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Heavy metal concentrations in soils and plants in the vicinity of Arufu lead-zinc mine, Middle Benue Trough, Nigeria

Nganje T.N.[1*], Adamu C.I.[1], and Ukpong E.E.[2]
[1] *Department of Geology, University of Calabar, P.M.B. 1115, Calabar*
[2] *Formerly Department of Geology, University of Calabar P.M.B. 1115, Calabar*
* *Corresponding author, E-mail: theresenganje@yahoo.com*

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Abstract This study focused on the influence of base metal mining on heavy metal levels in soils and plants in the vicinity of Arufu lead-zinc mine, Nigeria. Soil samples (0–15 cm depth) and plant samples were collected from cultivated farmlands in and around the mine, the unmineralized site and a nearby forest (the control site). The samples were analyzed for heavy metals (Fe, Zn, Mn, Cu, Pb, Cr and Cd) by Atomic Absorption Spectrophotometry (AAS). The physical properties of soils (pH and LOI) were also measured. Results showed that soils from cultivated farmlands have neutral pH values (6.5–7.5), and low organic matter contents (<10%). Levels of Zn, Pb and Cd in cultivated soils were higher than the concentrations obtained from the control site. These heavy metals are most probably sourced from mining and agricultural activities in the study area. Heavy metal concentrations measured in plant parts decreased in the order of rice leaves>cassava tubers>peelings. In the same plant species, metal levels decreased in the order of Zn>Fe>Mn>Cu>Pb>Cr>Cd. Most heavy metals were found in plant parts at average concentrations normally observed in plants grown in uncontaminated soil, however, elevated concentrations of Pb and Cd were found in a few cassava samples close to the mine dump. A stepwise linear regression analysis identified soil metal contents, pH and LOI as some of the factors influencing soil-plant metal uptake.

Key words heavy metal; soil; plant; lead-zinc mine; Benue Trough; Nigeria

1 Introduction

Metal contaminations in soils have become a worldwide concern since rice paddy fields irrigated with waste from a zinc mine caused excessive cadmium (Cd) intake which caused adverse health effects on farmers who had consumed the rice grown on the contaminated soil (Kobayashi, 1978). This first observation of human disease caused by heavy metals in the general environment has stimulated research on the potential adverse effects of heavy metals in soils and in agricultural and dietary systems.

Base metal mining can be an important source of heavy metals in the environment owing to various mining activities including processing and transportation of ores, disposal of tailings and waste water around mines (Chon et al., 1997; Lee et al., 2001). The elevated levels of heavy metals discharged from mine wastes can be found in nearby streams, agricultural soils and food crops, and eventually, they may impose a potential health risk on residents in the vicinity of mines. Heavy metal uptake by plants grown on polluted soils has been studied to a considerable extent (Jung and Thornton, 1996; Jung and Chon, 1998; Jung et al., 2002; Yung et al., 2004; Lee et al., 2001; Salami et al., 2006). All findings have shown that elevated levels of metals in soils may lead to increased uptake by plants. However, there is generally no strong relationship between the concentrations in soils and those in plants because it depends on many different factors, such as soil metal bioavailability, plant type, age and parts (Xian, 1989; Kabata-Pendias and Pendias, 2002; Chukwuma, 1995; Chon et al., 1997; Jung et al., 2002). So even the concentrations of heavy metals in plants grown in various unpolluted environments show quite large variations (Sauve et al., 1997; Dudka et al., 1996; Alloway; 1990).

In Nigeria, base metal mines producing Pb-Zn were distributed within the Benue Trough and had been actively operated until the early 1970s. Since then, however, base metal production has declined and most of the mines were abandoned mainly due to economic reasons and exhaustion. Upon the abandoning of the mines, improper disposal of mineral waste piles and untreated mine drainage have become the most important sources of heavy metals in nearby environments. Previous works have reported the elevated levels of heavy metals from soils, water and sediments as well as food crops from metaliferrous mines within the trough (Adiku-Brown and Ogezi, 1991; Chukwuma, 1995).

