ABSTRACT

Industrialization has been the major source of waste generation leading to heavy metal accumulation in soils. It has become major environmental challenge especially as it affects soil quality, biota, ecosystem processes and ground water pollution. This study was targeted at determining the contributions of human activities on heavy metal levels of soils in Owerri, eastern Nigeria, by applying Index of geo-accumulation (I-geo) and Contaminant factor (Cf) models on heavy metal levels at major locations. Soil samples were collected during the rainy season with the aid of a hand-dug soil auger (2.50 cm diameter) from three different composite sites: refuse dumps, auto-mechanic villages and a control sample from a virgin land at depths of 0-15 cm, 15-30 cm and 30-45 cm respectively. These were subjected to standard chemical analyses. There were significant variations (P<0.05) in heavy metals concentrations at different depths. There were no significant variations (P>0.05) in pH. pH (5.9-5.2) was acidic at different soil depths, which might

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favor ionization of metals; electrical conductivity at the contaminated sites was higher (300.4-248.4 µS/cm) than at the control site (195.8-150.6 µS/cm) which might imply high concentration of ionized dissolved chemicals from waste dumpsites. Copper (Cu) from contaminated areas ranged from 19.1-16.4 mg/kg against control value that was insignificant (P>0.05); Lead (Pb) ranged from 58.2-28.2 mg/kg against 0.4-0.2 mg/kg; Iron (Fe) ranged from 5.6-2.6 mg/kg against 5.6-2.6 mg/kg; Zinc (Zn) ranged from 140.3-119.4 mg/kg against 3.3-2.5 mg/kg of control. I-geo for all metals fell in the categories of 4-5 and >5. All categories with values within 4-5 were soils that were highly polluted to very highly polluted, while those with values greater than 5 were soils that were very highly polluted. The Cf values of metals were quite >1.5 which implied that the activities at the mechanic villages and waste dumpsites contributed significantly to the heavy metal burden of the soils. Anthropogenic impacts on heavy metal levels on the samples were between 97.6 and 99.9%. Anthropogenic activities should be controlled to prevent heavy metal pollution of soils and sustain quality soil health.

**Keywords:** Waste disposal; heavy metals; modeling; anthropogenic contamination; soil health.

1. INTRODUCTION

According to Adewole and Uchegbu [1], one of the major sources of increase in heavy metal concentration of the ecosystem in Nigeria is auto mechanic activities. These auto mechanic workshops are found in clusters of open plots of land near urban towns and cities [2,3]. In many auto-mechanic workshops, there are accidental or deliberate releases or discharges of petrol waste, diesel, solvents, grease, and lubricants on the land and the atmosphere [4]. Many of these petroleum products are organic chemicals that can be highly toxic and hazardous to soil fauna, flora and human health. The use of automobiles has also led to trace element and heavy metals-contaminated soil, which have serious consequences for soil dwelling organisms [5]. According to Gupta and Gupta [5], the toxicity or effects of heavy metals are consequences of an organism’s position in the food chain, while in others; they are based on the genetic abnormalities due to physiological impairments.

According to Iqbal et al. [6], uncontrolled dumping of municipal waste is the most common and overlooked cause of water pollution. It is of utmost public health and safety importance to continually monitor physicochemical characteristics, including heavy metals concentrations of natural water bodies [7]. The increasing chances of leachate movement to the ground water might be influenced by increased degradation time and longer retention of the heterogeneous waste [6].

According to Pouyat et al. [8], soil as an important component of the lithosphere and biosphere, is a vital natural resource on which the life supporting systems and socio-economic development depend. Groffman et al. [9], Pouyat et al. [10], White and McDonnell [11] and Lehmann and Stahr [12] further stated that soils form the foundation for many ecological processes, interactions, such as nutrient cycling, distribution of plants and animals, moderating the hydrologic cycle through absorption, storage, and supply of water, and ultimately location of human habitation. Soil acts as an important reservoir of nutrients and moisture [13], heavy metals [14]; as a result its quality has significant influence on the health and productivity of an ecosystem and the related environment [15].

