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Potentially Toxic Elements (PTEs) in Surface Soils of Gemstones and Lead Mining Communities: Differential Mineral Phases and Exposure Risks

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Abstract

Proper human health risk assessments are rarely conducted for mining communities, consequently when there is need for remediation to be conducted and compensation to be paid to impacted population they are typically done, assuming equal exposure risks from varying mining activities. This study was conducted to compare the distribution of potentially toxic elements in surfaces soils of gemstone and lead mining towns in Nigeria, and their anticipated exposure risks. To achieve this thirty-two surface soil samples were collected from Eggon, a gemstone mining town in Nasarawa State and Enyigba, a lead mining town in Ebonyi State. The samples were air dried, disaggregated and sieved through a 2000 µm mesh with the <2000 fraction retained for aqua regia digestion. Potentially toxic elements bound in the samples were quantified with Agilent Technologies 4210 (MP-AES). Mineral phases were identified and quantified using using a inXitu's portable transmission XRD/XRF instrument (Terra) with a miniature X-ray tube and a CCD detector for collection both XRD signatures. The PTEs, As, Cd, Mn, Pb and Zn were higher in surface soils of lead mining town. At the gemstone mining town most the PTEs indicated similar concentrations at both mine site and residential areas, while at the lead mining town of the PTEs indicated higher concentrations at mine sites and mine road. At the lead mining town the estimated daily exposure doses for Pb were above threshold values irrespective of the exposure scenario. Mineralogy data indicated most of the PTEs in mineralized phases.

Keywords: Surface soils • Gemstones • Lead • Mining • Mineralogy • Exposure risk

Introduction

Several emerging and industrial cities in developing countries are suffering from the impact of old technologies associated unacceptable emissions and wastes. Due to emerging environmental advocacy groups, governments are under intense pressure to remediate impacted land areas and compensate local human population. The desire by responsive governments to alleviate pollution impact is limited by the lack data relating to pollution legacies different industrial outfits. This study aims to generate data related to impacts anticipated on communities exposed to gemstones and lead mining activities. Both industries are classed as mining but data related their ranking is not available.

Previous studies [1-4] employed several geostatistic, multivariate statistical analysis tools like ANOVA, cluster analysis, inter-metal correlations and others for the identification of both natural and anthropogenic activities that introduce potentially toxic metals in surface soils. In addition to the use of statistical tools, mineralization has also been employed to tracing sources of potentially toxic metals in soil and underground media [5]. In this study surface soil from residential setting, mining site and mine wastes were sampled from two mining towns, Enyigba in Ebonyi state and Eggon in Nasarawa state both in Nigeria were analyzed for acid digestible fraction of arsenic, cadmium, lead, manganese and zinc and their likely mineralization.

Study area

Enyigba is located in south eastern Nigeria bordered by Abakiliki and Ikwo in the north and south, respectively (Figure 1). The economy of the town has been dominated by artisan mining for Pb and Zn, and farming. The population of Enyigba was estimated to be around 8,000 and that of Eggon 110, 613 based on the 2006 census figure [6,7]. Eggon is located in northern Nigeria bordered by Akwanga and Lafia in the north and south, respectively (Figure 1). The economy of Eggon has been dominated by gemstone mining and farming. The major ore deposits at Enyigba are suggested to be Pb-Zn minerals, while deposits at Eggon are quartz, mica and granite [8]. There is a potential of both locations in the future becoming major mining town with associated metalliferous wastes, since Nigeria is desperately working to diversify its economy due to dwindling foreign exchange from crude oil export.

Materials and Methods

Sample collection

A total of 32 surface soil and mine waste samples were collected from Eggon in Nasarawa State and Enyigba in Ebonyi State with a stainless steel trowel. Sampling focused upon mine sites, mine roads, streets and residential areas in both towns. Each sample consisted of 3 closely spaced subsamples, which were bulked to produce composite. In the laboratory the samples were air dried, disaggregated and sieved through a 2000 µm mesh with the <2000 fraction retained for digestion.