The current study is to investigate the relationships between heavy metal contents of some food crops and those in agricultural soils in the Arufu Pb-Zn mining district within the Middle Benue Trough, Nigeria.

2 Description of the study area

The Arufu mining district of the Middle Benue Trough is located between longitudes 9º10′ and 9º20′E and latitudes 7º40′ and 7º45′N (Fig. 1). The area which is undulating lies roughly between 200–300 m above sea level. Laterite scarps which have resulted from prolonged dissection of former laterite sheets are dominant features in the area. Many small seasonal streams including the Pii, Kutaji, Kiri and Ubaver rivers, which are all tributaries of the Benue River, control the drainage in the area. The streams are structurally controlled and generally join together in a dendritic drainage pattern.

The area is characterized by a tropical wet-dry climate and its rainfall is generally moderate, about 100–120 cm per annum. The temperature ranges from 30–5℃ and the relative humidity is in the order of 30%–60% (Ileoje, 1981). The vegetation in Arufu is of the savannah wood type, typified by tall grasses, lots of shrubs and a few tall trees ranging in height from 3–6 m. The bedrock in which the mineralization occurs is limestone with intercalations of shale and sandstone. The minerals are galena (PbS) and sphalerite (ZnS).

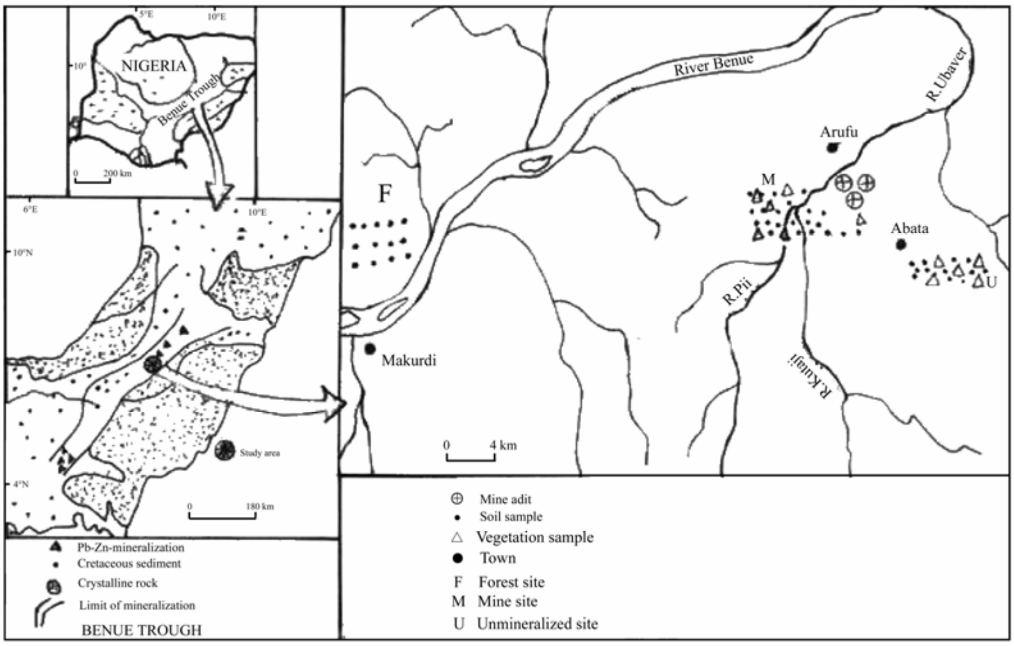

Fig.1. Sketch map showing the study area and sample localities.

3 Materials and methods

Surface soil samples (0–15 cm depth) were collected using a hand auger (2.5 cm in diameter) in cultivated farmlands near the mine dump and a rural unmineralized area (Abata) about 5 km south of the mine as well as in a government forest near Makurdi (capital of Benue State) to serve as control (Fig. 1).The forest is restricted from public use and assumed to have suffered little anthropogenic inputs. Each soil sample comprised a composite of 10 sub-samples taken on a 1×1 meter square. Random samples of plants were taken from the cultivated fields including rice leaves (*Orzya sativa*) and cassava (*Manihot esculenta*). Samples were placed in polythene bags and properly labelled.

