ASSESSMENT OF HEAVY METAL DISTRIBUTION IN ROCKS FROM ENYIGBA PB-ZN DISTRICT, SOUTH EASTERN NIGERIA

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ABSTRACT: The environmental and geochemical studies at Enyigba Pb-Zn mining district near Abakaliki South eastern Nigeria were undertaken to characterize the distribution of mobile heavy metals in rocks. A total of twenty-one (21) samples were collected from rock samples in the area. The distribution and determination of total concentration were estimated. The geochemical studies include sampling, partial leach test and chemical analysis for As, Cd, Ca, Co, Cu, Mn, Ni, Pb and Zn using UV-Spectrophotometer. The result revealed high concentration of some of the metals in the sampled media with decreasing concentrations with distance from known Pb/Zn mining sites for mostly Cd, Pb, As, and Ni. Their potential distribution and environmental risk were evaluated using enrichment and contamination factor. Indices of geoaccumulation, contamination and metal enrichment showed generally high values for Cd, As, Pb, and Cu compared with permissible limits and standard. Generally the concentration is in order: Cd > Pb > Cu > As > Co > Zn > Zn; for rock samples. Compared to permissible limits, the degree of enrichment is very high for Cd and moderate for Pb, and Cu in the rocks samples. Also the calculated contamination factor shows low to very high contamination status. These levels of contamination and values indicate that under the prevailing conditions and environmental regulations in Nigeria, the mining district would face major and hazardous discharges of these metals to soil and water sources.

KEYWORDS: Heavy Metal Distribution, Rocks, Nigeria.

1 INTRODUCTION

The study area (Figure 1), 14 km southeast of Abakaliki, covered the Pb-Zn mining district of Enyigba and its surrounding villages of Ameka, Ameri and Ohankwu all in Ebonyi State, southeastern Nigeria. The major occupation of the people living in the area is farming and mining activities. The deposit of Pb-Zn sulphides (galena and sphalerite) in Nigeria have been known for a long time but have only been exploited in the past on a very small scale. The lead-zinc field covers over 48,000/sqkm in extent with lead-zinc mineralization at many centres. Deposits are localized in the Cretaceous sediments along 600/km long belt within the Benue Trough, a sediment filled intracratonic basin extending from Ishiagu (South of Abakaliki) North-eastward to Gombe (Farrington 1952, Olade, 1976, Orazulike 1994). The occurrence of lead–zinc in the Benue valley has attracted a lot of attention. Mining of the ore has been carried out for a long time by both the natives, for local uses as cosmetics, and foreign companies, for export. Production of the ore started in the year 1925 (Offodile, 1989) but commercial production started in 1947 (Kogbe, 1989).

Since the discovery of and mining of Pb-Zn deposits in Abakaliki and its environs in the early 1900s, not much data exist on the impact of their mining on the environment.

Metal contamination that occurs as a result of mining characterized by elevated toxic metal concentrations and acid rock and mine drainage, continue several years after the cessation of mining activities. Heavy metal effluents from the weathering of the mineral deposits and mine dumps affect both the surface and underground water quality and soil. These level of contamination in the area may lead to low agricultural production, and other biological communities if present at anomalously high level. This study is to assess the levels of heavy metals distribution in rocks as may have resulted from lead-zinc mining in the area.
Figure 1. Geological map of lead-zinc deposits of Enyigba district, near Abakaliki, Lower Benue Trough. The area is underlain by Abakaliki shales (Modified from Orajaka, 1965).
The content of these heavy metals in average shale, the crust and normal soil were used as control.

1.1 PHYSIOGRAPHY

In the study area, the topography is undulating plain alternating with running of ridges and hills from east to west. The plains are underlain by shale and some mudstones. The Enyigba, Ameri and Ameka are marked by undulating range of shale outcrops, which serves as the host for Pb-Zn mineral ore bodies. The whole area formed the “Abakaliki antichronium” and generally underlain by shales. The area had about 60m as its highest elevation and 30m as its lowest elevation above sea level. The area falls within the tropical rainforest belt of South East Nigeria, and characterized by an average rainfall of 1750-2000 mm per annum. The rainy season and dry season are the two major seasons that prevail in the area. The vegetation cover in Enyigba is controlled by its climatic condition. The highlands are characterized by drought resistance grasses, along stream and rivers. Among the vegetation includes economic mangoes trees, orange trees, and palm and coconut trees. The drainage system of the area is dendritic in pattern, which is a function of the lithology. The area is majorly drained by Ebonyi River. All the drainage systems flow eastward to join the Cross River somewhere outside the area.

