Uptake of trace elements by food crops grown within the Kilembe copper mine catchment, Western Uganda

By: Abraham R. Mwesigye, Scott D. Young, Elizabeth H. Bailey and Susan B.Tumwebaze

Key words: trace elements, food contamination, copper mine

Abstract

The mining and processing of copper from the Kilembe mine between 1956 and 1982 left behind millions of tons of cupriferous and cobaltferrous mine tailings within the Kilembe mine catchment. Subsequent erosion and deposition of the tailings into adjacent areas led to increased concentrations of Cu, Co, Ni, Zn, and Pb in the catchment soils. The Kilembe catchment is utilised for subsistence farming, producing mainly food crops, but there are also a number of settlements in the contaminated area. A study was conducted in 2016 to establish the concentrations of trace elements in a range of food crops grown within the catchment. Samples of maize, bananas, cassava, sweet potatoes, ground nuts, amaranthus, onions, beans and yams were collected, washed and oven dried at 80°C. The dried foods were finely ground, microwave-digested in nitric acid and analysed using inductively coupled plasma mass spectrometry (ICPMS). All the foods grown in contaminated soils showed instances of higher concentrations of Cu, Co, Ni, Zn, and in some cases Pb, compared with controls grown in uncontaminated soils. Amaranthus accumulated a range of trace elements with 26% of the samples exceeding EC thresholds for Cu in vegetables of 26 mg kg^{-1}.Other crops with elemental concentrations exceeding recommended thresholds in some of the samples included beans (Zn), yams (Zn and Pb) and ground nuts (Zn). The concentrations of trace elements in onions, cassava, sweet potatoes, bananas and maize were not significantly different from controls. However, strong and positive correlations between the trace elements were found in beans, yams, amaranthus, maize and ground nuts, suggesting a common source of trace metals. There was strong evidence of soil dust retention on leaf vegetables (Amaranthus) despite washing. The accumulation of trace elements in the edible parts of vegetables and foods could have a direct impact on the health of local people, because the foods produced from gardens are mostly consumed locally.

1. Introduction

Pollution by heavy metals in the natural environmental is an issue that has become a global problem and is a common feature of industrial development (Nagajyoti et al., 2010). Increasing industrialization has been accompanied throughout the world by the extraction and distribution of mineral substances from their natural deposits (Singh, 2001). The contamination of agricultural soils is often a direct, or indirect, consequence of anthropogenic activities (McLaughlin *et al*., 1999). Sources of anthropogenic metal contamination in soils include urban and industrial wastes, mining and smelting of non-ferrous metals and metallurgical industries agricultural inputs, and fallout of industrial and urban emissions (Singh, 2001; Wilson and Pyatt, 2007). Excessive accumulation of trace metals in agricultural soils may have consequences for food quality and safety. Chunilall et al. (2005) found that the concentrations of elements in plant tissues were affected by the concentrations of the heavy metals in the soil. Accumulation of heavy metals by plants can be via the root uptake or deposition on foliar surfaces (Sawidis et al., 2001). So, it is essential to monitor food quality, given that plant uptake is one of the main pathways through which heavy metals enter the food supply (Antonious and Kochhar, 2009).

In the Kilembe mine area, Uganda, mining and processing of copper was active between 1956 and 1982. Previous studies (e.g Mwesigye et al., 2016) showed that the Kilembe catchment soils were contaminated with Cu, Co and Ni, in many cases exceeding recommended thresholds for agricultural soils. Food crops, including *Amarathus* vegetables, cassava and bananas, also had elevated concentrations of these three metals. Mwesigye and Tumwebaze (2017) found that the mine water and leachate flowing through Kilembe catchment soils contained elevated quantities of Cu, Co, Ni. Despite the high concentration of trace elements in soils and catchment waters, subsistence farming of food crops remains an important means of livelihood within the Kilembe catchment. However, previous studies on food quality in the area have examined only a limited range of food crops.. This study was therefore conducted to investigate the accumulation of trace elements in a wider range of crops grown in contaminated areas of the Kilembe catchment and to further assess the potential risks posed to consumers of locally grown foods.

1.1 Methods

A total of 97 food samples were collected between October 2016 to February 2017 from Kilembe catchment, Kasese district, Uganda. The area previously housed the (now defunct) Kilembe copper mines where copper ores were mined and processed. The wastes from copper processing (mine tailings) were dumped within the Kilembe mine valley, an area that is now predominantly agricultural but also residential. The food crops collected included beans (n=21), yams (n=14), onions (n=4), cassava (n=13), sweet potatoes (n=4), bananas (n=13), amarant (n=18), maize (n=7) and ground nuts (n=3). The foods were sampled from locations within the Kilembe mine catchment which had earlier been confirmed to have high concentrations of trace elements in top soils, especially Cu, Co and Ni (Mwesigye et. al, 2016). The food samples were collected from household gardens. In addition, at least 5 control samples of each specific food crop were collected in un contaminated soils within Kilembe mine catchment.

