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Source Apportionment and Health Risk of Heavy Metals in Contaminated Agricultural Soil from Egi, Rivers State, Nigeria

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Abstract: The study was to identify the source and evaluate the health risk of heavy metals in contaminated agricultural soil from Egi community (Oboburu, Obagi and Ogbogu), Niger Delta, Nigeria. Multivariate and health risk equations were employed to achieve this. The mean of heavy metals in samples of Oboburu indicated high levels for Fe (271.0±156.6 mg/kg), Pb (111.2±94.97 mg/kg), and Mn (27.92±23.80 mg/kg), Obagi was high for Fe (248.4±175.9 mg/kg), Mn (80.46±12.40 mg/kg), and Co (116.3±193.7 mg/kg), and Ogbogu was high for Fe (160.9±150.3 mg/kg), Co (240.1±412.1 mg/kg), and Mn (42.90±16.82 mg/kg) respectively. Geo-accumulation index indicated that Pb, Cd and Co was of high contamination and the enrichment factor showed that metals were of anthropogenic sources. The Principle component analysis revealed three (3) components accounting for 88.448%, which revealed that the contamination in the sample was more of anthropogenic than biogenic. The Cancer Risk estimate for children population was of high risk for Ni, Cr, Cd and Pb (10^{-5} to 10^{-6}) while adult population was 10^{-6} and lower. The non-carcinogenic risk indicated highest value for children in Co (2.75), Pb (1.70E-1) and Fe (1.32E-1), while adult in Co (1.007) and Fe (3.29E-2). The findings suggest human activities have contributed greatly to the contamination of the agricultural soil and usage of such may be of risk to the users.

Keywords: Heavy Metals, Source Apportionment, Risk, Egi

1. Introduction

Incessant release of heavy metals from anthropogenic sources causes significant changes in the biogeochemical cycle of those elements. Soil contamination is mostly due to contaminants released from point sources, as it has been shown in studies concerning developing countries that have petroleum and natural gas as their main natural resource [1]. Although diverse sources contributes to the contamination of the environment, crude oil exploitation and production activities are seen as the leading source of related contaminant into the soil [2]. Diffuse sources can also contribute to the contaminant in the soil. Gas flaring has been a major cause of contaminant into the soil [3]. The study was to identify the source and evaluate the human health risk of heavy metals in contaminated agricultural soil from Egi oil producing communities (Oboburu, Obagi and Ogbogu) in Ogba/Egbema/Ndoni local government Area of Rivers State, Niger Delta, Nigeria.

2. Materials and Method

2.1. Description of the Study Area

The soil samples were collected from four different selected contaminated farmlands from Oboburu Obagi, and Ogbogu all in Ogba/Egbema/Ndoni local government area (ONELGA) in Rivers State, Nigeria. The local government stretches from longitude 6°28’ 13”E through longitude 6°47’ 34”E and latitude 5°9’ 42” N through 5°44’ 3”N [1]. The study map is shown in figure 1 below.

