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Research Paper

Seasonal Variations of Cyanide and Heavy Metals in Groundwater around Enyimba Dumpsite, Aba Southeastern Nigeria.

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The concentration of Cynide and heavy metals in groundwater resources around an active dumpsite (Enyimba) were determined using Atomic Absorption Spectrophotometer with the aim of assessing the suitability of the groundwater for drinking purposes. Fifty groundwater samples were collected in both rain and dry seasons. The mean concentrations of Cyanide and heavy metals (mg/L) in the analyzed groundwater samples in rain season are in this order: Cd (0.03) < Cr (0.134) < As (0.142)< Ni (0.235) <Pb (0.358) < Fe (0.573) < Hg (0.894) < Cu (1.256) < CN (1.862) < Al (22.390). For the dry season, the mean values are in this order: As (0.017) < Fe (0.119) < Ni (0.206) < Cu (0.300) <Pb (0.448) < Cr (0.874) < Hg (0.972) < CN (1.168) < Cd (1.19) < Al (32.730). The result show that the mean concentrations of all the heavy metals and Cyanide were above the WHO standard. Geo-statistical analysis using Pearson correlation and hierarchical cluster analysis for Cyanide and the heavy metals revealed that the likely source of cyanide and heavy metals is from two main sources which include natural (crustal) and anthropogenic (industrial) sources.

Keywords:Cyanide, Heavy Metals, seasonal variation, groundwater*

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I. INTRODUCTION

Increase in human population has led to daily generation of thousands of tons of complex hazardous waste which is not properly separated from Municipal Solid Waste (MSW) (Cilinskis&Zaloksnis, 1996) before they are being disposed. MSW contains both degradable and non-degradable substance.

The appropriate method of refuse disposal is the sanitary landfill. But in most developing countries like Nigeria, refuse are disposed using the open dump method and they are far from standard recommendations. (Magda &Gaber, 2015 ;Esakku, *et al*., 2006 and Oyalemi, *et al*., 2013). Dumpsites are hollow excavations arising from abandoned burrow pits that are unlined and shallow as well as from quarry sites without any environmental impact assessment studies. (Amadi*et al* (2012). Therefore they are point source for pollution as they serve as host for leachate from the dumpsites. (Amadi&Nwankwoala, 2013).

In the dumpsite, refuse undergoes the natural process of decomposition, producing leachate that leaches into the aquifer where they contaminate the groundwater (aquifer) if the soil below the dumpsite is permeable.The contamination of groundwater by heavy metals and other contaminants can cause adverse effects on human health.Heavy metals don't break down like organic waste during decomposition and they have toxic effects on living things when they exceed certain concentration. Though some heavy metals like copper are essential to human health they can still be toxic at increased concentration. (Ali, *et al*., 2014; Ibraheem, *et al*., 2019).Cyanide is not an element, but a compound that is very reactive. It can react very quickly with different kinds of element to form different compounds entirely. The free cyanide is more toxic and harmful than the complex cyanide.. Its half-life in water is in weeks and they are relatively stable unless they are oxidized. (Kwaansah-Ansah*et al*,. 2017, Johnson, 2015; USEPA, 1984; WHO, 2007). USDHHS, 2006 specified Cyanide as a contaminant in the environment and its source is from waste and waste water from industries, exhaust gas from automobiles as well as fires from industries (Dzomback*et al*., 2005; Kuyucak and Akcil, 2013; Jaszezak*et al*., 2017).

In Aba, the study area, risk of exposure to groundwater contamination from leachate produced from dumpsite may be high due to the geological formation of the area (Benin formation).This formation present highly susceptible pathways for exposure to contamination. Therefore, large plume of leachate-contaminated groundwater can penetrate deep into the aquifer and can move laterally several hundreds of meters along the paths of groundwater flow. Transport by groundwater flow in the sand will cause the zone of contamination to greatly expand.

This quest for proper groundwater management has led to evaluate the concentration of Cyanide and heavy metals in groundwater around Enyimba Dumpsite in view of the health and environmental problems resulting fromthe use of polluted water. This research will investigate the seasonal variation in the concentration of Cyanide and heavy metals in groundwater resources around Enyimba Dumpsite in order to ascertain their suitability for domestic purposes.