Soil samples were dried in an air-circulating oven at 25℃, disaggregated and sieved to <2 mm. After quartering, the samples were ground to 80 mesh (<180 μm) in a stainless steel blender. Plant samples were thoroughly washed with deionized water and dried in an air-circulating oven at 25℃, ground to fine powder and repackaged in a sealed plastic bag. The cassava tubers were carefully peeled and rinsed with tap water to remove surface dirt before drying the tubers and peelings. After the preparation stage, 0.5 g of soil sample was digested in 4:1 ratio of concentrated nitric perchloric acids and the solution heated to dryness. The residue was leached with 5 mL of 2 M HCl (Alloway, 1990) and the solution made up to 10ml with deionized water in already calibrated and labelled test-tubes. The samples were analyzed for Fe, Zn, Mn, Cu, Pb, Cr and Cd using Bulk Scientific Atomic Absorption Spectrophotometer (AAS) model ZGP 210 (Gallemcent manufactured). 0.5 g of powdered plant sample was digested in fuming HNO_3 , followed by $Mg(NO_3)_2$, leached with the same procedure as that used for soil and also analyzed using the same Atomic Absorption Spectrophotometer (AAS) for Fe, Zn, Mn, Cu, Pb, Cr and Cd. Soil pH was determined by a pH meter in the field. Soil organic matter contents were estimated from loss-on-ignition (LOI) (Chukwuma, 1995).

A vigorous quality control programme was used to assess the accuracy and precision of the chemical data. The programme included reagent blanks, replicate and duplicate samples as well as in-house reference materials. Samples were spiked with a known amount of heavy metal, prepared and analyzed in the same way as the unspiked samples. Recovery in spiked samples ranged from 90%–95% for soil (extractable heavy metals) and 95%–100% for plant parts. Comparison of the levels of Fe, Zn, Mn, Cu, Pb, Cr and Cd in reagent blanks with their instrumental and analytical detection limits (IDL and ADL respectively) for soils and plants are given in Table 1. Analytical variances of the data obtained were <10%.

Table 1. Results of QA/QC* in chemical analysis for soils (mg/kg) and plants (mg/kg dry wt) in the Arufu lead-zinc mine

Metal	Reagent blank mean[a]		IDL[b]	ADL[c]	
	Soil	Plant		Soil	Plant
Fe	1.924	0.453	6.00	3.292	1.066
Zn	0.846	0.718	0.50	0.385	0.208
Mn	0.802	0.362	6.00	1.240	0.876
Pb	0.464	0.182	1.00	0.692	0.085
Cu	0.228	0.044	0.10	0.636	0.096
Cd	0.016	0.002	0.20	0.138	0.014
Cr	0.047	0.026	0.10	0.306	0.173

* QA/QC=Quality assurance/Quality control. a. Mean of 4 values; b. instrumental detection limit (IDL) was given by analytical laboratory; c. analytical detection limit (ADL) was calculated by multiplying the standard deviation of the mean reagent blank value by 2.353. This value was derived from t-table using 3 degrees of freedom at 95% confidence limit.

4 Results

4.1 Soil pH and LOI

The descriptive statistics summary of heavy metal concentrations, pH and organic matter contents (loss-on-ignition) of soil samples for the study area as well as previous related data are presented in Table 2. In the same table, the percent extraction rates of heavy metals evaluated from soil extraction in 0.1 M HCl divided by the extraction in 4 M HNO_3 are listed. The present study revealed the neutral pH values and low organic matter contents of soils in cultivated fields close to mine dump and the rural unmineralized area (Abata). The soil pH in both sites ranged from 6.5 to 7.5 with averages of 6.99%±0.2% and 6.92%±0.1%, respectively. Soil pH from the control site (forest) was slightly acidic and averaged 5.3%±1%. The organic matter contents of cultivated soils were generally <10% (loss-on-ignition, LOI). Whereas the forest soil had relatively high organic matter contents with an average of about 13%±2%.

