



# Lateral distribution of potential toxic elements from lead–zinc mine sites within Enyigba, Southeastern Nigeria

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## Abstract

Artisanal mining activities in Enyigba community and its environs have become a major environmental problem, which exposes the water, soil, and vegetation within the environment to potential toxic elements (PTEs). However, the lateral extent that the PTEs have spread within the environment has not been ascertained. It is important to determine the lateral extent of the PTEs within the environment to establish safe zones for drinking water usage and for crop cultivation. For this reason, about nine samples each for soil, sediment, vegetation, surface and groundwater were collected within and beyond the mine sites catchment area. The physicochemical properties of these samples were determined following standard procedures, whereas the concentration of PTEs such as cadmium, arsenic, zinc, and lead in them were determined using atomic absorption spectroscopy method. Statistical analysis, biogeochemical analysis, and geospatial information system (GIS) analytical tools were employed to interpret the field and laboratory results. The results revealed that proximity to mine site contributed significantly to the concentration level of PTEs within the environment. The concentration level of the PTEs was higher in water, soil, sediment, and vegetation samples that were collected close to the mine sites, which gradually reduces farther away from the potential pollutant source points. The environmental mobility index and soil–plant bioconcentration factor revealed that cadmium, arsenic, and zinc are more mobile and bioavailable than lead, thus their toxicity within the study area environment are more likely than that of lead.

**Keywords** Potential toxic elements · Toxicity · Mobility · Bioconcentration · Enyigba · Statistical analysis

## Introduction

Mining activities have been a significant source of environmental pollution in most places of the world (Chlopecka et al. 1996; Yang et al. 2008; Anju and Banerjee 2011). In some developed countries, mining activities are adequately planned to contain the tailings and residual effluents or run-offs which might emanate from the mine site. However, in less developed countries where there are no strong environmental policies, mining activities are carried out unprofessionally by mostly artisans without prior considerations of the resultant effect on the environment (Nnabo 2015; Igwe et al. 2017). This unprofessional mining activity is the case in Enyigba, Southeastern Nigeria, where mostly artisans are mining lead–zinc deposits. Previous environmental

assessments which have been carried out within this mine site revealed the presence of some potential toxic elements (PTEs) such as lead, zinc, chromium, cadmium, and arsenic in the prevailing soils and surface waters (Igwe et al. 2012; Okolo et al. 2018). Similar to the environmental report of Okolo et al. (2018) on the Enyigba mine site, Zhang et al. (2012) reported the high concentration of lead, zinc, copper, and arsenic on surface soil, sediments, and surface water samples within the proximity of mine sites in China. Likewise, Johansen et al. (2008), reported that water and sediment samples which were collected from Tunnelev River and delta at about 7 km and 15 km, respectively away from a Pb–Zn mine site tailings in East Greenland show elevated concentration of Pb and Zn.

Dissemination and distribution of PTEs away from their source depend on the climate, topography, transportation medium, and their degree of mobility. Containment of mine tailing and tailing dams in mine sites located in the tropical regions, which are characterized by high temperature and precipitation are less likely than those in arid and Polar

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regions. In conjunction with high relief, high rate of precipitation may lead to overflow of tailing dams and shallow mine pits thereby contaminating the resultant runoff (Gallart et al. 1999; Johansen et al. 2008). The contaminated runoff flowing downhill may also ingress in the adjacent mine tailing, leaching and eroding the available PTEs in them; thus resulting to increase in the contaminant concentration of the runoff (Benito et al. 2001). These resultant runoffs usually end up on land and are absorbed by the underlying soils which serve as a sink for the PTEs (Rowan et al. 1995). In some cases, the PTEs contaminated runoff reaches the nearby stream, which further distributes the contaminants away from their sources (UNEP 1996; Benito et al. 2001). PTEs that are in particulate matter form (mine dust and industrial emissions), which was generated as a result of mining and refining activities are also distributed further away from their source by air; hence are dependent on atmospheric conditions (Dubey et al. 2012; Singh and Li 2014).

Though PTEs may be distributed from their point source to the surrounding environment, their bioavailability depends on the mobility of the PTE; thus lead and copper have been observed to be less mobile than zinc, arsenic, and cadmium (Rieuwerts 2007; Caporale and Violante 2016). However, the degree of bioavailability of these PTEs depends on several factors such as; pH, organic matter composition, soil type, and Fe-Al oxide composition of the prevailing soil or the aqueous environment (de Matos et al. 2001; Fontes and Gomes 2003; Rieuwerts 2007). Plants absorb bioavailable PTEs; hence their relatively high concentrations in plant tissues (Yang et al. 2008; Shia et al. 2019). These PTEs may not be harmful to plants at the concentration that they are mostly found in plant tissues. However, as a result of bioaccumulation and subsequent biomagnification, their concentration level was elevated in animal tissues which feed on contaminated plants (Qu et al. 2012; Ashe 2012). Most cases of PTE poisoning in humans are as a result of consumption of contaminated fishes, meats, vegetables, and roots of plants (Zhuang et al. 2009). Though, PTEs contamination in humans is not only through direct consumption of contaminated meats and foods, but also through inhalation of contaminated air, and drinking of contaminated water. Cases of Lead poisoning was linked to the presence of lead ore mining and smelting facilities which produce fine particulate matter that are composed of Pb, Ar, and Cd in relatively high concentration (Singh and Li 2014), while Taylor et al. (2019) revealed that the prevalence of respiratory diseases in Port Pirie, South Australia was as a result of inhalation of lead. High mortality incidences have been reported in Ecuador (Betancourt et al. 2005), South Africa (Tutu et al. 2008), and Nigeria (Lo et al. 2012), as a result of direct consumption of surface waters which are contaminated by heavy metals from nearby mine sites. Contaminated groundwater in most mining towns have also been linked with acid

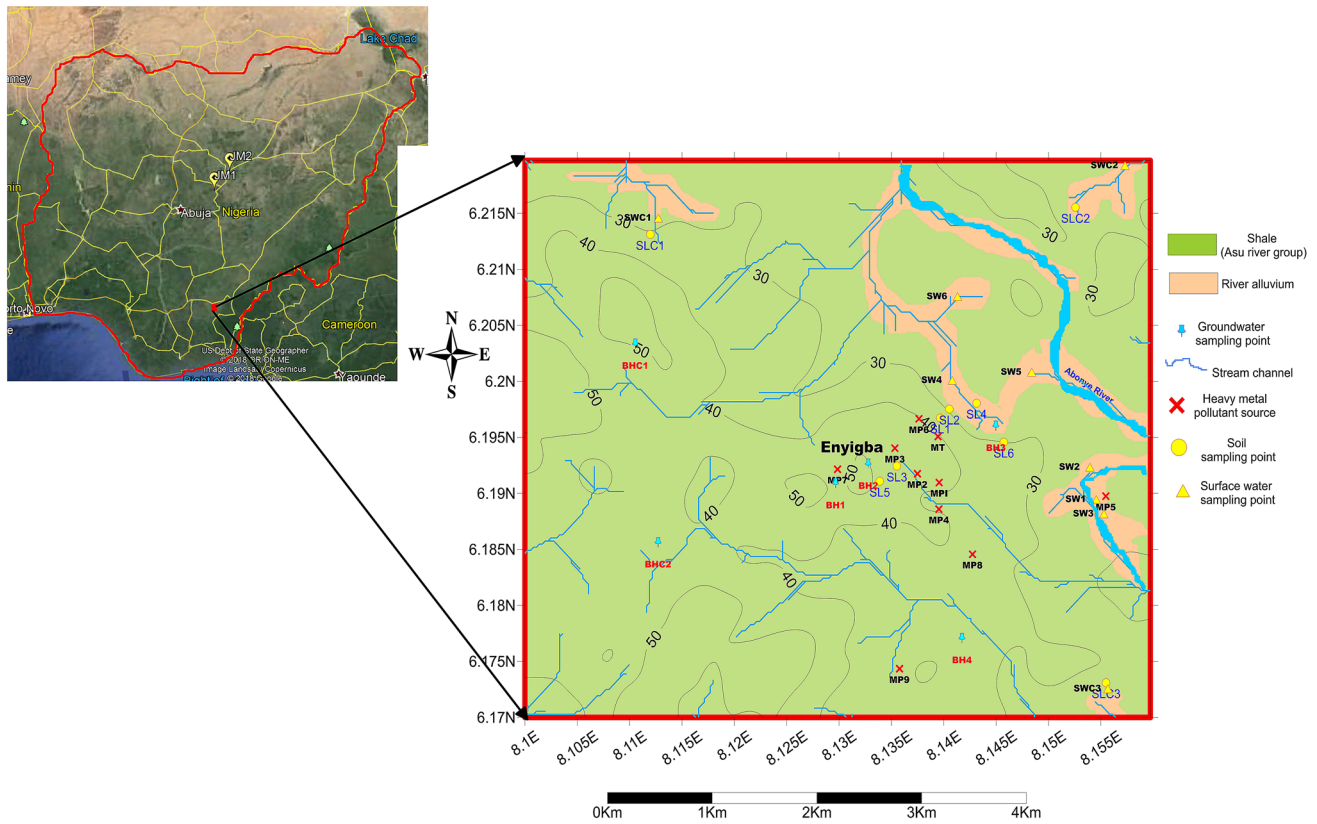
mine drainage into the surrounding aquifers through open fractures in the rock bodies; thus leading to a reduction in the groundwater quality (Tutu et al. 2008; Tame et al. 2017).