2 GEOLOGY

The Abakali shale of lower Cretaceous age is exposed in the area. The sedimentary rocks are predominantly black calcareous (calcite-cemented) shale with occasional intercalation of siltstone (Figure 2). The shale formation belongs to the Asu-River Group of the Albian Cretaceous sediments. The Asu River group which consists of alternating sequence of shales, mudstone and siltstone with some occurrence of sandstone and limestone lenses in some places and attains an estimated thickness of 1500 meters (Agumanu 1989, Farrington, 1952). Kogbe (1989) described the sediments as consisting of rather poorly-bedded sandy limestone lenses. The shales in some places are highly weathered and ferrugenized. The rocks are extensively fractured folded and faulted. From field observations, the rocks of the area consist of variably coloured shale and mudstone that has been imbedded by lead – zinc vein mineralization, baked intrusive shale as well as ironstone along veins. The ironstone occur as inter-beds within the shale and as vein filling. The vein mineralization is hosted within the dark shale (Nnabo et al. 2011). The geology and mineral resources are the major factors responsible for availability of the heavy metals in the area. While the sulphide mineralization have high concentration of these metals, the shale host are capable of retaining them from ancient sea (Nnabo et al. 2011).

3 METHODOLOGY

Several techniques and scientific methods were employed to achieve and fulfill the aim and objectives of this research work. Unmined, mineralized, unmineralized and altered rock samples were collected from rock outcrops. The samples are typically composite chip samples collected at mining sites. Single grab samples were collected where compositing was not possible. A total of twenty-one (21) rocks samples were collected with their descriptions appropriately recorded (Figure 3). Rock samples were collected from each sampling point and were then crushed to get fine grain sizes. The choice of fine-grained material for this analysis is because higher metals concentration is generally found on smaller grain of rocks due to higher metals surface area to grain size ratio (Kabata-Pendias, 1995). A positive correlation usually exist between decreasing grain-size and higher metal concentration. Moreover this is to say that fine-grained sediments have greater absorption surface area than coarser particles, especially clay minerals, Organic matter and Fe-Mn oxyhydroxide complexes are able to absorb larger quantities of metal through cation exchange processes (Kabata-Pendias, 1995).
Fig. 2: General geologic map of Southeastern Nigeria showing Abakaliki basin in the Lower Benue Trough (Modified after Hoque, 1984)

Figure 3. Map of Enyigba Area showing the Rock Sample Points
In the laboratory, the rock samples were reduced to about 0.5 cm fragments, crushed and then split. For each sample, an approximately 100g portion was pulverized with ceramic mortar and with pestle. The sample was grounded and then sieved until 100% passed an 80-mesh screen (< 180 µm). The sample was mixed to ensure homogeneity, and then served for chemical analysis. The remaining portion was archived for subsequent analysis. To ensure thorough cleanliness to avoid any level of cross-contamination especially when ore-grade samples were prepared, a small amount of the next sample to be prepared was crushed and discarded with the crusher scrubbed out thoroughly prior to preparing the whole sample.

The solid samples from outcrops were digested in the laboratory using the passive leach method that provides a measure of reactions in nature. In this method 100g of the samples was measured and placed in beaker with 200 ml of deionized water, stirred slightly and initial pH and temperature were measured. At 24 hours, the pH and temperature of the leach were again measured and a 60 ml aliquot was taken with a syringe and filtered. The leachate solutions were acidified with 5 drops of 1.1 ultrapure nitric acid (HNO₃) to stabilize metal in solution. The leachate was sent for analysis. A total of nine elements were analysed for and they include As, Ca, Cd, Co, Cu, Mn, Ni, Pb and Zn.

4 RESULTS

The result of analysis for As, Ca, Cd, Co, Cu, Mn, Ni, Pb, and Zn in rock samples for leach test and geochemical test are presented in Tables 1 and 2 below. The summarized basic statistics for rock samples geochemical data are also included.