4.2 Concentrations of heavy metals in soils

The levels of heavy metals in soils in cultivated farmlands close to the mine and in unmineralized areas are not significantly different with the mean values of 154 and 178 mg/kg (Fe), 171 and 150 mg/kg (Zn), 137 and 130 mg/kg (Mn), 40 and 52 mg/kg (Pb), 20 and 17 mg/kg (Cu), 2 and 2 mg/kg (Cr), and 4 and 3 mg/kg (Cd), respectively. Relatively low concentrations of Zn (mean 60 mg/kg), Pb (mean 36 mg/kg) and Cd (mean 0.6 mg/kg) were found in soils from the control area underlain by the same lithology but suffering minimal anthropogenic inputs. The concentrations in the control site are close to world average contents, of 60 mg/kg (Zn), 30 mg/kg (Pb) and 0.7 mg/kg (Cd) over unmineralized soil (Siegel, 2002).

4.3 Concentrations of heavy metals in plants

Mean concentrations and standard deviation values of heavy metals, based on dry weight, in crop plants are listed in Table 3. The concentrations of heavy metals were different in the same plant species but similar among sample locations except for Zn in rice leaves with a mean concentration of 74 mg/kg (dry wt) in farmlands close to mine dump and 35 mg/kg (dry wt) in farmlands in the unmineralized site. For average concentrations, Cd was lowest; 0.3 mg/kg (dry wt) in rice leaves and 0.7 mg/kg in cassava parts. While Zn was highest; 74 mg/kg (dry-wt) in rice leaves and 60 mg/kg (dry wt) in cassava parts. In addition, metal levels varied with plant parts decreasing in the order of rice leaves> cassava tubers >peelings.

5 Discussion

The relatively high organic matter contents in the forest soil were probably due to the decomposition of plant parts in the soil (Jopany and Young, 1993). Other workers have reported acidic (low) pH values from other Pb-Zn mine sites and have attributed it to sulphide oxidation (Salami et al., 2006; Ofulume et al., 2004; Lee et al., 2001). The high pH values in the present study may indicate some interaction with the carbonates which host mineralization in the area (Abimbola and Akande, 1996; Hwang and Kim, 1998) and the buffering influence of aluminosilicates (Siegel, 2002) as well as application of biocides to cultivated soils (Cox, 1995; Agbenin, 1998). The elevated levels of Zn, Pb and Cd in soils in cultivated fields compared to the levels in the control site are indicative of anthropogenic contributions most probably from mining and agricultural practices. The highest values of Zn (706 mg/kg), Pb (320 mg/kg) and Cd (10 mg/kg) were recorded from a few samples close to the mine dump site due most probably to heavy metal immobility in the neutral soils with low organic matter contents. Concentrations could therefore be higher in the tailings which were not considered in the present study.

Table 2. Heavy metal concentrations (mg/kg), pH and LOI (%) of soil samples

Parameter	Mine site (Arufu)	Unmineralized site (Abata)	Forest soil (Makurdi)	Enyimba mine site (Nigeria)[b]	Sambo mine site (Korea)[c]	Tamar mine site (Wales)[d]
Fe	154+44(5)[a]	178+60 (3)	171+20	ND	ND	ND
Zn	171+82(18)	150+107(14)	85+34	2610+182	87	108+15
Mn	137+51(4)	130+28 (5)	ND	ND	ND	
Pb	40+28 (16)	52+35 (6)	36+30	ND	243	75+116
Cu	20+14 (8)	17+13 (2)	54+31	43+2	22	134+36
Cr	2+015 (8)	2+1 (7)	ND	ND	ND	
Cd	4+1 (23)	3+1 (24)	0.6+.2	ND	3.4	0.8+0.08
LOI	5+	4+0.1	13+2	8+3	ND	ND
pH	6.99+.2	6.92+0.2	5.3+1	5.9+0.6	6.3	ND
N	40	40	20	22	72	18

Note: a. Extraction rate (%); b. after Chukwuma, 1995; c. after Jung and Thornton, 1996; d. after Fuge et al.,1989; *N*. number of samples; ND. not determined.