These incidences of PTE toxicity are common in developing countries where land-use activities are carried out without prior environmental impact assessment. In Nigeria, the high rate of population increase has led to rapid urbanization, which subsequently resulted in increased pressure on the existing natural resource such as arable lands, quality drinking water, and clean air. This increased pressure on the available natural resources in conjunction with the high rate of soil susceptibility to erosion (Emeh and Igwe 2017) in the Southeastern region of the country, with a relatively very small landmass and high population density (Okafor 1987), has made land-use a major environmental concern within the region. This is because natural resources such as arable lands, surface water, and groundwater which are not previously harnessed due to their proximity to contaminant source points (mine sites), are now in demand. For this reason, it becomes imperative to assess the presence of PTEs such as cadmium, arsenic, zinc, and lead in the environment that is within the catchment area of the Pb–Zn mine sites. Thus, the aim of this research was to determine the lateral extent of cadmium, arsenic, zinc, and lead concentration in soils, sediments, groundwater, surface water, and vegetation as a result of their proximity to the Enyigba Pb–Zn mine sites. Previous authors like Zawadzki and Fabijanczyk (2013), Fabijańczyk et al. (2017, 2019) have successfully determined the lateral and geospatial distribution of PTEs from potential pollutant source points in the environment by integrating geochemical, geophysical, and geostatistical methods. However, this research will employ the use of biogeochemical, inferential statistical, and geospatial analytical methods to achieve its aim. The result from the research will help to ascertain the quality of the soil, surface water, and groundwater within the study area for agricultural and domestic uses. It will also assist the government in advising on the toxicity potential which may arise as a result of possible land-use in the area.

## Materials and methods

### Study area

The study area lies within latitude 6.17–6.22 N and longitude 8.1–8.16 N in the Southeastern part of Nigeria, and covers a total area of about 36 km<sup>2</sup> (Fig. 1). It is being underlain by the Asu river group, which is an Albian–Cenomanian deposit that comprises of shales and siltstones with intercalations of limestone and arkosic sandstones (Akande et al. 2011). These geologic deposits are relatively old and have undergone several tectonic events during the Turoonian period (Nwajide 1990). During the Turoonian period,



**Fig. 1** Geology and sample location map of the study area

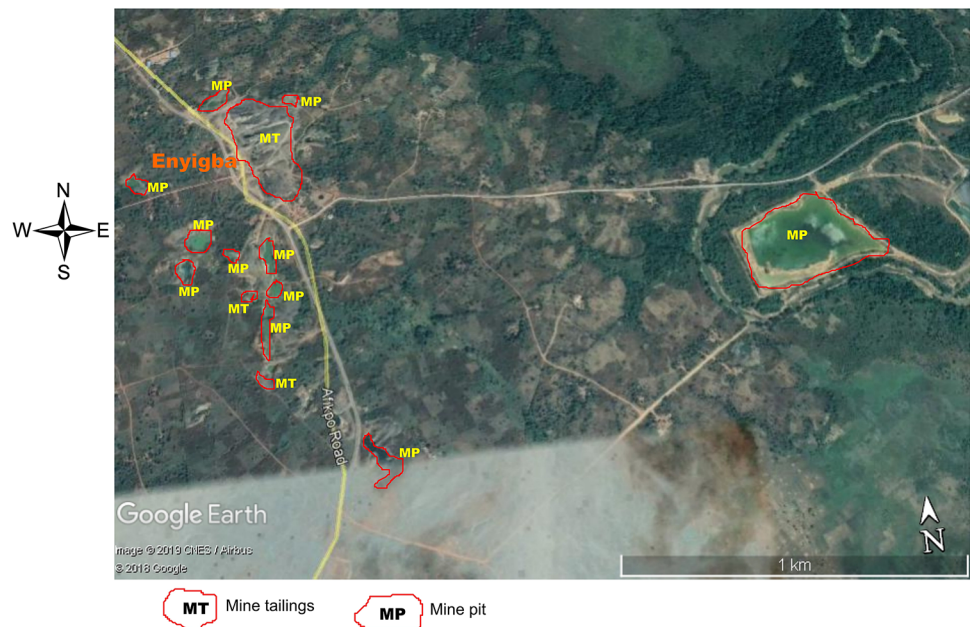
lead–zinc mineralization, which is presently mined, was believed to have occurred (Nwajide 1990; Obiora and Umeji 2004). Over the years, weathering processes have generated a thick layer of residual soil which supports rich vegetation. Due to the prevailing tropical weathering, the resultant soils are lateritic, with a high percentage of aluminium and iron sesquioxides, and a relatively low percentage of essential soil nutrients (Igwe et al. 1999). The soils are mainly clayey soils, with relatively low organic matter composition, and are characterized by high porosity and low permeability (Akamigbo and Asadu 1983; Okolo 2014). However, some of the soils along the major stream channels were found to be clayey-sandy loam, with relatively high permeability than the adjacent in situ soils. These thin layered alluvial soil deposits are believed to have been deposited by fluvial processes and are restricted few meters from the major stream channels (Fig. 1).

The area is characterized by a tropical climate with two major seasons, the wet and the dry season. The mean annual temperature is 28 °C, which peaks to 31 °C in March, and drops to its lowest point to about 16 °C in December. Similar to the temperature, the total annual rainfall is 1581 mm/year, with the highest amount (350 mm/month) occurring in July, which drops almost to 16 mm/month in February

(Eze 2007). The topography of the area is undulating, and is characterized by thick vegetations in the valleys which progressively thin out uphill. Numerous meandering streams dissect the area, and they are being drained by the major headstream, the Abonye River, which flows in the south-east direction of the study area (Fig. 1). Most of the surface water bodies are ephemeral streams, and ponds, which are formed as a result of abandoned lead–zinc mine pits (Fig. 2). During the peak of the rainy season, June–August, these shallow mine pits usually overflows into the surrounding environment. However, in the dry season, most of this ponds and streams dry up, leaving behind greener vegetations along their flow channels. This vegetation which grows along the stream channels serves as a source of forage for grazing animals during the dry season.

The area is originally moderately populated with mostly farmers, which their primary occupation is to produce food crops such as cassava, yam, maize, and vegetables in subsistence quantity. However, due to the presence of lead–zinc deposits, artisanal miners have added significantly to the population of the area. Mining activities are carried out by mostly artisans, who have learned few skills from the mining companies that have previously operated within the environment; thus leading to the proliferation of illegal mines.

**Fig. 2** Satellite image of the study area showing road accessibility, mine pits and mine tailings



These illegal mining activities have resulted in numerous abandoned shallow mine pits and unplanned mine tailing dumpsites, which are potential sources of heavy metal contaminants. Schools, homes, and agricultural farmlands are close to these mine pits and mine tailing dumpsites. The educational, domestic, and agricultural activities within this area depend solely on the nearby surface and groundwater for their water needs; hence are at risk of heavy metal toxicity.

### Field survey and sampling

Before the field investigation, the area was surveyed using remotely sensed information. The satellite imageries of the area were acquired through Google earth (Pro version). The acquired images revealed the location of the mine pits and the mine tailing dumpsites (Fig. 2), which are of major concern to this investigation. The topographical characteristics and stream flow direction of the area were determined by analysing the digital elevation model (DEM) data which was acquired from the United States Geological Survey (USGS) archives with the aid of Surfer (version 11). Integrating the satellite imageries, the topographic map, and the geologic map of the study—which was sourced from the Nigerian Geological Survey Agency (NGSA), helped in the production of the study area map (Fig. 1). The study area map that was produced helped in the location of the potential sampling points prior to field investigations.

