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Chemical Characteristics of Discharges from Two Derelict Coal Mine Sites in Enugu Nigeria: Implication for Pollution and Acid Mine Drainage

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Abstract

In this study, the chemical composition of water and sediments contiguous to two abandoned coal mines in southeastern Nigeria, were assessed to determine the impact on the geoenvironment and potential for acid mine drainage (AMD). Parameters including the pH, anions and cations, and the heavy metals were measured. These were used to evaluate contamination/pollution using hybrid factors including Pollution Load Index, factors, enrichment factors, pollution load index and index of geoaccumulation. The pH range of 3.4 to 5.9 classified the water as weakly to strongly acidic, typical of AMD. The SO42– ion, which indicates pollution by mine waters, showed moderate to high concentrations. Iron, zinc lead and copper were the most abundant heavy metals. Pollution Load Index values were greater than unity which shows progressive deterioration in water and sediment quality. The Enrichment Factor values of up to 1 indicated enrichment through lithogenic and anthropogenic sources. The mine dumps serve as pools that can release toxic heavy metals into the water bodies by various processes of remobilization. Based on the lithology, mineralogy, chemical concentrations and environmental factors, the study has shown that there exists a potential for the generation of AMD. The heavy metals enriched mine flow, especially iron, empty into the nearby water bodies which serve as sources of municipal water supply. Consumption of untreated water over a prolonged period from these water sources may be detrimental to health. Remedial measure and continuous monitoring are recommended for good environmental stewardship.

1. Introduction

Coal is a natural fossil fuel that occurs in many Cretaceous rocks all over the world, and it is used as a major source of energy. It is classified as one of the strategic minerals of Nigeria with an estimated deposit of about 2.8 billion tonnes. The earliest major discovery of coal in Nigeria was in Enugu town in southern Nigeria. In the present, coal production has ceased completely, and all these premier mines have been abandoned and left un-reclaimed. As long as coal remains buried in host rocks in the subsurface, it poses no immediate environmental problem until when it is excavated and mined. During the life cycle of mines, they are continuously flooded by the ingress of water that requires pumping to keep them drained and workable. However, when mines become moribund, pumping is discontinued which results in flooding that forms perennial mine ponds. Untreated water from these mine ponds and those from the underground mine are drained into the surrounding agricultural lands, and both adjoining surface and ground water bodies. The presence of water, together with oxygen trigger reactions that drain the rocks and produce undesirable environmental outcomes, principally acid mine drainage. Acid mine drainage (AMD) refers to acidic water that forms in abandoned metal mines or coal mines when sulphide minerals like pyrite that often occur in association are exposed to air

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and water and, through a natural chemical reaction, eventually producing sulphuric acid.

The formation of acid drainage and the acid load generated from mine sites is thought to be complex, controlled by physical, chemical and biological factors including chemistry of pyrite weathering, microbiological controls, depositional environment, acid/base balance of the overburden, lithology, mineralogy and mine site hydrologic conditions. The cyclic propagation of acid generation by iron takes place very rapidly and continues until the supply of ferric iron or pyrite is exhausted according to the well-known pyrite reactions [1].

$$\text{2 FeS2 + 7 02 + 2 H2O} \rightarrow \text{2 Fe2+ + 4 SO4 + 4 H} \qquad (1)$$

$$\text{4 Fe 2+ + O2 + 4 H+} \rightarrow \text{4 Fe3+ + 2 H2O} \qquad (2)$$

$$\text{4 Fe3+ + 12 H2O} \rightarrow \text{4 Fe (OH)3 + 12 H} \qquad (3)$$

$$\text{FeS2 + 14 Fe3+ + 8 H2O} \rightarrow \text{15 Fe2+ +2 SO42- + 16 H} \qquad (4)$$

These reactions are enhanced by the bacteria *thiobacillus ferroxidans* which are widespread in the environment, and their activity is pH dependent with optimal conditions in the range of pH 2 to 3.

Rocks formed in brackish water conditions are generally most prone to acid production. The acid generation process consists of three phases: the initiation phase where pyritic materials are exposed to an oxidizing environment to generate relatively small acid load; the propagation phase when acid production increase rapidly but declines in the termination phase. Mineralogically, the conditions most conducive to acid formation are high pyrite contents with little base material present. The potential for acidic mine drainage to release toxic elements from mine wastes and impact negatively on contiguous farmlands, surface water, hand dug wells and shallow aquifers exist where mining occurs. These water sources are prone to contamination when these toxic-substances laden waters flow into them. This worldwide problem is associated with sulphide-rich metalliferous ore deposits, viz., copper, lead, zinc, gold, nickel, tin, mines [2] and coal mines [3] which contain abundant reactive pyrite. Earth excavations of these deposits have the potential to accelerate the AMD process by exposing the sulphide minerals to water and air. The ever-present effects of AMD on the quality of water, soil, plants and human beings can be summarized to include acidity, metal toxicity, metal precipitation and salinization with the degree of impact ranging from non-detectable to complete destruction of the normal flora and fauna. AMD decreases the quality of receiving waters by lowering the pH and increasing the dissolved metal content of surface and groundwater [4-6] making these waters harmful to fish and aquatic life because of the transport and persistence of toxic products of the mines. These adverse impacts of mine sites on environmental media have been discussed in surface water bodies [7-9] groundwater [10-12]), soils [13], stream sediments [9], plants [14-15], and microbial community [16].

The active interactions between AMD-contaminated river water and groundwater in mining areas has long term adverse impacts that are difficult to reverse [12]. However, for rivers, there are composite interacting factors, including climate, geology and hydrological pathways e.tc, that control heavy metal concentrations from polluted waters and the extent to which they have a detrimental effect on the ecosystem of a river. These factors are both complex and site specific [17]. When soil is affected by the AMD, the soil may be acidified and its chemical composition may also be changed. These acidic conditions can cause nutrients to be leached from soils resulting in nutrient depletion that can lead to increased susceptibility to stress that affects the survival and growth of plants. Also, when agricultural soils are polluted by AMD, heavy metals are taken up by plants and consequently accumulate in their tissues that could impair metabolism in human beings when passed unto them through the food chain. The effective treatment of acid drainage from mine areas reduces material damage, allows resource reuse and recovery, and enables successful post-mine land use [13]. AMD prevention and/or remediation are essential in order to reclaim the areas of land and water that are plagued by the AMD problem include five main treatments: carbonate neutralization, ion exchange to absorb from the AMD aeration which lowers the concentration of CO_2 and decomposes H_2CO_3 for the neutralization of more acid and precipitation of metal by making water more alkaline to raise water pH and supply hydroxides (OH^-) so dissolved metals in the water will form insoluble metal hydroxides and settle out of the water. Contaminated water constitutes health risk, as this could result to excess accumulation of heavy metals in biotic organisms, including humans that take in these waters leading to toxic effects. Human exposure to these metals includes by ingestion inhalation, dermal contact or through the food chain.

In the present study area, poor environmental consciousness has left the abandoned mines not properly closed according to best practice for the health of the environment. There is the possibility that acid mine drainage may be active, but may not be detected resulting in continuous contamination of surface and groundwater resources. For example, assessmentstudies of the mines by [18-20] identified the onset of AMD. Therefore, regular monitoring of drainage characteristics to manage drainage problems is a concern at operating mine sites and after mine closure. Hydrogeological investigations [21] identified the severity of continuous ingress of groundwater in sandstone rocks underlying the mines and the attendant acid mine drainage problems. Continuous monitoring of abandoned mines is essential to update knowledge on the geoenvironmental status of the surroundings. This knowledge may be applied in the effective monitoring and treatment of acid drainage from mine areas to reduce material damage, allow resource reuse and recovery, and enable successful post-mine land use [22]. The present study is to investigate the chemistry of discharges from these moribund mines, the contamination risks of the heavy metal's concentrations in the soil, water and sediments in their vicinity and implications for acid mine drainage. Water from these mines flow into nearby rivers and streams which are the major sources of water supply to Enugu metropolis.

2. Description of the study area.

In 1909, sub-bituminous coal was discovered in Enugu and mining started in 1916 in a drift mine. Subsequently, other mines like the Ribadu, Onyeama and Okpara were opened. The management of the two underground mines: Okpara and Onyeama mines were consolidated into one entity called the Nigerian Coal Corporation in 1950. The mines increased production at the beginning in 1916 from 24,511 tonnes, peaking at 925,000 tonnes in 1958 and declined drastically to just about 17,000 tonnes in 1999 [23]. The discovery of oil in the Niger Delta, coupled with improvements in technology killed the use of coal in railway engines and power generation in preference to diesel, and effectively left the mines derelict. Coal production from the mines ceased completely in 2002. Since then, all the mines have been abandoned and left un-reclaimed.

Geologically, the mines are underlain by three uconformable Cretaceous geologic formations, from oldest to youngest namely; Enugu Shale Formation (Campanian in age), Mamu Formation (Lower Maastrichtian in age), and Ajali Sandstone Formation which is Upper Maastrichtian in age (Figure 1). The Enugu shale consists of carbonaceous grey black shales and coals with interbeds of very fines and sandstone/siltstone. The Mamu Formation overlies the Enugu shale conformably and contains sandstone, shale mudstone, sandy-shale with coal seams in various horizons [24] all the coal seams tapped by Onyeama mine are from Mamu Formation [25]. The coal occurs in association with pyrite and siderite. The Ajali Formation is characterized by thick, friable, poorly sorted, white coloured sandstone. These sandstones are overlain by tropical lateritic red soils. The topography consists of hills underlain by Mamu Formation and Ajali Sandstone and lowlands underlain by Enugu Shale. Enugu is a tropical area which experiences temperatures in the range of 23 - 30°C, and rains are usually heavy [26]. Hydrogeologically, the highly fractured Ajali Formation contributes the groundwater entering the coal mines. Mining by excavation along the coal seam created an artificial stream along which mine waters flow from the highlands into streams and rivers that run through the immediate lowlands. Shallow water table in the area increases the aquifer susceptibility to contamination. The coal sequence is an interconnected multi-aquifer system with the sandstone and coal aquifers alternating with shale aquitards [27].