Study Area Description

Aba is an industrialized and commercial city located between latitude 5.07°N 7.22°SE and longitude 5.177°N 7.367°E in the South Eastern region Nigeria. Aba as a town is a major economic contributor in the country Nigeria because it hosts the textiles, pharmaceuticals, plastics, timbers, cosmetics and shoe manufacturing industries. All these are of different scales of small and large. There is also the Ariaria International market which is a major market in the Eastern part of Nigeria. The population of Aba according to 2006 census is 534,265 having an average population density of 3000 inhabitants per square kilometer (Adindu*et al*, 2012). Aba lies within the rain forest zone in the Southeastern Nigeria. It has two climatic conditions, the dry season and rainy season in a year. The rainfall regime is bimodal and its peaks are in July and September with little dry season known as August break in-between. The rainy or wet season begins about February or March and last till October or early November. The mean annual rainfall of Abia State is between 2550mm and 2890mm. The dry Season starts from December to about February or March although it varies due to seasonal changes. The mean temperature of the city is between 24°C to 34°C and relative humidity of 70% in dry season and 90% in rainy season.

The geology of a place should be well understood before any meaning work on soil and water contamination can be done. This is because the dispersion of contaminants is influenced by the soil porosity as well as the rate of water movement. While some materials may allow contaminants to infiltrate through it to groundwater, others may allow the contaminants to build up around it to a significant level. Sometimes, geology may be the source of contamination. Therefore, the geology of the study area will be understood as this will help in its geo-environmental risk assessment.

Aba lies within a geological area known as the Niger Delta Basin. Short and Stauble (1967); Adegoke, O.S., (1969); Nwajide, C.S., (2013); geologically, classified the study area into three basic Formations having the Akata Formation at its base, the Agbada Formation at the middle and the Benin Formation which is recent at the top. The Benin Formation is the Formation that outcropped in the study area. It is a fresh water bearing formation consists of coastal plain sand which is unconsolidated and dominantly sandy. In Aba and its environs the depth to water table is between 8m – 26m and the aquifer thickness is about 1500m (Adindu*et al*., 2012). The hydraulic conductivity is about 1.13 x 10^{-4} to 5.70 x 10^{-3}m/s with a specific yield of 14.2m/year and an average linear groundwater velocity of 53.46m/year (Agharanya& Dim, 2018).

II. MATERIALS AND METHODS

Sampling was done within the months of December, January and February to represent dry season and the months of May, June and July to represent rainy season for water samples. Soil samples were only collected in the rainy season. The sampling was done at About 5 – 10m radius round the dumpsite to cover industrial, commercial, and residential areas. The water samples were collected directly from boreholes after 5 minutes of pumping from boreholes around the dumpsite. They were collected with labeled polyethylene bottles and kept away from the sun and heat in Styrofoam boxes with ice and then transferred to the laboratory within 12 hrs of collection. Before the bottles were used to collect water samples, they were rinsed with the water sample to be collected to minimize the chance of any contamination. During sampling care was taken to ensure that no air bubble remained inside the bottle, hence the glass stopper was inserted under water. The water samples for the heavy metal investigation were then acidified on collection with 2-3 drops of 1:1 dilution ratio of HNO3. This is to prevent the precipitation of heavy metals (APHA 1995).Distilled water was then added to the flask and stored in 125 ml polypropylene bottle for Atomic Absorption Spectrophotometer (AAS) analyses. Different heavy metals cathode lamps were used to detect various heavy metals in the samples.

The concentrations of total cyanide in the water samples were determined by distillation of the sample to which 10 mL of conc. HCl and 10 mL of 12 % w/v hydroxylamine hydrogen chloride solution has been added to generate hydrogen cyanide gas (HCN), which was absorbed into 2 M NaOH solutions. The resulting sodium hydroxide solution was further diluted to 250 mL out of which 100 mL was titrated against standardized 0.1 M AgNO3 solution using 5 mL of *p*-dimethylaminobenzalrhodanine indicator to the salmon blue end point.

During the analysis and sample preparation, adequate measures were taken to prevent samples contamination and instrumental errors. The borehole sampling points were Geo-referenced using Garmin GPS map 75. A total 50 water samples were collected.