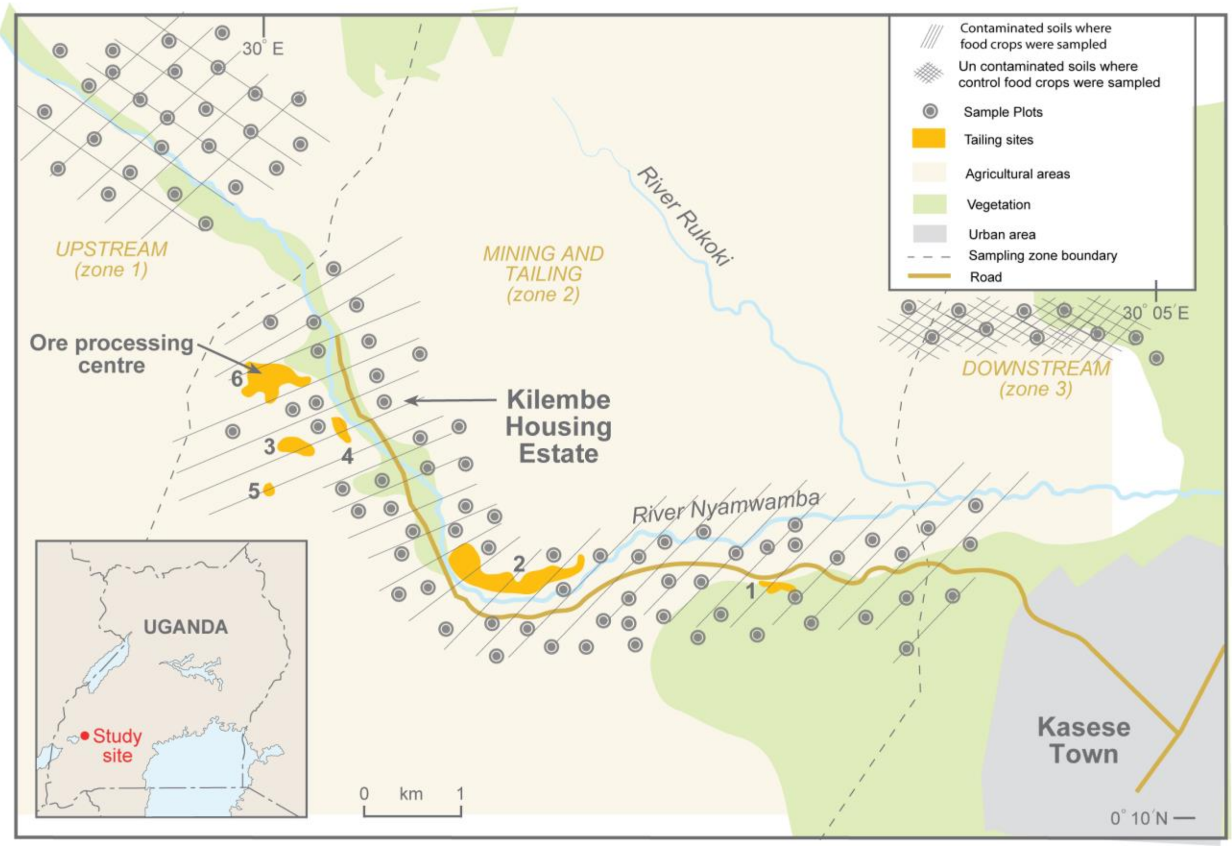


Figure 1: Map of Kilembe mine catchment showing the study area and sampling sites

Preparation of food samples involved rinsing vegetables twice using tap water and finally washing with distilled water before oven-drying at 80°C for 24 hours. After washing, yams, cassava, sweet potatoes, bananas and ground nuts were peeled using a stainless-steel knife and sliced into small pieces before being oven dried at 90°C for a period of 24 hours. Grains of maize and seeds of beans were washed in distilled water and oven dried as other crops; onions were washed then cut into smaller pieces and oven dried. Fresh and dry weights of each food crop sample were taken before and after oven drying to determine fresh-to-dry weight conversion factors.