Various quantitative indices have been employed to assess the impact of human activities on the concentration toxic trace metals in soil namely: Index of geo-accumulation (I-geo) and Contamination factor (Cf). The I-geo enables the assessment of contamination by comparing current and pristine concentrations of the contaminants [16-18]. The second approach is using the Contamination factor (Cf) and the degree of contamination. According to Fagbote and Olanipekun [19], Cf can be used to differentiate between the metals originating from anthropogenic activities and those from natural processes and to assess the degree of anthropogenic influence.

Analysis of soil provides environmentally significant information and its characterization is needed for understanding the natural and anthropogenic influence on it. When the human societies become increasingly urbanized, fewer people have intimate contact with the soil, and individuals tend to lose sight of the many ways in which they are dependent upon soils for their survival and prosperity. Our reliance on soil resources will increase rather than decrease, day by day.
This study therefore aimed at the applications of Index of geo-accumulation (I-geo) and Contaminant factor (Cf) models to assess the degree of anthropogenic impact of waste dumpsites and automobile mechanic activities on the soil in Owerri.

2. MATERIALS AND METHODS

2.1 Study Area

Owerri is situated at Latitude 5.48º North and Longitude 7.03º East. It is a big town in Nigeria and has a population of about 215,038 inhabitants. Major sources of heavy metals include refuse disposal sites in Owerri metropolis (Umuguma market, Old Nekede road and Relief market) and the wastes were generated from domestic, commercial and industrial activities. Another major source of heavy metal contamination of soil in Owerri includes activities of auto-mechanic at Nekede and Orji Mechanic Villages.

2.2 Soil Sample Collection

Soil samples were collected with the aid of a hand-dug soil auger (2.5 cm diameter) from three different sites: Sample A, composite soil sample from 3 refuse dump sites (Umuguma market, Old Nekede road, and Relief market); Sample B, composite soil sample from Nekede Mechanic and Orji Mechanic Villages; and a control sample from a virgin land at the Federal University of Technology, Owerri (Sample C), at depths of 0-15 cm, 15-30 cm and 30-45 cm respectively.

2.3 Chemical Analyses

2.3.1 Determination of pH

Twenty grams of air dried soil samples were weighed into a 50 mL beaker and 20 mL distilled water was added and allowed to stand for 30 mins. The solution was filtered and the filtrate used to determine the conductivity of soil sample. Hach conductivity meter was used to determine the conductivity. The conductivity meter was calibrated using conductivity calibration solution. The electrode of the meter was dipped into the filtrate and the conductivity meter readings taken to the nearest 0.05 unit.

2.3.2 Determination of conductivity

Twenty grams of air dried soil samples were weighed into a 50 mL beaker and 20 mL distilled water was added and allowed to stand for 30 mins. The solution was filtered and the filtrate used to determine the conductivity of soil sample. Hach conductivity meter was used to determine the conductivity. The conductivity meter was calibrated using conductivity calibration solution. The electrode of the meter was dipped into the filtrate and the conductivity meter readings taken to the nearest 0.05 unit.

2.3.3 Determination of heavy metals

Heavy metals in the soil samples were determined according to the method described by Ogunmohede et al. [20]. One gram of the dried fine soil sample was weighed and transferred into an acid washed, round bottom flask containing 10 cm$^3$ concentrated nitric acid. The mixture was slowly evaporated over a period of 1 hour on a hot plate. Each of the solid residue obtained was digested with a 3:1 concentrated HNO$_3$ and HClO$_4$ mixture for 10 min at room temperature before heating on a hot plate. The digested mixture was placed on a hot plate and heated intermittently to ensure a steady temperature of 150°C over 5 hours until the fumes of HClO$_4$ were completely evaporated [21]. The mixture was allowed to cool to room temperature and then filtered using Whatman No.1 filter paper into a 50 cm$^3$ volumetric flask and made up to the standard mark with deionized water after rinsing the reacting vessels, to recover any residual metal. The filtrate was then stored in pre-cleaned polyethylene storage bottles ready for analysis. Heavy metal concentrations were determined, each with a specific lamp using an Atomic Absorption Spectrophotometer (AAS) AA600 Series.