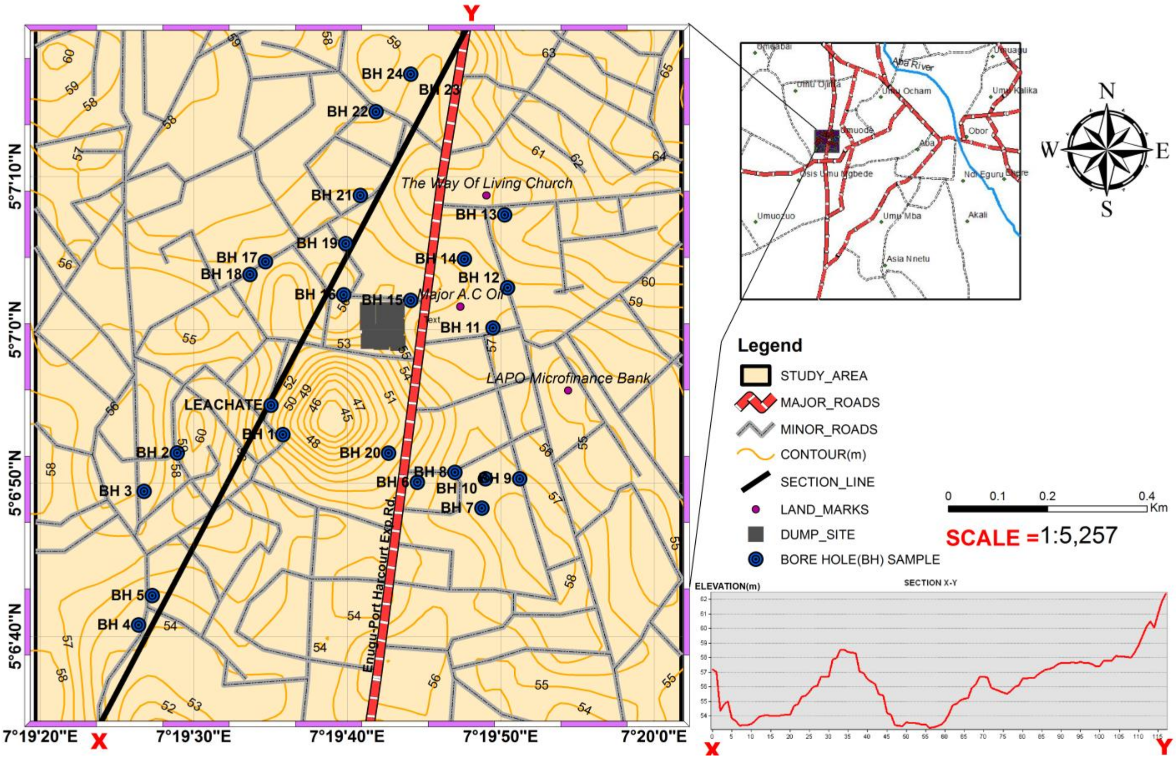

Figure 1: Map of the study area showing the groundwater sampling points.

III. RESULTS AND DISCUSSION

The summary of the descriptive statistics for Cyanide and heavy metals are presented in Table 1. The concentration of Cadmium in groundwater in rain season range from 0.000 to 0.160 mg/L with a mean value of 0.036 mg/L. In dry season, Cadmium concentration range from 0.000 to 0.338 mg/L, with a mean value of 0.119 mg/L.The concentration of Cd is some locations were above the WHO (2011) maximum limit of 0.003 mg/L in both seasons. This acute contamination can be attributed to its availability in the environment as is among the metals with high mobility in aqueous medium. Also it can remain in water as water soluble complexes with dissolved anions and dissolved organic matter and its concentration is not reduced by precipitation as is the case with other metals (Kubier*et al*., 2019). Cadmium solubility in water is enhanced by the acidity of the medium to a large extent (Ros&Slooff, 1987). Its major source in the environment is from phosphate fertilizers and sewage sludge (WHO 2011). Cadmium has been reported to have neuro-toxic effects on human and can be substituted for Zinc in enzymes. It can substitute Ca in bone producing a bone disease known as *itaiitai* (Oni and Hassan, 2013, Stoessel, 2004, Kubier*et al.*, 2019).