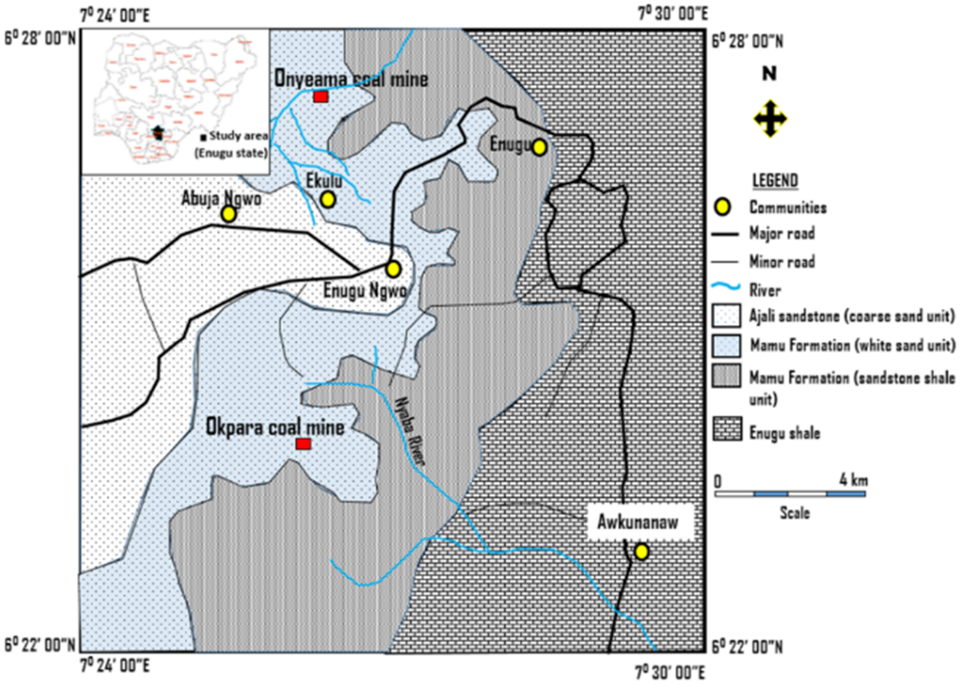

Figure 1. Geological map showing location of the mines (Modified from Nwajide 2002).

3. Materials and methods

Water and soil/sediment samples were obtained from abandoned mine spoils and tunnels, adjoining streams and river tributaries, boreholes and shallow hand dug wells in the vicinity of the mine sites (Figure 2).

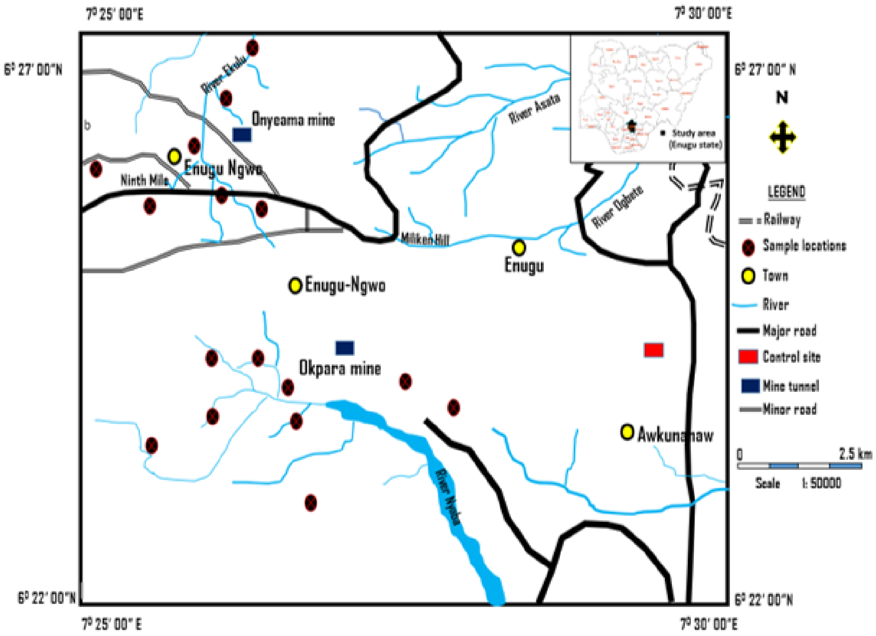

Figure 2 Location of the mines and sampling points.

Surface water sampling points in rivers and streams were selected to reflect sources of inflowing, intermediate and out flowing water. A total of (18) water samples were collected from different media within the mine sites. These media were reddish to yellowish brown in colour, and indication of iron precipitation as shown in Figure 3. Surface water samples were collected below the river/stream surface and filtered using 0.45 µm Millipore filters into sterilized low-density polyethylene bottles, each for cationic and anionic analyses. The bottles were rinsed thoroughly with the water to be sampled before collection to avoid the introduction of foreign substances in the samples. They were labelled according to their sources as Ek= Ekulu River, NY Nyaba River, ONMT = Onyeama Mine tunnel, OKMT = Okpara Mine Tunnel, BH = Borehole, HDW = Hand Dug Well, RW – River water. Water samples for the determination of cations were stabilized by adding few drops of diluted HCl after collection and those for heavy metal analysis were preserved by acidifying with a few drops of HNO_3 acid to achieve a pH of ≤ 2. To maintain the integrity of the analysis, physico-chemical parameters sensitive to environmental changes such as pH and electrical conductivity were measured in-situ using portable digital meters. The samples used for laboratory analyses were preserved in a refrigerator prior to the analysis to exclude microbial activity.


(a) (b) (c) (d)

Figure 3. View of (a) Onyeama mine tunnel (b) Ekulu river tributary (c) Okpara mine tunnel (d) Nyaba river.

Soils and sediments were also collected from very old mine spoils, mine tunnels, along the water channels, and near stream confluences, in order to cover the drainage network within the mine's catchment. At each sampling point, 3 samples were taken and mixed thoroughly into a homogenous sample. The background samples were taken from areas far removed from the influence of the mines. The sediments samples were dried in the oven at 105 °C and digested. The analytes for water included pH, electrical conductivity (EC), total alkalinity (TA), total dissolved solids (TDS), salinity, inorganic ions/salts. Also, heavy metals (As, Cu, Ni, Fe, Pb, Zn, Cr, Cd, Mn) were analysed for both water and sediment samples. For every metal investigated, standards and blanks were prepared and used for calibration before samples were aspirated. The samples were analyzed for chemical parameters according to the methods summarized in Table 1. All samples analysis was carried out in Zetta Allied Digital Energy Limited (ZADEL) Laboratory in Port-Harcourt, Nigeria.

Relationships between physico-chemical parameters, metals and other controlling factors were determined by bivariate correlation using the Pearson's coefficient in a two-tailed test ($r < 0.01$ and 0.05).

Table 1 Summary of analytical methods used

Analyte	Type of test	Measurement/Analytical Method
pH	In-situ	Digital pH meter
Temperature	In-situ	Mercury-in-glass thermometer
Conductivity	In-situ	Digital conductivity meter
Ca^{2+}, Mg^{2+}, Alkalinity	Laboratory	Titration method
K^+, Na^+	Laboratory	Digital flame photometer
Cu^{2+}, Ni^{2+}, Fe^{2+}, Pb^{2+}, Zn^{2+}, Cr^{2+}, Cd^{2+}, Mn^{2+}, As^{2+}	Laboratory	Atomic absorption spectrophotometer
Total Dissolved Solids	Laboratory	Filtration and evaporation method
SO_4^{2-}	Laboratory	Gravimetric Method
Cl^-	Laboratory	Argentometric method
NO_3^-	Laboratory	Brucine method

3.1. Data analysis

3.1.1. Determination of pollution factors

In order to assess heavy metal enrichment and degree of contamination in water and sediments, the heavy metals data were used to determine pollution factors and indices using established empirical relationships as follows:

3.1.2 Contamination factor (CF) and pollution load index (PLI)

The contamination factor was determined by the contamination/pollution index according to [28] method as,

$$CF=\frac{Cn}{Bn} \qquad (1)$$

Where Cn is the concentration of the metal in the sample; Bn is the background concentration. For the sediments, Bn is taken as concentration values obtained from control samples.

The contamination factor of the metal was used to derive the PLI and assess the degree of contamination, using the [29] relationship,

$$PLI = [CF_1 \times CF_2 \times CF_3 \times \ldots \times CF_n]^{1/n} \qquad (2)$$

where, CF= contamination factor; and n = number of metals.

The significance of the Contamination factor (CF) is that values less than unity or greater than unity (1) indicate contamination or pollution respectively.

3.1.3. Enrichment factor (EF)

Enrichment Factor was used to assess the relative contributions of natural and anthropogenic heavy metal sources to sediments/water based on the [30] method where:

$$EF = \frac{[\frac{X}{Fe}]sample}{[\frac{X}{Fe}]background/control} \qquad (3)$$

In this method, $[X/Fe]_{sample}$ is the ratio of heavy metal [X] to Fe in the sample and $[X/Fe]_{background/control}$ is the natural background/control value of the metal-Fe ratio.

Fe was chosen as the geochemical normalizer because of its conservative nature during diagenesis [31], and is also used as the reference element at [32] value of 0.3 mg/L for sediments. The EF values close to unity indicates crustal origin, those less than 1.0 suggest a possible mobilization or depletion of metals, whereas EF greater than 1.0 indicates that the element is of anthropogenic origin [33]

3.1.4 Geo-accumulation index (I_{geo})

The index of geo-accumulation enables the assessment of contamination by comparing the current levels of metal concentrations and the original pre-industrial concentrations in the sediments and water, and was determined using the [34] relation.

$$I_{geo} = Log_2 \frac{[Cn]}{[1.5Bn]} \qquad (4)$$

where, C_n is the measured concentration of the element in the sediment and water samples and B_n is the geochemical background/control values of that metal.

The control sample was used as the background for the sediments and [32] standard for water. According to [35], the constant 1.5 is introduced to minimize the effect of possible variations in the background values which may be attributed to lithologic differences.

3.2. Results and discussion

3.2.1. Physico-chemical parameters

Statistical values of the physical parameters, major inorganic ions/salts and heavy metals composition of surface and groundwater are summarized in Table 2 and the concentration of heavy metals in the sediments are summarised in Table 3.

The range of pH values for waters in the Onyeama and Okpara mines are 3.4 - 5.8 and 3.5 - 6.1 respectively, indicating acidic to weakly acidic conditions. The pH value of water is a measure of the hydrogen ion activity in water and indicates whether the water is acidic or alkaline. All the waters sampled in both mines yielded pH of less than 7.0, demonstrating the generally acidic nature of the water which provides a favourable environment for the oxidation of sulphides in pyrites that occur in association with the host rocks, thus triggering the onset of acid mine drainage. In the Onyeama mine impact area, the highest acidity obtained from Ekulu river (RW4 and RW5) and the lowest acidity occurred in the shallow hand well (HDW). The degree of acidity in the water samples decreases as follows; river > mine tunnel > BH > HDW. In Okpara mine environment the highest acidity was obtained from River Nyaba (RW2 and RW3) and the lowest acidity from the borehole (BH2). The low pH of the waters is in agreement with the earlier results of [27]. Acidic waters are susceptible to dissolution of minerals and release of toxic heavy metals into the aquatic environment [36]. The trend of variation in the acidity suggests that the surface water bodies have been impacted more than groundwater.