Six samples each for soil, sediments, surface water, vegetation, and four groundwater samples were collected within the catchment area of the potential toxic elements sources (mine tailings dumps and abandoned mine pits). Three samples each for soil, sediments, surface water, vegetation, and

two groundwater samples were collected beyond the catchment area of the potential toxic elements sources. These samples which are collected beyond the catchment area of the potential toxic elements sources were used as control samples for the investigation. This was on the assumption that samples which are relatively far from the possible pollutant sources will not be affected by the PTEs. Samples were collected at predetermined lateral intervals (sample spacing) from the PTEs sources (Table 1). This sample spacing was designed to determine the variation in the concentration of PTEs with successive distance away from their potential sources.

Soil and vegetation samples were collected at the same location in the nearby farmlands. The reason for collecting soil and vegetation samples in the same location was to ensure that the concentration level of the contaminants in the vegetation samples will reflect what is found in the soil. Soil samples were collected at a depth of 0–20 cm using a hand auger, while cassava (*Manihot esculanta* Crantz) leaves at the same farmland was hand-plucked as representative vegetation samples. Following similar reason with that of the soil and vegetation sampling, surface water and sediment samples were also collected at the same location in the nearby stream banks. The reason for sampling the water at the stream bank is because it is a common practice by most villagers to fetch water for domestic purposes at the stream bank. Sediments samples were collected at a depth of 0–20 cm using a hand auger, while surface water samples were collected in triply filtered 1-L plastic bottles. Groundwater samples were collected in the nearby wells and boreholes within the area. Unlike the other samples, the locations of the boreholes were not predetermined because the wells



**Table 1** Sampling spacing from PTEs potential contaminant source

S/N	1	2	3	4	5	6	7	8	9
Sample code	Distance from potential heavy metal contaminant source (m)								
	Mine catchment					Control area			
SL	0	100	200	300	400	500	3600	2900	2900
VG	0	100	200	300	400	500	3600	2900	2900
SW	0	50	200	500	800	1200	3600	3800	3100
SD	0	50	200	500	800	1200	3600	3800	3100
GW	130	350	500	700			3100	2900	

SL soil, VG vegetation, SW surface water, SD sediment, GW groundwater

already exist. However, wells, where the groundwater samples were collected, were chosen in such a way that reflects spatial variability (Table 1). Field sampling was carried out in the month of September 2018, and the samples were collected in triplicates at each sampling location. They were packaged in well-labelled containers and were preserved following standard procedures for each sample type prior to laboratory analysis.

### Laboratory analysis

The soil and sediment samples were allowed to air dry for 2 weeks in the laboratory. Due to the clayey nature of the soil and sediment aggregates, they became compacted when dry. These compacted aggregates were pulverised in a hardened plastic mortar into finer particles, which was then sieved using an electrically controlled sieve shaker. Particles less than 2 mm were used for the soil and sediment chemical analysis. To determine the pH of the soil and sediment samples, about 10 g of soil or sediment samples that are less than 2 mm were mixed with 50 mL of deionised water in a glass beaker. The mixture was slightly stirred for 2–3 min and was then allowed to settle for about 2 min. After 2 min, a portable pH meter (Hanna, HI 9813-5) which has been properly calibrated was dipped into the mixture to determine the pH. Organic matter composition of the soil was determined following the Walkley and Black method that was described by Nelson and Sommers (1982).

Heavy metal concentration in soil and sediment samples was determined by atomic absorption spectroscopy (AAS) method using Shimadzu atomic adsorption spectrometer with ROM version 1.01. Prior to the heavy metal analysis, soil and sediment samples were digested by weighing about 0.3 g of soil or sediment samples that are less than 2 mm into a digestion flask, and 30 mL of aqua regia was added into it. The mixture was digested into a fume cupboard until a clear solution was obtained. It was further cooled, filtered, and then made up to 50 mL mark in a standard volumetric flask with deionised water. Standard solutions of 2 mg/L, 4 mg/L and 6 mg/L were prepared from 1000 mg/L of the above stock solution which was thereafter analysed using

Shimadzu atomic adsorption spectrometer with ROM version 1.01. The calibration curve was plotted automatically for the metal of interest (Pb, Zn, Cd, and As) which was analysed using its respective wavelength. After that, the concentration of each metal was generated from the standard graph by the instrument.

The water samples temperature, total dissolved solids (TDS), electrical conductivity (EC), and pH were determined in situ. This was done by dipping the multipurpose portable temperature, pH, EC, and TDS meter (Hanna, HI 9813-5)—which has been properly calibrated, into 100 mL of a water sample that was collected in a transparent 250 mL plastic beaker. The corresponding temperature, EC, TDS, and pH were thus read. PTEs concentration in water samples were determined by AAS method after the water samples have been digested following a similar method that was used in soil analysis which was described in the previous paragraph. The water chemical analytical procedures were in accordance with APHA (2005) procedures, unless otherwise stated.

The fresh plant leaves which was collected from the field was washed using deionised water to remove any impurities and was allowed to air dry away from sunlight. The dried leaf samples were then cut into smaller pieces using a knife, which was further blended to granules with a blender. 1.00 g of the dried leaf granules was weighed into a 250 mL Pyrex conical flask and 20 mL of digestion solution (a mixture of 2:2:1 nitric acid, sulphuric acid, and perchloric acid) was added following APHA 3030-I (2005) modified procedure. The vegetation sample-acid mixture was heated in a fume hood at 250 °C until complete digestion was achieved. The digested solution was removed from the fume hood and was allowed to cool. 20 mL of distilled water was added and allowed to cool further. The digested solution was sieved using a Whatmann 42 filter ashless filter paper into 100 mL glass volumetric flask. The digested filtrate in the volumetric flask was then made up to the 100 mL mark on the flask and thereafter was subjected to AAS following APHA 3111-B (2005) procedures to determine the heavy metal concentration in plant tissues using atomic absorption spectrometer (PG instruments, AA500, software: AAWin 3.0).

## Data analysis

The data which were generated from the field investigations and laboratory analysis was summarized statistically in such a way that presents their central and dispersion tendency. *T* test comparative analysis was carried out on the data to determine if there are significant lateral variations between the concentration level of PTEs in samples collected within the mine catchment area and those from the control area. Linear regression analysis was also carried out on the data to determine the relationship between the concentration of PTEs in soil and plant tissue, as well as in sediment and the corresponding surface water. The PTEs environmental mobility index (EMI) in soil, sediment, and surface water were determined by calculating the ratio of heavy metal concentration in soil, sediment or surface water obtained from the furthest distance, to the concentration in the closest distance. Similarly, soil–plant bioconcentration factor (BCF) was determined by calculating the ratio of heavy metal concentrations in the plant to their concentration in soil. All statistical data analysis, such as summary statistics, *T* test comparative analysis, linear regression analysis, and graphs, were done with Microsoft Excel 2007. The potential contaminants susceptibility maps were generated by plotting the normalized average concentration of PTEs in soil or surface water against their corresponding geographic coordinates with the aid of Surfer (version 11)—a geospatial analytical package.

## Results and discussion

### Potential toxic elements concentration in soil and plant tissues

The soils within the study area are acidic with an average pH value of 5.7, which ranges from 5.1 to 6.1 (Table 2). The minimum pH value of 5.1 was found in samples within 5 m radius from the mine tailing dumps, while the maximum pH

of 6.1 was found in soil which was collected about 400 m away from the sources of the possible pollutants. The lateral variation that was observed in the soil acidity could be attributed to the contamination of sulphide minerals (Sphalerite and Galena) from the nearby mine tailings. These lateral variations between the samples collected within the mine catchment area and those of the control area were revealed by the *T* test statistical comparative analysis to be statistically insignificant ( $p \geq 0.05$ ) (Table 2). Thus the proximity of mine pits or mine tailing dumpsite does not have a significant contribution to the acidity of soil within the study area. Similar pH has been recorded by Akamigbo and Asadu (1983), Igwe et al. (1999) and Okolo (2014) in soil within the study area. Igwe et al. (1999) attributed the general acidic pH of the soils within this area to the prevailing lateritic conditions, which are also common in soils within most tropical region.

Similarly, the soils organic compositions (SOC) were low with an average value of 1.16% (Table 2), which range from 0.61% in soils samples that were collected within 5 m radius from the mine tailing dumps, to 1.59 in soils collected at about 500 m away from the mine tailings dump. This lateral variation was statistically insignificant ( $p \geq 0.05$ ) (Table 2), which also imply that distance of farmlands from mine site may not have a significant contribution to the fertility of the soil, thus its SOC. These observed variations in the SOC of the soils were attributed to variation in soil biomass due to land use management (Okolo 2014).