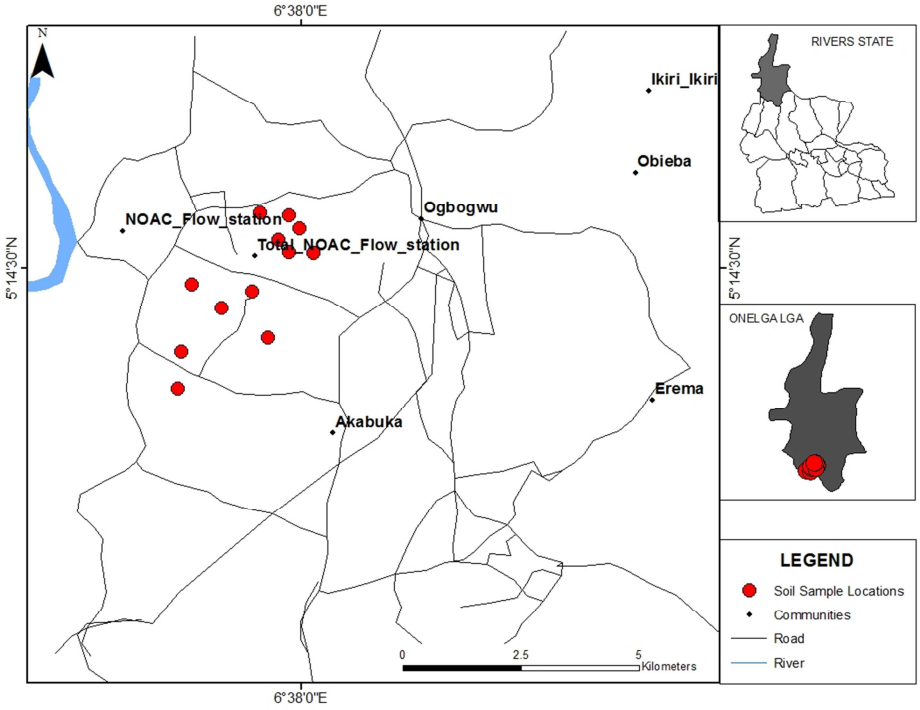

Figure 1. *Sample locations across the Study area in Egi.*

2.2. Sample Collection

In each of the selected sites, six quadrats were marked. In each quadrat, four core soil samples were collected randomly at depth 0–20 cm using a soil auger and put together to form a composite sample. The composite samples were well-mixed and sub-samples taken.

2.2.1. Sample Preparation

The soil samples were first air and sun dried, and then placed in electric oven at a temperature of 40^{o}C approximately for 30 minutes. Thereafter, homogenized having been previously ground and sieved through sieves of stainless steel 2mm mesh, to get the particle size needed for further analysis.

2.2.2. Digestion of the Soil Sample

To determine total heavy metal content, 2 g of the treated samples were weighed placed into digestion vessels and digested with HCl, HNO_3, HF, and $HClO_4$ by means of graphite furnace digestion instrument. Then the solutions were diluted into a final volume of 50 ml with 2% (v/v) HNO_3. Selected heavy metals (Mn, Fe, Cr, Zn, Pb, Ni, Cd, Cu and Co) concentrations were determined by Atomic absorption spectrometry [4].

2.3. Contamination Index

2.3.1. Geo-accumulation Index

The heavy metal contamination level based on baseline concentrations were calculated using the method proposed by Muller [5], known as geoaccumulation index (Igeo). This method assesses the metal pollution in terms of seven (0 to 6) enrichment classes ranging from background concentration to very heavily polluted, as follows:

$$Igeo = \log_2 (Cn/1.5Bn) \quad (1)$$

Cn is a heavy metal concentration in the sample of the study area, Bn is the geochemical background value in average shale of element [6], the shale value were 1.5 is the background matrix correction in factor due to lithogenic effect.

2.3.2. Enrichment Factor

This is an index in which the metal concentrations of the reference metal in the sample are compared to the world average shale values, as indicated in the formula below [7].

$$EF = (M/Fe)_{sample}/(M/Fe)_{background} \quad (2)$$

Where M stand for metal ion concentration in both the sample and background while Fe is for iron concentration in both the sample as well as the background. Zhang and Liu [8], opines that EF values from ½ to 1.5 shows the metal is of crustal or natural processes, while values more than 1.5 is of anthropogenic source in soils.

2.4. Health Risk Assessment

2.4.1. Cancer Possibility Evaluation

The preliminary risk assessment was based on equations described by the US Environmental Protection Agency [9].

$$CDIm = (Msoil \times IR \times EF \times ED \times CF)/(BW \times AT) \quad (3)$$

where CDIm = metal daily intake (mg/kg/day); Msoil = metal concentration in soil (mg/kg); IR = the ingestion rate of soil (mg/day); EF = exposure frequency (day/year); ED = the exposure duration (year); BW = the body weight

(kg); AT = the averaging time (days); and CF = the conversion factor (10^{-6}kg/mg). The above equation assumes 100% bioavailability of the ingested soil-borne contaminant. However, contaminant bioavailability is dependent on the metal speciation and soil properties such as pH, texture, and organic matter. Equation 2 can therefore be adjusted to include a relative bioavailability factor as follows [10].

$$\text{Risk} = \text{CDIm} \times \text{SF} \quad (4)$$

where Risk = probability of carcinogenic effect (unitless) and SF= cancer slope factor $(\text{mg/kg/day})^{-1}$.