Table 3. Heavy metal concentrations in plant parts (mg/kg, dry wt) in the study area

Plant parts	Fe	Zn	Mn	Cu	Pb	Cd	Cr	*N*
(a) Mine site (Arufu)								
Rice leaves	40±13	74±34	26±10	4±2	3±0.5	0.3±0.1	0.5±.1	9
Cassava tubers	18±2	31±3	13±5	11±1	0.5±.1	0.5±.2	0.7±.3	8
Cassava peelings	3±.4	19±8	5±4	3±.8	0.2±.7	0.2±0.08	0.6±0.4	8
(b) Unmineralized site (Abata)								
Rice leaves	37±12	35±43	28±14	5±2	3.3±1	0.3±0.2	0.4±0.2	12
Cassava tubers	21±6	25±14	14±2	13±1.4	0.4±.1	0.25±0.1	0.8±0.4	11
Cassava peelings	4±0.8	10±10	5±3	6±3	0.24±0.06	0.16±0.07	0.5±0.2	11

Chukwuma (1995) reported higher values in tailings and soil contaminated by old Pb-Zn mines in southern Benue Trough with mean levels of 2610 mg/kg (Zn), 480 mg/kg (Pb) and 43 mg/kg (CU), respectively. In Korea, Jung and Thornton (1996) reported mean levels of heavy metals in soils from the Pb-Zn mine of 87 mg/kg (Zn), 234 mg/kg (Pb), 22 mg/kg (Cu) and 3.4 mg/kg (Cd). Similar values were reported from the Tamar Pb-Zn mineralization valley in Wales by Fuge et al. (1989) (Table 2). Variations in the levels of heavy metals in soils among different Pb-Zn mine sites might partly be due to different climatic conditions as well as differences in sampling and analytical methods employed.

Generally in soils, most heavy metals were found within the concentrations normally determined worldwide (Li and Thornton, 1993; Kabata-Pendias and Pendias, 2002; Siegel, 2002) and also within the levels proposed by various countries for agricultural soils (Kabata-Pendias and Pendias, 2002) as well as world average natural contents (Cox, 1995; Siegel, 2002,) expect for Pb and Cd. This indicates that despite the close proximity of cultivated fields close to mine dump, surface (0–15 cm depth) agricultural soils do not seem to have been seriously contaminated. This is thought to be related to the properties of soil; sandy texture (low clay), neutral pH, low organic matter content as well as Fe and Mn which are known to absorb and retain metals (Siegel, 2002; Adamu et al., 2003). Possible leaching into the subsurface, a function of metal phase, is a model for further geoenvironmental study in the area. It can be expected that the high levels of Cd, Pb and to a lesser degree Zn as well as indiscriminate applications of fertilizer, biocides and manure may influence metal uptake into crop plants (Chukwuma, 1995).

It is well known that heavy metal concentrations vary with plant species (Alloway, 1990; Jung and Thornton, 1996; Xian, 1989; Lee et al., 2004) and that leaves tend to accumulate higher concentrations than roots or tubers and grains or fruits (Jung et al., 2002). Results from this study showed that metal concentrations were higher in rice leaves than in cassava tubers confirming that leaves tend to concentrate more heavy metals than tubers. Though, it may reflect differences in accumulation between plant species.

Concentration levels of heavy metals determined in plant parts from the present study were comparable to those found in plants parts in the lower Benue Trough (Chukwuma, 1995) but were remarkably lower than levels in plants in the upper Benue Trough (Adiku-Brown and Ogezi, 1991). It is well known that heavy metal levels in crop plants often show large variations even at the same location in the field. This has been attributed to variable emission rates, atmospheric transport and deposition processes, and plant uptake. Furthermore, bioavailability and variations in growth rate may add to the variability (Voutsa et al., 1996; Chon et al., 1997; Ofulume et al., 2004). Although soils in the study area are not highly contaminated (see Table 2), metal levels in plant parts are relatively high. For instance, Pb contents in plants grown in uncontaminated and unmineralized areas fall within a relatively narrow range, from 0.1 to 10 mg/kg (dry wt) with an average of 2 mg/kg (dry wt) (Siegel, 2002). In this study, levels of Pb in plants averaged 3 mg/kg (dry wt) in rice leaves. Similarly, the average level of Zn (74 mg/kg (dry wt)) in rice leaves in farmlands close to mine dump is higher than the usual range of 1.2 to 73 mg/kg in edible plants from uncontaminated soil (Kabata-Pendias and Pendias, 2002).