Electrical Conductivity (EC) is a measure of the capacity of a water sample to conduct electric current as well as the relative level of dissolved salts in the water. EC is an important factor that influences the level of dissolution of heavy metals and desorption. EC ranged from 0-159.4 µS/cm in Onyeama mine and 0-349 µS/cm in Okpara mine with mean values of 31.05 and 74.20 µS/cm respectively. The conductivity of fresh water ranges from 10 to 1000 µS/cm, but may exceed 1000 µS/cm in polluted waters or those receiving large quantities of land run– off or AMD runoff [31]. [37] reported EC value of 1550 µS/cm in surface water from the Okpara coal mine. However, in the present study, the highest EC value in both Okpara and Onyeama mines was 349 µS/cm, which is far less than the 1000 µS/cm prescribed as limits by [32]. Total Dissolved Solids (TDS) is a measure of both anion and cation concentration in a water body. The low values of TDS were possibly due to low dissolution of sulphate in the presence of buffers such as calcium, carbonates and bicarbonates or due to dilution [36]. TDS correlates strongly with EC values in both mines (r= 0.99) indicating their association and common provenance. The Enugu shale which is the host rock for the coal is calcareous. Dissolution of the carbonates increases the alkalinity of the solvent and attenuates dispersion of certain metals. Compared with results obtained by other workers (Table 4), there is significant increase in the concentration of the physicochemical parameters in this study. The quality of any water resource for suitability for the intended use is a function of its physical and chemical characteristics which in turn depends on the geology of the area and impacts of human activities.

Table 2: Summary results of surface and groundwater in the area.

Chemical parameter	Onyeama mine			Okpara mine			WHO (2017)	NIS (2007)	NESREA (2009)	
	Range	**Mean**	**SD**	**Range**	**Mean**	**SD**	**(mg/l)**	**(mg/l)**	**Surface water**	**Land Application**
Alkalinity(mg/L)	1-18	4.00	5.73	2-8	4.10	2.02	400.00			
Temperature (^{o}C)	26.3-28.8	27.89	0.80	17.1-28.4	26.11	4.57	Ambient	Ambient	<40	<40
TDS (mg/L)	0-115.5	21.28	40.70	0-242	47.60	82.25	1000.00	500.00	2000	2000
pH	3.39-5.84	4.05	1.01	3.54-6.5	4.56	2.48	6.5-8.50	6.5-8.5	6.59	5.5-9.0
EC (µS/cm)	0-159.4	31.05	5.86	0-349	74.20	15.80	1000.00	1000.00	2000	2000
Salinity (mg/L)	0-72.7	13.13	25.89	0-164	31.35	56.14				
Potassium	3.4-40.08	15.40	11.89	1.1-55.03	21.28	19.17	200.00			
Magnesium	0.32-103.3	15.30	35.79	0.3-5.03	1.99	1.53	50.00		200	
Calcium	5.2-17.608	14.18	3.79	3.1-19.762	13.41	5.46	75.00		200	
Sodium	6.8-16.278	14.08	3.03	2.5-17.108	13.09	5.61	200.00	200.00		
Sulphate	113.9-210.1	134.95	7.98	4.6-270.2	144.01	8.70	200.00	100.00	500	1000
Nitrate	4.12-35.1	19.12	11.66	7.5-269	44.28	79.27	50.00	50.00	20	
Chloride	0.14-10	4.16	3.45	2.01-13.1	5.10	3.51	250.00	250.00	600	
Nickel	0.001-0.058	0.02	0.02	0.001-0.067	0.02	0.02		0.02	<1.0	
Iron	0.11-20.5	9.21	7.80	0.02-10.01	1.95	3.22	0.30	0.30	20	
Copper	2.21-31.14	14.45	8.39	0.01-35.2	11.85	10.83	0.50	1.00	<1.0	
Arsenic	0.001-8.5	1.07	3.00	0.001-8.5	0.86	2.68		0.01	0.1	
Lead	0.002-55.1	16.96	28.62	0.001-27.31	4.32	9.37	0.004	0.01	<1.0	
Manganese	0.01-1.21	0.66	1.06	0.01-0.15	0.05	0.04	0.10	0.20	200	
Chromium	0.001-0.211	0.08	0.07	0.001-0.267	0.09	0.08		0.01	<1.0	
Zinc	3.01-17.11	15.42	5.62	7.72-50.8	21.64	13.20	5.00	3.00	<1.0	
Cadmium	0.001-0.001	0.00	0.00	0.001-0.001	0.00	0.00	0.003	0.003	<1.0	

NIS – Nigerian Industrial Standard; WHO- World Health Organisation NESREA = National Environmental Standards and Regulations Enforcement Agency

Table 3 Statistical summary results of heavy metals concentration in sediments

Heavy metals(mg/kg)	Okpara mine			Onyeama mine			Control Site
	Min	Max	Mean	Min	Max	Mean	
Fe	253.4	420.1	342.2	270.1	420.1	326.64	265.53
Cu	12	19.9	15.2	11.7	20.1	16.12	13.18
As	10.5	29.1	18.1	15.1	37.1	22.26	10.12
Pb	3.12	21.5	8.41	15.9	41.5	30.32	2.3
Mn	15.3	100.1	49.44	65.1	150.3	100.38	68.7
Cr	7.8	22.1	13.4	8.2	23.5	12.44	12.1
Cd	0.03	0.09	0.05	0.03	0.04	0.03	0.03
Ni	13.4	50.3	26.12	10.3	21.7	14.26	15.57
Zn	15.8	21.1	18.58	14.9	25.3	18.98	17.53

Table 4 Results from this study compared with others.

Chemical Parameter	This study		Ezeigbo & Ezeanyim (1993)		Other related studies		
	Onyeama mine	Okpara mine	Onyeama Mine	Okpara mine	Nganje *et al.* (2010)	Utom & Odoh, (2013)	Sikakwe *et al.*, (2015)
pH	4.05	4.56	2.80	2.3	4.22	4.70	4.22
Temperature (^{0}C)	27.89	26.11	na	na	na	20.7	26.84
TDS	21.28	47.6	330.00	785	157.77	210.5	157.76
EC (µS/cm)	31.05	74.2	300.00	1550	269.67	293.10	269.7
Alkalinity (HCO3-)	4.00	4.10	na	na	na	na	Na
Salinity	13.13	31.35	na	na	nd	na	Na
Potassium	15.40	21.28	nd	na	12.01	4.10	12.01
Magnesium	15.30	1.99	158.08	85.12	6.80	0.40	6.77
Calcium	14.18	13.41	4.00	6.41	9.90	21.8	8.79
Sodium	14.08	13.09	na	na	14.00	6.20	14.00
Sulphate	134.95	144.01	310.00	420.00	119.11	129.2	108
Nitrate	19.12	44.28	1.20	30.00	167.67	14.50	567.67
Chloride	4.16	5.1	10.42	1.99	138.89	5.40	136.67
Nickel	0.02	0.02	na	na	53.1	na	0.051
Iron	9.21	1.95	8.40	25.76	5.14	1.60	5.14
Copper	14.45	11.85	na	na	13.47	na	0.008
Arsenic	1.07	0.86	na	na	0.337	na	na
Lead	16.96	4.32	na	na	6.38	na	0.006
Manganese	0.66	0.05	na	na	3353.8	0.40	3.35
Chromium	0.08	0.09	na	na	12.03	na	0.001
Zinc	15.42	21.64	na	na	253.82	na	0.254
Cadmium	0	0			0.35		0.0003

All concentrations are in mg/L unless otherwise stated. na = not available

The general trend in the mean values of the cations (mg/l) in the waters from Onyeama mine area shows that $K^+ > Mg^{2+} > Ca^{2+} > Na^+$ and in the Okpara mine, $K^+ > Na^+ > Ca^{2+} > Mg^{2+}$. The cationic trend reveals that potassium, an element with low geochemical mobility, is the dominant cation in the water sources from both mining areas. The dominant anion levels in the waters from Onyeama mine are in the order: $SO_4^{2-} > NO_3^- > Cl^-$, while in the Okpara mine it is $NO_3^- > SO_4^{2-} > Cl^-$. The trends show that sulphates and nitrates are the dominant anions in Onyeama and Okpara mines respectively. Sulphate ion is an indicator of AMD pollution because sulphates are not affected by changes in pH. The mean sulphate concentrations in the waters appear lower than expected of AMD conditions (34.95 mg/L in Onyeama; 44.01 in Okpara), even as the values obtained from the mine tunnels which range from 210.1mg/L to 270.2 mg/L. The level of concentration of sulphate in the waters can be attributed to insufficient pyritization. The longer rocks are exposed to weathering the more the pyrite oxidation which in turn influences the average sulphate concentration in relation to other constituents.

Heavy metals have an abundance trend in the order of Pb>Zn>Cu>Fe>As>Mn>Cr>Ni>Cd and Fe>Mn>Pb>As>Zn>Cu>Ni>Cr>Cd in water and sediments from Onyeama mine, and Zn>Cu>Pb>Fe>As>Cr>Mn>Ni>Cd and Fe Mn>Ni>Zn>As>Cu>Cr>Pb>Cd for water and sediments from Okpara mine. Pb, Zn and Cu are the dominant heavy metals in all the water samples while Fe constituted the most abundant metal in all the sediments. This is expected because of the occurrence of pyrite and siderite which are ores of iron in the host rocks. The trend of heavy metals concentration in water in this study agree closely with results in other studies [36-38] except the high concentrations of Mn and Zn obtained by [19]. Ni concentration in the sediments in the river bodies within Onyeama mine, and similarly Pb concentrations in Okpara mine are elevated, exceeding the levels of concentration in the corresponding media in the control site. This implies that the leached heavy metals generated by the mining activities have been mobilized and concentrated in the nearby water body channels during transportation. In the sediments, higher heavy metals values were obtained in the present study compared to those obtained by [38-39], but lower than those obtained by [6] as shown in Table 5.