All the dried samples were ground into fine powder using a centrifugal mill with a titanium screen (Retsch ZM 200) and stored in plastic zip lock bags. Following exportation to the United

Kingdom, approximately 0.2 g of each sample was microwave digested in nitric acid (70%, Trace Analytical Grade-TAG), and the resultant solution was diluted with Milli-Q water (18.2 MΩ cm) before analysis using ICP-MS. All laboratory tests were conducted at the University of Nottingham School of Biosciences.

A survey was conducted, comprising 21 households and 3 schools, to assess the type and quantity of foods consumed. In the homes, volunteers had the food apportioned for their consumption measured during lunch time meals. Within schools, childrens' lunch time meals were characterized and weighed. In all cases, permission was sought after explanations of the importance of the exercise. Local people were informed that they were free to decline to participate and those who declined were omitted from the study.

1.1.1 Sample analysis

The concentrations of 31 elements in food samples, including Zn, Cu, Co, Ni, As, Cd, Cr, As and Pb were measured using ICP-MS (Model iCAPQ; Thermo Scientific, Bremen) with 'in-sample switching' between three operational modes; standard mode, hydrogen cell mode and kinetic energy discrimination with He as the cell gas to reduce polyatomic interferences. Internal standards included Sc (10 $\mu g\ L^{-1}$), Ge (10 $\mu g\ L^{-1}$), Rh (5 $\mu g\ L^{-1}$) and Ir (2 $\mu g\ L^{-1}$) in 2% trace analytical grade (TAG) HNO_3. External multi-element calibration standards (Claritas-PPT grade CLMS-2, Certiprep) included elements in the concentration range 0–100 $\mu g\ L^{-1}$.

1.1.2 Estimation of soil dust in foods

Although food crops were washed in water, it was considered that they could still have some soil dust particles on the substrate. This is a particularly important consideration on contaminated sites partially denuded of vegetation, such as minespoil areas. Some trace elements have very poor bioavailability and can be used to estimate the likely proportion of the metal content of plants arising from external contamination from soil dust. Thus, vanadium (V) may be a reliable indicator of extraneous contamination with soil dust because (i) vanadate (VO_4^{3-}) ions are poorly available to plants in soil, (ii) vanadium is unlikely to follow a similar uptake path to that of Fe^{3+} or Fe^{2+} but (iii) trivalent V^{3+} ions substitute for Fe^{3+} in soil iron hydrous oxide particles and vanadate anions are strongly adsorbed by iron oxides (Joy et al., 2015). Thus, a strong correlation between Fe and V concentrations is more likely to reflect the inclusion of Fe oxide particles from soil dust within the foods rather than systemic uptake of V and Fe via the plant root system. The levels of soil dust contamination in foods were therefore estimated, for each element (M), from the soil V concentration and knowledge of the M:V ratio in the surrounding soil (Eq. 1; Joy et al., 2015) The average soil V and other elemental concentrations for the area were obtained from Mwesigye et. al, (2016).

$$\mathrm{Py}\ (\%) = \frac{\mathrm{Vp\ xMs)\ 100}}{\mathrm{Vs\ x\ Mp}}\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots 1$$

In Eq. 1, Py (%) is the percentage contamination from soil dust for a given element (M) in a plant sample, Vp and Vs are the vanadium concentrations in the plant and in the local soil, Mp and Ms are the concentrations of the test element in the plant and the local soil respectively. It must be stressed that this approach provides only an approximate estimate of Py because it assumes (i) no systemic uptake of V and (ii) that the ratio of M:V in the local soil also applies to fine dust particles embedded in plant tissue.

1.1.3 Risk assessments of foods

Hazard quotients (HQs) have been widely used to express 'non-cancer' health risk from consumption of foods grown in contaminated soils (e.g. Hough et al., 2004). Values of trace element specific HQ were calculated according to Eq. (3) (Datta and Young, 2005):

$$\mathrm{HQ} = \frac{\mathrm{Cp\ x\ ADI\ x\ Fwc}}{\mathrm{RfD\ x\ BW}}\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots 2$$

where Cp is the trace element concentration in the edible portion of vegetables or food (mg kg^{-1} dry weight-DW), ADI is the average daily intake (fresh weight) of vegetable and foods (established from the survey to be 0.74 kg day^{-1} for children and 0.94 kg day^{-1} for adults), FWC is a dry-to-fresh weight conversion factor, obtained as a ratio of dry weight to fresh weight of the same food type. Fresh and dry weights of each food crop were taken before and after drying the food samples. The reference dose (RfD) is a numerical estimate of the daily exposure to the human population, including sensitive subgroups, that is not likely to cause adverse health effects during a lifetime (EPA, 2002). The average body weight (BW in Eq. (2) was obtained from a past study in the area (Mwesigye et. al, 2016) where average body weight of children between 7-18 years was measured at 29.6 kg while average body weight of adults above 18 years was 65.5 kg.