Generally, most elements were found in plant parts in the study at concentrations found in crops grown in uncontaminated soils (Cox, 1995). The concentrations in the study represent the fraction of element remaining in plant tissues after the usual household preparation of vegetable and cassava. Washing certainly removed some heavy metals. An additional fraction had been removed from cassava tubers by peeling (Table 3). The fraction leached from plant parts by washing was not determined, but is variable between heavy metals depending on their function or metabolic association. For example, Pb is mainly a superficial deposit in plants, especially in leafy surfaces, whereas Zn, Cu and Cd showed greater root penetration (Chon et al., 1997). Thus, plant parts accumulated more heavy metals than what was reported in this study.

5.1 Relationships between heavy metal concentrations in soils and those in plants

Extraction rate (defined as the ratio of heavy metals extracted from soil sample by 0.1 M HCl to that extracted by 4 M HNO_3, expressed as percent) provides a useful indication of the soluble and available portion of heavy metals from soils to plants (Davidson et al., 1994; Xian, 1989). The results of extraction rate presented in Table 1 depict the following trends of solubility:

(1) Mine site; Cd> Zn> Pb>Cu>Fe>Mn>Cr

(2) Unmineralized site; Cd>Zn>Cr>Pb>Mn>Fe>Cu

The trends predict Cd and Zn as the most soluble and available heavy metals from soils to plants in the sampled areas. The rates are also different between the sample areas, most probably, reflecting different sources and geochemical controls. Relationships between heavy metals and plant parts are shown in Figs. 2 and 3. In general, heavy metal concentrations in plant parts are highly comparable with those of soil, although the gradient can differ between plant species and parts. It is well known that total metal content in soils is an important factor influencing the uptake of metals into plants (Alloway, 1990). However, the uptake is affected by so many factors including soil texture, pH, Eh, CEC, organic matter content, Fe-Mn-Al-oxides and hydroxides, presence of other metals, amount and rate of fertilizer and biocide applications as well as plant species, parts, cultivars and age (Voutsa et al., 1996; Li and Thornton, 1993), accounting for the variable relationships between soil heavy metal contents and plants. Stepwise multiple linear regression analysis was applied in the study to find the dominant factors influencing metal uptake in plants and to also predict metal levels in plants. A step-by-step procedure was employed to obtain the best fit regression equation. The first independent variable was always total metal content in the surface soils. The next major factor was found from the correlation matrix and the regression equation was calculated using statistical package for the soil sciences (SPSS). At each stage, the significance of the equation was tested by the coefficient of determination (r^2) and probability ($P<0.05$). The equation was considered significant for r-squared values ≥ 0.50 at $P<0.05$.

The results of linear multiple regressions are presented in Table 4. It can be seen that total metal concentrations in surface soils are the main factor affecting those in plants, being correlated positively with metals in plants at each occasion except for Cd in cassava tubers in the unmineralized site. In addition, where pH correlated positively with metals in plants LOI correlated negatively on each occasion but for Cd in cassava tubers in the unmineralized site. Thus, pH and LOI also contributed to the prediction of heavy metal concentrations in plants in this study.

5.2 Environmental implications

The study area is a rural agricultural area where more than 90% of the inhabitants are farmers. Land is intensively cultivated and animals are reared for food production. The common diet of the animals is locally grown crops. Metals are directly ingested by these animals as they feed on unwashed crops. Soil ingestion ranges between 1%–15% of the dry matter intake by cattle and may be as large as 20%–30% in sheep and even >30% in swine which grazes close to ground

(Alloway, 1990). It was not possible to sample animal tissues from the study area due to limited resources. It is likely that animals in the study area might be accumulating heavy metals through ingesting tailings and through a diet grown on contaminated soils. This may have some health implications on the animals, though this may not result in direct increasing intake in human beings. Contamination of useable water is a more direct way of ingestion by human beings (Cox, 1995).