Aqua regia digestion and analysis

Aqua regia digestions ($HCl:HNO_3$ in the ratio 3:1 v/v) were

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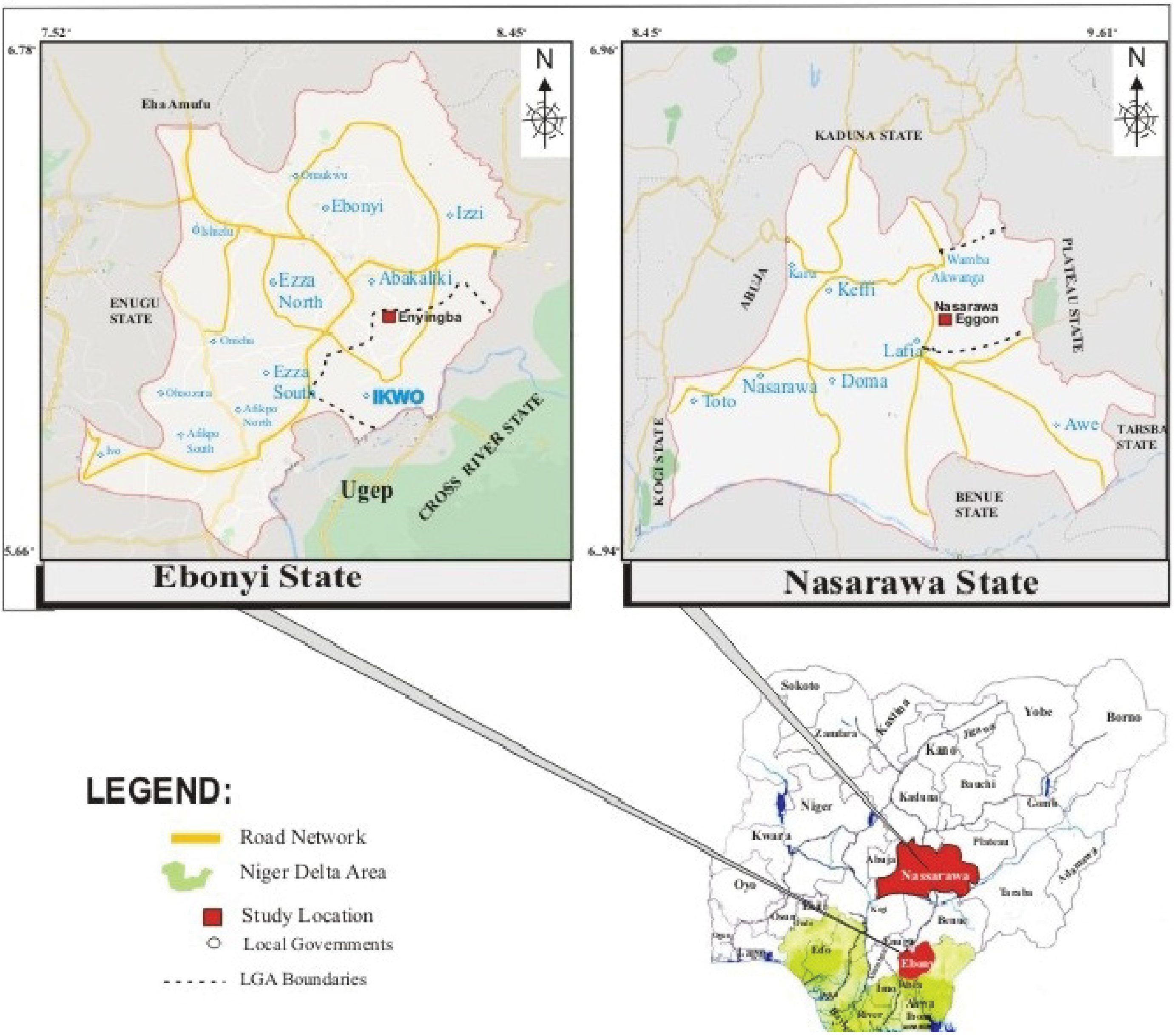

Figure 1. Map of the two study locations, Eggon and Enyigba in Nigeria.

performed on 0.5 g sub-sample in digestion tubes mounted on hot plate in fume cupboard. The filtrates obtained from the digestion were stored in the refrigerated prior to analysis with Agilent Technologies 4210 (MP-AES).

Soil ingestion exposure assessment at Eggon and Enyigba

It is possible to estimate human exposure risk associated with the ingestion of surface soils at the different study locations. PTEs daily intake (PTE DI) from ingestion of surface based on total concentrations of the different PTEs of interest in this study were obtained using the equation.

$$\text{PTE DI}=(\text{EC} \times \text{SIR} \times \text{ED})/1000\text{BW}$$

Where , EC is the concentration of PTEs (μg/g), SIR is the soil ingestion rate (mgd^{-1}) which for this study was set at 200 mgd^{-1} for 2-6 year child against that (100 mgd^{-1}) suggested by USEPA [9] because of dusty nature of tropical Africa. ED (unitless) is the exposure duration. ED was set at different values, 1 for residential [10]. In Nigeria artisan working mother that are nursing children below 6 year go along with such kids. Also most casual workers in developing countries work for at least 12 hrs per day. The absence in most cases of functional roads and lack motorized means of transport at rural setting in developing countries, suggest that workers with their children spend between 4-hours on roads per day. Consequently the ED for children at mine site and street/road were set at 0.5 and 0.17, respectively. BW is the body weight of the children between 2-6 years. The default BW for children of the age grade was set at 17.8 kg [9].

X-ray diffraction analysis

To identify possible origins of the potentially toxic elements in streets and residential areas a limited number of samples (n=15) were selected for mineralogy. Subsamples were prepared and scanned using a inXitu's portable transmission XRD/XRF instrument (Terra 575, USA) with CoKa (1.7903000 A) α radiation. The instrument is fitted with a miniature X-ray tube and a CCD detector for collection both XRD signatures. The scan was run from 5 to 55° (2-theta scale), with increments of 0.02° and a counting time of 1.0 second per step. The operating conditions were 40 kV and 40 mA. Peak identification was carried out with the Match, 2003-2019 CRYSTAL IMPACT, software package (Bonn, Germany).

Quality control

Analytical accuracy of aqua regia extractions was assessed against a reference material, BCR 143R (aqua regia certified sewage sludge amended soil) and the inclusion of blank digestion for every run. Acceptable results were obtained for the aqua regia extractable concentrations of PTEs compared to certified values.