2.4.2. Non-Carcinogenic Risk

For non-cancer risk, the hazard quotient (HQ) was calculated
using Eq. 5 [9].

$$\text{HQ} = \text{CDIm}/\text{RfD} \quad (5)$$

where RfD = reference dose (mg/kg/day).

2.5. Statistical Analysis

Excel 2010 (Microsoft Office, Microsoft,) was employed the descriptive statistics, and multivariate analyses, which calculated the Principal Component Analysis (PCA) and correlational matrix.

3. Results and Discussions

3.1. Concentration of Heavy Metals

The mean of the heavy metal levels from the three communities is shown in table 1 below. The mean concentration of heavy metals in soil samples from Oboburu community indicates concentrations as follows; iron (271.0±156.6 mg/kg), lead (111.2±94.97 mg/kg), manganese (27.92±23.80 mg/kg), copper (23.30±18.56 mg/kg), chromium (17.27±10.31 mg/kg), nickel (5.853±2.190 mg/kg), cadmium (5.650±5.749 mg/kg), cobalt (11.28±16.33 mg/kg) and zinc (6.980±2.800 mg/kg), Obagi community results also shows heavy metals concentration as follows; iron (248.4±175.9 mg/kg), cobalt (116.3±193.7 mg/kg), manganese (80.46±12.40 mg/kg), lead (64.75±59.94 mg/kg), copper (34.15±29.57 mg/kg), chromium (11.51±8.385 mg/kg), zinc (8.728±5.845 mg/kg), nickel (3.973±2.843 mg/kg) and cadmium (0.770±0.830 mg/kg) while Ogbogu community also had metals concentration like cobalt (240.1±412.1 mg/kg), iron (160.9±150.3 mg/kg), manganese (42.90±16.82 mg/kg), lead (10.63±10.64 mg/kg), chromium (3.820±3.886 mg/kg), copper (2.495±2.516 mg/kg), zinc (1.630±0.724 mg/kg), nickel (1.458±1.547 mg/kg) and cadmium not detected.

Table 1. *Mean of heavy metals (mg/kg) in contaminated soil samples of studied communities.*

H M	Oboburu	Obagi	Ogbogu	Ase-Azaga	WHO Limits
Manganese (Mn)	27.92±23.80	80.46±12.40	42.90±16.82	ND	2000
Iron (Fe)	271.0±156.6	248.4±175.9	160.9±150.3	2.769	50000
Chromium (Cr)	17.27±10.31	11.51±8.385	3.820±3.886	ND	100
Zinc (Zn)	6.980±2.800	8.728±5.845	1.630±0.724	0.346	300
Lead (Pb)	111.2±94.97	64.75±59.94	10.63±10.65	0.745	10
Nickel (Ni)	5.853±2.190	3.973±2.843	1.458±1.547	ND	50
Cadmium (Cd)	5.650±5.749	0.770±0.830	ND	ND	3
Copper (Cu)	23.30±18.56	34.15±29.57	2.495±2.516	0.045	100
Cobalt (Co)	11.28±16.33	116.3±193.7	240.1±412.1	ND	3

[11]

This results indicates high level of contamination and higher than values from the control site aze-azaga in Ndoni area of Ogba/Egbema/Ndoni LGA of Rivers State. These values were lower than works by Fosu-mensah *et al*., [12] Pb (184.44 mg/kg), Cd (103.66 mg/kg), Cu (202.99 mg/kg) and Ni (72.00 mg/kg), but higher than reports by Farid *et al*., [13] Cd (0.00-0.111ppm), Pb (0.87-8.97pmm) and Ni (0.017-1.72 ppm). The mean from the three study communities were high. This level of contamination could be attributed to oil and gas production activities in the study area. The high amount of iron is ascribed to the iron pipes use in drilling and oil pipeline transport from well head to the various oil installation and storage facilities. This shows that generally on the average there is not much surface soil contamination at the locations.