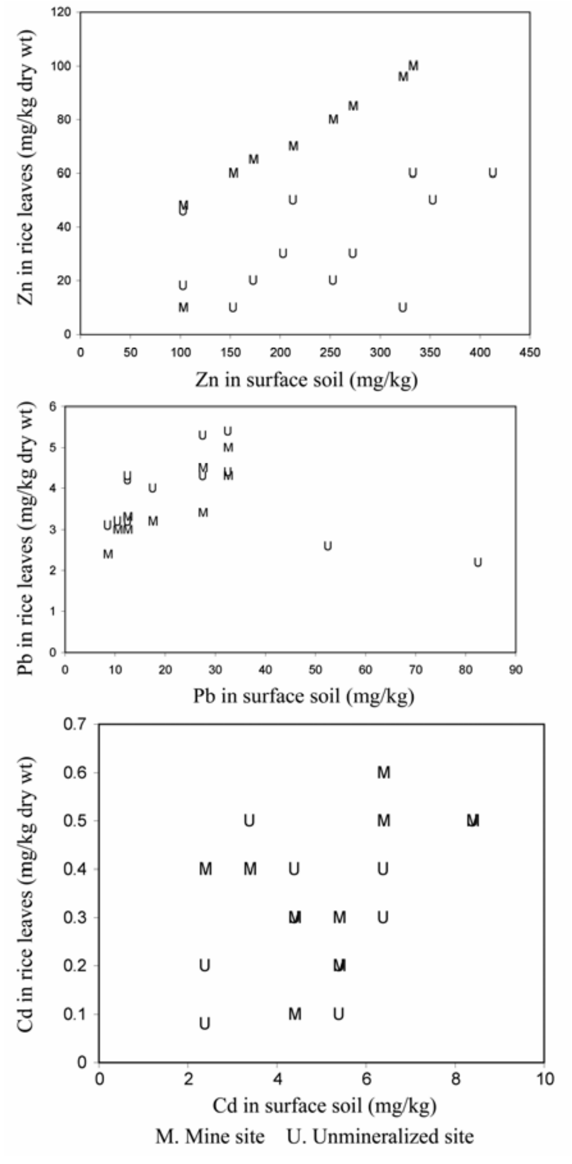

Fig. 2. Relationships between metal concentrations in surface soil and those in rice leaves.

Residents of the study area consume crops grown on these feeds and use water from the mines. The long-term metal exposures by regularly consuming locally grown crops imposes potential health problems on both animals and human beings in the vicinity of the mine dump, although crops investigated appeared to contain a normal range of heavy metals. It was not the total levels that were measured in this study. Also the deep-rooted crops are likely to take up and accumulate more heavy metals from the subsurface than from crops considered in the study.