2.3.4 Calculation of Index of geo-accumulation (I-geo)

The degree of metal pollution at different depths was assessed in terms of seven contamination classes in order of increasing numerical value of the index as shown in Table 1 [19,22]. It was used to unravel levels of accumulation of heavy metals at the different depths of soils [20].

$$I_{geo} = \log_2\left(\frac{C_n}{1.5B_n}\right)$$

Where $C_n$ was the concentration of the heavy metal in the contaminated sample and $B_n$ was the concentration of the metal in the unpolluted (control) samples. The factor 1.5 was introduced to minimize the effect of the possible variations in the background or control values which may be attributed to lithogenic variations in the soil [19].
Table 1. Seven classes of geo-accumulation index

<table>
<thead>
<tr>
<th>Category</th>
<th>Value of soil quality</th>
</tr>
</thead>
<tbody>
<tr>
<td>&lt;0</td>
<td>Unpolluted</td>
</tr>
<tr>
<td>0-1</td>
<td>Unpolluted to moderately polluted</td>
</tr>
<tr>
<td>1-2</td>
<td>Moderately polluted</td>
</tr>
<tr>
<td>2-3</td>
<td>Moderately polluted to highly polluted</td>
</tr>
<tr>
<td>3-4</td>
<td>Highly polluted</td>
</tr>
<tr>
<td>4-5</td>
<td>Highly polluted to very highly polluted</td>
</tr>
<tr>
<td>&gt;5</td>
<td>Very highly polluted</td>
</tr>
</tbody>
</table>

2.3.5 Calculation of contaminant factor (Cf)

The second approach is using the Contamination factor (Cf) and the degree of contamination. In calculating Cf, the equation (2) suggested by Hakanson [23] and Dasaram et al. [17] was used.

\[
Cf = \frac{C_{i01}}{C_{in}}
\]  

(2)

Where \(C_{i01}\) is the mean content of metals from at least 5 sample sites and \(C_{in}\) is the pre-anthropogenic concentration of individual metals. Cf can be used to differentiate between the metals originating from anthropogenic activities and those from natural processes and to assess the degree of anthropogenic influence.

Table 2. Categories of contamination factors

<table>
<thead>
<tr>
<th>Contamination factor</th>
<th>Category</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cf &lt;1</td>
<td>Low contamination factor indicating low</td>
</tr>
<tr>
<td>1&lt;Cf &lt;3</td>
<td>Moderate contamination factor</td>
</tr>
<tr>
<td>3&lt; Cf &lt; 6</td>
<td>Considerable contamination factor</td>
</tr>
<tr>
<td>6&lt; Cf</td>
<td>Very high contamination factor</td>
</tr>
</tbody>
</table>

[23,17]

The third approach using the quantification of anthropogenic concentration of metal employs the concentration in the control samples to represent the lithogenic metal [24]. This was calculated in accordance with Equation (3):

\[
\text{Quantification of anthropogenic metal} = \frac{X - Xc}{X} \times 100\%
\]  

(3)

Where \(X\) = average concentration of the metal in the soil under investigation, and \(Xc\) = average concentration of the metal in the control samples [25].

In this study, the concentration of the control samples is taken to represent the pre-anthropogenic concentration as suggested by Victor et al. [24].

3. RESULTS AND DISCUSSION

3.1 Results

The results of chemical analyses were as shown in Table 3.

3.1.1 pH

The pH did not show any significant variation at different depths, sites and with control (P>0.05). In the composite sample, pH ranged from 5.9±0.1-5.4±0.1 (0-15 cm), 5.4±0.2-5.2±0.1 (15-30 cm) and 5.3±0.1-5.0±0.1 (30-45 cm); while the control varied thus; 5.4±0.3 (0-15 cm) to 5.4±0.1 (15-30 cm) and 5.7±0.2 (30-45 cm).