3.2. Contamination Indexes

The geo-accumulation index result showed no contamination for most of the heavy metals except for Pb which had moderate contamination for soil from Oboburu (1.89) and Obagi (1.11). The others were Cd with heavy contamination for soil samples from Oboburu (3.650) and moderate contamination for samples from Obagi (0.775). Cobalt was of moderate to heavy contamination for Obagi (2.107) and heavily contaminated for samples from Ogbogu (3.153).

Table 2. *Geo-accumulation Indexes of heavy metals in contaminated soil.*

	Geo-accumulation Index		Ogbogu
	Oboburu	Obagi	
Mn	-5.422	-3.986	-4.893
Fe	-8.029	-8.155	-8.781
Cr	-2.967	-3.552	-5.143
Zn	-4.352	-4.029	-6.450
Pb	1.890	1.110	-0.451
Ni	-1.241	-1.409	-6.128
Cd	3.650	0.775	-
Cu	-1.534	-0.983	-4.758
Co	-1.259	2.107	3.153

Table 3. *Enrichment Factors of heavy metals in contaminated soil.*

	Enrichment	Factor	
Mn	5.724	17.99	14.81
Fe	1	1	1
Cr	31.86	23.17	11.87
Zn	12.88	17.57	5.065
Pb	1026	651.7	165.2
Ni	21.60	15.99	9.061
Cd	3228	487.4	1425
Cu	85.98	137.5	155.1
Co	104.1	1170	3927

The enrichment factor indicated that all the contaminants were of anthropogenic origin.

3.3. Source Apportionment

The table below indicates the principle component loadings of the different heavy metals from the study area. Principal component analysis helped in identifying the factors and sources responsible for the variation in heavy metals in soil sample. Result revealed that 3 principal components were extracted and that accounted for 88.448% of the total variance in the observed variables with component 1 accounting for 48.316% while component 2 and 3 was 25.692% and 14.44% respectively. It showed that the contamination in the soil sample was greatly influenced by Pb, Ni, Cd, Cr and Zn for component 1, Mn and Fe for principal component 2 and Co for principal component 3. This result indicated that metals from PC1 are of same sources which are anthropogenic like combustion (coal, wood, diesel, petrol and kerosene), incineration and vehicle exhaust. The component 2 indicated possible natural sources which could be from seepages, erosion, geogenic and others. These findings correlate to work by Nuamah [14] on sources of heavy metals in soil from Akuse area, Ghana with Ni and Pb from anthropogenic sources and Cu from natural sources. The third component indicated a mixed source of both anthropogenic and biogenic sources which had Co in abundance. It can be clearly stated that since majority of the heavy metals are of anthropogenic it can be due to oil and gas related activities which is the major industrial or emission activity in the area of study.

Table 4. *Principal component loadings of heavy metals in contaminated soil sample from the study area.*

Heavy metals	Principal Components		
	PC1	PC2	PC3
Mn	-0.253	0.886	0.079
Fe	0.129	0.751	0.271
Cr	0.921	0.066	-0.028
Zn	0.786	0.415	-0.406
Pb	0.966	-0.133	-0.086
Ni	0.941	0.111	-0.192
Cd	0.839	-0.522	0.007
Cu	0.513	0.656	-0.443
Co	-0.124	0.232	0.902
Eigen value	4.348	2.312	1.300
Loading %	48.316	25.692	14.44
Cumulative %	48.316	74.008	88.448

3.4. Cancer Risk Assessment

This work specially targets at the adult and children exposure scenario. The juvenile scenario was considered to establish the likely health effect on children exposed to similar conditions. The result in below shows the carcinogenic risk for some heavy metals in the soil from the study area. Table 4 below shows the result of the carcinogenic risk of some heavy metals from the contaminated soils. The slope factors were used to evaluate the carcinogenic risk (CR). Average predictable life time risk to cancer due to heavy metal intake estimated in Oboburu study area were as follows; Chromium had a value of 2.84E-5, Nickel was 3.26E-5 and the least metal was Cadmium at 7.07E-6 while the risk for adults were high in the following metals; Nickel (8.18E-6), Chromium (2.36E-6) and least in Lead (7.77E-7).