Results and Discussion

Mineralogy

XRD data provide an indication as to the PTE-bearing mineral assemblage within surface soil at mining roads, mine site, residential areas and streets at both Eggon (Nasarawa state) and Enyigba (Ebonyi State) (Table 1). At Eggon the data suggest the presence of As, Cu, Mn, Pb and Sb bearing minerals like; Nordstromite Coronadite, Altaite, Cuprite and Antimony lead manganese oxide (Table 1). The other minerals at Eggon were principally gemstone materials. The list includes Quartz; a vitreous trigonal crystal system, Muscovite; a hydrated phyllosilicate commonly found in granite, Argentopyrite with monoclinic crystal system, and Brianroulstonite; with a trigonal crystal system. Published literature has not previously indicated existence of Brianroulstonite mineral in Nigeria. The mineral phases observed in Enyigba surface soils were, Arsenopyrite, Berlinite, Cadmium lead (IV) oxide, Manganese lead antimony selenide, Magnesite, Pyrite, Quartz, Ramsdellite, Wulfenite and Zincochromite (Table 1). Over 50% of the mineral consisted of PHEs like As, Cd, Cr, Mn, Pb, Sb and Zn (Table 1). The XRD results indicated As-bearing minerals in the form of Aluminium arsenide (4.8%) for Eggon in Nasarawa State and Arsenopyrite (7.3%) for Enyigba in Ebonyi State. This finding addresses the doubt expressed by section of environmental scientists and engineers that there are no natural sources of As in soil matrices in Nigeria. The % heavy metals separated for Nordstromite (86.6%) Coronadite (9.1-10.5%), Altaite (3.0%) at Eggon in Nasarawa State suggests the town may also be a viable site for Pb mining. The varying mineralized phases of PTEs of interest in this study obtained for the different towns also suggest that even when PTEs doses equate for the two towns risk may be consistent.

PTEs concentrations in surface soils at Eggon and Enyigba

The mean concentrations of As at mine roads, mine sites, residential areas and streets at Eggon in Nasarawa State of Nigeria were 14.352 mg/kg, 13.450 mg/kg, 56.075 mg/kg and 36.250 mg/kg, respectively (Figure 2). The means of As reported in this study are consistent with 40 mg/kg previously reported by Patrick, et al. [11] for the same town, though the employed XRF for their analysis. At Eggon mine roads indicated lowest mean concentration of As and residential areas indicated the highest mean concentration of As. The mean concentrations of As at mine roads, mine sites, residential areas and streets at Enyigba in Ebonyi State of Nigeria were 114.800 mg/kg, 143.041 mg/kg, 143.075 mg/kg and 111.825 mg/kg, respectively (Figure 2). Our results for as at Enyigba (111.835-143.075 mg/kg) (Figure 2) is about 20 times higher than the maximum (5.3 mg/kg) reported for the Enygba mine area soils [12]. However, since they did not indicate the sample treatment method employed in their paper that may explain the large variance. Consistent with our higher As concentration (Figure 2), previous workers have highlighted large scale As pollution of surface soils (35.27 mg/kg) [13] and surrounding lakes and rivers at Enyigba [14]. Unlike Eggon, at Enyigba streets indicated lowest mean concentration of As and mine sites indicated the highest mean concentration of As. The relatively lower concentrations obtained for Eggon may be due the fact this here they mine mainly for gemstone and mica [15] and this mineral are typically not associated minerals bearing significant amounts of PTEs.

Mean concentrations of 2.025 mg/kg, 1.500 mg/kg, 1.65 mg/kg and 1.825 were obtained for mine road, mine site, residential and street areas of the gemstone mining town. Relatively higher mean concentrations of 8.443 mg/kg, 13.371 mg/kg, 10.550 mg/kg and 11.100 were obtained for mine road, mine site, residential and street areas of the Pb-Zn mining town. The highest mean concentrations were for mine roads and mine sites, for Eggon and Enyigba towns, respectively. Enyigba consistently indicated higher concentrations irrespective of the exposure scenarios, this may be due the fact the mineralized phases for As (arsenopyrite) at Enyigba was higher than As (aluminium arsenide) for Eggon (Table 1).

Table 1. Minerals and associated PTEs at Eggon and Enyigba.

Town	% Range HM separated	Mineral phases	PTE
Eggon	9.7-12.2 (n=2)	Argentopyrite	-
	4.8 (n=1)	Aluminum arsenide	As
	86.6 (n=1)	Nordstromite	Cu, Pb
	76.5-93.6 (n=7)	Quartz	-
	11.0 (n=1)	Tridynite	-
	9.6 (n=1)	Muscovite	-
	15 (n=1)	Bianroulstonite	-
	9.1-10.5 (n=2)	Coronadite	Pb, Mn
	3.0 (n=1)	Altaite	Pb
	3.8 (n=1)	Cuprite	Cu
	2.9 (n=1)	Antimony lead manganese oxide	Mn, Pb, Sb
Enyigba	7.3 (n=1)	Arsenopyrite	As
	62.8-93.4 (n=8)	Quartz	-
	12.2-12.8 (n=2)	Pyrite	-
	13.5 (n=1)	Berlinite	-
	18.2 (n=1)	Ramsdellite	Mn
	13.5 (n=1)	Magnesite	-
	0.5 (n=1)	Cadmium lead (IV) oxide	Cd, Pb
	3.4 (n=1)	Wulfenite	Pb
	3.9 (n=1)	Manganese lead antimony selenide	Mn, Pb, Sb
	3.6 (n-1)	Magnetoplumbite	Pb
	2.0 (n=1)	Zincochromite	Cr, Zn

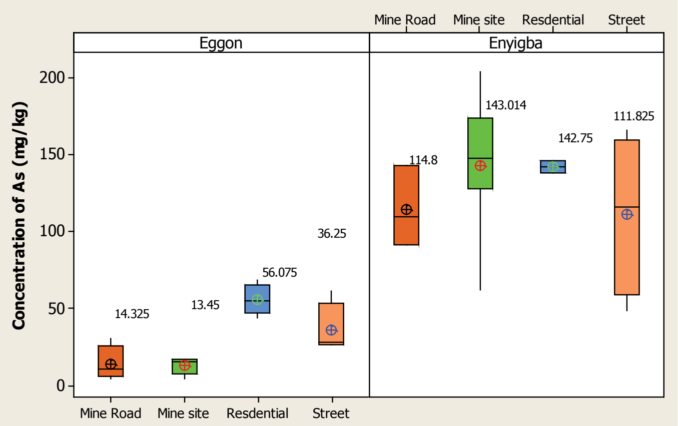

Figure 2. Distribution of As at Eggon and Enyigba.