3.1.2 Electrical conductivity (EC)

There were significant variations (P<0.05) between electrical conductivity values at different depths and with the control values. The values of electrical conductivity in the composites sample varied from 300.4±1.2-248.4±1.1 µS/cm (0-15 cm) to 295.8±1.1-294.7±1.0 µS/cm (15-30 cm) and 280.9±1.0-280.6±1.0 µS/cm (30-45 cm); while the control varied thus: 155.4±0.9 µS/cm, 195.8±1.1 µS/cm and 150.6±1.0 µS/cm at depths of 0-15 cm, 15-30 cm and 30-45 cm respectively.

3.1.3 Heavy metals

There were significant variations (P<0.05) between heavy metal concentrations at different depths and with the control values at different sites.

The concentration of copper in the composite samples varied from 18.6±0.2 -17.9±0.1 mg/kg, 17.4±0.2-16.4±0.1 mg/kg and 19.1±0.1-17.4±0.1 mg/kg at depths of 0-15 cm, 15-30 cm and 30-45 cm respectively; while the control was not significantly detected at varying depths.

The concentration of lead in the composite sample varied from 45.9±0.1-35.7±0.1 mg/kg (0-15 cm), 40.7±0.2-30.8±0.2 mg/kg (15-30 cm) and 58.2±0.1-28.2±0.1 mg/kg (30-45 cm); while the
control varied thus: 0.4±0.01 mg/kg (0-15 cm) to 0.3±0.01 mg/kg (15-30 cm) and 0.2±0.01 mg/kg (30-45 cm).

Iron content in the composite samples varied from 380.2±1.1-360.8±1.0 mg/kg, 345.5±1.2-340.6±1.2 mg/kg and 410.5±1.1-338.2±1.0 mg/kg at depths 0-15 cm, 15-30 cm and 30-45 cm respectively; while the control varied thus 5.6±0.01 mg/kg, 3.4±0.01 mg/kg to 2.6±0.01 mg/kg at depths 0-15 cm, 15-30 cm and 30-45 cm respectively.

In the composite sample, zinc varied as follows: 138.8±1.2-120.4±1.0 mg/kg (0-15 cm), 129.6±0.9-98.5±1.1 mg/kg (15-30 cm) to 140.3±1.0-119.4±0.8 mg/kg (30-45 cm); while the control varied thus: 3.3±0.03 mg/kg (0-15 cm), 3.2±0.01 mg/kg (15-30 cm) to 2.5±0.01 mg/kg (30-45 cm).

I-geo for all metals (Table 4) fell in the categories of 4-5 and >5. All soils with values within 4-5 were soils that were highly polluted to very highly polluted, while those with values greater than 5 were soils that were very highly polluted.

Note: The C values in different depths and for different parameters were used as Bn values for respective metals.

The results of Contaminant factor computation (Table 5) showed that anthropogenic impacts on heavy metal discharge from both waste dumpsites and auto-mechanic activities on the soils were between 97.6 and 99.9%.

3.2 Discussion

According to Chukwulobe et al. [25], solid wastes are sources of environmental pollution through the introduction of chemical substances above their threshold limit into the environment. Heavy metals might be major components of waste streams of industrial and domestic activities and might end up in dumpsites. Dumping of wastes might be responsible for the increased levels of heavy metals in contaminated soils.

The decay and solubility of waste materials on dumpsite might be responsible for heavy metal deposits at dumpsites [25]. The pH values from different soil samples and at different depths were acidic. However, the control samples were also acidic at different depths; this might imply that the soil was naturally acidic. The soil might also be made up of higher content of clay which might be attributed to retention of the heavy metals [25]. Acidic pH values encourage ionization and solubilization of heavy metals in the soil. This might have aided the easy accumulation and concentration of these metals at both waste dumpsites and auto-mechanic villages.

The electrical conductivity at the contaminated sites was higher than at the control site. This might imply high concentration of ionized dissolved chemicals. Increased discharge of waste with high ionic content might encourage levels of EC. The acidic pH value and clay content levels might also encourage increased electrical conductivity levels in dumpsite [26].