Table 1: Result of partial leach test of rock samples

<table>
<thead>
<tr>
<th>S/N</th>
<th>Sample No</th>
<th>Initial pH</th>
<th>pH @24hrs</th>
</tr>
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<td>6.4</td>
</tr>
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<td>PN/R/14</td>
<td>9.2</td>
<td>8.9</td>
</tr>
<tr>
<td>6</td>
<td>PN/R/15</td>
<td>8.5</td>
<td>8.4</td>
</tr>
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<td>PN/R/16</td>
<td>8.9</td>
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<td>8.6</td>
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<tr>
<td>11</td>
<td>PN/R/40</td>
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<td>PN/R/41</td>
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<td>4.6</td>
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<td>PN/R/45</td>
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<td>4.3</td>
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<td>PN/R/108</td>
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<td>21</td>
<td>PN/R/149</td>
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</table>
Arsenic was evenly distributed in rocks around Enyigba and its environs. The mean concentration of As was 7.3 mg/kg. The maximum concentration of As was 31.09 mg/kg with range of concentration of 0.66-31.09 mg/kg. Ca was well dispersed in the rocks in the area due to its mineral forming-element, calcite. The total mean concentration of Ca was 98 37.34 mg/kg with the range of 2244.00-15744.00 mg/kg. Its highest occurrence was record in Enyigba around the mining area, while its lowest concentration was recorded in Ohankwu.

The Cd concentration in rocks around Enyigba was not much when compared with other heavy metals. The mean concentration was 4.73 mg/kg while the maximum and minimum concentration was 16.23mg/kg and 0.28 mg/kg respectively. The concentration of Co ranges from 0.84-41.00 mg/kg with the distribution almost equal in most of the samples. It ranges in concentration from 1528.00 mg/kg to 6.00 mg/kg. The mean concentration of Cu was 140.55mg/kg and it ranges from 8.60 to 486.90 mg/kg. Total mean concentration was 16.07mg/kg while Pb total mean concentration was 60.53 mg/kg. The distribution of Pb in the rocks of Enyigba was not much when compared with Ca and Cu respectively Zn was evenly distributed in the rocks of Enyigba and its environs. The total mean concentration Zn was 49.25 mg/kg. The minimum and maximum concentrations were 2.90 mg/kg and 168.78 mg/kg respectively.

5 Discussion

The assessment of heavy metal distribution in rocks from Enyigba and environs was carried out using the Geoaccumulation Index, Metal Enrichment Index (MEI) and Enrichment Factor (EF). These parameters were employed to access the pollution of individual metals of interest in rocks.
5.1 Geoaaccumulation Index

Geoaaccumulation Index (Igeo) is the enrichment on geological substrate and described by equation of Muller,1988 (cited in Yao 2008). The classes of intensity based on Igeo is as given in Table 3. The values of Igeo in rocks of the study area are given in Table 4.

\[
(Igeo) = \log_2 \left( \frac{Cn}{1.5 \times Bn} \right)
\]

(1)

Where Cn is the measured concentration of the element and Bn is the geochemical background value (Average Shale) in earth’s crust.

The concept of Igeo has been widely used to evaluate the degree of metal contamination in terrestrial, aquatic and marine environment, (Yao, 2008; Kumar and Edward, 2009). The geoaaccumulation index compares the measured concentration of the element in the sample fraction; Cn with the geochemical background value, Bn. For samples of mineralized rocks, Bn was considered as average concentrations of the elements in the bed rock unit.

Table 3. Classes of concentration intensity of heavy metal based on Igeo

<table>
<thead>
<tr>
<th>CONTAMINATION INTENSITY</th>
<th>Practically unpolluted</th>
<th>Slightly polluted</th>
<th>Moderately polluted</th>
<th>Moderately to strongly polluted</th>
<th>Strongly polluted</th>
<th>Strongly very strongly polluted</th>
<th>Very strongly polluted</th>
</tr>
</thead>
<tbody>
<tr>
<td>Igeo Class</td>
<td>&lt; 0</td>
<td>&gt; 0 &lt; 1</td>
<td>&gt;1. &lt;2</td>
<td>&gt;2&lt;3</td>
<td>&gt;3&lt;4</td>
<td>&gt;4&lt;5</td>
<td>&gt;5</td>
</tr>
</tbody>
</table>