The mean concentrations of Cd at mine roads, mine sites, residential areas and streets at Eggon in Nasarawa State of Nigeria were 2.025 mg/kg, 1.500 mg/kg, 1.650 mg/kg and 1.825 mg/kg, respectively (Figure 3). At Eggon the mine road indicated the highest concentrations and mine site indicated the lowest concentrations, suggesting for Eggon that the mine site is not a major contributor of Cd to the surrounding environment. This argument is consistent with mineralogy data (Table 1) that did not indicate the presence of any Cd mineralized phase. The mean concentrations of Cd at mine roads, mine sites, residential areas and streets at Enyigba in Ebonyi State of Nigeria were 8.433 mg/kg, 13.371 mg/kg, 10.550 mg/kg and 11.100 mg/kg, respectively (Figure 3). Irrespective of the exposure scenarios this location indicated at least four time higher concentrations than Eggon in Nasarawa State. At Enyigba the highest mean concentration for Cd (13.371 mg/kg) was recorded for the mine site suggesting that mine activities may have also been a major source of Cd in the locality. The suggestion is supported by the presence of a Cd mineral at Enyigba (Table 1). The elevated concentrations of Cd recorded at Enyigba is not unexpected taking into consideration that this location indicated Cd mineralized phase (Cadmium lead (IV) oxide) (Table 1). The range of means for different exposure scenarios (8.433-13.371 mg/kg) indicated in this study for Enyigba is relatively above the range (0.310 -11.190 mg/kg) previously reported by Chukwuma [16] and consistent with the range (7.00-15.00 mg/kg) by Obiora [12]. The differences in the concentration of Cd between this study and the one by Chukwuma [16] study suggest that over time enhancement may have occurred, since his study was conducted in 1993.

The mean concentrations of Cd at mine roads, mine sites, residential areas and streets at Eggon in Nasarawa State of Nigeria were 503.093 mg/kg, 339.625 mg/kg, 584.025 mg/kg and 628.925 mg/kg, respectively (Figure 4). At Eggon the streets indicated the highest concentrations and mine site indicated the lowest concentrations, suggesting for Eggon that the mine site is not a major contributor of Mn to the surrounding environment.. The mean concentrations of Mn at mine roads, mine sites, residential areas and streets at Enyigba in Ebonyi State of Nigeria were 1832.730 mg/kg, 3375.010 mg/kg, 291.250 mg/kg and 1877.300 mg/kg, respectively (Figure 4). At Enyigba means recorded for mine road and streets are consistent with a range reported in 2016 by Obiora [12]. At Enyigba mine site

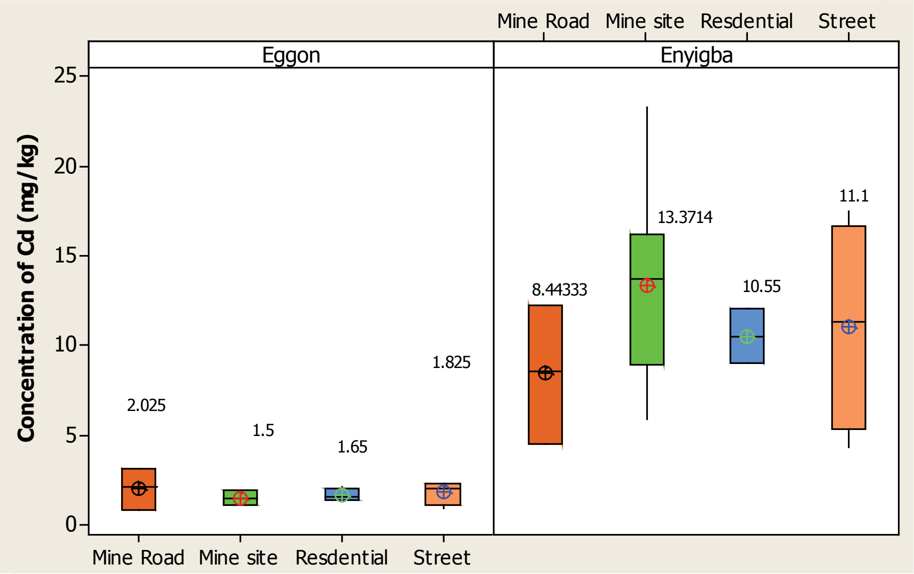

Figure 3. Distribution of Cd at Eggon and Enyigba.

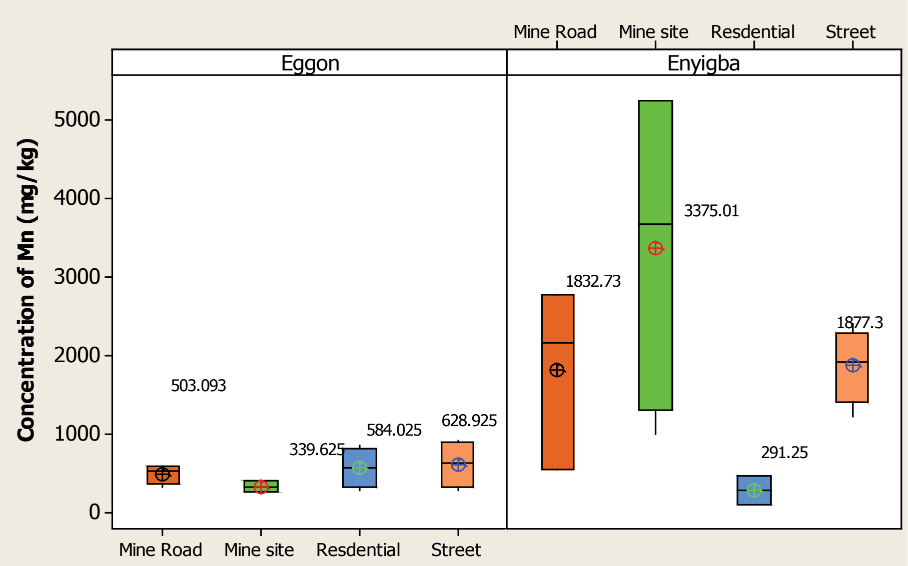

Figure 4. Distribution of Mn at Eggon and Enyigba.