Table 5. *Carcinogenic risk of heavy metals in contaminated soil samples of Egi study area.*

Children			
Heavy Metals	**Oboburu**	**Obagi**	**Ogbogu**
Cr	2.84E-5	1.85E-4	3.70E-5
Pb	3.10E-6	1.81E-6	1.80E-6
Ni	3.26E-5	2.23E-5	4.90E-5
Cd	7.07E-6	9.61E-7	-
Adult			
Cr	2.36E-6	4.73E-6	1.57E-6
Pb	7.77E-7	4.52E-7	7.43E-8
Ni	8.18E-6	5.54E-6	2.04E-6
Cd	1.76E-6	2.41E-7	-

Obagi showed high values for two metals Chromium (1.85E-4) and Nickel (2.23E-5) while the least metal was cadmium (9.16E-7) for children and adult were Nickel had 5.54E-6 followed by chromium at 4.73E-6 and least was cadmium with a value of 2.41E-7. Ogbogu were as follows, for the children Chromium was 3.70E-5 and Nickel was 4.90E-5 while the least was Lead (1.80E-6). Cadmium was nil because it was no detected in the analysis. Chromium showed the highest level of risk followed by Nickel, while lead and cadmium were least from the result as depicted above.

In similar study Afrifa *et al.,* [15], had cancer risk value of 10^{-7} and $10^{—6}$ for lead in children and adults due to heavy metal Exposure from Soil Dust at Selected Fuel Filling Stations in Accra which is lower than this work. The result from this work is higher than reports by Yang *et al.,* [16] on heavy metals in the soil of an urban recreational area amended with Composted Sewage Sludge, in China. The result also shows a higher value compared with report by Gyimah *et al.,* [17] (10^{-6} to 10^{-10}), but lower than report by Ekpete and Owhoeke [18] (10^{-2} to 10^{-4}). The values were The heavy metal of concerns are Chromium and Nickel, though others are of low risk but there is need to check the rate of effluence discharge and oil activities within the area of study. The result from these sites though not high enough is an indication of contamination which could be of risk to adults and children within the areas. It is relevant that actions be

taken on the possible way to remediate spillages properly as to see an end to pollution of these areas.

3.5. Non Carcinogenic Risk

For non-carcinogenic risk, the HQ values of these heavy metals and HI values for children and adults varied considerably across the three communities. The HQ was of the following order Co>Pb>Fe>Cr>Cd for children in Oboburu community and for adult was of the order as follows; Co>Pb>Fe>Cd. The table also shows that Ogbogu community has heavy metals with HQ of the order Pb>Fe>Cr>Cu>CD for the children and HQ for the adult population was high as follows Co>Pb>Fe>Mn. The HQ for Ogbogu community with that of the children high in the order as follow; Co>Fe>Pb>Cr and adults hazard quotients were high in Co>Fe>Pb>Cr respectively. The THQ was greater than 1 in Cobalt, also high but less than 1 in some other heavy metals like Pb>Cr>Fe>Cd>Cr. The entire result shows a hazard quotient less than 1 which is safe for inhabitants within the area. The HQ for Cobalt was greater than one which is high and not safe for those living within the area of study.

