turkey has been linked to metal pollution in soil, fruits and vegetables (Turkdogan *et al*., 2003).

Among the medicinal plants commonly seen in home gardens at Enyimba city are *Azadiractha indica*, *Citrus sinensis*, *Psidium guajava*, *Mangifera indica*, *Vernonia amygdalina*, *Occimum gratissimum*, and *Carica papaya*. These plants extracts has undergone extensive pharmacological screening and found to have several pharmacological activities due to presence of several active ingredients. Notwithstanding this, there is increasing concern in environmental connection with human health. Consequently, there is need to investigate the level of concentrations of metals in medicinal plants around the industrial area in Enyimba city. This will enhance the knowledge of the people on the possible health hazards of using medicinal plants contaminated with heavy metals.

2.0. Materials and Methods

2.1. Study area

The study was carried out at Enyimba city. The Enyimba city is the major industrial and commercial hub of Abia State and it is located in the lowland rainforest zone of Nigeria (Keay, 1959). It lies on latitude 5° 1'N and longitude 7° 35' E and characterized by heavy rainfall and short dry season. The mean annual rainfall is 150 to 186 mm, and annual relative humidity is over 80% while the mean annual temperature exceeds 21°C.

2.2. Sampling collection and analysis

A simple factorial experiment in a randomized complete block design was used to carry out the study. Twenty three (3) surface soil samples (0-15 cm) each were collected randomly from seven (7) different sampling positions (1, 2, 3, 4, 5, 6, and 7) at industrial and residential areas and stored in cellophane bags, labelled well, and taken to the laboratory for pre-treatment and analysis. The residential area where there was no industry served as control. Twenty three samples from each sampling positions were bulked separately (e.g. 23 samples of 0-15 cm at sampling position 1), homogenized and air dried in circulating air in an oven at 30°C to constant weight and passed through a 2 mm sieve. The procedure described by MAFF (1981) was used for the digestion of soil samples. Exactly 1 g of sub samples were placed in a 100 ml beaker and 10 ml HNO3 acid and 3 ml HClO4 were added and the solution was heated until fuming. Sample solution was obtained by processing with 4 ml hot E mol/Hec, filtered and diluted with water in a 50 ml standard flask. Triplicate digestion of each sample together with a blank was also carried out and metallic content of digested samples were determined with flame atomic absorption spectrophotometer (UNICAM 919 Model) after calibration.

Fresh leaves were sampled from twenty one (21) stands each of *Azadiractha indica, Citrus sinensis, Psidium guajava, Mangifera indica, Vernonia amygdalina, Occimum gratissimum*, and *Carica papaya* from where soil samples were collected at industrial and residential areas. The fresh leaves were collected randomly from the various branches of the medicinal plants (except for *Carica papaya* that does not have branches). The samples from each plant species were placed in large paper bags, labelled well and transferred to the Laboratory for pre-treatment and analysis. The plant samples were thoroughly rinsed with distilled-deionized water to remove dust and pollen particles and placed in large crucibles and oven dried at 70°C for 72 hrs. The dried samples from each species were bulked together and milled with a Thomas Wiley machine. Sub samples were collected from the milled samples and analysed for heavy metals. The procedure described by Allen *et al*. (1976) was used for sample digestion. Exactly 0.2 g of each samples were weighed into 100 ml standard flask and 1 ml per chloric acid and 5 ml conc. HNO_3 added and digested at 80-90°C on hot plates until white fumes evolved. The digest was allowed to cool and then filtered into 50 ml standard flask with watchman No 1 filter paper. Triplicate digestion of each sample together with a blank was also carried out and metallic content of digested samples was determined with flame atomic absorption spectrophotometer (UNICAM 919 Model) after calibration. Data from the result of laboratory analysis were subjected to analysis of variance (ANOVA) and means were separated with Duncan Multiple Range Test.

2.3. Experimental design and statistical analysis

A Single Factor Experiment in a randomized complete block design (RCBD) was used to carry out the experiment. The data generated from laboratory analysis were subjected to analysis of variance (ANOVA) using Statistical Package for Social Sciences (SPSS) version 15.0. Analysis of variance was performed using a One-way ANOVA and Duncan Multiple Range Test (DMRT) was used to test if significance difference existed between mean concentrations in physical parameters and heavy metals in water, fish and sediment from the different sampling stations.