surface soils indicated the highest concentration of Mn compared to other exposure scenarios. This implies that the mine may be significant source of Mn in the town. Excluding the residential areas at both towns, irrespective of the exposure scenarios Enyigba indicated at least three time higher concentrations than Eggon in Nasarawa State for Mn.

Data (Figure 5) indicate widespread occurrence of elevated concentrations of Zn at both Eggon and Enyigba irrespective of the exposure scenario. At Eggon the highest and lowest concentrations were recorded for mine roads and streets (Figure 5), respectively. At Enyigba the lowest mean (750 mg/kg) was recorded for the residential areas and the highest (8915.20 mg/kg) for streets. The range of means obtained at Enyigba (750.000-8915.200 mg/kg) is relatively above the range (265.300-4073.300 mg/kg) reported three years earlier by Obiora [12]. The lower mean concentrations recorded for Eggon town may be due the fact that presently mining is focused on gemstones and not lead, meaning in spite of Pb mineralization at the town the Pb may not have been excavated to the surface.

Results (Figure 6) indicate widespread occurrence of elevated concentrations of Pb at both Eggon and Enyigba irrespective of the exposure scenario. At Eggon the highest and lowest concentrations were recorded for mine roads and streets (Figure 6), respectively. At Enyigba the lowest mean (148.750 mg/kg) was recorded for the residential areas and the highest (612.857 mg/kg) for streets. The range of means obtained at Enyigba (750.000-8915.200 mg/kg) is relatively above the range (117.000-214.000 mg/kg) reported three years earlier by Obiora [12]. The lower mean concentrations recorded for Eggon town may be due the fact that no Zn mineral was identified for Eggon (Table 1), compared to a Zn mineral (zincochromite) recorded for Enyigba town.

Soil ingestion exposure assessment in Eggon and Enyigba and implications

We sought to quantify exposure to children in the age range of 2-6 years, given their hand-to-mouth habit and their susceptibility toxic substances [16]. Calculations were made using mean concentrations of PTEs in samples for the different exposure scenarios, mining roads, mine sites, residential areas and streets at gemstone mining town (Eggon) and lead mining town (Enyigba).

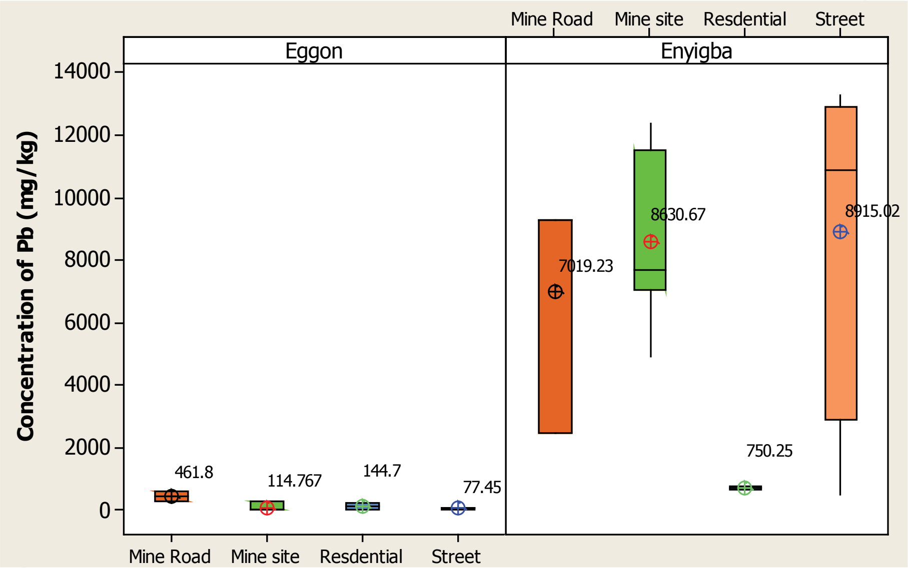

Figure 5. Distribution of Pb at Eggon and Enyigba.

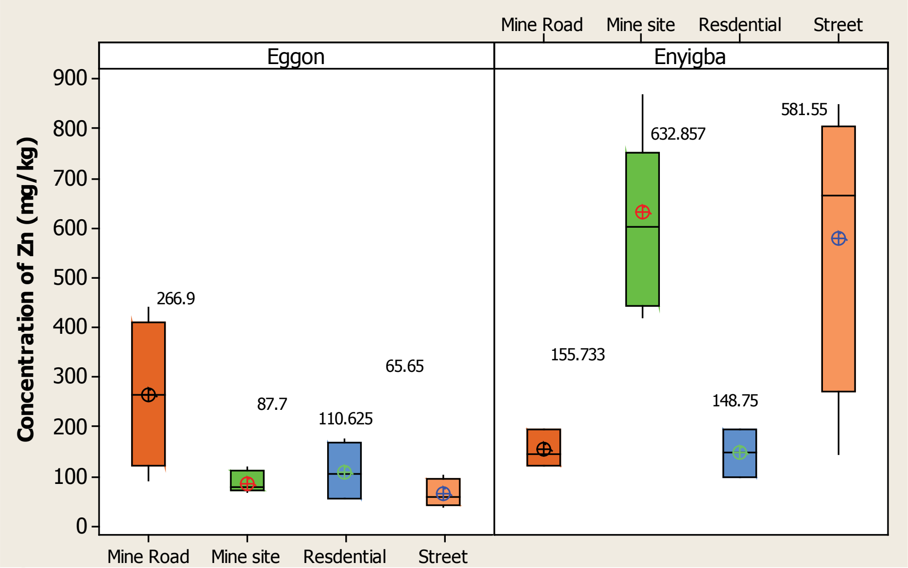

Figure 6. Distribution of Zn at Eggon and Enyigba.