Table 4. Geoaaccumulation indices (Igeo) of heavy metals in rocks from Enyigba geoaaccumulation indices of heavy metals in rock samples from the Enyigba environs

In terms of Igeo, As, Ca, Co, Mn and Zn values in rocks showed practically uncontaminated condition (Table 4) and made little contribution to I(tot). The content of As in rocks showed only moderate enrichment factor and health risk level varying from 0 to 2.
Cd in rock reached very strong enrichment conditions in many parts of the area. It also contributed significantly to the high I(tot) recorded. In all, Cd showed strong to very strong enrichment. As, Ca, Co, Mn, Ni and Zn, showed practically unpolluted conditions. The main polluting trace metals are Cd, Pb and Cu, followed by Zn, As Mn, Ni and Co.

5.2 Enrichment Factor (EF)

The enrichment factor (EF) was based on the standardization of the tested element against a reference. A reference element is the one characterized by low occurrence variability. The most common reference elements are Si, Mn, Ti, Al and Fe (Kumar and Edward 2009). Mn was used as the reference metal using the formula below. This was justified based on high correlation of Mn with some of the other heavy metals.

\[
EF = \frac{C_n (Sample)}{C_{ref} (Sample)} / \frac{B_n (Background)}{B_{ref} (Background)}
\]

Where \(C_n\) (sample) is the concentration of the examined element in the examined environment, \(C_{ref}\) (Sample) is the content of the reference element in the examined environment and \(B_{ref}\) (background) is the content of the reference element in the reference environment. For this analysis, the average shale was taken as the reference environment while \(C_{ref}\) was taken as the average Mn content of the analysed rock samples (604.67mg/kg, see Table 2) of the Enyigba and environs. The enrichment of heavy metals in rocks of the area is as given in Table 5. Six pollution conditions and health risk levels recognized on the basis of the enrichment factor as shown in Table 6. The health risk level was determined by the value of enrichment factor, so that the enrichment factor represents the impact level of the metal on the environment.

Table 5. Enrichment Factor (EF) of Heavy Metals in Rocks from Enyigba

<table>
<thead>
<tr>
<th>Sample Code</th>
<th>As</th>
<th>EF</th>
<th>Ca</th>
<th>EF</th>
<th>Cd</th>
<th>EF</th>
<th>Co</th>
<th>EF</th>
<th>Cu</th>
<th>EF</th>
<th>Ni</th>
<th>EF</th>
<th>Zn</th>
<th>EF</th>
<th>Pb</th>
<th>EF</th>
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<td>4.04</td>
<td>1.00</td>
<td>10740.00</td>
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<td>1.35</td>
<td>6.38</td>
<td>4.00</td>
<td>0.33</td>
<td>40.28</td>
<td>1.11</td>
<td>5.34</td>
<td>0.10</td>
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<td>0.21</td>
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<td>2.63</td>
<td>310.90</td>
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<td>0.13</td>
<td>0.28</td>
<td>1.32</td>
<td>0.86</td>
<td>0.07</td>
<td>8.60</td>
<td>0.24</td>
<td>1.10</td>
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<td>8.41</td>
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<td>53.10</td>
<td>1.46</td>
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<td>0.44</td>
<td>2.46</td>
<td>11.62</td>
<td>8.04</td>
<td>0.66</td>
<td>73.92</td>
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<td>6.80</td>
<td>0.56</td>
<td>66.60</td>
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<td>0.88</td>
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<td>3.02</td>
<td>14.27</td>
<td>6.00</td>
<td>0.50</td>
<td>56.30</td>
<td>1.55</td>
<td>12.08</td>
<td>0.22</td>
<td>31.78</td>
<td>0.48</td>
<td>152.8</td>
<td>12.63</td>
</tr>
<tr>
<td>PNR/103</td>
<td>2.19</td>
<td>0.36</td>
<td>11735.00</td>
<td>0.66</td>
<td>7.32</td>
<td>34.57</td>
<td>22.04</td>
<td>1.82</td>
<td>219.30</td>
<td>6.04</td>
<td>29.29</td>
<td>0.54</td>
<td>76.16</td>
<td>1.15</td>
<td>51.26</td>
<td>4.24</td>
</tr>
<tr>
<td>PNR/107</td>
<td>2.87</td>
<td>0.47</td>
<td>15744.00</td>
<td>0.89</td>
<td>9.01</td>
<td>42.57</td>
<td>30.06</td>
<td>2.49</td>
<td>294.90</td>
<td>8.13</td>
<td>39.30</td>
<td>0.72</td>
<td>102.12</td>
<td>1.54</td>
<td>68.77</td>
<td>5.69</td>
</tr>
<tr>
<td>PNR/108</td>
<td>24.44</td>
<td>4.00</td>
<td>13050.00</td>
<td>0.73</td>
<td>8.17</td>
<td>38.60</td>
<td>24.70</td>
<td>2.04</td>
<td>244.60</td>
<td>6.74</td>
<td>32.55</td>
<td>0.60</td>
<td>84.65</td>
<td>1.27</td>
<td>56.99</td>
<td>4.71</td>
</tr>
<tr>
<td>PNR/147</td>
<td>4.77</td>
<td>1.00</td>
<td>12682.000</td>
<td>0.71</td>
<td>1.59</td>
<td>7.51</td>
<td>4.82</td>
<td>0.40</td>
<td>47.45</td>
<td>1.31</td>
<td>6.30</td>
<td>0.12</td>
<td>16.43</td>
<td>0.25</td>
<td>11.07</td>
<td>0.92</td>
</tr>
<tr>
<td>PNR/149</td>
<td>5.06</td>
<td>1.00</td>
<td>13442.00</td>
<td>0.76</td>
<td>1.68</td>
<td>7.94</td>
<td>5.03</td>
<td>0.42</td>
<td>50.47</td>
<td>1.39</td>
<td>6.70</td>
<td>0.12</td>
<td>17.47</td>
<td>0.26</td>
<td>11.76</td>
<td>0.97</td>
</tr>
</tbody>
</table>