The concentration of lead in groundwater ranges from 0.000 to 5.604 and 0.146 to 0.860 mg/L for rainy and dry seasons respectively. Their mean values of 0.358 and 0.448 mg/L during rain and dry seasons are above World Health Organization (WHO, 2011) and Standard Organization of Nigeria (NSDQW, 2007) permissible limits of 0.01 mg/L. The presence of Pb in groundwater around the dumpsite may be attributed to industrial waste that contain lead batteries, pipes, and Pb based paints, also high volume of fairly used electronics such as computers and their accessories, TV sets, fax machines, cell phones, etc. are disposed at the dumpsite and when they decompose, toxic metals like Pb are leached to the aquifer (Schmidt, 2006).Lead interferes with the formation of hemoglobin in the body hence it inhibits iron uptake, kidney dysfunction and permanent brain damage when water contaminated by lead is taken in high dosage. Even when the concentration of lead is low, exposure to it in a high dosage may lead to diminished intellectual capacity (Mortada*et al.*, 2001).

The mean value of copper concentration in the studied area in both rain and dry seasons are 1.256 mg/L and 0.300 mg/L respectively. The rainy season data range is from 0.000 mg/L to 21.546 mg/L while the dry season range is from 0.000 mg/L to 1.401 mg/L. The mean results in rain and dry season are below the WHO (2011) standard of 2mg/L. Copper is an essential nutrient for human body as well as a contaminant found in groundwater.Because of the low mobility of Cu, its reactions with water is also low as they produce a thin protective metallic shield when exposed to water which reduces its reactivity with water (Obasi and Akudinobi, 2020, Nriagu and Pacyna, 1988). Though Cu is not classified as a human carcinogen, high intake of Cu can cause liver and kidney damage (Obasi and Akudinobi, 2020).

The mean concentration of Nickel in groundwater samples around Enyimba dumpsite is 0.235 mg/L in rainy season and 0.206 mg/L in dry season. The values range from 0.000 to 0.150 mg/L in rainy season while thedry season ranges from 0.000 to 0.441 mg/L.

The concentration of Arsenic in groundwater in the rainy season ranged from 0.000 to 0.504 and a mean value of 0.142 mg/L. Groundwater concentration of Arsenic in dry season ranges from 0.000 to 0.049 mg/L having a mean value of 0.017 mg/L. Arsenic a toxic element that can exists naturally in the earth crust formations e.g. rocks, volcanic emissions, and forest fires and they are mobilized into the system through weathering, volcanic emission etc. Anthropogenic activities like mining, combustion of fossil fuel and the use of pesticides, domestic and industrial waste water and herbicides that contain arsenic also introduce arsenic into the environment at higher concentrations especially in drinking water (Biswas, *et al*., 2008; Violante, *et al*., 2006; and Hassan, 2018).Low level exposure to As can cause abnormal heartbeat, pricking sensation in hands and legs etc. while long term exposure can cause diabetes mellitus, hypertension, skin lesions and internal cancers etc. (Obasi and Akudinobi, 2020).

The concentration of Iron in the groundwater sample around Enyimba dumpsite in rain season ranged from 0.000 to 3.780 mg/L with a mean value of 0.573 mg/L. Groundwater concentration of Iron in dry season ranged from 0.000 to 0.338 mg/L with a mean value of 0.119 mg/L. Iron is an essential human nutritional need for up to about 10 – 50 mg/L. It is not considered as a health risk; therefore no health-based guideline value is proposed for Fe (WHO, 2011). It can only affect the taste and colour of water in high concentration.

Chromium concentration in groundwater in the vicinity of the dumpsite ranged from 0.000 to 0.543 mg/L in rainy season with a mean value of 0.134 mg/L. Its concentration in the dry season ranged from 0.360 to 1.410 mg/L with a mean value of 0.874 mg/L. Sources of Cr is mainly through human activities at large like in leather tanning industry where Cr and its salts are used, the manufacture of catalysts, pigments and paints, fungicides, the ceramic and glass industry, and in photography, and for chrome alloy and chromium metal production, chrome plating, and corrosion control (WHO, 1996, Kazakis*et al*., 2017).Ingestion of Cr cause stomach and intestine ulcers, when inhaled can lead to nose ulcer and cancer (Obasi&Akudinobi, 2020). Extreme exposure on high dose may result in serious cardiovascular, respiratory, neurological, gastrointestinal effects and death (Engwa, *et al*., 2018).