Table 6. *Hazard Quotients of heavy metals in contaminated soil samples for children and adults.*

Children				
Heavy Metals	**Oboburu**	**Obagi**	**Ogbogu**	**Total Hazard Quotient**
Mn	6.56E-5	2.64E-4	1.01E-3	1.34E-3
Fe	5.24E-2	4.81E-2	3.11E-2	1.32E-1
Cr	1.89E-2	1.20E-2	4.17E-3	3.51E-2
Zn	7.63E-5	9.57E-5	1.79E-5	1.90E-4
Pb	1.01E-1	5.92E-2	9.69E-3	1.70E-1
Ni	9.60E-4	6.55E-5	2.48E-3	3.50E-3
Cd	1.86E-2	2.53E-3	-	2.11E-2
Cu	2.05E-3	3.03E-3	2.22E-4	5.30E-3
Co	1.24E-1	1.27E-3	2.63000	2.75527
Adults				
Mn	1.63E-3	4.72E-4	2.52E-4	2.35E-3
Fe	1.31E-2	1.20E-2	7.76E-3	3.29E-2
Cr	1.58E-3	3.15E-3	1.05E-3	5.78E-3
Zn	1.91E-5	2.39E-5	4.47E-6	4.66E-5
Pb	2.54E-2	1.48E-2	2.43E-3	4.26E-2
Ni	2.40E-4	1.63E-4	6.00E-5	4.63E-4
Cd	4.64E-3	6.35E-4	-	4.70E-3
Cu	5.19E-4	7.59E-4	5.54E-5	1.33E-3
Co	3.09E-2	3.19E-1	6.57E-1	1.0069

THQ >1 the population is considered unsafe, THQ < 1 the population is considered safe.

This result was a bit lower than works by Aluko *et al.,* [19], in soil within areas of iron mines of Itakpe and Agbaja, Kogi State. The implication of the findings is that the risk associated with non-carcinogenic contaminants is not much except for Cobalt which showed a value greater than 1. The presence of cobalt could be dangerous if they accumulate in the body system, taken by plants or washed by into water bodies.

3.6. Correlational Matrix

Table 7 showed the relationship between heavy metals in soil samples. Result revealed significant negative relationship between the concentration of Mn and Cd (r =-0.652, $p<0.05$) while Cr concentration in soil sample was found to have significant positive relationship with Pb concentration (r =0.848, $p<0.01$), Ni (r =0.822, $p<0.01$) and Cd (r= 0.848, $p<0.01$) and Cr (r =0.883, $p<0.05$). The concentration of Pb shows significant positive relationship with Ni (r =0.908, $p<0.01$) and Cd (r=0.884, $p<0.01$) while the concentration of Ni shows significant positive relationship with Cd (r = 0.722, $p<0.01$) and Cu (r =0.623, $p<0.01$). Results obtained between other heavy metals were not significant ($p>0.05$).

Table 7. *Correlation matrix for heavy metals in soil samples.*

		1	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**
1	Mn	1								
2	Fe	0.511	1							
3	Cr	-0.146	0.155	1						
4	Zn	0.182	0.192	0.739**	1					
5	Pb	-0.333	-0.02	0.848**	0.759**	1				
6	Ni	-0.195	0.220	0.822**	0.848**	0.908**	1			
7	Cd	-0.652*	-0.289	0.725**	0.443	0.884**	0.722**	1		
8	Cu	0.375	0.348	0.498	0.883**	0.425	0.623**	0.083	1	
9	Co	0.334	0.229	-0.161	-0.307	-0.225	-0.284	-0.221	-0.221	1

*significant at 5% ($p<0.05$), **significant at 1% ($p<0.01$).

4. Conclusion

The levels of pollution from the metals showed that of concern across the areas studied since human activities goes on unabated. Principle component analysis applied in this study proved useful in the characterization of heavy metal sources in soils from Egi communities in Rivers State. It showed that majority of the heavy metals were of anthropogenic origin, two metals of biogenic and one of mixed origin. As revealed in this study, for both adults and children, the carcinogenic and non-carcinogenic risk of the heavy metals are not high except for cobalt which showed slight risk for children and adult in the non-carcinogenic estimation. Considering the health hazards from the accumulation of heavy metals, it is quite needed to properly monitor drilling operations in the areas. The present study provides a good basis for further research on the impact of mining and its various processes to the environment.

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