Av. Shale 10 2.510* 0.3 20 50 80 90 20
Table 6 Pollution condition and health risk (HR) level based on class of enrichment factor

<table>
<thead>
<tr>
<th>EF</th>
<th>Pollution condition</th>
<th>Health Risk level</th>
</tr>
</thead>
<tbody>
<tr>
<td>EF&lt;1</td>
<td>Deficient</td>
<td>0</td>
</tr>
<tr>
<td>EF=1-2</td>
<td>Minimal</td>
<td>1</td>
</tr>
<tr>
<td>EF=2-5</td>
<td>Low</td>
<td>2</td>
</tr>
<tr>
<td>EF = 5-20</td>
<td>Significant</td>
<td>3</td>
</tr>
<tr>
<td>EF = 20-40</td>
<td>Very High</td>
<td>4</td>
</tr>
<tr>
<td>EF &gt; 40</td>
<td>Extremely high</td>
<td>5</td>
</tr>
</tbody>
</table>

Enrichment factor (EF) of heavy metal in rock from Enyigba and environs Ca has extremely high enrichment in four sample points, with the highest at Ameri PN/R/45 (76.69). Cu show significant enrichment in six sample points with highest of 13.42 at Ameri, and this location corresponds to site of high Ca enrichment Pb was significantly enriched with the highest value of 12.63 at Ameri at sample point PN/R/52. Co is significantly enriched in only four sample point with the highest of value 3.39. The level of enrichment is significant in only two sample points with highest value of 5.0 at Ohankwu (PN/R/10). the concentration of Zn enrichment is significantly high in only one sample point with a value of 5.54 which correspond to site of high Ca enrichment.

5.3 Contamination Factor (CF)

The assessment of contamination of rocks of Enyigba and environs was also carried out using the contamination factor. This calculation was used to evaluate the potential risk of the heavy metal to the enrichment using the formula below (Kumar and Edward, 2009).

\[
CF = \frac{C_{i_1}}{C_n}
\]

Where \(C_{i_1}\) is the mean concentration of the metal from sampling sites of Enyigba and environs (at least five) and \(C_n\) was taken as the average concentration of elements in the earth’s crust as a reference value. Four categories of contamination factor have been distinguished in Table 7. Table 8 shows the calculated contamination factor of heavy metals in rock of the area.