The average concentration value (mg/kg) of lead, zinc, cadmium, and arsenic in soil samples within the mine catchment area were 2.83, 3.89, 0.38, and 5.02, respectively. These values range from 0.03–6.79, 1.17–9.89, 0.10–0.91, and 1.75–10.83 for Pb, Zn, Cd, and As, respectively, in the soil samples (Table 2). The trend in the line graph (Fig. 3) revealed that the concentration levels of most PTEs that were collected within a 0–5 m radius from the potential pollutant source points are relatively high. These values gradually reduce further away from the PTEs source points up to the distance of 500 m. It was observed that the concentration

**Table 2** Summary statistics of pH, SOC, and PTEs concentration in the soil within and beyond the mine site catchment area

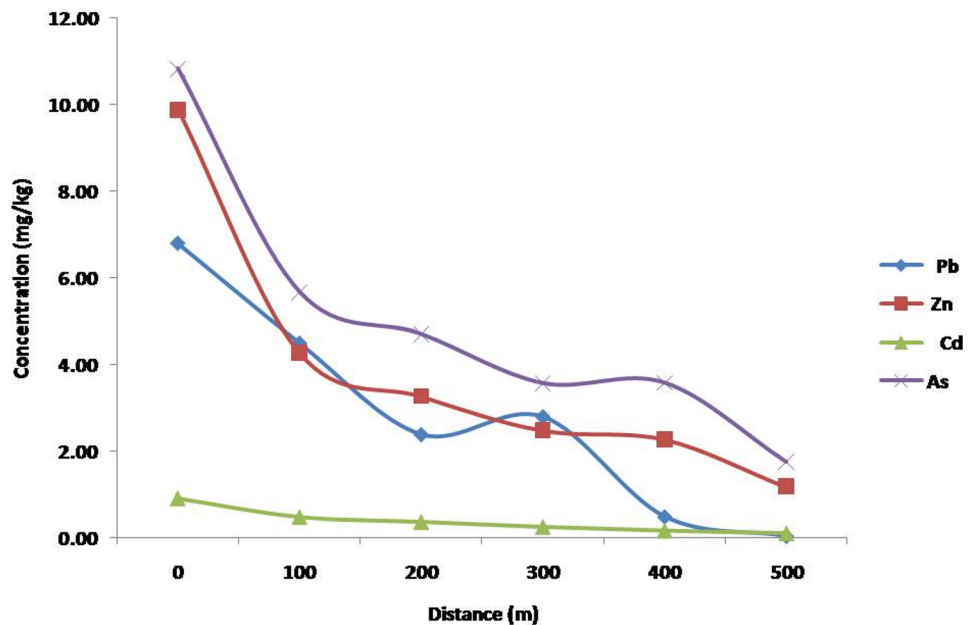
Parameter	Mine catchment					Control area					<i>P</i> value	EMI	Perm. limit
	N/O	Mean	SD	Max	Min	N/O	Mean	SD	Max	Min			
Pb (mg/L)	6	2.83	2.53	6.79	0.03	3	0.02	0.01	0.03	0.01	0.04	0.004	200 <sup>a</sup>
Zn (mg/L)	6	3.89	3.12	9.89	1.17	3	0.23	0.19	0.40	0.03	0.03	0.118	1100 <sup>a</sup>
Cd (mg/L)	6	0.38	0.29	0.91	0.10	3	0.01	0.01	0.02	0.00	0.03	0.110	0.43 <sup>a</sup>
As (mg/L)	6	5.02	3.13	10.83	1.75	3	0.00	0.00	0.00	0.00	0.01	0.162	20 <sup>b</sup>
pH	6	5.70	0.34	6.10	5.10	3	5.97	0.21	6.20	5.80	0.19		
SOC (%)	6	1.16	0.33	1.59	0.61	3	1.45	0.22	1.67	1.24	0.16		

SD standard deviation, EMI environmental mobility index, N/O number of observation, Perm. permissible

<sup>a</sup>Source from NYS DEC (2006)

<sup>b</sup>Source from BGS (2005)

**Fig. 3** Variation in heavy metal concentration in soil with distance from mine site



levels of most PTEs that were collected at about 500 m away from the potential source points were similar to the concentration levels of samples collected at the control area (Table 2). Thus, this could imply that the lateral movement of most of the PTEs in soil is still restricted within 500 m radius from potential heavy metal source point (mine pit and mine tailing dumps). These lateral variations that were observed in the concentration levels of the PTEs in soils were statistically significant ( $p \leq 0.05$ ) for all the PTEs that were analysed (Table 2). This implies that proximity to mine pit or mine tailing dumpsite has a significant contribution to the concentration level of the PTEs in the soil. Similar observation were made by Zawadzki and Fabijanczyk (2013) and Fabijańczyk et al. (2019) where they revealed that proximity to industrial activities that are associated with production of heavy metals and rare earth elements have strong influence on the concentration level of PTEs in the surrounding soil.

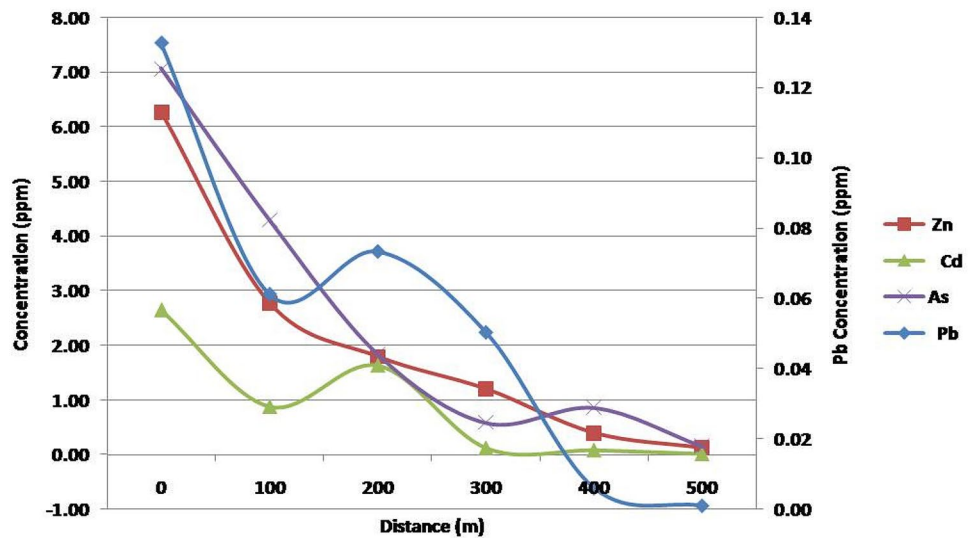
The environmental mobility index (EMI) revealed that the sequence of PTEs mobility in the soil was  $As > Zn > Cd > Pb$  (Table 2); hence Pb is by far less mobile than Zn, Cd, and As. Rieuwertz (2007) and Caporale and Violante (2016) have attributed the mobility of metals to their sorbent nature and as well as their degree of solubility. Eriksson (1989) revealed that Cd and As are more soluble and bioavailable in sandy soils, whereas Pb has strong affinity to clay fractions of the soil (Andersson 1979). Since the soil within the study area is predominantly composed of clay fractions, Pb in runoff which emanate from mine pits and mine tailings dumps will be adsorbed to the soil particles whereas Zn, Cd, and As which are relatively more insoluble could easily be transported down slope in particulate form to relatively far locations. Therefore this could be the reason

why the concentration levels of Zn, Cd, and As are relatively higher in soils further away from the source points (Fig. 3), thus resulting in their relatively higher EMI.

Similar to what was observed in the soil, the concentration level of the PTEs in plant tissues decrease with increase in distance away from the heavy metal potential contaminant sources (Fig. 4). These lateral variations between the concentration level of PTEs in plant tissues within the mine catchment area and those from the control area are statistically significant ( $p \leq 0.05$ ) for Pb, Zn, and As, but insignificant ( $p \geq 0.05$ ) for Cd (Table 3). Thus, this implies that proximity to mine pit and mine tailing dumps has a significant contribution on the concentration level of Pb, Zn, and As in plant tissues, but not on the concentration level of Cd in plant tissues. This observed lateral variation in the concentration level of PTEs in plant tissues could be attributed to their degree of bioavailability in soils. Cd has been shown to be more bioavailable to plants than Pb, Zn, and As (Yang et al. 2008; Shia et al. 2019); hence the observed variations.