3.0. Results and Discussion

3.1. Heavy metal in soil

The result of the concentrations of heavy metals in soil is summarized in Table 1. The result indicates that the highest and lowest concentrations of metals in soils were observed at the industrial area and the residential area, respectively. The lower concentration of metals at the residential area (i.e. control) is attributed to the absence of industry (ies) in that location. The highest concentrations of Zn (142.06 ± 2.91 mg/kg), Pb (18.06 ± 1.30 mg/kg), and Cd (27.055 ± 2.468 mg/kg) were obtained at the sampling points of 2, 5 and 7, respectively at the industrial area and these values are significantly ($P < 0.05$) higher than their highest corresponding values (35.59 ± 0.69, 8.99 ± 1.03, and 1.060 ± 0.042 mg/kg) at the residential area, respectively. The high values of Zn, Pb, and Cd at the industrial area are attributed to various industrial activities taken place in the location. Mobilization of metals into the atmosphere as a result of anthropogenic activities is an important process in the geochemical cycling of heavy metals. This is acutely evident in urban areas where various stationary and mobile sources release large quantities of metals into the atmosphere and soil (Bilos *et al*., 2001).

The highest concentration of Zn, Pb and Cd at the industrial area is 3.99, 2.01 and 25.52 times higher than their highest concentrations at residential area, respectively. According to Logan and Miller (1983), soil is said to be contaminated when concentrations of an element in soils were two-to-three times higher than the control. The soils at the industrial area is considered to be contaminated base on the findings that Cd, Zn and Pb concentrations in the control soil samples are significantly lower compared to their corresponding values at the industrial area. The concentrations of Zn and Cd in soils of the industrial area of Enyimba city, Nigeria were 13.77 ± 1.35 to 142.06 ± 2.91 and 0.695 ± 0.106 to 27.055 ± 2.468, respectively for Zn and Cd which are above the accepted limits (i.e. target value) of 140 mg/kg (Zn) and 0.8 mg/kg (Cd) as described by Dutch criteria for soil (Wikipedia, 2013) and the maximum permitted levels of 60 mg/kg (Zn) and 0.1 mg/kg (Cd) established by the Codex Alimentarius Commission (FAO/WHO, 2001) (Table 2). The concentration of Pb (4.91 ± 1.26 to 18.06 ± 1.30 mg/kg) in soils at the industrial area is below the accepted limits (i.e. target value) of 85 mg/kg (Pb) as described by Dutch criteria for soil (Wikipedia, 2013) and the maximum permitted levels of 50 mg/kg (Pb) established by the Codex Alimentarius Commission (FAO/WHO, 2001) Table 2). The values of Zn range from 9.22 ± 1.14 mg/kg at the residential area to 142.06 ± 2.91 mg/kg at the industrial area, which is relatively lower than 114.0 to 162.0 mg/kg for Zn in soils at industrial sites in Isfahan, Iran (Fallahzade *et al*., 2013) but higher than 59.85 ± 0.002 to 64.5 ± 0.014 mg/kg for Zn in soil at Kaduna, Nigeria (Ogundele *et al*., 2015), 0.01 to 1.26 ppm for Zn in soils around Superphosphate factory at Assiut city, Egypt (El-Desoky and Ghallab, 2000) but well below 93.0 to 2841 mg/kg for Zn in soils around a smelter at Slovenia (Glavac *et al*., 2017). The values of Pb in this study range from 1.02 ± 0.04 mg/kg at the residential area to 18.06 ± 1.30 mg/kg at the industrial area, which is higher than 0.102 to 1.082 ppm for Pb (El-Desoky and Ghallab, 2000) but substantially lower than 45.0 to 4132 mg/kg for Pb (Glavac *et al*., 2017) and 27.0 ± 0.005 to 238.0 ± 0.003 mg/kg for Pb in soil (Ogundele *et al*., 2015) while the values of Cd range from 0.275±0.332 at the residential area to 27.055 ± 2.468 mg/kg at the industrial area, which is higher than 0.033 ± 0.003 to 0.100 ± 0.002 mg/kg for Cd in soil (Ogundele *et al*., 2015) and 0.002 to 0.140 ppm for Cd (El-Desoky and Ghallab, 2000) but well below 0.40 to 74.7 mg/kg for Cd (Glavac *et al*., 2017). Generally, the concentrations of heavy metals in soil followed a decreasing order: Zn > Cd > Pb.