The data (Figures 7a and 7b) suggest that on average exposure dose for lead mining town (Enyigba) and gemstone mining town (Eggon) indicated 0.217 μg As Kg^{-1} BW d^{-1} and 0.027 μg As Kg^{-1} BW d^{-1}, 0.807 μg As Kg^{-1} BW d^{-1} and 0.077 μg As Kg^{-1} BW d^{-1}, 1.605 μg As Kg^{-1} BW d^{-1} and 0.630 μg As Kg^{-1} BW d^{-1}, and 0.215 μg As Kg^{-1} BW d^{-1} and 0.070 μg As Kg^{-1} BW d^{-1}, for mine roads, mine sites, residential and streets, respectively. The exposure doses for lead mining town (Enyigba) were at least two time the doses for the gemstone mining town (Eggon) irrespective of the exposure scenarios and may be due the presence of the different minerals and concentrations (aluminium arsenide 4.8% for Eggon and arsenopyrite 7.4% for Enyigba) (Table 1) identified at the lead mining town. Calculated doses for Cd at mine site and residential areas at Enyigba and residential areas for Eggon were above recommended tolerable daily intake (0.3 μg As Kg^{-1} BW d^{-1}) (Figures 7a and 7b).

The mean exposure doses for Enyigba and Eggon indicated 0.017 μg Cd Kg^{-1} BW d^{-1} and 0.05 μg Cd Kg^{-1} BW d^{-1}, 0.074 μg Cd Kg^{-1} BW d^{-1} and 0.005 μg Cd Kg^{-1} BW d^{-1}, 0.012 and 0.020 μg Cd Kg^{-1} BW d^{-1}, and 0.022 μg Cd Kg^{-1} BW d^{-1} and 0 μg Cd Kg^{-1} BW d^{-1}, for mine roads, mine sites, residential and streets, respectively (Figures 8a and 8b). At both towns to sites indicated doses above the recommended Tolerable daily intake (0.36 μg Cd Kg^{-1} BW d^{-1}) (Figures 8a and 8b). The lead mining town Enyigba indicated more dosage, this may be due the Cd mineral (Table 1) found there.

The mean exposure doses for Enyigba and Eggon indicated 13.406 μg Pb Kg^{-1} BW d^{-1} and 0.882 μg Pb Kg^{-1} BW d^{-1}, 48.483 μg Pb Kg^{-1} BW d^{-1} and μg Pb Kg^{-1} BW d^{-1} 0.54 μg Pb Kg^{-1} BW d^{-1}, 8.43 μg Pb Kg^{-1} BW d^{-1} and 1.625 μg Pb Kg^{-1} BW d^{-1}, and 17.027 μg Pb Kg^{-1} BW d^{-1} and 0.15 μg Pb Kg^{-1} BW d^{-1}, for mining roads, mine sites, residential areas and streets, respectively (Figures 9a and 9b). All exposure scenarios at Enyigba (lead mining town) indicated doses above recommended (3.6 μg Pb Kg^{-1} BW d^{-1}) (Figure 9a). The exposure doses are consistently lower for the gemstone mining town (Eggon) irrespective of the exposure scenario. It is important however, to note the higher doses may not always equate to higher risk/toxicity because of large variance in mineralogy (Table 1) the two mining town. It has previously been highlighted by Boisa, et al. [10] mineralogy has the

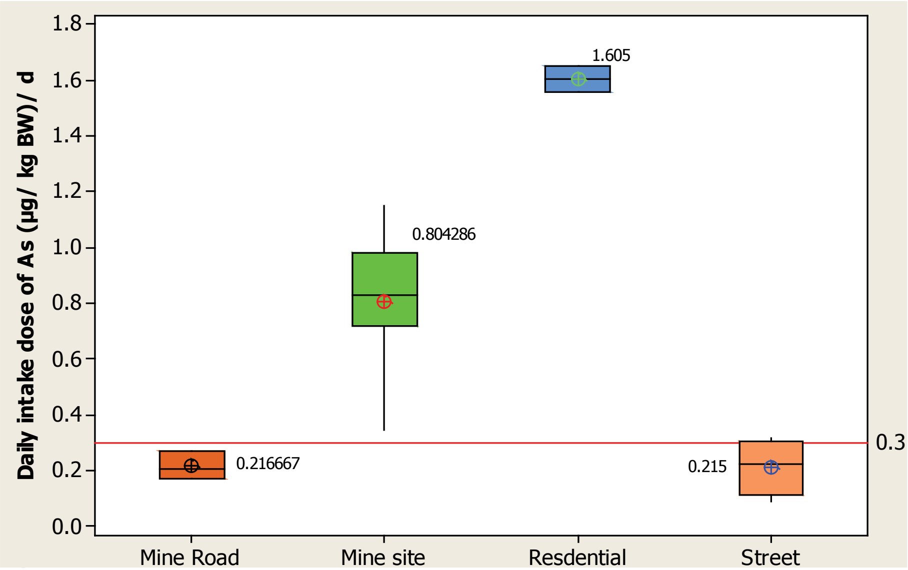

Figure 7a. Soil ingestion dose for As at Enyigba.