Evidence from the linear regression analysis (Fig. 5) revealed that the concentration level of PTEs in soils correlated well with their corresponding concentration in plant tissues, with correlation coefficient ( $R^2$ ) values of 0.88, 0.99, 0.98, and 0.91 for Pb, Zn, Cd, and As, respectively. These values imply that the more the concentration of heavy minerals in soils, the more likely they are to be concentrated in plant tissues. However, de Matos et al. (2001), Fontes and Gomes (2003), and Rieuwertz (2007) have revealed that other factors such, pH, sorbent nature, amount of organic and inorganic ligands, and redox reaction within the soil environment controls the soil–plant bioconcentration of PTEs rather than just their concentration level in soil. The

**Fig. 4** Variation in heavy metal concentration in plant tissues with distance from mine site

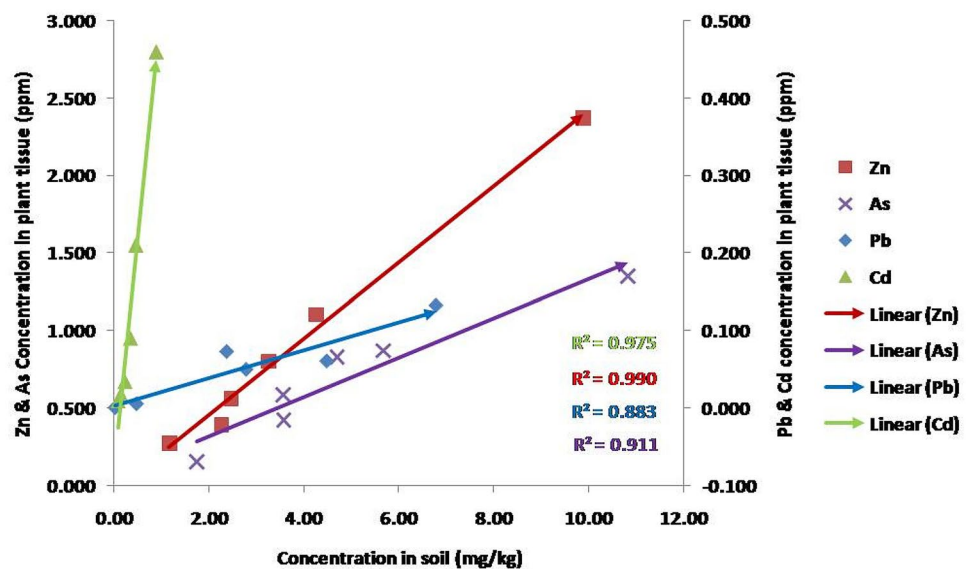


**Table 3** Summary statistics of the PTEs concentration in plant tissues

Parameter	Mine catchment					Control area					p value	WHO permissible limit
	N/O	Mean	SD	Max	Min	N/O	Mean	SD	Max	Min		
Pb (mg/kg)	6	0.054	0.048	0.133	0.0010	3	0.0004	0.0008	0.0013	0.00	0.042	2
Zn (mg/kg)	6	0.915	0.772	2.370	0.2700	3	0.0300	0.0265	0.0600	0.01	0.038	0.60
Cd (mg/kg)	6	0.137	0.175	0.460	0.0090	3	0.0004	0.0008	0.0013	0.00	0.115	0.02
As (mg/kg)	6	0.701	0.415	1.350	0.1503	3	0.0003	0.0006	0.0010	0.00	0.009	–

SD standard deviation, N/O number of observation

**Fig. 5** Relationship between concentration of heavy metals in soil and in plant tissues



sequence of soil–plant bioconcentration factor (BCF) within the study area (Table 4) was Cd > Zn > As, >> Pb, with average BCF values of 0.25, 0.23, 0.14, and 0.02, respectively. A similar sequence in the BCF of PTEs has been observed by

Chopra et al. (2009), and Ndeda and Manohar (2014). These authors explained that the observed variation in the BCF of the PTEs is as a result of their inherent chemical properties, which subsequently affect their mobility and bioavailability



**Table 4** Summary statistics of soil–vegetation bioconcentration factor

Parameter	Pb	Zn	Cd	As
N/O	6	6	6	6
Mean	0.02	0.23	0.25	0.14
SD	0.01	0.03	0.18	0.03
Max	0.03	0.26	0.51	0.18
Min	0.01	0.17	0.09	0.09
Skewness	0.63	− 1.69	0.68	− 0.45

SD standard deviation, N/O number of observation

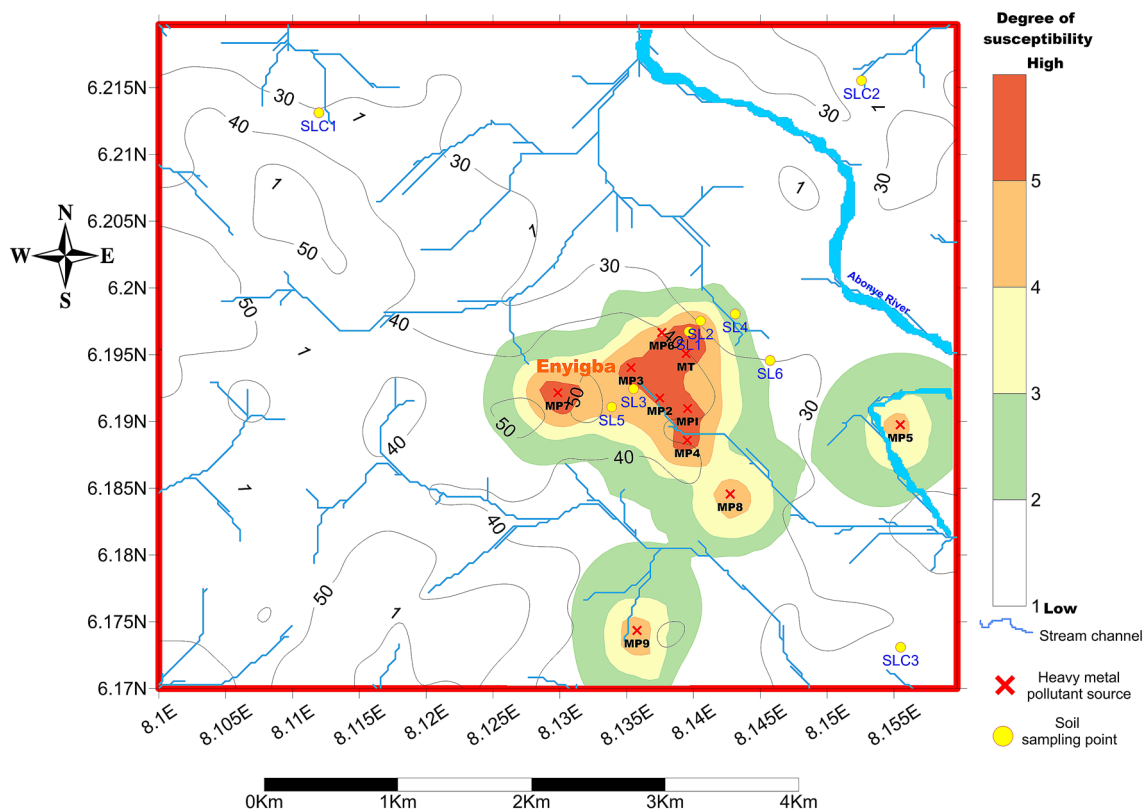
to plants. Therefore plants within the study area are more susceptible to cadmium contamination than lead.

The average concentration values of Zn and Cd in plant tissues that were collected within the mine catchment area are beyond the WHO permissible limit of 0.60 mg/kg and 0.02 mg/kg (WHO 1996), respectively, while that of Pb is within WHO permissible limit of 2 mg/kg (Table 3). It was however, observed that while the mean concentration level of most of the PTEs in plant tissues that were collected within the mine catchment area are beyond WHO permissible limits, their concentration level in plant tissues that were collected beyond 400 m way from the mine sites and those of the control area are generally within the permissible

limit. This observation was further revealed by the topsoil potential to PTEs susceptibility map (Fig. 6), which shows the spatial distribution of PTEs in soils within the study area. Thus, crops which are grown in farmlands within 500 m radius from the mine sites are more likely to be susceptible to PTEs contamination than those beyond. However, their degree of susceptibility to heavy metal contaminant would depend on their proximity to the mine sites (Fig. 6).