1.1.4 Quality control

All samples were prepared, digested and analysed in duplicate. The reagents used for sample preparation were trace analysis grade (TAG) supplied by Fisher Scientific, UK. Operational (digestion) blanks were run to determine limits of detection (LODs). A certified reference material (NIST 1573a; tomato leaves) was included in each run; average recoveries (%) for the CRM were As (140), Cd (100), Co (107), Cu (89), Fe (105), Mn (115), Ni (99), Zn (110).

1.1.5 Statistical analysis

The analytical data was processed using Minitab to determine correlations between the elements in food crops. A two-sample t-test was used to assess the significance of differences in trace element concentrations between Kilembe catchment food samples and their controls. Statistical analyses were conducted to generate means, medians and standard deviations for all food sampled. All statistical tests were conducted at a 95% confidence level.

2. Results and discussion

Most of the food crops sampled from the Kilembe valley where concentrations of trace elements in soils are high (Mwesigye et. al, 2016) contained higher concentrations of trace elements compared with controls which were collected from uncontaminated soils. Table 1 shows the median and range of elemental concentrations in Kilembe crops and the median value for the control crops. The trace elements in soils originated from contamination of the area with mine tailings and eroded mine water. Mwesigye et.al (2016) found that tailings in Kilembe Mine area contained Co in the range of 80-152 mg ^{kg} compared with average world crust of 1-15mg ^{kg,} Ni ranged between 101-164 mg ^{kg} compared with world crust average of 20 mg ^{kg,} Copper ranged between 101-10200 mg ^{kg,} compared with World crust average of 25-75 mg ^{kg,} and these were eroding by wind and water into surrounding soils. The soils were also highly contaminated with Co in the range of 8-52 mg ^{kg} compared with world average of 10 mg ^{kg} in soils. Ni in Kilembe contaminated soils was in the range of 19-102 mg ^{kg} compared with the normal range of 13-37 mg ^{kg} while Cu in Kilembe soils ranged between 7-399 mg ^{kg} compared with world range of 14-109 mg ^{kg}. The sulphides of Co, Ni and Cu were associated with Zn, As, Cd and Pb. Therefore Kilembe soils were highly contaminated with trace metals that were geologically associated with Cu and the sulphides in the mining area. The trace metals in contaminated soils were being taken up and accumulated by crops during growth.

Table 1. Trace elements in foods grown in Kilembe mine catchment contaminated soils (mg kg^{-1})

	Co	**Ni**	**Cu**	**Zn**	**As**	**Cd**	**Pb**
Food crops	(mg kg^{-1})	(mg kg^{-1})	(mg kg^{-1})	(mg kg^{-1})	(mg kg^{-1})	(mg kg^{-1})	(mg kg^{-1})
Yams n=13							
range	0.04-5.8	0.2-3.3	4.5-12.5	11-159.3	0.003-0.07	0.001-0.05	0.01-0.04
median	0.21	0.71	8.23	28.14	0.006	0.02	0.03
Controls median (n=6)	0.07	0.23	4.32	18.4	<LOD	0.004	0.01
Maize n=7							
range	0.008-0.15	0.09-0.78	1.2-3.25	18.5-64.22	0-0.003	0.001-0.006	0.0-0.1
median	0.03	0.3	2.3	24.13	0.001	0.002	0.002
Controls median	0.007	0.06	1.01	9.9	<LOD	0.001	0.02
Cassava n=13							
range	0.08-3.4	0.8-3.7	1.4-11.3	3.4-16.1	0-0.005	0.001-0.008	0-0.08
median	0.44	2.15	2.42	6.32	0.002	0.008	0.01
Controls median (n=5)	0.03	2.31	3.13	9.91	<LOD	0.03	0.08
Sweet potatoes n=4							
range	0.5-3.3	1.2-3.81	5.4-7.6	6.1-10.34	0.001-0.004	0.002-0.004	0.004-0.03
median	1.34	1.41	6.63	8.36	0.003	0.003	0.006
Controls median (n=5)	0.04	0.16	3.01	7.84	0.001	0.001	0.03
Bananas n=13							
range	0.003-	0.1-3.2	2.4-6.5	5.5-9.9	0 -0.006	0-0.002	0-0.06