Table 1: Heavy metal concentration (mg/kg) in soil

	Zn	Pb	Cd
Industrial area	$58.66^{c} \pm 3.34$	$10.7^{de} \pm 0.05$	$1.160^{d} \pm 0.113$
	$142.06^{a} \pm 2.91$	$16.35^{ab} \pm 1.20$	$9.210^{c} \pm 1.683$
	$58.66^{c} \pm 2.20$	$4.91^{g} \pm 1.26$	$1.490^{d} \pm 0.156$
	$26.57^{e} \pm 2.17$	$11.08^{d} \pm 0.37$	$1.170^{d} \pm 0.042$
	$105.30^{b} \pm 4.38$	$16.41^{ab} \pm 1.68$	$27.055^{a} \pm 2.468$
	$13.77^{g} \pm 1.35$	$12.97^{c} \pm 1.18$	$0.695^{e} \pm 0.106$
	$57.08^{c} \pm 0.45$	$18.06^{a} \pm 1.30$	$14.005^{b} \pm 0.290$
Residential area	$18.55^{ef} \pm 2.19$	$3.07^{gh} \pm 0.22$	$0.550^{ef} \pm 0.651$
	$27.41^{e} \pm 1.83$	$1.64^{i} \pm 0.31$	$1.005^{d} \pm 0.007$
	$11.76^{g} \pm 1.07$	$1.02^{ij} \pm 0.04$	$0.275^{f} \pm 0.332$
	$9.86^{h} \pm 1.20$	$5.68^{f} \pm 0.48$	$1.060^{d} \pm 0.042$
	$21.56^{ef} \pm 2.21$	$8.99^{e} \pm 1.03$	$0.790^{e} \pm 0.099$
	$9.22^{h} \pm 1.14$	$3.59^{gh} \pm 0.53$	$0.305^{f} \pm 0.035$
	$35.59^{d} \pm 0.69$	$5.20^{fg} \pm 0.57$	$0.520^{ef} \pm 0.085$

a, b, c, d, e, f, g, h, i, j, means in a column with different superscript are significantly different (P < 0.05). Values are mean ± standard deviation of 3 replications

Table 2: Comparison of our result with International Standard (Dutch criteria and FAO/WHO Codex Alimentarius Commission

	Dutch criteria (target value) mg/kg	FAO/WHO 2001 Codex Alimentarius Commission (mg/kg)	NESREA 2011 Standard (mg/kg)
Zn	140	60	421
Pb	85	50	164
Cd	0.8	0.1	3

3.2. Heavy metal accumulation in plants

The result of heavy metal accumulation in medicinal plants is summarized in Table 3. The result shows that samples of medicinal plants collected from the environment of the industrial area were more contaminated with the metals (Cd, Pb and Zn). Some pollution studies in different sites in the world, such as Rome, Naples and Sydney, showed that air, soil or plants adjoined to source of pollutants had higher content of metals than a control area (Imperatoa *et al*., 2003; Moreno *et al*., 2003; Birch and Snowdon, 2004; Davila *et al*., 2006). The result also indicate that the concentrations of metals in the medicinal plants varied with plant species and metal contaminants with Zn being the most and Cd the least accumulated.