Mercury concentration in groundwater ranged from 0.000 to 2.688 mg/L in rainy season with a mean value of 0.894 mg/L. Groundwater concentration in dry season ranged from 0.046 to 1.694 mg/L with a mean value of 0.972 mg/L. This elevated concentration of Hg could be as from waste emanating from different anthropogenic and industrial activities of chemical production going on in the study area and these waste are disposed mostly in the dumpsite as anthropogenic activities are the main source of mercury (ATSDR, 1999). As a neurotoxin, it can affect the kidney, digestive system, immune system, the lungs, skin and eyes.

Aluminum is the third largest metal after oxygen and is very reactive (Momot&Synzynys, 2005). Concentration of Al in rainy season ranged from 4.500 to 40.260 mg/L with a mean value of 22.390 mg/L. In dry season its range is from 3.640 to 76.360 mg/L with a mean value of 32.730 mg/L. The elevated values of Al in the water samples analyzed can be as a result of industrial and commercial activities influence in the production of leachate from Enyimba dumpsite. Since Al is pH dependent and the presence of heavy metals lowers groundwater pH values to acidic environment, it means that more Al is dissolved and are available in the water resources around the study area.Al is not carcinogenic but it can cause neurotoxicity that can lead to the increase risk of neurological disorders like dementia. Epidemiological studies from several countries have shown that there is a correlation between Al in drinking water and Alzheimer's disease (Popugaeva, 2019).

Cyanide is not an element, but a compound that is very reactive. USDepartment of Health and Human Services(2006), specified Cyanide as a contaminant in the environment. Concentration of cyanide in groundwater during the rainy season ranged from 0.084 to 2.840 mg/L with a mean value of 1.862 mg/L. In dry season, its concentration ranged from 0.780 to 2.210 mg/L with a mean value of 1.168 mg/L. These values are above the WHO (2011) and NSDQW (2007) permissible standard of 0.070 and 0.010 respectively. Depending on their occurrence, cyanides are acting poison because they disrupt the process of cellular respiration, causing damage to the nervous system. Cyanide is a toxic species that a teaspoonful of cyanide solution containing 50 to 200 mg of cyanide ingested by an adult human being will result in premature death (Obiri, *et al*., 2007).

Table 1: Cyanide and heavy metals variations in water samples for both rainy and dry seasons.

Metals	Rain	Season	(n=25)	Dry	Season	(n=25)	NSDQW 2007	WHO 2011
	Min.	Max.	Mean	Min.	Max.	Mean		
Cadmium	0.000	0.160	0.036	0.000	0.338	0.119	0.003	0.003
Lead	0.000	5.604	0.358	0.146	0.860	0.448	0.010	0.010
Copper	0.000	21.546	1.256	0.000	1.401	0.300	1.000	2.000

Nickel	0.000	0.150	0.235	0.000	0.441	0.206	0.020	0.007
Arsenic	0.000	0.504	0.142	0.000	0.049	0.017	0.010	0.010
Iron	0.000	3.780	0.573	0.000	0.338	0.119	0.300	0.100
Chromium	0.000	0.543	0.134	0.360	1.410	0.874	0.050	0.050
Mercury	0.000	2.688	0.894	0.046	1.694	0.972	0.001	0.006
Aluminium	4.500	40.260	22.390	3.640	76.360	32.730	0.200	0.100
Cyanide	0.084	2.840	1.862	0.780	2.210	1.168	0.010	0.070

Statistical Analysis

Correlation coefficient is a statistical evaluation tool used to ascertain relationships between variables. It helps to understand how one variable can help predict the other by establishing an association between them. If correlation coefficient (r) between two variables is +1, it shows that they are perfectly related linearly, meaning that they have the same source, a coefficient of -1 show a negative linear relationship showing that the variables are from different sources. r = 0 indicates that no relationship exist between the two variables (Chinemelu&Nwankwor, 2019).

The Pearson correlation Matrix for the heavy metal levels in groundwater were evaluated at a significant level of 5%. For the rainy season (Table 2), the correlation matrix showed a strong positive correlation between Cu and Pb with a value of 0.972. This means that these two metals are from the same anthropogenic source. High positive correlations were observed among Fe and Cd, Hg and Cr then Al and Cd with coefficient correlation values (r) of 0.629, 0.610 and 0.566 respectively.