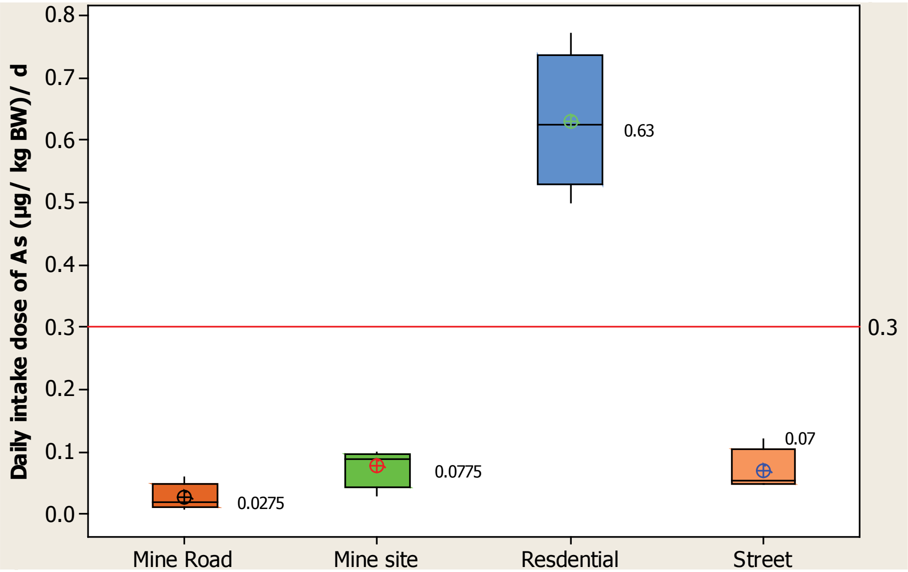

Figure 7b. Soil ingestion dose for As at Eggon.

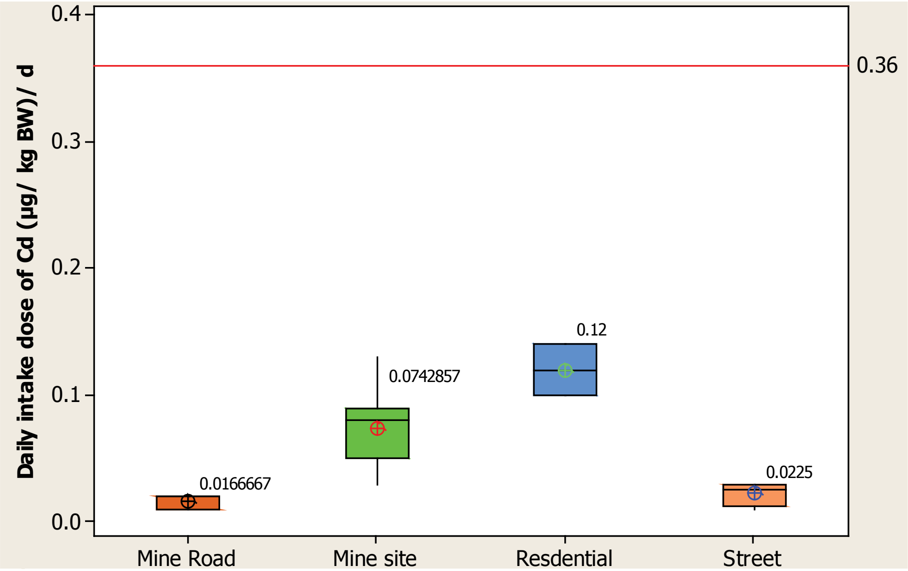

Figure 8a: Soil ingestion dose for Cd at Enyigba.

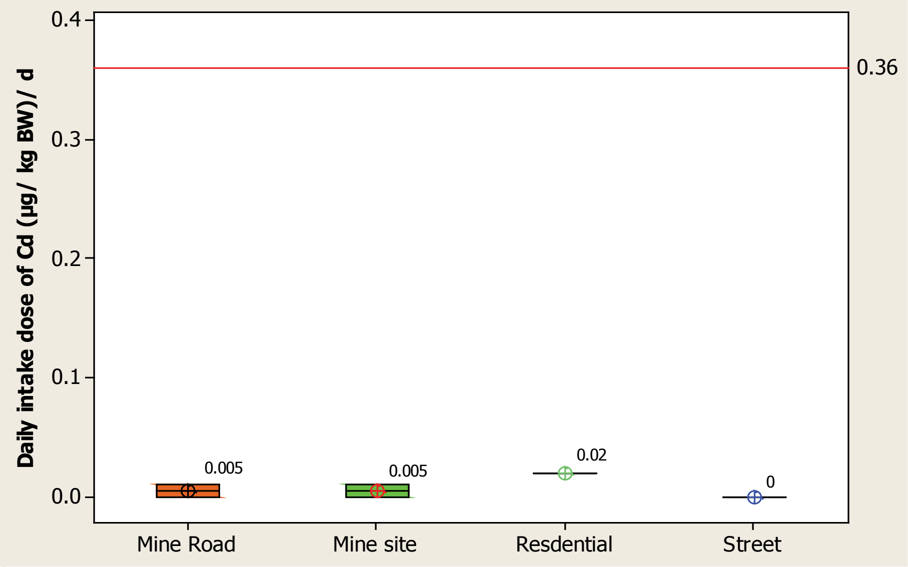

Figure 8b: Soil ingestion dose for Cd at Eggon.