Table 5 Comparative results of mean heavy metals concentration

Heavy Metals (mg/kg)	This study		Other related studies				
	Onyeama mine	Okpara mine	Adaikpoh et al., (2005)	Ameh, (2013)	Sikakwe et al., (2015)	*WHO (2017)	NESREA (2009)
Fe	326.64	342.20		195.4	104.91	050	20.00
Cu	16.12	15.20		0.50	30.21	0.017	<1.00
As	22.26	18.10				010	0.10
Pb	30.32	8.41	0.017	0.50	16.18	0.065	<1.00
Mn	0.30	49.44	0.39		12.00	0.20	200.00
Cr	12.44	13.40	0.26		131.89	0.55	<1.00
Cd	0.03	0.05	0.043	0.60	0.001	0.01	<1.00
Ni	14.26	26.12	0.067	4.30	8.22	1.40	<1.00
Zn	18.98	18.58		1.50	64.14	0.20	<1.00

*WHO maximum permissible levels in irrigation water

[40] Obtained similar trend of results in a mine in Bosnia and Herzegovina. Apart from Ni and Cd in both mining areas and Mn in Okpara mine, the waters have mean concentrations of all other metals in exceedance of maximum permissible limits of [32] and [41] for potable water. They also exceed the Nigerian legislation for groundwater [42] permissible discharge limits into surface water for industries in engaged in the mining and processing of coal. However, [42] has no permissible limits of concentration of heavy metals for land application.

The mean concentration of all the heavy metals in surface soil sediments from Onyeama and Okpara mines apart from Nickel were higher than values obtained from samples of the control site, similar to Mn and Cr in Okpara mine. This shows that there is significant enrichment of heavy metals in the soils as a result of poor management of the abandoned mining sites. This chemical profile of the mine area in this study agrees with the geochemistry of acid mine drainage from coal mining areas in Brazil [43]. However, the in Enugu area, the concentration of iron, calcium and sulphate are generally about 10 times lower than the values reported by [3] in mines with intense AMD in Brazil.

3.2.2. Pollution assessment

In order to assess the heavy metal contamination in water and sediments in the study area, contamination factor (CF), pollution load index (PLI), enrichment factor (EF) and index of geo-accumulation (I_{geo}) were empirically determined from results of the concentrations of the heavy metals. According to [28], C/p values range from <0.1 to 1.0. The higher the value, the higher is the grade of pollution. Similarly, the PLI of [29] ranges from 1 (indicating perfection} to >1 (implying deterioration) of soil or water quality. The use of these different approaches for evaluating contamination of water and sediments with heavy metals facilitates a comprehensive interpretation of the water and sediments characteristics in terms of the background influences. Contamination generally refers to the introduction of non-native materials into the environment. Pollution on the other hand is when contaminant concentration is objectionable, implying that it is above permissible limits and has adverse impact on living organisms when it is in bioavailable form.

3.2.2.1. Contamination in Onyeama mine area

In Onyeama mine, the mean contamination factor (CF) shows that Pb, As and Fe have the highest CF which indicates that there is high contamination of the waters (Table 6 and Figure 4). It also shows that the water is severely polluted with Cu, Cr and Zn, moderately polluted with Mn and is slightly polluted with Ni. Cd shows the least CF which implies moderate water contamination. The sediments and water are severely polluted with Pb, moderately polluted with As, slightly polluted with Fe, Cu and Mn, but very severely contaminated with Cr, Cd, Zn and Ni (Table 7 and Figure 5). Apart from water samples from hand dug wells (HDW) and boreholes (BH), all others gave PLI greater than 1, indicating progressive deterioration in water quality, with river water (RW) sample as the most affected (Figure 6).

Table 6: Contamination factor (CF) and PLI of heavy metals in surface and groundwater resources

Onyeama mine			Okpara mine		
Location	**CF range**	**PLI**	**Location**	**CF range**	**PLI**
RW1	MC-EP	Deteriorated	RW1	VSC-VSP	Deteriorated
RW2	MC-EP	Deteriorated	RW2	SC-EP	Deteriorated
RW3	MC-EP	Deteriorated	RW3	MC-EP	Deteriorated
RW4	SC-EP	Deteriorated	RW4	SC-EP	Perfection
RW5	MC-EP	Deteriorated	RW5	SC-EP	Deteriorated
OYMT	SC-EP	Deteriorated	OKMT	SC-EP	Deteriorated
HDW	SC-VSP	Perfection	HDW1	MC-EP	Deteriorated
BH	SC-EP	Perfection	HDW2	MC-EP	Deteriorated
			BH1	VSC-MP	Perfection
			BH2	VSC-MP	Perfection

MC - Moderate contamination; EP - Excessive pollution; SC - Slight contamination;
VSP - Very severe pollution; VSC - Very severe contamination; MP - Moderate pollution;
OKMT – Okpara Mine Tunnel; BH – Borehole; HDW – Hand Dug Well; RW – River

Table 7. Contamination factor (CF) and PLI of heavy metals in sediments from Onyeama and Okpara coal mine.

Onyeama mine			Okpara mine		
Location	**CF range**	**PLI**	**Location**	**CF range**	**PLI**
OYMT	VSC-SP	Deteriorated	OKMT	VSC-SP	Deteriorated
EK1	SC-VSP	Deteriorated	NY1	MC-MP	Deteriorated
EK2	SC-VSP	Deteriorated	NY2	SC-MP	Deteriorated
EK3	SC-VSP	Deteriorated	NY3	SC-MP	Deteriorated
EK4	SC-SP	Deteriorated	NY4	VSC-MP	Deteriorated

MC - Moderate contamination; EP - Excessive pollution; SC - Slight contamination;
VSP - Very severe pollution; VSC - Very severe contamination; MP - Moderate pollution;
SP - Slight pollution

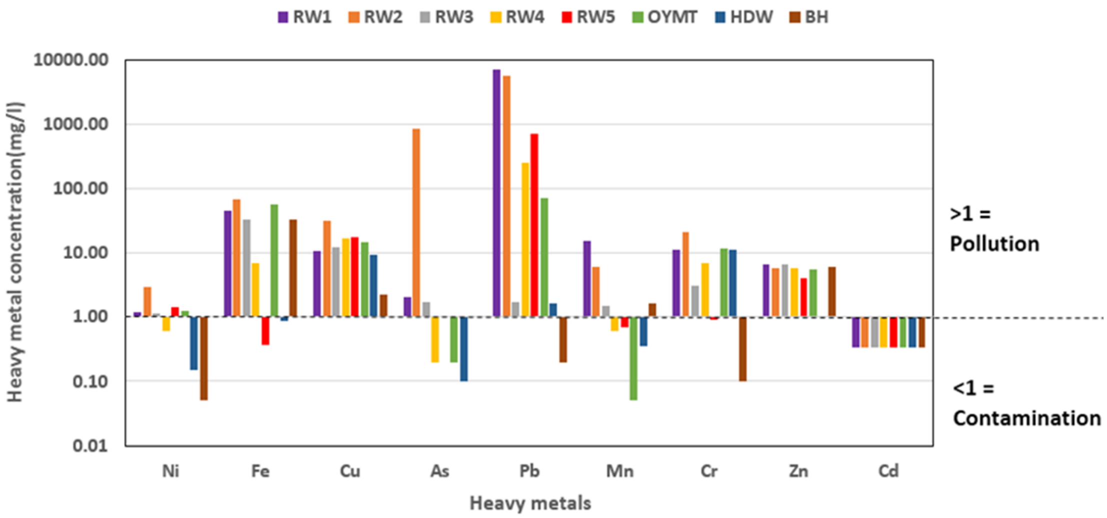

Figure 4. Contamination factor (CF) of heavy metals in water sources from Onyeama mine

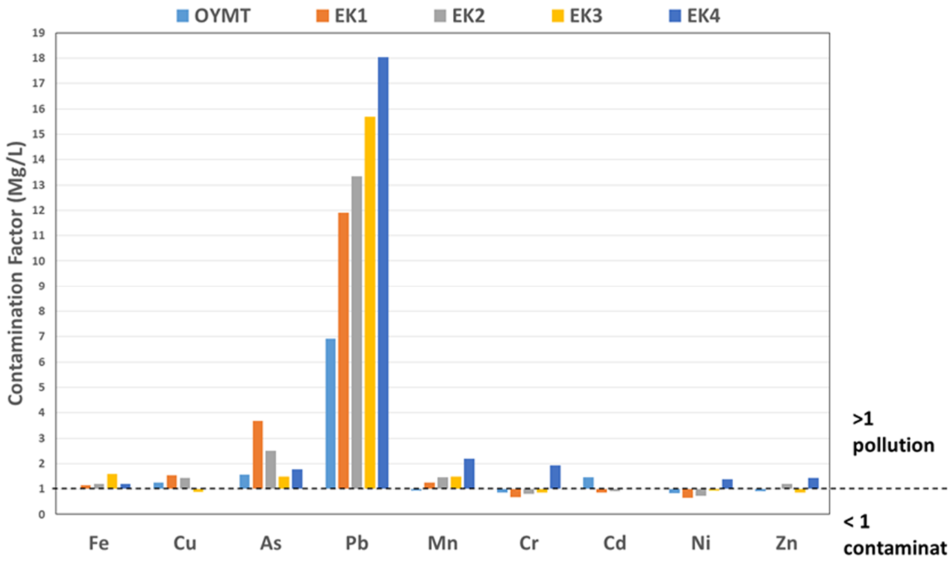

Figure 5. Contamination factor (CF) of heavy metals in soils from Onyeama mine. Baseline boundary between pollution and contamination = 1.

PLI values also indicated progressive deterioration in soils quality (Figure 7) in all sampled locations as shown by CF. The mean enrichment factor (EF) values reveal that the waters are significantly enriched in all the other heavy metals (Figure 8) apart from Ni and Mn which show no enrichment. This suggests a possible mobilization of metals. The waters are extremely severely enriched, moderately severely enriched and minimally enriched in Pb, Cu and As, Cr and Zn respectively. The sediments are only severely enrichment in Pb (Figure 9). These results indicate that the elements Pb, Cu, As, Cr and Zn in waters and As, Cu, Mn and Pb in the sediments emanates from the abandoned mine, hence they are of anthropogenic origin. According to the method of [44], EF value of <1 indicates no enrichment and >1 indicates extremely severe enrichment. In all the water samples, the I_{geo} values summarised in Figure 10 are generally low (< 1) compared to high EF values. The waters fall in two I_{geo} classes in line with [45] interpretation: uncontaminated to moderately contaminated with As, Pb, Cd and Ni and uncontaminated with Fe, Mn, Cu, Cr and Zn (Figure 14). In the sediments three categories of metal contamination were delineated: uncontaminated with Fe, Cu, Mn, Cr, Cd, Ni and Zn, uncontaminated to moderately contaminated with As and strongly contaminated with Pb (Figure 11). The Igeo of [45] is used in the sense that values fall between <0 and >5 indicating practically uncontaminated to extremely contaminated conditions.