Table 2: Rain Season Pearson Correlation Matrix

	Cd	Pb	Cu	Ni	As	Fe	Cr	Hg	Al	CN
Cd	1.000									
Pb	-0.210	1.000								
Cu	-0.265	*0.972	1.000							
Ni	-0.143	-0.125	-0.134	1.000						
As	0.301	-0.023	-0.040	-0.040	1.000					
Fe	*0.629	-0.170	-0.137	0.081	0.136	1.000				
Cr	0.156	-0.060	-0.176	0.158	-0.022	-0.036	1.000			
Hg	-0.146	0.069	0.010	0.120	0.039	-0.254	*0.610	1.000		
Al	*0.566	0.043	-0.013	0.354	0.203	0.312	0.383	0.173	1.000	0.319
CN	0.320	-0.120	-0.262	0.195	0.356	0.214	0.308	0.159	0.319	1.000

* Correlation is significant at the 0.05 level (2-tailed).

The Pearson correlation matrix of cyanide and heavy metals in dry season are presented in Table 3. The result revealed that a low positive correlation exist between Hg and Fe with coefficient value (r) of 0.405. This shows that these two metals may have come from the same source. However, moderate negative correlation of r = -0.619 exist among Fe and Cu signifying that they are from different source. Also, Hg and Cu had a low negative correlation of r = -0.479.

Table 3: Dry Season Pearson Correlation Matrix

	Cd	Pb	Cu	Ni	As	Fe	Cr	Hg	Al	CN
Cd	1.000									
Pb	-3.5e-05	1.000								
Cu	-4.0e-01	4.6e-02	1.000							
Ni	-1.4e-01	-7.5e-02	-0.081	1.000						
As	-2.2e-02	5.4e-02	0.232	0.196	1.000					
Fe	-3.9e-02	-2.0e-01	* -0.619	-0.289	-0.285	1.000				
Cr	3.0e-01	-2.5e-01	-0.023	-0.158	-0.105	0.351	1.000			
Hg	2.9e-01	9.4e-02	* -0.479	-0.088	-0.351	*0.405	0.308	1.000		
Al	-1.8e-01	2.5e-01	0.231	-0.088	0.239	-0.134	0.011	-0.259	1.000	0.157
CN	1.5e-01	5.0e-02	-0.040	0.099	-0.179	-0.063	0.269	0.246	0.157	1.000

* Correlation is significant at the 0.05 level (2-tailed).

Cluster Analysis

The hierarchical cluster analysis (HCA) of cyanide and heavy metalsare presented in Fig. 2 and Fig. 3. This analysis helps in organizing the components into different classes and different sources. This was done in accordance to Ward's method and the distance apart is a reflection of the extent of the relationship among the components.

For rain season, the dendrogram indicates that there are two main clusters (Fig. 3). Cluster one has Pb and Cu while cluster two has two sub cluster in which the first cluster contains Ni, Cr and Hg while the second cluster involves Al, Cd and Fe in one group and As and CN in another group. The relationship between Pb and Cu agrees with the results of the correlation for rain season.