Table 7. Categories of contamination based on contamination factors.

<table>
<thead>
<tr>
<th>CF</th>
<th>Category of contamination</th>
</tr>
</thead>
<tbody>
<tr>
<td>(C_{i_1}/C_n &lt; 1)</td>
<td>Low contamination factor indicating low concentration</td>
</tr>
<tr>
<td>(1/C_{i_1}/C_n = 1-3)</td>
<td>Moderate contamination factor</td>
</tr>
<tr>
<td>(C_{i_1}/C_n = 3-6)</td>
<td>Considerable contamination factor</td>
</tr>
<tr>
<td>(C_{i_1}/C_n &gt; 6)</td>
<td>Very high contamination factor</td>
</tr>
</tbody>
</table>

Table 8: Calculated contamination factor in rock from Enyigba

<table>
<thead>
<tr>
<th>Elements</th>
<th>Mean</th>
<th>Contamination factor CF</th>
</tr>
</thead>
<tbody>
<tr>
<td>As</td>
<td>7.31</td>
<td>2.92</td>
</tr>
<tr>
<td>Cd</td>
<td>4.73</td>
<td>59.13</td>
</tr>
<tr>
<td>Co</td>
<td>12.45</td>
<td>0.47</td>
</tr>
<tr>
<td>Cu</td>
<td>140.55</td>
<td>5.21</td>
</tr>
<tr>
<td>Mn</td>
<td>604.67</td>
<td>0.60</td>
</tr>
<tr>
<td>Pb</td>
<td>60.53</td>
<td>5.50</td>
</tr>
<tr>
<td>Zn</td>
<td>49.25</td>
<td>0.68</td>
</tr>
<tr>
<td>Ni</td>
<td>16.07</td>
<td>0.27</td>
</tr>
<tr>
<td>Ca</td>
<td>9837.34</td>
<td>0.32</td>
</tr>
</tbody>
</table>

The contamination factor (CF) of Co, Mn, Zn, Ni and Ca is less than one i.e (CF < 1). This shows low contamination. The contamination factor of As is greater than 1 and less than 3 that is, (CF > 1<3.) This show that As is of moderate
contamination in the rock of Enyigba area. Cu and Pb show considerable contamination factor of 5.21 and 5.5 respectively. The Cd has a very high contamination factor of 59.13 which is the highest value of contamination factor among other heavy metals around Enyigba area.

5.4 The effect of the establishment metal distribution in the rock

It has been established that most of the rocks around Enyigba are moderately to very highly contaminated with heavy metal such as Cu, Pb, Cd, and As. This may be as a result of mining activities going on in the area. The weathered rock formed soil in which plants grow. In most natural setting, heavy metal accumulation in organisms are not very serious because of the natural contaminations of these metals are low in soil it begins when human activities locally disturb the natural cycle. This results in distribution of this heavy element in the rock and soil, where weathering and erosion may also contribute to the dispersion of these heavy elements in the soil. By this process contaminants are introduced into new areas, thereby becoming available for ingestion by greater number of microorganisms. A feature that the heavy metals have in common is that they tend to accumulate in the bodies of organisms that ingest them. Therefore, the concentrations increase up a food chain.

The effect of heavy metal on plants and animals depend on ingested dosage and may cause serious problem to the living organisms. This may lead to:

1. Low agricultural production
2. Decreases among animals
3. Diseases among human beings
4. Death of plants and animals as the case may be.

6 Conclusion

The Enyigba area presents a good case of heavy element distribution and with high concentration of As, Ca, Cd, Co, Mn, Cu, Ni, Pb, Zn in rock were recorded in most the sample points. These are location where active mining took place or still taking place at present. Geoaccumulation index environment factor and contamination factor were used to assess the level of heavy metal distribution in the rock of Enyigba area. Cd shows very high contamination factor of 59.13 which is toxic for both animals and plants. As also shows moderate contamination in the rock. Three parameters were used to evaluate the contamination status of rocks in Enyigba show low to very high contamination.

The comparison between the three methods indicates that the rocks fall, between uncontaminated to highly contaminated status. These levels of metal concentration may have direct toxicity on plants and animals in contact with the soil.
REFERENCES