The highest concentration of Zn was obtained in *Azadiractha indica* (27.09 ± 1.44 mg/kg) and the value is significantly (P < 0.05) higher than its corresponding values in *Psidium guajava* (19.77 ± 1.17 mg/kg), *Mangifera indica* (15.45 ± 0.92 mg/kg), *Vernonia amygdalina* (11.43 ± 0.87 mg/kg), *Carica papaya* (9.36 ± 0.95 mg/kg), *Citrus sinensis* (7.97 ± 1.33 mg/kg) and *Ocimum gratissimum* (5.06 ± 0.35 mg/kg) at the industrial area as well as the values recorded for plants at the residential area (Table 3). The highest concentration of Cd was also obtained in *Azadiractha indica* (2.000 ± 0.156 mg/kg) and the value is significantly (P < 0.05) higher than values observed in *Psidium guajava* (1.72 ± 0.17 mg/kg), *Mangifera indica* (1.190 ± 0.099 mg/kg), *Vernonia amygdalina* (0.055 ± 0.021 mg/kg), *Carica papaya* (0.008 ± 0.001 mg/kg), *Ocimum gratissimum* (0.005 ± 0.002 mg/kg) and *Citrus sinensis* (0.002 ± 0.001 mg/kg) at the industrial area as well as the values recorded for plants at the residential area. The high concentration of Zn and Cd in *A. indica* may be attributed to inherent ability of the plant (*A. indica*) to absorb and translocate more Zn and Cd to the aerial plant parts (leaves) than other plants. Some plants can tolerate high heavy metals concentration from soil (McGrath *et al*., 2001) by binding metals to cell walls, compartmentalizing them in vacuoles or complexing them to certain organic acids or proteins (Reeves and Baker, 2000). The concentration of Zn increased from 1.15 ± 0.09 mg/kg (*Ocimum gratissimum*) at the residential area to 27.09 ± 1.44 mg/kg at the industrial area (*Azadiractha indica*). The level of Zn in this study is higher than 2.42 ± 0.2173 to 8.93 ± 0.0264 ppm for Zn in *Acacia nilotica*, *Bacopa monnieri*, *Commiphora wightii*, *Ficus religiosa*, *Glycyrrhiza glabra*, *Hemidesmus indicus*, *Salvadora oleoides*, *Terminalia bellirica*, *Terminalia chebula* and *Withania somnifera* from north western India (Kulhari *et al*., 2013) but well below 23.2 to 799.5 mg/kg for Zn in *Urtica dioica*, *Hypericum perforatum*, *Achillea millefolium*, and *Plantago lanceolata* around a smelter at eight Meza Valley locations, Slovenia (Glavac *et al*., 2017), 12.65 to 146.67 mg/kg for Zn in *Petroselinum crispum*, *Ocimum basilicum*, *Salvia officinalis*, *Origanum vulgare*, *Mentha spicata*, *Thymus vulgaris*, and *Matricaria chamomilla* in the United Arab Emirates, UAE (Dghaim *et al*., 2015), and 83.74 to 433.76 for Zn µg/g in *G. glabra*, *O. bracteatum*, *V. odorata*, *F. vulgare*, *C. cyminum*, *C. sativum*, and *Z. officinalis* from Karachi city, Pakistan (Hina *et al*., 2011). The level of Zn in our study is below the permissible limit (PL) 50 mg/kg set by Codex Alimentarius Commission, FAO/WHO (2006) for medicinal plants and herbs but if the concentration of the metal in the plants at the industrial area continue to increase, it could pose significant health hazard to the population who consume the medicinal plants grown there.

Table 3: Heavy metal concentration in medicinal plants

Location	Medicinal plants	Zn	Pb	Cd
Industrial area	*Carica papaya*	$9.36^{e} \pm 0.95$	$2.56^{bc} \pm 0.21$	$0.008^{d} \pm 0.001$
	Azadiractha indica	$27.09^{a} \pm 1.44$	$2.77^{b} \pm 0.21$	$2.000^{a} \pm 0.156$
	Vernonia amygdalina	$11.43^{d} \pm 0.87$	$0.76^{d} \pm 0.21$	$0.055^{d} \pm 0.021$
	Citrus sinensis	$7.97^{g} \pm 1.33$	$2.40^{c} \pm 0.28$	$0.002^{d} \pm 0.001$
	Psidium guajava	$19.77^{b} \pm 1.17$	$2.28^{cd} \pm 0.51$	$1.720^{b} \pm 0.170$
	Ocimum gratissimum	$5.06^{gh} \pm 0.35$	$2.55^{bc} \pm 0.35$	$0.005^{d} \pm 0.002$
	Mangifera indica	$15.45^{c} \pm 0.92$	$4.58^{a} \pm 0.51$	$1.190^{c} \pm 0.099$
Residential area	*Carica papaya*	$4.05^{h} \pm 0.21$	$0.40^{de} \pm 0.14$	$0.004^{d} \pm 0.001$
	Azadiractha indica	$9.00^{ef} \pm 0.57$	$0.25^{e} \pm 0.07$	$0.008^{d} \pm 0.001$
	Vernonia amygdalina	$1.66^{j} \pm 0.54$	$0.15^{f} \pm 0.07$	$0.002^{d} \pm 0.001$
	Citrus sinensis	$3.21^{i} \pm 0.57$	$0.75^{d} \pm 0.21$	$0.003^{d} \pm 0.001$
	Psidium guajava	$6.40^{gh} \pm 0.42$	$1.10^{d} \pm 0.28$	$0.002^{d} \pm 0.001$
	Ocimum gratissimum	$1.15^{j} \pm 0.09$	$0.27^{e} \pm 0.06$	$0.001^{d} \pm 0.000$
	Mangifera indica	$8.07^{f} \pm 0.06$	$0.86^{d} \pm 0.06$	$0.005^{d} \pm 0.002$

a, b, c, d, e, f, g, h, i, j, means in a column with different superscript are significantly different (P<0.05). Values are mean ± standard deviation of 3 replications