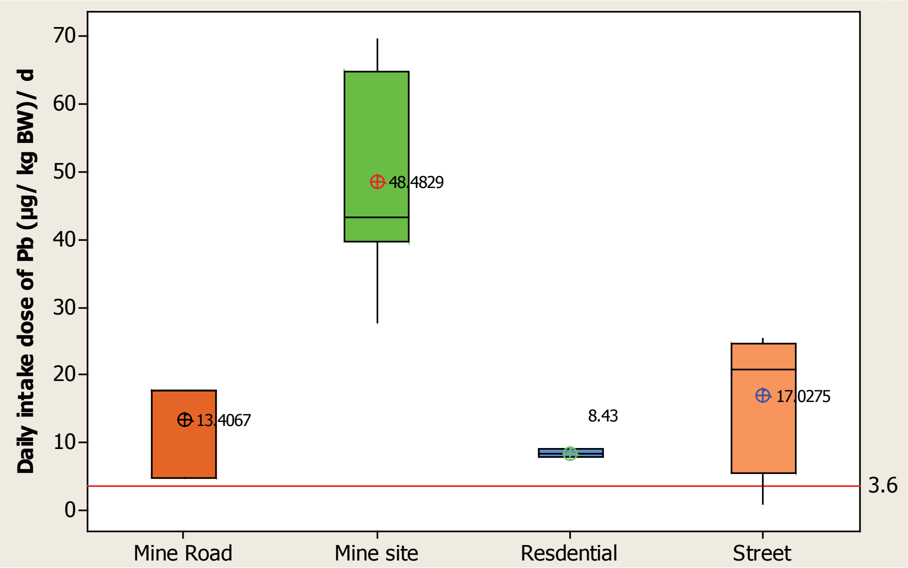

Figure 9a: Soil ingestion dose for Pb at Enyigba.

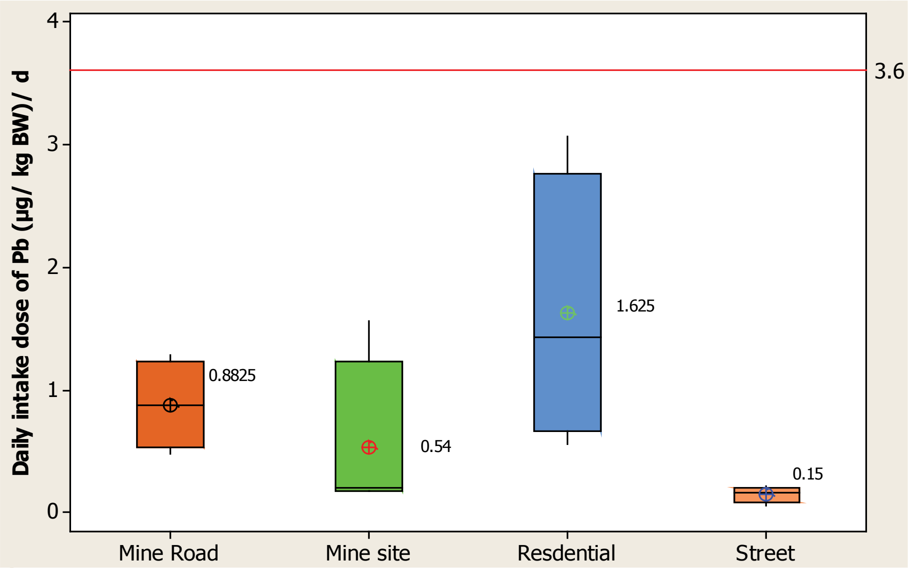

Figure 9b: Soil ingestion dose for Pb at Eggon.

capacity to influence bioaccessibility (a factor that influences uptake in humans) of PTEs in different matrices.

Conclusion

This study was conducted to compare the distribution of potentially toxic elements in surfaces soils of gemstone and lead mining towns in Nigeria, and their associated exposure risks. Thirty-two surface soil samples were collected from Eggon, a gemstone mining town in Nasarawa State and Enyigba, a lead mining town in Ebonyi State. At Eggon Nordstromite Coronadite, Altaite, Cuprite and Antimony lead manganese oxide mineral phases where identified. Gemstone mineral found at Eggon were Muscovite; a hydrated phyllosilicate commonly found in granite, Argentopyrite with monoclinic crystal system, and Brianroulstonite; with a trigonal crystal system. Published literature has not previously indicated existence of Brianroulstonite mineral in Nigeria. The mineral phases observed in Enyigba surface soils were, Arsenopyrite, Berlinite, Cadmium lead (IV) oxide, Manganese lead antimony selenide, Magnesite, Pyrite, Quartz, Ramsdellite, Wulfenite and Zincochromite.

Both mining towns indicated varying concentrations of As, Cd, Mn, Pb and Zn for the different exposure scenarios (residential, mine road, streets and mine site), with the lead mining town, Enyigba indicating higher concentrations than the gemstone mining town, Eggon. Consequently the different mining towns indicated varying potential exposure doses for the PTEs studied. As indicated estimated daily exposure doses above existing threshold value, (0.3 µg Cd Kg^{-1} BW d^{-1}) at both gemstone and lead mining communities at residential areas, suggesting the likelihood As poisoning to human population. Pb indicated estimated daily exposure doses above existing threshold value, (3.6 µg Cd Kg^{-1} BW d^{-1}) at both and lead mining community alone irrespective of the exposure scenario, suggesting the likelihood Pb poisoning to human population.

The results of this study have highlighted the need to conduct risk assessment for individual mining towns, since they may indicate varying concentrations for PTEs and also different mineral phases.

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