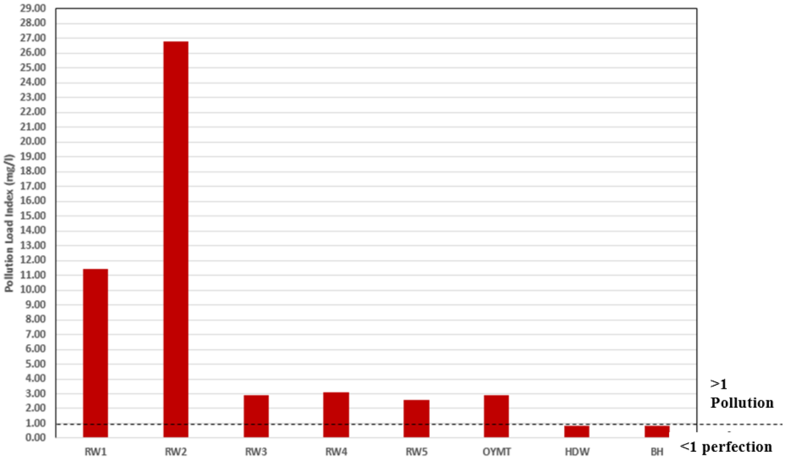

Figure 6. Pollution Load Index (PLI) for heavy metals in surface and groundwater from Onyeama mine area. Baseline boundary between pollution and perfection in water quality = 1.

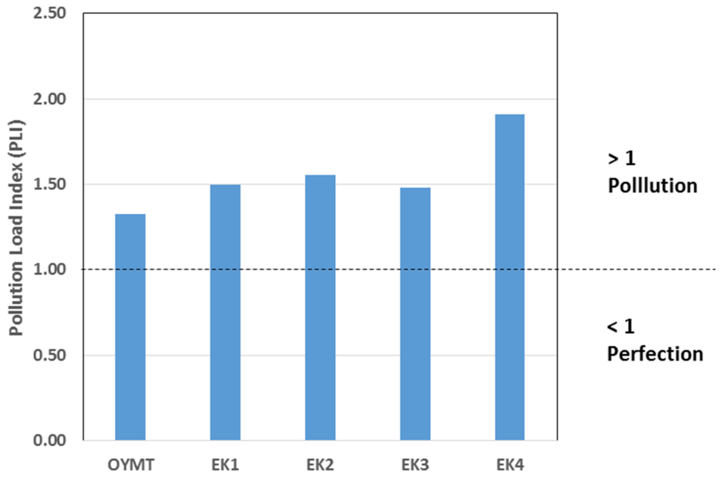

Figure 7. Pollution Load Index (PLI) in soils from Onyeama area. Baseline boundary between pollution and perfection in water quality = 1.

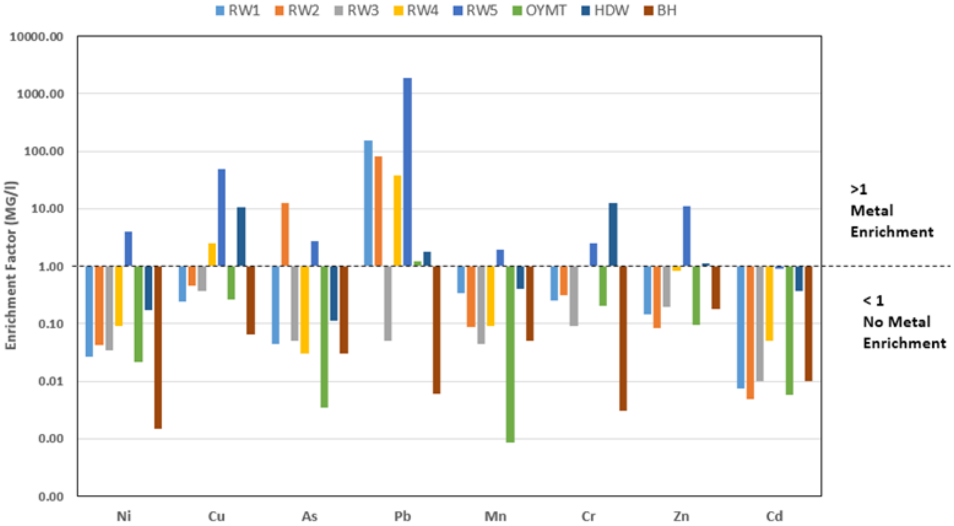

Figure 8. Enrichment factor (EF) in surface and groundwater from Onyeama mine area

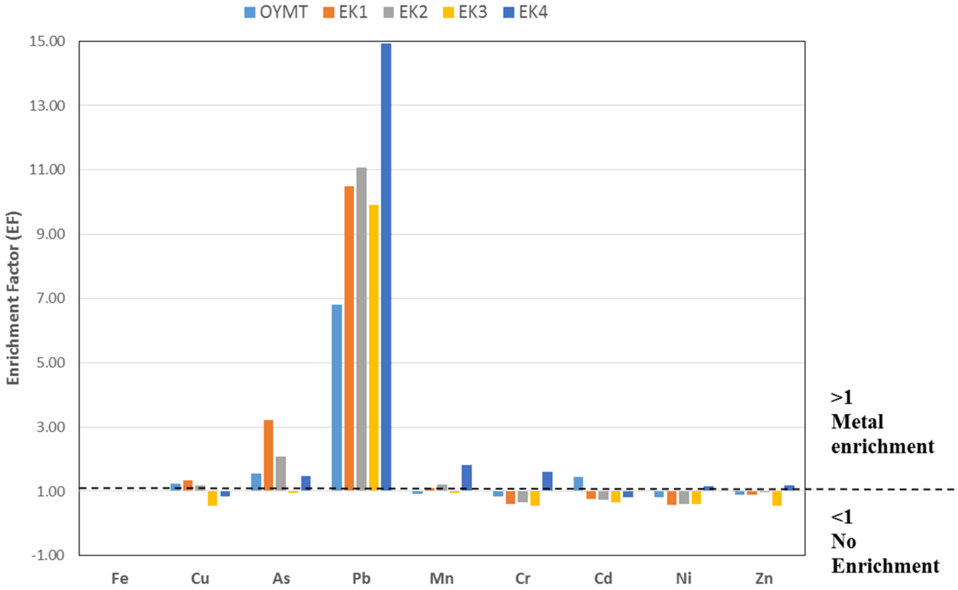

Figure 9. Enrichment factor (EF) of heavy metals in sediments from Onyeama coal mine

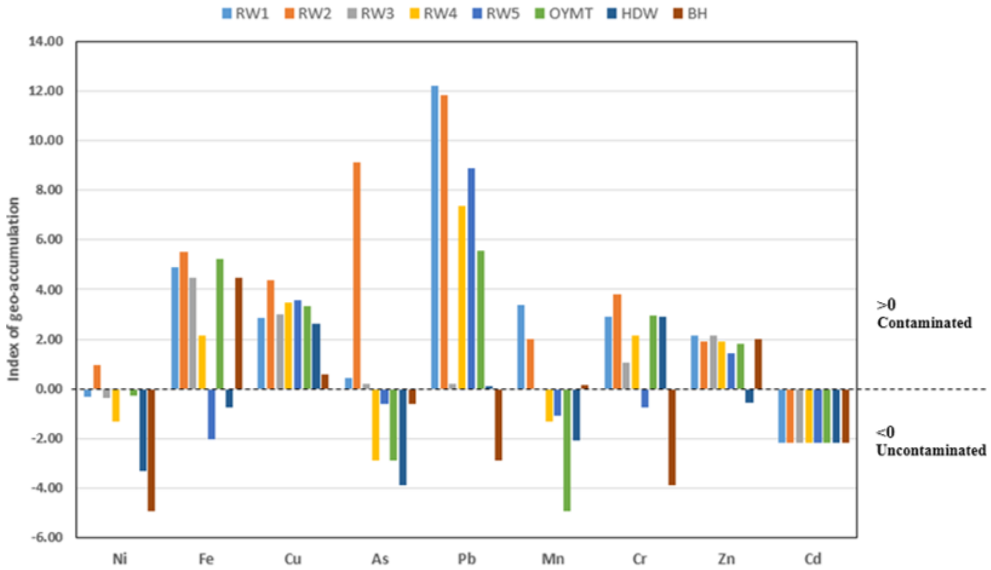

Figure 10. Index of geo-accumulation (I_{geo}) in surface and groundwater in Onyeama mine area

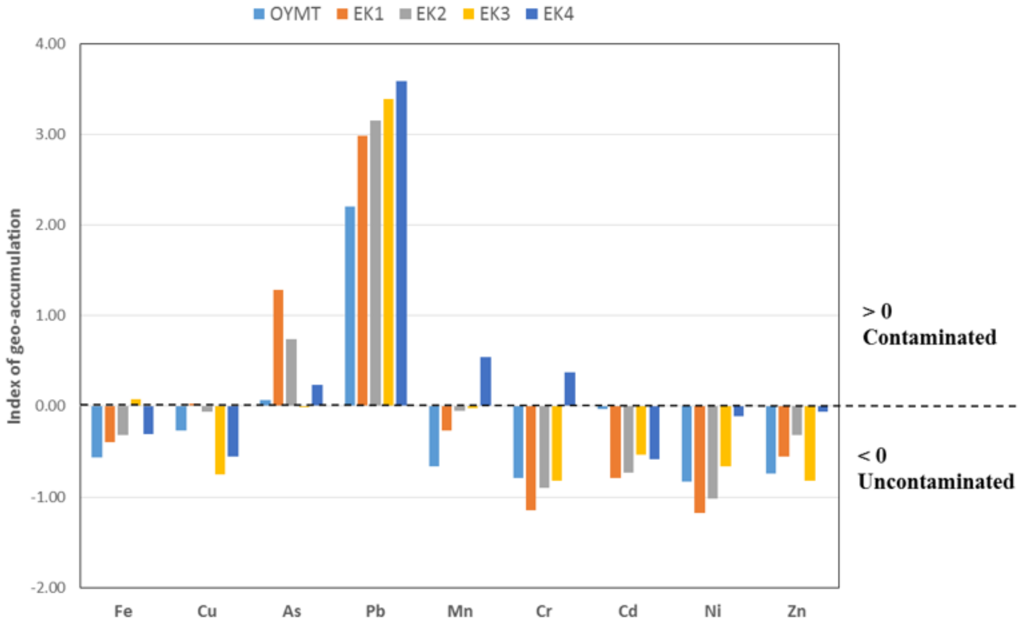

Figure 11. Index of geo-accumulatsion (I_{geo}) of heavy in soils around Onyeama mine area.

3.2.2.2. Contamination in Okpara mine area

In Okpara mine, the mean contamination factor (CF) reveals As and Pb have the highest CF which implies high contamination of the waters (Table 6 and Figure 12). The water is very severely polluted with Cu, Fe, Cr and Zn and very severely contaminated with Ni. Cd and Mn have the least CF implying moderate water contamination. The sediments are moderately polluted with Pb, slightly polluted with Fe, Cu, As, Cr, Cd, Ni and Zn and severely contaminated with Mn (Table 6). However, NY4 samples have the highest Pb pollution. CF of the sediments are generally greater than 1 (>1), implying that there is pollution. Mn shows slight pollution to slight contamination in the rest of the samples (Figure 13).