The concentrations of heavy metals in the dumpsites were quite higher than the control site values and when subjected to analysis of variance, there were significant variations (P<0.05) which showed that waste dumps might be responsible for the elevated values of the heavy metals. The concentrations of heavy metals reduced significant (P<0.05) as the depth increased which might imply possible loss of affinity of the soil to cations and reduced concentration of clay in the soil. The values of Cu varied from 19.1±0.1 to 16.4±0.2 mg/kg which was below the values obtained by previous works by Chukwulobe et al. [25] and Abdallah et al. [27] in similar study areas. This might imply that the concentrations of Cu in the waste streams from mechanic workshops and market dumpsites were lower when compared with referenced study areas. The concentrations of Cu obtained in this study were lower than the maximum allowable limit of 36 mg/kg set by Department of Petroleum Resources (DPR) [28] and the USEPA [29] permissible limit of 250 mg/kg. The mean concentration of Pb in the sites ranged from 58.2±0.1 to 28.2±0.1 mg/kg. This was in accordance with the result obtained in a similar study by Chukwulobe et al. [25] and Leke et al. [30] but lower than the concentrations obtained by Abdallah et al. [26] in some dumpsite soils within Kaduna metropolis. Moreover, the concentrations of Pb obtained in this study were lower than the USEPA [29] limit of 300 mg/kg and the DPR [28] maximum allowable limit of 85 mg/kg. The concentrations of Fe (410.5±1.1 to 338.2±1.0 mg/kg) were below the 1000 mg/kg permissible limit of DPR [28]. The amounts of iron ranked the highest compared to other heavy metals in the dumpsites. According to Adefemi et al. [31], this is expected because iron occurs at high levels in Nigerian soil.
Table 3. Values of chemical parameters at different locations and indifferent soil depths

<table>
<thead>
<tr>
<th>S/No.</th>
<th>Parameters</th>
<th>A₁</th>
<th>A₂</th>
<th>A₃</th>
<th>B₁</th>
<th>B₂</th>
<th>B₃</th>
<th>C₁</th>
<th>C₂</th>
<th>C₃</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>pH</td>
<td>5.9±0.1</td>
<td>5.2±0.1</td>
<td>5.0±0.1</td>
<td>5.4±0.1</td>
<td>5.4±0.2</td>
<td>5.3±0.1</td>
<td>5.4±0.3</td>
<td>5.4±0.1</td>
<td>5.7±0.2</td>
</tr>
<tr>
<td>2</td>
<td>EC (us/cm)</td>
<td>300.4±1.2</td>
<td>295.8±1.1</td>
<td>280.6±1.0</td>
<td>248.4±1.1</td>
<td>294.7±1.0</td>
<td>280.9±1.0</td>
<td>155.4±0.9</td>
<td>195.8±1.1</td>
<td>150.6±1.0</td>
</tr>
<tr>
<td>3</td>
<td>Cu (mg/kg)</td>
<td>18.8±0.2</td>
<td>16.4±0.2</td>
<td>19.1±0.1</td>
<td>17.9±0.1</td>
<td>18.2±0.1</td>
<td>17.4±0.1</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>4</td>
<td>Pb (mg/kg)</td>
<td>45.9±0.1</td>
<td>40.7±0.2</td>
<td>58.2±0.1</td>
<td>35.7±0.1</td>
<td>30.8±0.2</td>
<td>28.2±0.1</td>
<td>0.4±0.01</td>
<td>0.3±0.01</td>
<td>0.2±0.01</td>
</tr>
<tr>
<td>5</td>
<td>Fe (mg/kg)</td>
<td>380.2±1.1</td>
<td>340.6±1.2</td>
<td>410.5±1.1</td>
<td>360.8±1.0</td>
<td>345.5±1.2</td>
<td>338.2±1.0</td>
<td>5.6±0.01</td>
<td>3.4±0.01</td>
<td>2.6±0.01</td>
</tr>
<tr>
<td>6</td>
<td>Zn (mg/kg)</td>
<td>120.4±1.0</td>
<td>98.5±1.1</td>
<td>140.3±1.0</td>
<td>138.8±1.2</td>
<td>129.6±0.9</td>
<td>119.4±0.8</td>
<td>3.31±0.03</td>
<td>3.20±0.01</td>
<td>2.5±0.01</td>
</tr>
</tbody>
</table>