**Potential toxic elements concentration in sediment and surface water**

Results from the laboratory analysis revealed that the sediments within the study area are generally acidic with an average pH value of 5.80 (Table 5), which ranges from 5.20 in samples collected within a 0–5 m radius from the mine sites to 6.10 in samples collected 800 m away from the potential pollutant source points. The lateral variation between the pH of samples that were collected within the mine catchment area and those of the control area was statistically insignificant ( $p \geq 0.05$ ). This implies that proximity to mine site does not have a significant contribution to the pH of the sediments within the study area. Increase in pH could be associated with sulphide minerals such as galena, and sphalerite, which are mined at the nearby mines. Noting that



**Fig. 6** Top soil potential to heavy metal susceptibility map

**Table 5** Summary statistics of pH, SOC, and PTEs concentration in the sediment within and beyond the mine site catchment area

Parameter	Mine catchment					Control area					<i>p</i> value	EMI
	N/O	Mean	SD	Max	Min	N/O	Mean	SD	Max	Min		
Pb (mg/L)	6	1.91	1.51	4.13	0.09	3	0.000	0.001	0.001	0.00	0.03	0.02
Zn (mg/L)	6	11.36	10.54	30.08	0.78	3	0.099	0.118	0.230	0.00	0.05	0.03
Cd (mg/L)	6	4.49	3.93	11.23	0.62	3	0.030	0.052	0.090	0.00	0.04	0.06
As (mg/L)	6	5.70	5.10	14.78	0.93	3	0.087	0.125	0.230	0.00	0.04	0.06
pH	6	5.80	0.33	6.10	5.20	3	6.033	0.115	6.100	5.90	0.17	
SOC (%)	6	0.85	0.12	0.97	0.67	3	1.027	0.064	1.100	0.98	0.02	

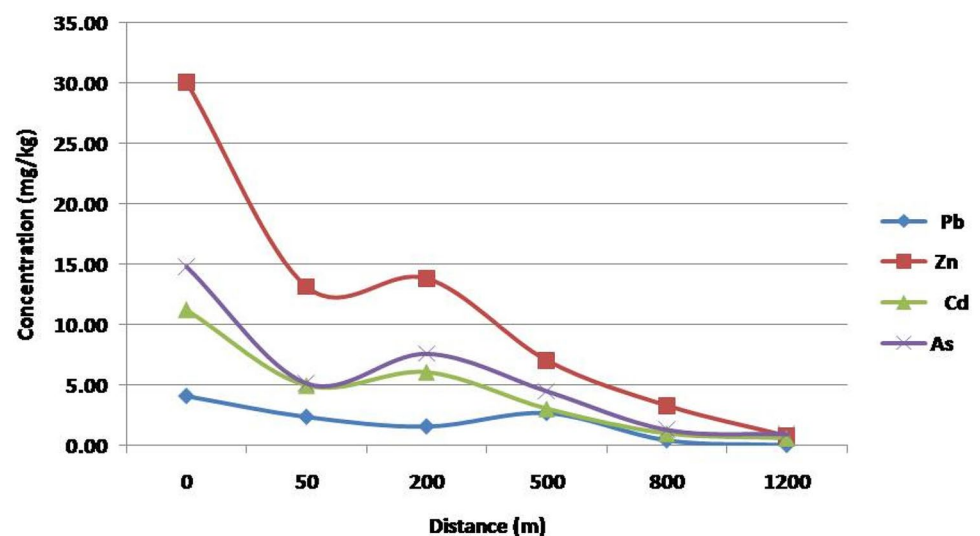
SD standard deviation, EMI environmental mobility index, N/O number of observation

dissolution of sulphates which resulted from the subsequent oxidation of sulphides produces acidic solutions (Shu et al. 2001). Similar to the pH of the sediments, the average SOC of the sediments that were collected within the mine site catchment area (0.85%) was lower than those of the control area (1.03%). Unlike the pH, the observed lateral variation in SOC of the sediments was statistically significant ( $p \leq 0.05$ ). This implies that the proximity to mine site has a significant effect on the organic matter composition of the sediments.

Figure 7 reveals that the heavy metal concentrations in the sediments within a 0–50 m radius from the mine sites were higher than those beyond this limit. These concentration levels of PTEs slightly fluctuate between the distances of 50–500 m radius from the mine sites, and then gradually decreased further away. The minimum values (Table 5) of heavy metal concentration in sediments that were collected within the mine catchment area were obtained from samples at the distance of 1200 m away from the mine sites. These concentration levels of the PTEs in sediments at 1200 m away from the mine sites are almost similar to those of the control area. However, the observed lateral variations in the concentration levels of PTEs in sediments were all statistically significant ( $p \leq 0.05$ ). The implication of these results

is that proximity to mine sites has a significant contribution to the concentration levels of the PTEs in sediment within the study area, and it is in agreement with the results of Stafilov et al. (2010) and Johansen et al. (2008). Evidence from the EMI result reveals that the sequence of mobility of PTEs in sediments within the study area was  $Cd > As > Zn > Pb$  (Table 5). The reason for this observed lateral variation in the degree of the mobility of the heavy metal in sediments samples could be attributed to their chemical properties; hence their reactivity (Caporale and Violante 2016). de Matos et al. (2001), Udom et al. (2004) and Violante et al. (2008) have asserted that Zn, Cd, and As are more mobile in the environment than Pb. Noting that Pb has more sorbent affinity for clay minerals (de Matos et al. 2001) whereas As, Cd, and Zn has more affinity to organic substances (Violante et al. 2012). Moreover, the prevailing sediments are primarily composed of clay particles (minerals) with relatively low SOC.

Similar to the pH of the sediments, the pH of the surface waters that was collected within the mine catchment area are all acidic with an average pH value of 5.98, which ranges from 5.30–6.60. The maximum pH values were obtained in streams within a 0–5 m radius from the mine sites, while

**Fig. 7** Variation in heavy metal concentration in sediments with distance from mine site

the minimum values were obtained in streams 1200 m away from the mine sites (Table 6). The pH of the surface water samples that were collected at the control area was also slightly acidic with average pH values of 6.23. This lateral variation that was observed in the pH values of the surface waters was observed to be statistically insignificant ( $p \geq 0.05$ ). This implies that the proximity of surface water to mine site may not have a significant contribution to its pH. The EC and TDS values of the surface water samples that were collected within the mine site catchment area were slightly higher than those of the control area (Table 6). These observed lateral variation in the concentration level of TDS and EC of the surface waters were statistically insignificant ( $p \geq 0.05$ ); hence implying that proximity to mine site has no significant contribution to the EC and TDS composition of the surface waters within the study area. The relationship between TDS and pH of runoff samples has been described by Emeh and Igwe (2018) to be strongly correlated, which increase with increasing acidity and alkalinity. Thus, increase in the acidity of the surface waters—which is caused by the possible dissolution of sulphide minerals from the mine sites (Ezeh and Anike 2009; Zhang et al. 2012), will subsequently result to increase in their ability to dissolve solutes. Similar pH, EC, and TDS values have been reported by Okolo et al. (2018) and Kacmaz (2020) in surface water samples from the same study area and in a mining district in Turkey, respectively.

The average concentration level (mg/L) of PTEs in surface waters within the mine catchment area is 0.10, 8.09, 1.54, and 3.77, for Pb, Zn, Cd, and As, respectively (Table 6). These values range from 0.001–0.23, 0.88–20.74, 0.26–3.83, and 0.49–9.81, for Pb, Zn, Cd, and As, respectively. The maximum values were obtained from samples collected within a 0–5 m radius from the potential pollutant sources, whereas the minimum values are collected in surface water samples 1200 m away from the mine sites (Fig. 8). These concentration levels of PTEs in surface

waters within the mine catchment area compare favourably with the results obtained from other works which have been carried out within the study area (Igwe et al. 2015; Nnabo 2015; Okolo et al. 2018). However, these authors did not consider the lateral variations in their sampling techniques. Thus PTEs contaminant limits in surface water within the environment could not be established. It was further observed that the average concentration level of PTEs in the surface water samples that were collected in the control areas was relatively low compared with those of the mine catchment area (Table 6). This observed lateral variation was statistically significant ( $p \leq 0.05$ ), for all the PTEs. The implication of this lateral variation could be that the concentration levels of the PTEs in surface water within the study area are significantly determined by the proximity to the mine site. This is because the PTEs concentration gradually dilutes further away from the mine site (Fig. 8) probably due to influx of other surface waters into the main stream, and also due to adsorption of PTEs in the stream sediments (Welch et al. 1988). The surface water potential to PTEs susceptibility map (Fig. 9) further revealed that most surface waters within 1200 m radius from the mine sites are likely to be contaminated by PTEs. However, their degree of susceptibility to PTEs contamination is more likely to occur in areas closer to the mine sites, which then gradually reduces further away from the PTEs pollutant sources.