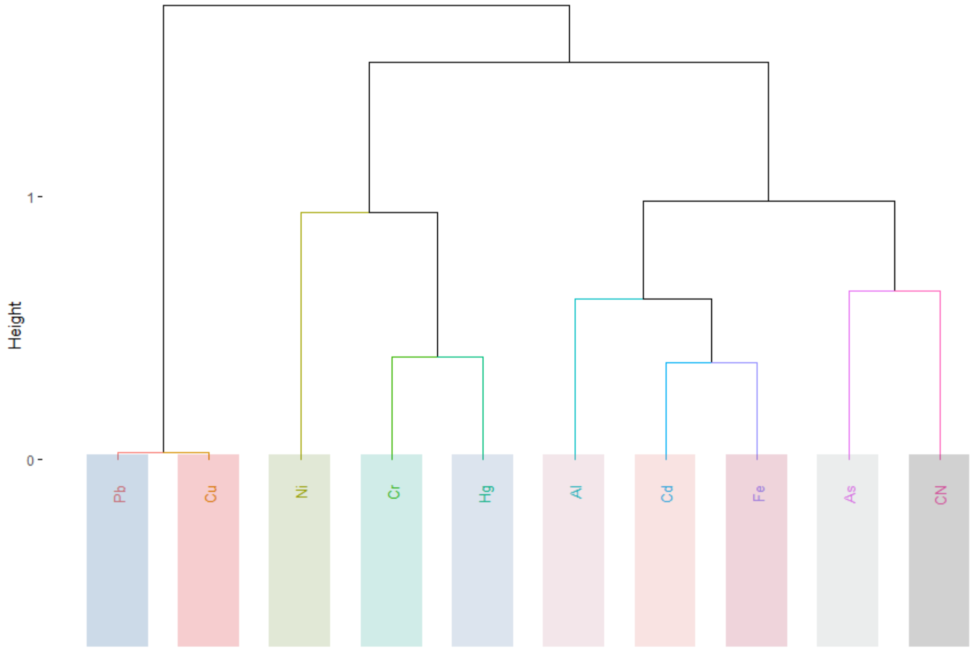

Figure 2: Dendrogram of cyanide and heavy metal pollution in rain season in the study area

The dendrogram for dry season also produced two main clusters. CN, Cd and Cr are in one cluster. The second cluster contains two sub clusters with one containing As, Hg, Cu and Fe while Ni, Pb and Al are in another cluster. These cluster associations is an indication that the contaminants are from two sources. They are from natural and anthropogenic activities which involve industrial and commercial activities going on in the study area and the waste from these activities are disposed indiscriminately at the dumpsite.

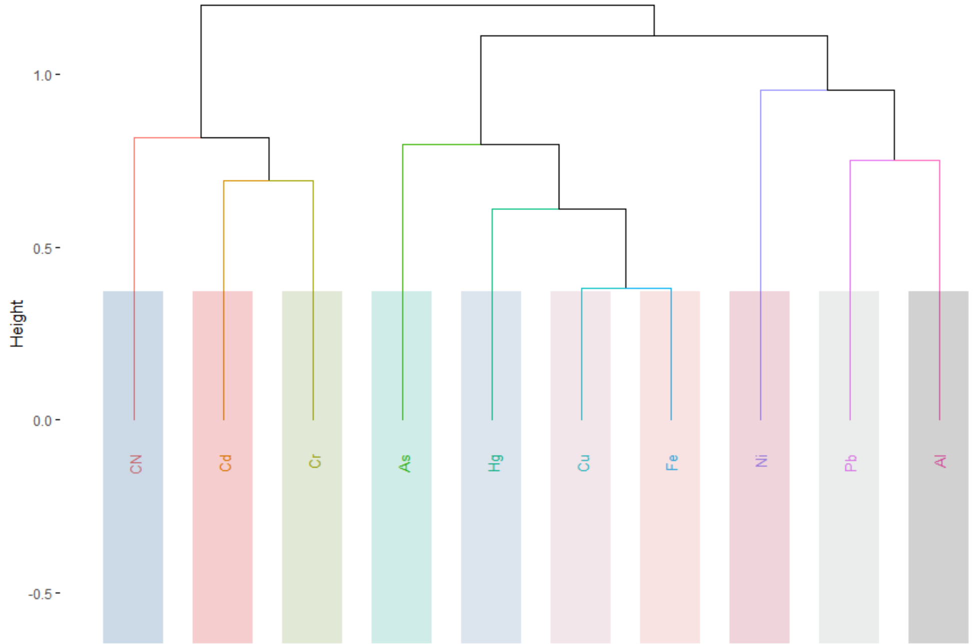

Figure 3:Dendrogram of cyanide and heavy metal pollution in dry season in the study area

IV. CONCLUSION

This study determined the level of concentration and the source of cyanide and heavy metals in groundwater around the vicinity of Enyimba dumpsite Aba, Southeastern Nigeria. The mean concentration of these identified contaminants Cd, Pb, Cu, Ni, As, Fe, Cr, Hg, Al, CN were found to be higher than the permissible limits in both seasons. The correlation analysis PCA and HCA revealed association of cyanide and metal under consideration. The result showed that there are two main sources of pollution in the study area. They include natural from the earth's crust as well as from industrial activities going on in the study area. It was recommended that sanitary landfill should be used instead of dumpsite in other to protect the groundwater resources. It is also advised that groundwater from the study area should be treated before consumption and the concentration of heavy metals and other contaminants should be monitored regularly. Awareness should be made to the populace on the effects of these contaminants on human health and also to the environment at large.

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