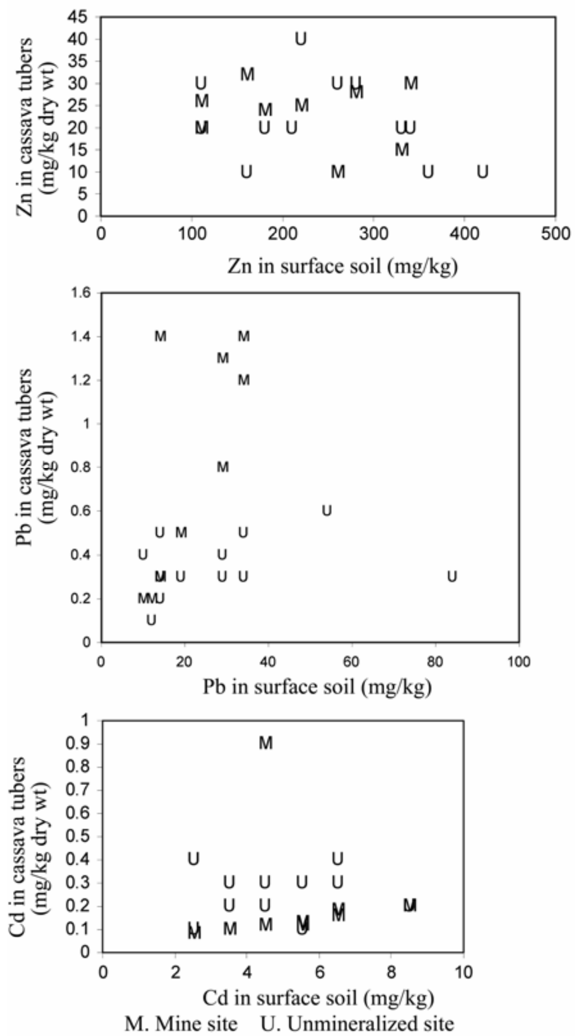

Fig. 3. Relationships between metal concentrations in surface soil and those in cassava tubers.

6 Conclusions

Soil samples from cultivated farmlands close to mine dumps and unmineralized rural soil are characterized by neutral pH (≥6.7≤7.5) , low organic matter contents (<10%, LOI) and similar levels of heavy metals, with the exception of few samples close to the immediate vicinity of the mine, which are characterized by relatively high concentrations of heavy metals. The concentrations of Pb, Zn and Cd in soils in the cultivated farmlands were higher than those in the control area (forest area), due to mining and agricultural activities. The concentrations of heavy metals in cultivated soil are not restricted to mining activities as the rural unmineralized soils have not affected by mining activities.

Most heavy metals were found in plant parts at levels normally observed in crop plants grown on uncontaminated soils except for Cd and Pb close to mine dumps, particularly for rice leaves. Release and transport of heavy metals as well as bioavailability were affected to varying degree by acid-neutralization processes. Hence, metal levels in soil were immobilized and not reflected in plant parts away from mine dumps. Heavy metal levels in plant parts decreased in the order of rice leaves > cassava tubers >peelings. In the same plant part, heavy metal leaves decreased in the order of Zn>Fe>Mn>Cu>Pb>Cr>Cd due partly to total metal levels in soil and bioavailability.

Table 4. Results of stepwise multiple regression analysis

Plant type	Multiple regression equation*	*r*2
Cassava tubers (*N*=8)	(Cd)p = 0.03 + 0.02 (Cd)s + 0.007pH-0.008 LOI	0.89
	(Pb)p = -0.14+0.05 (Pb)s – 0.042pH + 0.050 LOI	0.51
Rice leaves (*N*=9)	(Pb)P = 2.87 + 0.08 (Pb)s -0.175pH + 0.098 LOI	0.90
	(Zn)p = 46.47 + 0.26 (Zn)s – 5.149pH 0.991 LOI	0.84
	(Cu)p = 0.07 + 0.17 (Cu)s + 0.036pH -0.034 LOI	0.56
	Unmineralized site	
Plant type	Multiple regression equation*	*r*2
Cassava tubers (*N*=11)	(Cd)p = 0.73 -0.002 (Cd)s -0.029pH -0.077 LOI	0.80
Rice leaves	(Pb)P = 4.03 + 0.033 (Pb)s -0.364pH + 0.273 LOI	0.85
(*N*=12)	(Cu)p = 0.07 + 0.173 (Cu)s + 0.046pH – 0.034 LOI	0.56

Note: *N* . Number of samples; (Cu)p. copper concentrations in plants (mg/kg, dry weight); (Cu)s. copper concentrations in soils (mg/kg); LOI. loss-on-ignition of surface soil (%); pH. surface soil pH. * $p<0.05$.

In conclusion, this pilot study has provided the base-levels of heavy metals for soils and some stable food crops and their dispersion without which unsafe practices can compound the problem of heavy metal dispersion and increase the risk to the biosphere. The study has identified agriculture expansion based upon indiscriminate application of biocides and fertilizers as an unsafe practice.

Proper managing practice should, therefore, not be restricted to mining. Agriculture practice should be checked as well. Further studies should be carried out to evaluate spatial and temporal variations as well as speciation of heavy metals to establish the controls of their distribution in the study area.

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