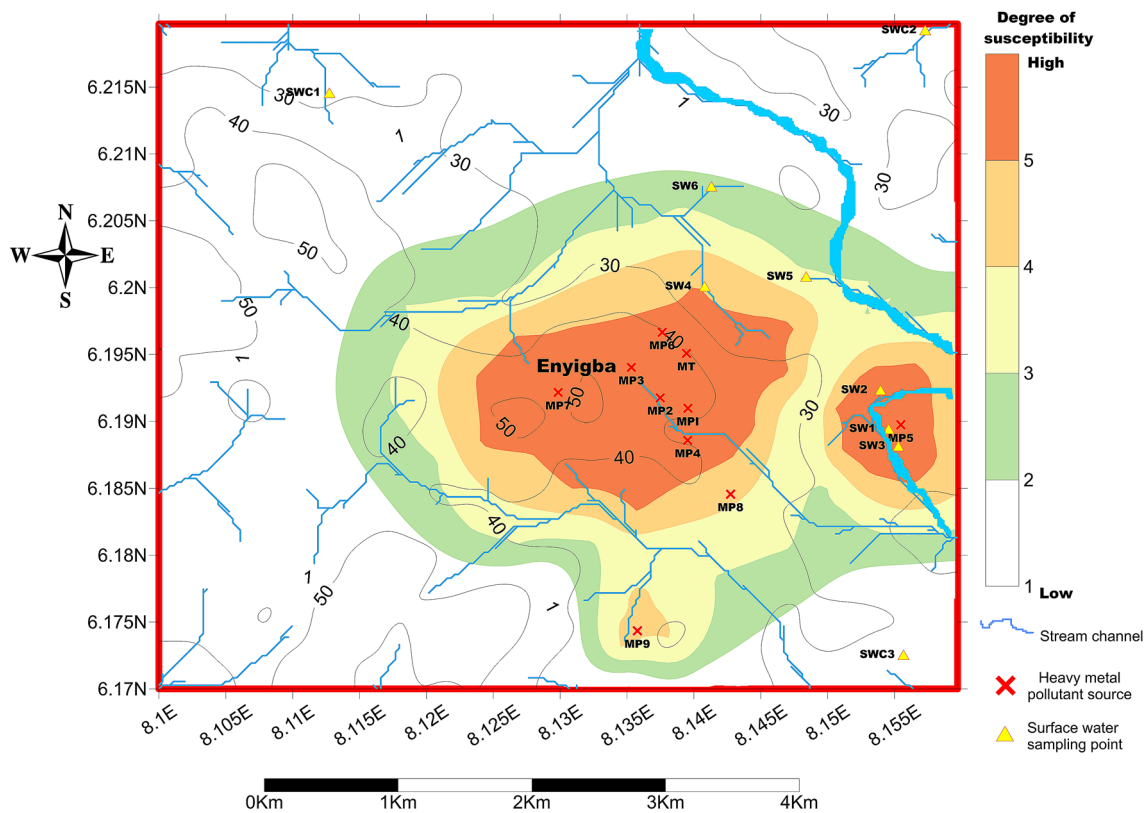
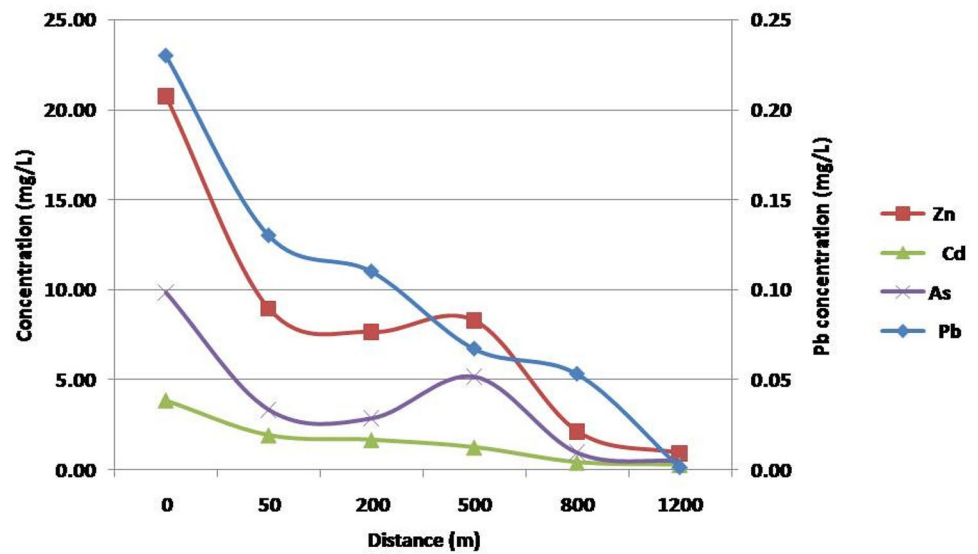
However, the concentration level of Pb and Zn in surface water samples that was collected beyond 800 m radius from the mine site are within the WHO permissible limit for drinking water, whereas those of Cd and As in surface waters within same radius are beyond WHO permissible limit. This observed variation could be as a result of difference in the mobility of the PTEs. Noting that the EMI of the PTEs in surface waters revealed that the sequence of mobility of the PTEs in the surface water was  $Cd > As > Zn > Pb$  (Table 6). The implication of this result is that surface water samples within 1200 m radius from the mine sites are not

**Table 6** Summary statistics of pH, EC, TDS, TEMP., and PTEs concentration in the surface water within and beyond the mine site catchment area

Parameter	Mine catchment					Control area					p value	EMI	WHO limit
	N/O	Mean	SD	Max	Min	N/O	Mean	SD	Max	Min			
Pb (mg/L)	6	0.10	0.08	0.23	0.001	3	0.001	0.002	0.003	0.000	0.03	0.004	0.01
Zn (mg/L)	6	8.09	7.04	20.72	0.897	3	0.387	0.392	0.800	0.020	0.04	0.043	5.00
Cd (mg/L)	6	1.54	1.30	3.83	0.260	3	0.011	0.012	0.023	0.000	0.03	0.068	0.003
As (mg/L)	6	3.77	3.41	9.81	0.487	3	0.012	0.014	0.027	0.000	0.04	0.050	0.01
pH	6	5.98	0.46	6.60	5.300	3	6.233	0.115	6.300	6.100	0.25		6.5–8.5
TDS (mg/L)	6	237.67	62.05	340.00	160.00	3	202.00	25.53	230.00	180.00	0.26		500
EC (µS/cm)	6	506.00	168.38	782.00	320.00	3	342.00	61.58	410.00	290.00	0.07		
TEMP (°C)	6	28.17	1.47	31.00	27.00	3	29.00	1.00	30.00	28.00	0.36		

SD standard deviation, EMI environmental mobility index, N/O number of observation, TDS total dissolved solute

**Fig. 8** Variation in heavy metal concentration in surface water with distance from mine site



**Fig. 9** Surface water potential to heavy metal susceptibility map

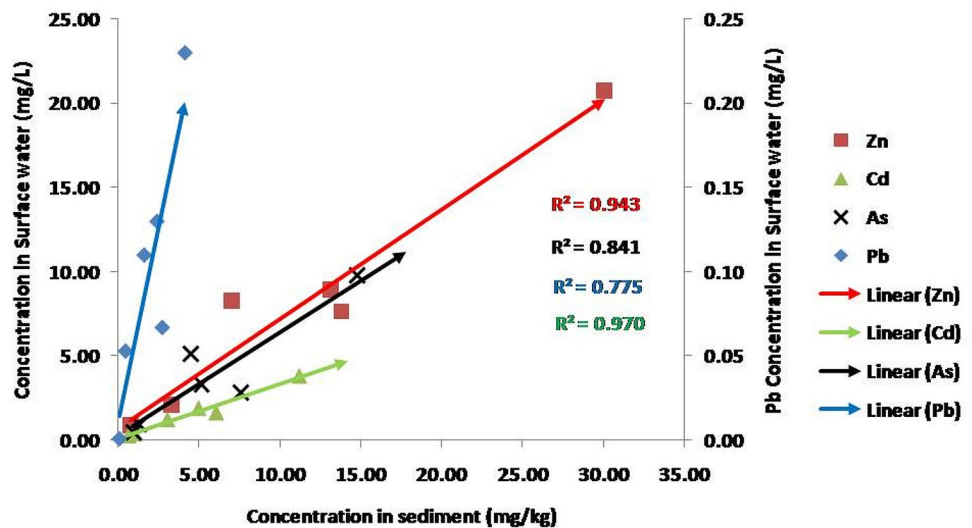
suitable for direct consumption without prior treatment due to the level of heavy metal toxicity. The toxicity of Pb and Zn in surface waters beyond this radius might not be harmful for direct consumption, but the concentration level of Cd and As in them renders it unsuitable for direct consumption since they are beyond WHO’s permissible limit. Thus Cd and As

toxicity in the surface water are more likely than Pb and Zn toxicity within the study area.