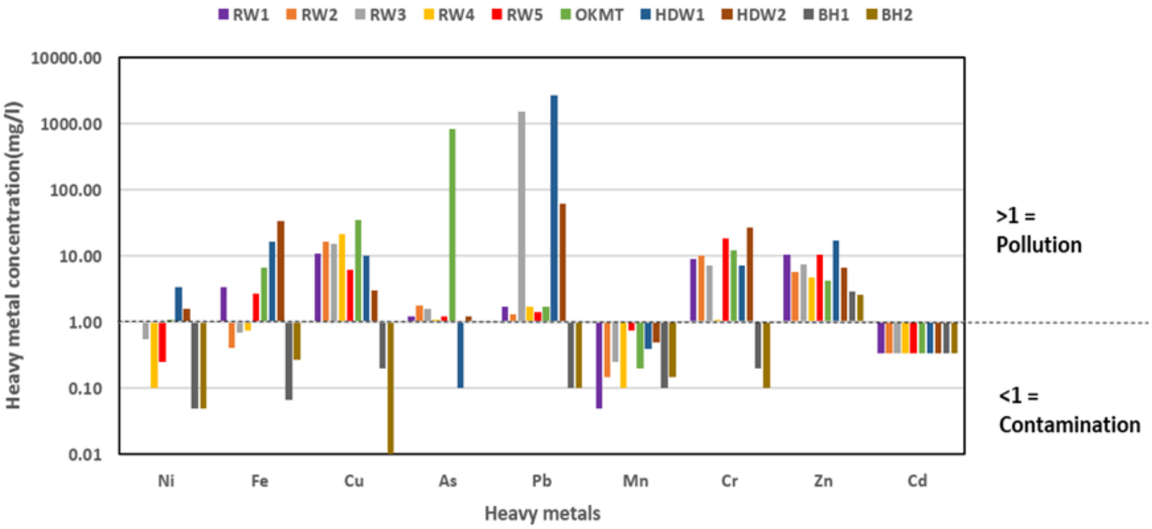

Figure 12. Contamination factor (CF) of heavy metals in surface and groundwater resources from Okpara coal mine.

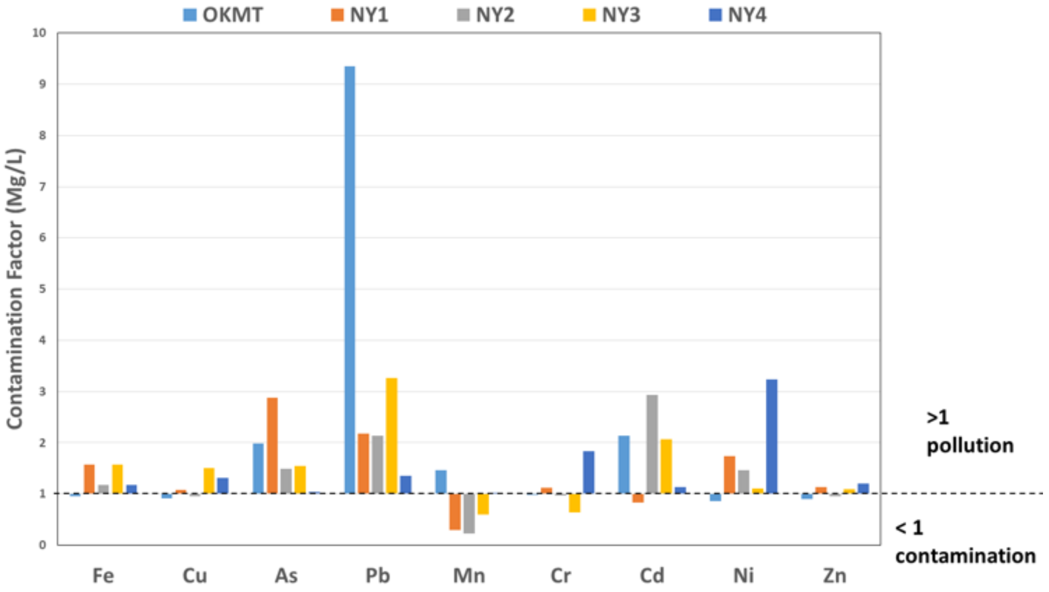

Figure 13. Contamination factor (CF) of heavy metals in sediments from Okpara mine.

Apart from water samples RW4, BH1 and BH2, the pollution load index (PLI) value shows that all other water sampled locations have PLI > 1, indicating progressive deterioration in water quality, with HDW1, OKMT and HDW2 as the most affected while locations RW3, RW5, RW1 and RW2 are the least affected by heavy metal contamination (Figure 14). Similarly, all the sediments analysed gave PLI values of greater than 1, implying progressive deterioration in quality (Table 7 Figure 15).

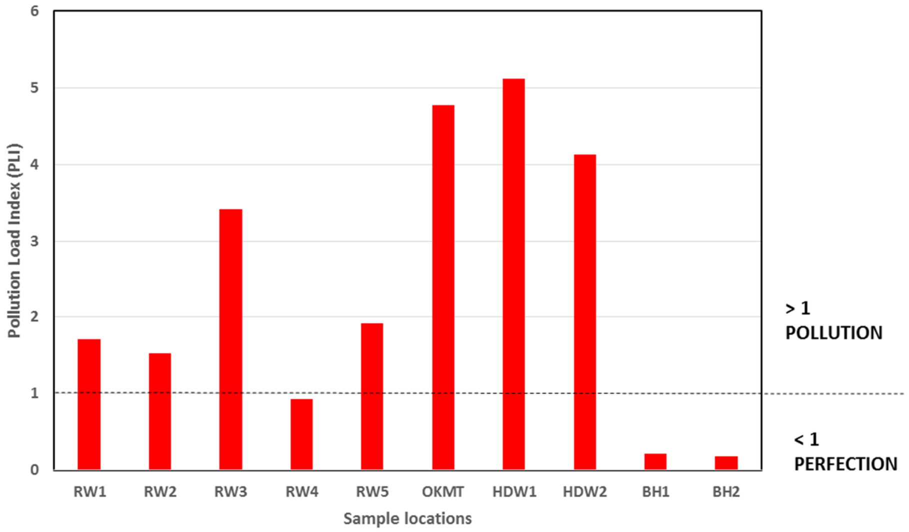

Figure 14: Pollution Load Index (PLI) for heavy metals in surface and groundwater resources from Okpara coal mine. Baseline boundary between pollution and perfection in water quality = 1.

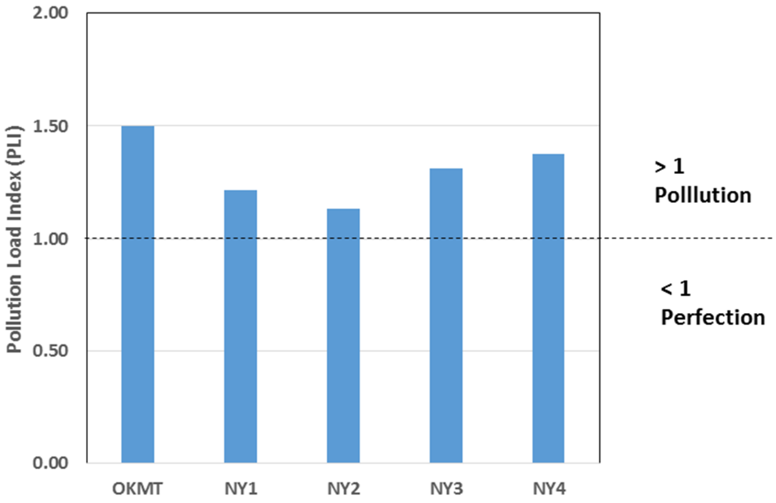

Figure 15. Pollution Load Index (PLI) for heavy metals in sediments from Okpara coal mine. Baseline boundary between pollution and perfection in water quality = 1.

The mean enrichment factor (EF) values reveals that apart from Ni, Cd and Mn which indicated minor enrichment and suggests a possible mobilization of metals, all the other heavy metals are significantly enriched in the waters (Figures 16 and 17). Lead indicates extremely severe enrichment, Cu and as shows severe enrichment and Cr and Zn indicates moderately severe enrichment in sediments. There is minor enrichment in As, Cd and Ni while Pb shows moderate enrichment. This indicates that the elements: Pb, Cu, As, Cr and Zn in the waters and As, Cd, Ni and Pb in the sediments originate from the abandoned mines, hence they are of anthropogenic origin. Unlike the EFs, the I_{geo} values in sediments are generally low (< 3), compared surface and groundwaters where As and Pb are high.

Four I_{geo} categories based on [45] method were identified in the water samples, as indicated in Figure 18. They are (i) moderately to strong contamination with Zn, (ii) moderately contaminated with Cu, Pb and Cr (iii) uncontaminated to moderately contaminated with Fe and As and (iv) uncontaminated with Ni, Mn and Cd. In the sediments, the metals fall into two categories; uncontaminated with Fe, Cu, Mn, Cr and Zn and uncontaminated to moderately contaminated with As, Pb, Ni and Cd (Figure 19).

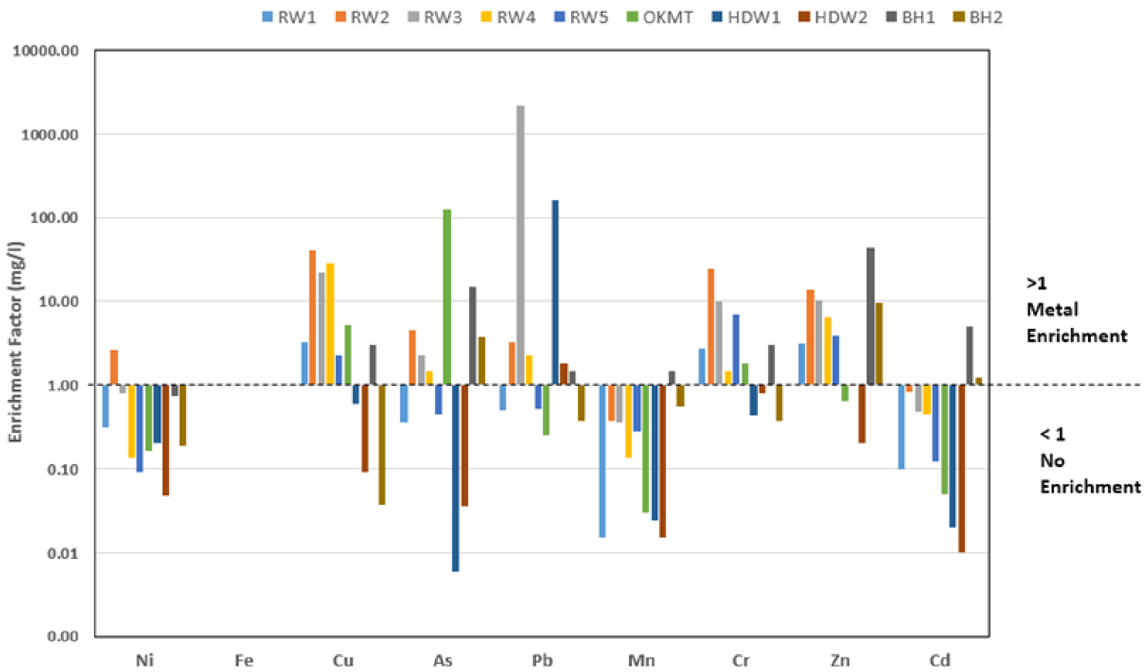

Figure 16. Enrichment factor (EF) of heavy metals in surface and groundwater resources from Okpara coal mine.