X = Sample A, B or C, X₁ = 0-15cm, X₂ = 15-30cm and X₃ = 30-45cm

Table 4. Index of geo-accumulation of metals at different locations in different depths

<table>
<thead>
<tr>
<th>Index of geo-accumulation for metals</th>
<th>A₁ Category</th>
<th>A₂ Category</th>
<th>A₃ Category</th>
<th>B₁ Category</th>
<th>B₂ Category</th>
<th>B₃ Category</th>
</tr>
</thead>
<tbody>
<tr>
<td>I-geo Cu</td>
<td>10.0</td>
<td>16.7</td>
<td>15.3</td>
<td>10.0</td>
<td>16.9</td>
<td>14.8</td>
</tr>
<tr>
<td>I-geo Pb</td>
<td>6.3</td>
<td>6.5</td>
<td>4.9</td>
<td>5.9</td>
<td>6.1</td>
<td>7.1</td>
</tr>
<tr>
<td>I-geo Fe</td>
<td>5.5</td>
<td>5.9</td>
<td>6.7</td>
<td>5.4</td>
<td>6.1</td>
<td>6.4</td>
</tr>
<tr>
<td>I-geo Zn</td>
<td>4.6</td>
<td>4.4</td>
<td>5.3</td>
<td>4.8</td>
<td>4.8</td>
<td>5.0</td>
</tr>
</tbody>
</table>

Table 5. Categorization of Contaminant factors of metals and quantification of anthropogenic metal in contaminated soils

<table>
<thead>
<tr>
<th>Metals</th>
<th>Contaminant factors (Cf) of metals</th>
<th>Category</th>
<th>% Anthropogenic impact</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu</td>
<td>4278.6</td>
<td>Very high contamination factor</td>
<td>99.9</td>
</tr>
<tr>
<td>Pb</td>
<td>113.2</td>
<td>Very high contamination factor</td>
<td>99.1</td>
</tr>
<tr>
<td>Fe</td>
<td>90.7</td>
<td>Very high contamination factor</td>
<td>98.8</td>
</tr>
<tr>
<td>Zn</td>
<td>41.6</td>
<td>Very high contamination factor</td>
<td>97.6</td>
</tr>
</tbody>
</table>
The concentrations of Zn (140.3±1.0 to 98.5±1.1 mg/kg) obtained in this study were lower than that obtained in a similar study by Chukwulobe et al. [25] and below the maximum allowable limit of 146 mg/kg [28]. The concentrations of these heavy metals in the waste dumpsites were indicative of the levels and types of industrial activities and the characteristics of wastes generated in Owerri metropolis as compared Kaduna metropolis.

Application of In-geo model at the contaminated sites indicated that the soils were highly to very highly polluted sites. These activities must have contributed so much soil burden of heavy metals in Owerri. The observed increased accumulation at deeper depths of soil might indicate high level of infiltration of these metals into the soil.

This accumulation might result to primary negative effects on soil health, soil biota and ground water quality. The secondary effects might include destruction of soil ecosystem processes and heavy metal poisoning of groundwater.

Contaminant factor values between 0.5 and 1.5 indicate that the source of the metals is natural process [32]. The Cf values of metals were quite >1.5 which implied that the activities at the mechanic villages and waste dumpsites contributed significantly to the heavy metal burden of the soils. The application of Contaminant factor further exposed anthropogenic impact of both activities on soil heavy metal levels. Anthropogenic impact on heavy metal levels of the soil contributed more than 90% of the soil heavy metal levels. The order of anthropogenic impact on heavy metal accumulation in investigated soils is Cu>Pb>Fe>Zn. This was in agreement with the work of Pam et al. [33].

4. CONCLUSION

The models applied in this study exposed the high level of anthropogenic influence on the levels of heavy metals in waste contaminated soils in Owerri, Nigeria.

COMPETING INTERESTS

Authors have declared that no competing interests exist.


32. Akoto O, Ephraim JH, Darko G. Heavy metals pollution in surface soils in the vicinity of abandoned railway servicing


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