The concentration of Pb increased from 0.15 ± 0.07 mg/kg (*Vernonia amygdalina*) at the residential area to 4.58 ± 0.51 mg/kg at the industrial area (*Mangifera indica*). The level of Pb in this study is higher than 0.25 ± 0.00088 to 2.34 ± 0.0173 ppm (Kulhari *et al*., 2013) but well below 1.1 to 195.9 mg/kg in *Urtica dioica*, *Hypericum perforatum*, *Achillea millefolium*, and *Plantago lanceolata* around a smelter at Slovenia (Glavac *et al*., 2017), 1.0 to 23.52 mg/kg in *Petroselinum crispum*, *Ocimum basilicum*, *Salvia officinalis*, *Origanum vulgare*, *Mentha spicata*, *Thymus vulgaris*, and *Matricaria chamomilla* in the United Arab Emirates, UAE (Dghaim *et al*., 2015), 19.50 to 121.3 mg/kg in *Plantago lanceolata* at polluted areas of Poland (Nadgorska-Socha *et al*., 2013), and 102.3 mg/kg in *Urtica dioica* at polluted areas of Macedonia (Gjorgieva *et al*., 2010). The level of Pb in our study is below the permissible limit (PL) 10 mg/kg set by Codex Alimentarius Commission, FAO/WHO (2006) for medicinal plants and herbs. Notwithstanding this, the use of these medicinal plants on a regular basis can increase the accumulation of Pb in human's body beyond the permitted limit, and this could pose serious health issues for them. Accumulation and magnification of heavy metals in human tissues through consumption of herbal remedies can cause hazardous impacts on health (Kulhari *et al*., 2013).

Cadmium concentration in this study range from 0.001 ± 0.00 mg/kg (*Ocimum gratissimum*) at the residential area to 2.00 ± 1.56 mg/kg at the industrial area (*Azadiractha indica*). The level of Cd in this study is well below 0.20 to 16 mg/kg in *Urtica dioica*, *Hypericum perforatum*, *Achillea millefolium*, and *Plantago lanceolata* around a smelter at Slovenia (Glavac *et al*., 2017), 5.70 to 13.8 mg/kg in *Plantago lanceolata* at polluted areas of Poland (Nadgorska-Socha *et al*., 2013) and 7.37 mg/kg in *Urtica dioica* at polluted areas of Macedonia (Gjorgieva *et al*., 2010) but higher than 0.1 to 1.11 mg/kg for Cd in *Petroselinum crispum*, *Ocimum basilicum*, *Salvia officinalis*, *Origanum vulgare*, *Mentha spicata*, *Thymus vulgaris*, and *Matricaria chamomilla* in the United Arab Emirates, UAE (Dghaim *et al*., 2015). The level of Cd in our study is well above the permissible limit (PL) 0.3 mg/kg set by FAO/WHO (2006) for medicinal plants and herbs. The level of Cd in the samples of medicinal plants is 6.67 times higher than the permissible limit (PL) 0.3 mg/kg set by Codex Alimentarius Commission, FAO/WHO (2006) for medicinal plants and herbs. The safety and benefits of medicinal plants are directly connected to its composition and or quality.

Consequently, the use of medicinal plants grown at the industrial area of Enyimba city can be a major route of entry of Cd in human body at the study area, which can be very deleterious to health. Kidney is the critical target organ in the exposed population and excretion of Cd is very slow and it accumulates in human kidney for a relatively long time, resulting in an irreversible impairment of the renal tract (Martin and Griswold, 2009; Li *et al*., 2012; Maobe *et al*., 2012; Dghaim *et al*., 2015). Generally, the concentrations of heavy metals in medicinal plants followed an increasing order: Cd < Pb < Zn.

4.0. Conclusion

The study shows that industrial activities are one of the anthropogenic sources of metals in the environment. Comparison with an international (Dutch criteria and FAO/WHO Codex Alimentarius Commission) standard i.e. the international scientific literature, shows that the level of metal contamination in soil and accumulation in plant is high. The *Azadiractha indica* clearly has an inherent potential to take up metals from the soil compared with the other plants tested and could constitute serious health risk when used by consumers.

Acknowledgements

The authors appreciate the Technologists in the Soil Laboratory of National Root Crops Research Institute (NRCRI) Umudike, Abia State, Nigeria for the analysis of the samples tested in this study.

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