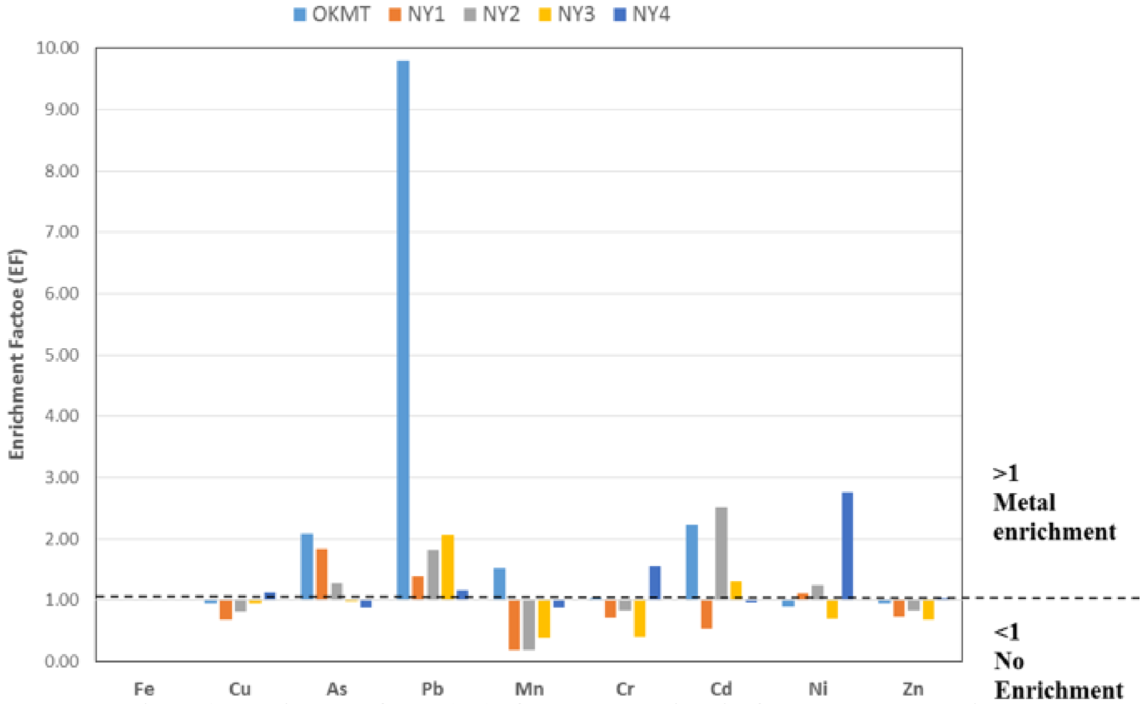

Figure 17. Enrichment factor (EF) of heavy metals in soils from Okpara coal mine.

3.2.3 Water type assessment

The relationship between pH and metal load, defined by [46] as the summation of total metals in each sample, was used to classify the water types. The metal load (the sum of the metals: Ni+Fe+Cu+As+Pb+Mn+Cr+Zn+Cd) and plotted against pH. Applying the classification scheme of [45], the three water types obtained in Okpara mine area (Figure 20) ranged from near neutral-high metal (BH2 and RW5) to acid-high metal in tributaries to the River Nyaba, abandoned mine tunnel, hand dug wells and borehole (BH1). In Onyeama coal mine axis, the two-water type obtained varied from near neutral-high metal in the borehole (BH) and hand dug well (HDW) to acid-high metal in the abandoned mine tunnel and River Ekulu River (Figure 21). Also, the water type found in the two river tributaries (RW1 and RW2) situated close to the mine tunnel indicates acid-extreme metal water type.

High levels of heavy metals are detrimental to the aquatic environment due to the toxicity and adverse effects on both plants and animals, including human beings. In particular, acute exposure to high concentration of heavy metals can kill organism directly, while long term exposures to lower concentration causes mortality or other effects, such as stunted growth, lower reproduction rates, deformities and lesions [47].

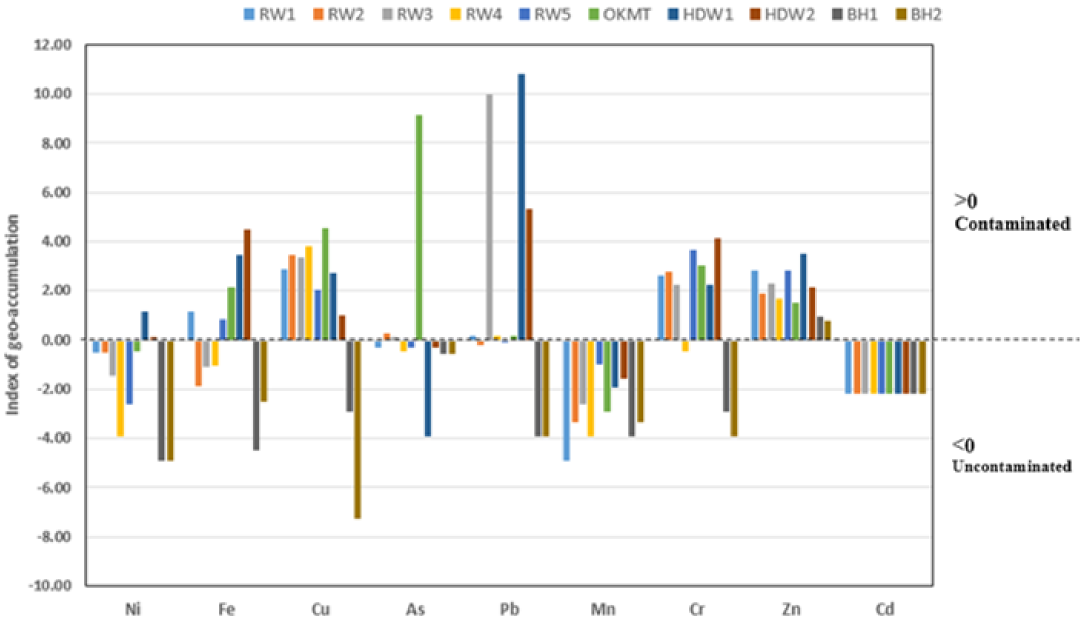

Figure 18. Index of geo-accumulation (I_{geo}) of heavy metals in surface and groundwater resources from Okpara mine.

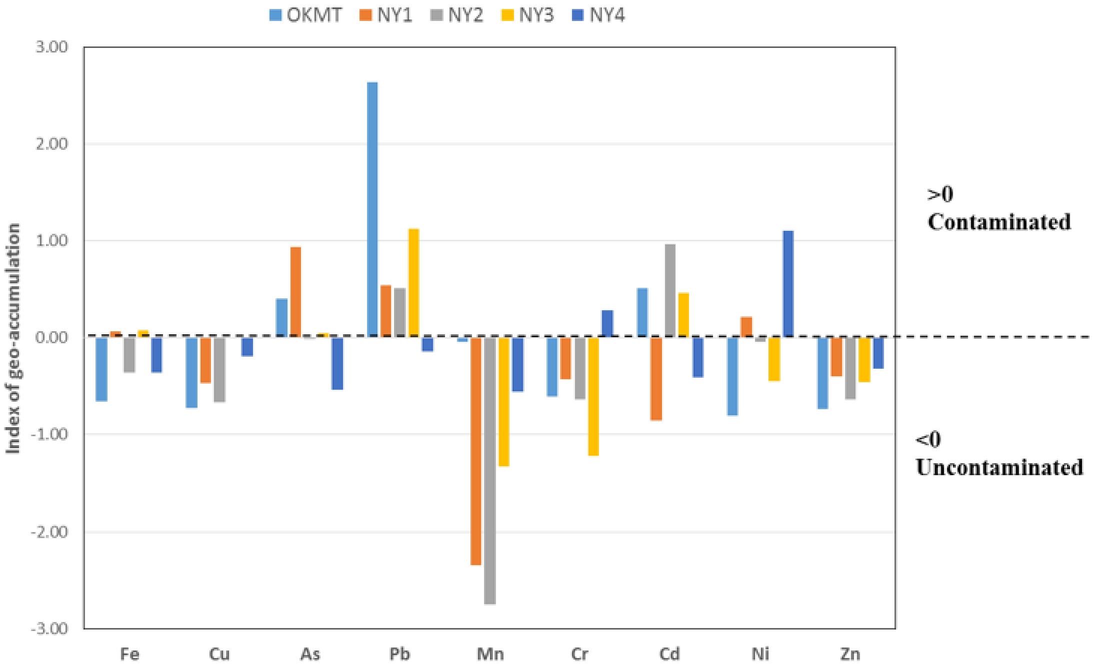

Figure 19. Index of geo-accumulation (I_{geo}) of heavy metals in soils/sediments from Okpara coal mine.