The graph of the linear regression analysis (Fig. 10) revealed that there was a good correlation between the concentration level of PTEs in sediments to those of the corresponding surface waters, with  $R^2$  values of 0.94, 0.84,



**Fig. 10** Relationship between concentration of heavy metals in sediments and in surface water



0.97, and 0.78 for Zn, As, Cd, and Pb, respectively. However, the concentration levels of the PTEs were higher in sediments than in the surface waters (Tables 5, 6). The reason for this difference could be because sediments serve as a sink for PTEs (Welch et al. 1988; Rowan et al. 1995); hence their higher concentration level in sediments than in surface water. Though the concentration levels of PTEs are relatively higher in sediments than in surface waters, their concentration level in sediments are within the permissible limits for agricultural soils (BGS 2005; NYC DEC 2006), whereas those of surface waters are beyond permissible limits for drinking water (WHO 2011). This result implies that the sediments along the stream channels within 1200 m radius from the mine sites might be suitable for agricultural crop production, whereas the surface waters within and beyond this radius are not suitable for consumption. Nevertheless, frequent environmental evaluation is recommended for the use of such sediments in crop production since bioaccumulation of PTEs in plant tissues could be a potential health risk

for humans (Qu et al. 2012; Ashe 2012). These results agree with similar studies by Asiam (1996) and Kumi (2007) on the effect of proximity of mine sites to PTEs concentration in sediments and surface waters.

**Potential toxic elements concentration in groundwater**

The groundwater samples that were collected within the mine catchment area were slightly acidic with average pH value of 6.6, which ranges from 6.1 to 6.8 (Table 7). The *T* test statistical analysis revealed that there was no significant variation between the pH of the samples collected within and beyond the mine catchment area. The average TDS and EC of the groundwater were approximately 1.65 mg/L and 276 μS/cm, respectively. Similar to that of the pH, there is also no significant lateral variation in the TDS and EC of the groundwater samples. This could imply that proximity to mine site does not have a significant influence on the pH,

**Table 7** Summary statistics of pH, EC, TDS, TEMP., and PTEs concentration in the groundwater within and beyond the mine site catchment area

Parameter	Mine catchment area					Control area					<i>p</i> value	WHO limit
	N/O	Mean	SD	Max	Min	N/O	Mean	SD	Max	Min		
Pb (mg/L)	6	0.02	0.01	0.03	0.002	3	0.001	0.001	0.002	0.00	0.13	0.01
Zn (mg/L)	6	1.75	1.27	3.17	0.47	3	0.13	0.18	0.26	0.00	0.09	5
Cd (mg/L)	6	0.40	0.43	0.94	0.04	3	0.00	0.00	0.00	0.00	0.16	0.003
As (mg/L)	6	0.37	0.41	0.82	0.00	3	0.00	0.00	0.00	0.00	0.17	0.01
pH	6	6.58	0.32	6.80	6.10	3	6.30	0.00	6.30	6.30	0.18	6.5–8.5
TDS (mg/L)	6	165.25	40.38	210.00	120.00	3	179.00	86.27	240.00	118.00	0.87	500
EC (μS/cm)	6	275.50	85.25	386.00	198.00	3	262.00	67.88	310.00	214.00	0.85	
TEMP (°C)	6	29.50	1.73	31.00	28.00	3	31.50	0.71	32.00	31.00	0.12	

*SD* standard deviation, *N/O* number of observation, *TDS* total dissolved solute

TDS, and EC of the groundwater samples. The reason for this could be that the groundwater level is deep; hence surface contaminants from the mine cannot affect its pH, TDS, and EC. Though the groundwater depth was not determined because the wells were not drilled for the purpose of this investigation, but rather an existing well which serves as a source of drinking water for the community; thus the depth cannot be accessed.

The average concentration level (mg/L) of the PTEs in the groundwater samples that were collected within the mine catchment area were 0.002, 1.74, 0.01, and 0.006 for Pb, Zn, Cd, and As, respectively (Table 7). Figure 11 revealed that the proximity of wells to the mine site resulted to increase in the concentration level of Zn and As. The concentration level of Cd fluctuates with proximity to the mine site, whereas the concentration level of Pb was relatively constant with proximity to the mine site. These lateral variations in the concentration level of PTEs in groundwater samples within the study area were revealed to be statistically insignificant. Thus, it implies that proximity to mine site does not have a significant contribution to the concentration levels of PTEs in groundwater within the study area. The reason for this insignificant variation could be as a result of the depth of the water table which is relatively deep (38 m) (Omonona et al. 2019); hence heavy metal contaminants from surface runoffs could not easily infiltrate into the groundwater. Therefore, the presence of the detected PTEs in the groundwater could have resulted due to geogenic factors, noting that PTEs are naturally occurring in rocks (Elueze et al. 2007; Jia et al. 2018). Meanwhile, among the PTEs which were detected in the groundwater samples that were collected within the mine catchment area, the average concentration levels of Pb, Zn, and As were within the WHO permissible limit for drinking water, whereas that of Cd was beyond the limit. However, in the control area, the concentration levels of the PTEs in the groundwater samples were all within the WHO permissible

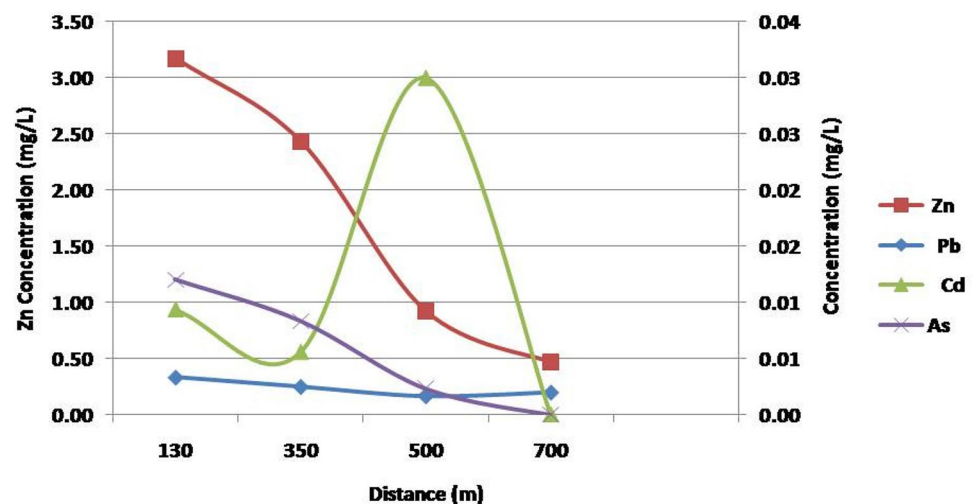
limit. These results are similar to what was observed by Gajowiec and Witkowski (1993), Camacho et al (2011) and Igwe et al. (2015), in similar mining area. They advised that while the concentration levels of most PTEs in groundwater are within WHO's permissible limits, there is a tendency of future enrichment. Thus groundwater within 700 m radius from the mine sites is a potential health risk for the immediate population due to PTEs (especially Cd) contamination, whereas those beyond 700 m radius are relatively safe from PTEs contamination.

## Conclusion

The results from this research have revealed that mining activities within Enyigba community contributed significantly to the concentration level of lead, zinc, cadmium, and arsenic in the environment. However, their degree of concentration in the environment depends on the proximity to the mine site. The spatial distribution of PTEs contaminants from the mine site in soil environment and in cassava plants was found to be restricted within 500 m radius. This is because their concentration level in the soil and plant tissues beyond this radius are normal with the background values. Thus plants which are grown within 500 m radius from the mine sites are more susceptible to heavy metal contamination than those grown beyond this radius. However Cd, As, and Zn were found to be more bioavailable than Pb; hence plants grown within 500 m radius from the mine sites are more susceptible to Cd, As, and Zn contamination than Pb.

Surface water within and beyond 1200 m radius from the mine sites was found to be unsafe for direct consumption without prior treatment. This is because the concentration level of PTEs in them are beyond WHO's permissible limit for drinking water. Though these metals are all beyond WHO's permissible limit for drinking water, the mobility

**Fig. 11** Variation in heavy metal concentration in groundwater with distance from mine site



of Pb and Zn in surface water was lower than those of Cd, and As. Thus Cd and As toxicity as a result of surface water consumption are more likely than Pb and Zn toxicity within the study area.

Heavy metal concentrations in sediments along the stream channels are relatively higher than those in the corresponding surface water. However their concentration levels in sediments within and beyond the mine catchment area are within BGS permissible limits for agricultural soils, thus may be suitable for agricultural purposes. Meanwhile, due to bioaccumulation and biomagnifications ability of PTEs in plant tissues over time, crops grown on these sediments may be a potential health risk to consumers.

Proximity to mine site was not found to have a significant influence on the concentration level of PTEs in groundwater within the study area. Pb, Zn, and As concentration in groundwater are within WHO's permissible limit for drinking water, whereas that of Cd is beyond. Therefore, Cd toxicity is the groundwater within the study area is more likely than Pb, Zn, and As toxicity. Thus treatment of the groundwater in order to reduce the Cd concentration is required prior to consumption.

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