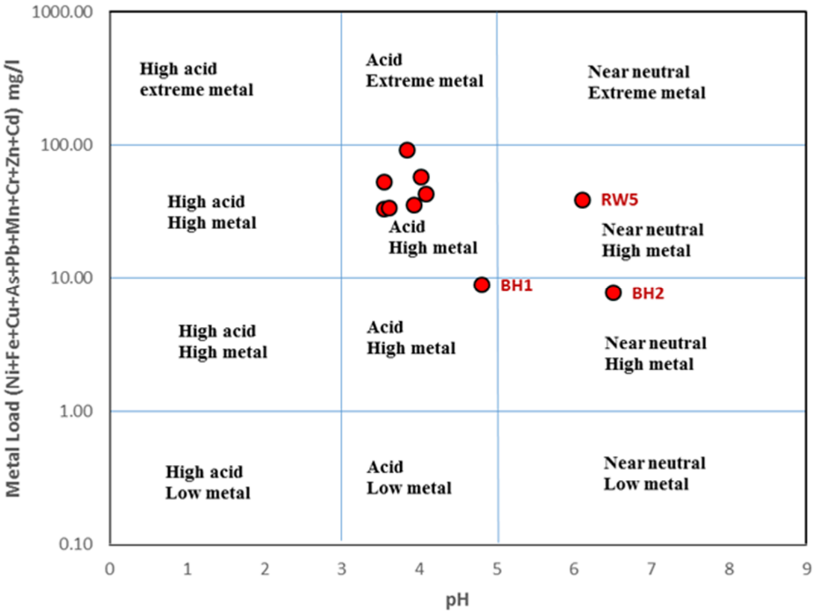

Figure 20. Classification of water samples in Okpara mine based on the plot of metal load and pH (modified from Edet *et al*. 2004).

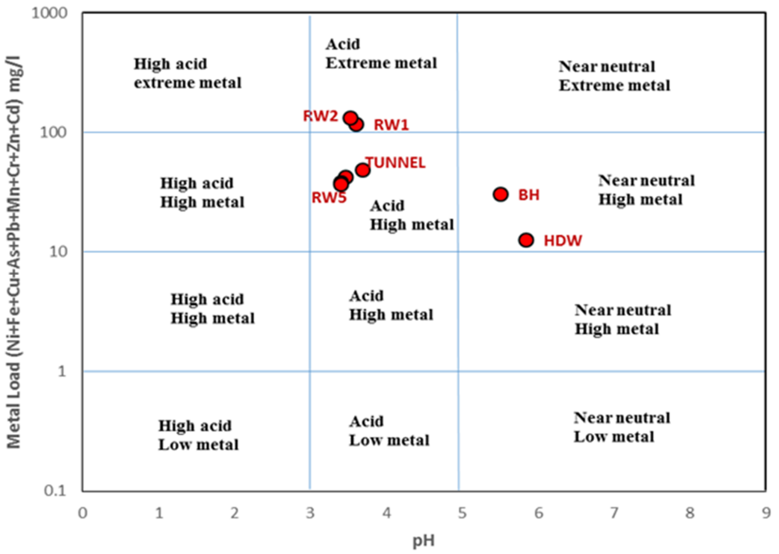

Figure 21. Classification of water samples in Onyeama mine based on the plot of metal load and pH (modified from Edet *et al.* 2004).

Data obtained was subjected to statistical evaluation using SPSS software (version 21.0). The Pearson's correlation coefficient test at 5% confidence level (p = 0.05) used to determine the similarity or association between the chemical parameters, and their possible sources or genetic relationships. The strength of the relationships between any two variables is described by a correlation coefficient (r) which rates as follows:

r = 0.0-0.19 indicates very weak relationship
r = 0.2-0.39 indicates weak relationship
r = 0.0-0.59 indicates moderate relationship
r = 0.60-0,79 indicates strong relationship
r = 0.80-1.0 indicates very strong relationship

The coefficients may be positive showing linear relationships, but could also be negative showing inverse relationships. The results show that moderate to very strong inter-correlations exist between the different pairs of metals. No relationship falls in the weak class. Generally, there is a dominance of strong positive inter-correlation existing between the heavy metals in both the water and sediments. This association is stronger in Onyeama mine sediments (r = 0.51-0.97) compared to 0.57 to 0.75 in the Okpara mine. These relationships may be interpreted that the metals have a common origin.

4. Implication for acid mine drainage

The rate and occurrence of coal-related AMD is controlled by chemical, physical and biological factors [48]. The geology of the Enugu mines area consists of layers of sandstones, siltstones and shale which host the coal seams. These host rocks contain pyrite and siderite which are iron bearing minerals. Weathering releases these minerals into the host rocks which eventually leach and concentrate the iron in them. The concentration of iron is one of the major characteristics of acid mining drainage. In the present study the concentration of iron ranges from 32 to 326 mg/kg in the sediments. These values are 2-3 times the values obtained by earlier studies in the area [36, 39]. Indicating the potential for mine drainage. In its two major oxidation states, a favourable pH dissolves into water as Fe^{2+} and eventually precipitates in a reddish-brownish yellowish colour. This is very obvious in the water as shown in Figure 3. The range of pH values for waters in the Onyeama and Okpara mines are 3.4 - 5.8 and 3.5 - 6.1 respectively, indicating acidic to weakly acidic conditions. These are ideal acidic conditions to initiate the dissolution of the iron. The presence of these solid precipitates causes a decrease of elements and their compounds which results in lower total dissolved solids. The soil and water in these mines also show significant heavy metal load comprising Fe, Cu As, Pb, Cr, Ni and Zn which typically occur in association with coal. A combination these metals and pH, causes the acidity of the water to increase. The high metal load follows the trend of results in mines characterized by AMD [49] and is similar to results obtained by [50] in some coal mines in Enugu. [51] Examined the influence of moisture content on the AMD producing potential of mineral sulphide wastes and confirmed that the presence of water plays an important role in the generation of AMD. In Enugu area, heavy precipitation (mean annual precipitation of about 1500 mm) during long rainy seasons provide enough moisture as AMD catalyst. EC values were higher in soil samples at the mine spoils which is expected due to the mineral concentration process [52]. The occurrence of sulphides together with abundant heavy metals which enhance the acid mine drainage process was noticed in this study. There is a high sulphate and iron ions in the water. The high concentration of sulphates (135-144 mg/L) in the water may be attributed to time of exposure of the rocks to the weathering agents and processes which have provided ambient conditions for the oxidation of the pyrites. However, it would appear that the occurrence of carbonate rocks in the rock sequences hosting the coal may have been dissolved by the acid drainage which has buffered the acid due to increased alkalinity. This may have attenuated the dispersal of the metals and aggressive progress of AMD.

Overall results indicate the potentials for acid mine drainage in the study area. However, the seasonal variation of the water table resulting in seasonal peak discharges from the mines only during the rainy season, and the high alkalinity, may have attenuated its progress.

5. Conclusions

This study was carried out to determine the chemical nature of discharges from two abandoned mines in the Enugu coalfield, the impact on soil and groundwater that interacts with the mine products and the occurrence of acid mine drainage. The coal host rocks are essentially sequences of sandstones and siltstones which contains pyrite, the ore mineral of iron. Environmental consciousness and protection around the mine areas is poor, hence the waters and soils impacted by the mine effluents show characteristics that may be averse to the health of plants and human beings. The water which is reddish to yellowish brown and indicating iron precipitation, is weakly to moderately acidic (3.3- 6.5). Soils and waters at the mine tunnel and spoils are more acidic and give the highest concentration of heavy metals especially Pb Cu Zn and Fe. The spoils serve as heavy metals pools that can be released into the water bodies by various processes of remobilization. The various parameters, including contamination factors pollution load index and enrichment factors that were used to determine the degree of pollution in the water bodies and sediments all show that the pollution ranges from mild to severe. The high alkalinity may be attributed to the occurrence of carbonate rocks with the host rocks. The overall effect is a buffering of the acidity of the waters. Based on the lithology, mineralogy, chemical concentrations and environmental factors, the study has shown that there is not just a potential for the generation of AMD, but its initiation or occurrence which has impacted the adjoining environment. Anomalous concentrations of heavy metals are being discharged into the nearby Ekulu and Nyaba River from these mines. Consumption of untreated water over a prolonged period from these water sources in which the concentration of these pollutants exceed drinking water criteria might be detrimental to heath.

There is therefore the need for the treatment of the mine water using appropriate remediation technology based on the site characteristics to remove or reduce AMD. This may include methods that contain or neutralize the amount of acid produced before the mine waters flow into Ekulu and Nyaba River. It is imperative to periodically monitor the quality of both surface and groundwater in the vicinity of these abandoned mines, to ascertain the potability, and to assure the protection of the health of the contiguous biotic communities who have over the years, depended on these water sources for their domestic and agricultural purposes. As a proactive measure at new mining sites, operators should be required to evaluate the potentials for acid mine drainage and provide measures to reduce it at all stages of operation using an environmental impact assessment that identifies predicts, evaluates and mitigates impacts of new projects on the environment.

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ویژگی‌های شیمیایی تخلیه از دو معدن متروکه، زغالسنگ در جنوب شرقی نیجریه: پیامدها برای آلودگی و زهاب اسیدی معادن

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چکیده:

در این مطالعه تاثیرات زیست محیطی دو معدن متروکه زغال‌سنگ در جنوب شرقی نیجریه و پتانسیل زهاب اسیدی ناشی از معدنکاری (AMD) بررسی و محاسبه شده است. پارامترهای pH، کاتیون و آنیون و میزان فلزات سنگین مورد بررسی قرار گرفته است. برای ارزیابی آلودگی از عوامل ترکیبی شامل شاخص بار آلودگی ، فاکتورهای غنی سازی ، شاخص بار آلودگی و شاخص انباشت زمین استفاده شده است. pH آب بین بازه‌ی ۳/۴ الی ۵/۹ طبقه‌بندی شده است که در بازه‌ی ضعیف تا قوی AMD قرار دارد. یون SO42، که نشان دهنده آلودگی آبهای معدنی است، غلظتی متوسط تا زیاد نشان داده است. فلزات آهن، سرب، روی و مس بیشترین فلزات سنگین در آب‌های معدن بوده‌اند. مقادیر شاخص بار آلودگی بیشتر از میزان مورد تایید بود که وخامت در کیفیت آب و رسوبات را نشان می‌دهد. مقادیر فاکتور انباشت آلودگی تا میازن ۱ نشانگر آلودگی از طریق منابع لیتوژنیک و انسانی است. انباشتگاه‌های باطله این معادن به عنوان استخرهایی عمل می‌کنند که می‌توانند با فرآیندهای مختلف و انتقال مجدد فلزات سنگین سمی را به داخل آب آزاد کنند. بر اساس ادبیات تحقیق، بررسی‌های معدنی و شیمیایی و محیط زیستی مشخص شد، این معادن متروکه دارای پتانسیل AMD هستند. جریان آب‌های معدن فلزات سنگین به ویژه آهن، را به درون آبهای مجاور خالی می‌کنند که منبع تأمین آب شهری هستند. مصرف آب تصفیه نشده برای مدت طولانی از این منابع آب ممکن است برای سلامتی مضر باشد. اقدامات مناسب و نظارت مستمر برای نظارت خوب در محیط زیست توصیه می‌شود.

کلمات کلیدی: زهاب اسیدی معدن، تخلیه، آلودگی، سوخت‌های فسیلی.