	0.6						
median	0.06	0.36	4.13	7.73	0.002	0.001	0.007
Controls median (n=6)	0.02	0.53	3.21	8.84	<LOD	0.001	0.012
Thresholds for foods	**-**	***67.9a***	***73.3b***	***99.1a***	**-**	***0.05c***	***0.3b***
Vegetables							
Amaranthus n=18							
range	0.034-72.01	0.24-13.26	5.8-41.06	39.4-271.16	0.01-0.13	0.03-0.37	0.07-3.9
median	1.3	1	13	56	0.04	0.08	0.2
Controls median (n=7)	0.14	0.34	7.51	58.33	0.03	0.03	0.55
Beans (n=21)							
range	0.29-5.53	0.74-9.51	5.2-11.64	22-139.11	0.005-0.32	0.002-0.007	0.002-0.06
median	0.84	2.64	7.44	27.24	0.01	0.004	0.02
Controls median (n=7)	0.25	2.93	6.91	31.19	0.001	0.002	0.02
Ground nuts n=3							
range	0.6-0.62	1.1-3.1	6.6-8.3	24-621.01	0.005-0.01	0.004-0.03	0.01-0.04
median	0.6	2.24	8.81	35.27	0.006	0.01	0.01
Controls median (n=5)	0.04	2.13	8.63	26.06	<LOD	0.01	0.11
Onions n=4							
range	0.08-0.15	0.3-0.43	3.5-4.07	13-19.21	0-0.004	0.007-0.01	
median	0.14	0.43	3.60	19.6	0.002	0.01	0.01
Controls median (n=5)	0.03	0.34	5.80	24.25	0.004	0.06	0.04
Thresholds for vegetables	***50a***	***66.9a***	***20b***	***99.4a***	**-**	***0.05c***	***0.3a***

a = WHO/FAO (2011); b = EC standards (2006), Codex Alimentarius Commission (CAC), 2001; c = General Standardisation Organisation (GSO) 2013 . <LOD : Less than limit of detection

2.1 **Trace elements in food crops**

Beans (*Phaseolus vulgalis*) appeared to accumulate mainly Co compared with controls (p=0.0014). Although Cu and Ni concentrations in beans were higher in Kilembe foods compared with controls, the differences were not statistically significant (p>0.05). Pearson's correlation of the elements revealed very strong and positive associations between Cu and Co (r=0.786, P<0.001) suggesting co-existence in the soil. These same correlation of elements were identified in earlier studies of soils in the Kilembe mine catchment (Mwesigye et. Al, 2016) and are associated with the area's mineralogy. The concentrations of Zn in only 5% of bean samples appeared to exceed the threshold of 99.4 set by WHO/FAO.

The concentrations of Cu, Co, Ni and As in yams (*Dioscorea species*) were generally higher than control samples, but only Ni (p=0.021), Cu (p=0.022) and As (p=0.02) were significantly higher than in control samples. Concentrations of Zn in 14% of yams appeared to exceed the thresholds for consumable foods set by WHO /FAO. Yams revealed very strong and positive correlations between all the trace elements again suggesting a common source.

Elemental concentrations in onions (*Allium sepa*) showed an unexpected trend because concentrations of Ni, Cu, Zn, and Pb in control samples were higher than in onions grown on Kilembe contaminated soils. Lead in the control samples was significantly higher than in Kilembe catchment onions (p=0.012). However the concentration of Co in Kilembe catchment onions was significantly higher than in controls (p=0.019). Onions showed a strong negative correlation between Co and Zn (r= -998, P= 0.002). This might suggest competition for uptake between Co and Zn. However, the relative concentrations of Co and Zn in the crop suggest that Co is very unlikely to influence uptake of Zn and so the negative correlation is more likely to reflect a negative correlation in soil metal loadings. The number of onion samples were quite limited (n=4) and extensive surveys of the crop in Kilembe mine catchment are required to generate meaningful inferences.

Cobalt concentration in cassava *(Manihot esculenta)* was significantly higher (P=0.025) than in controls. This finding is in accord with previous studies such as Kríbek, et al. (2014) who found that cassava cultivated in areas affected by mining contained higher concentrations of heavy metals and metalloids when compared with those grown in uncontaminated areas. Nester et al. (2015) also found that Co, Ni, and Zn were elevated in cassava grown on mine-contaminated soils in Ghana. Cassava appeared to show strong and positive correlations between Ni and Cu (r=0.733, P=0.004), Co and Cu (r=0.692, P=0.009) and, in contrast to onions, between Co and Zn (r=0.633, P=0.2).

Sweet potatoes (*Ipomea batatas***)** grown in Kilembe catchment soils generally showed higher concentrations of the trace elements known to be contaminants in Kilembe soils, i.e. Cu, Co and Ni, compared with controls. However, only Cu was significantly higher than in the controls (P=0.019). Strong positive correlations in potatoes was only observed between Ni and Co concentrations (r=0.975, P=0.025).

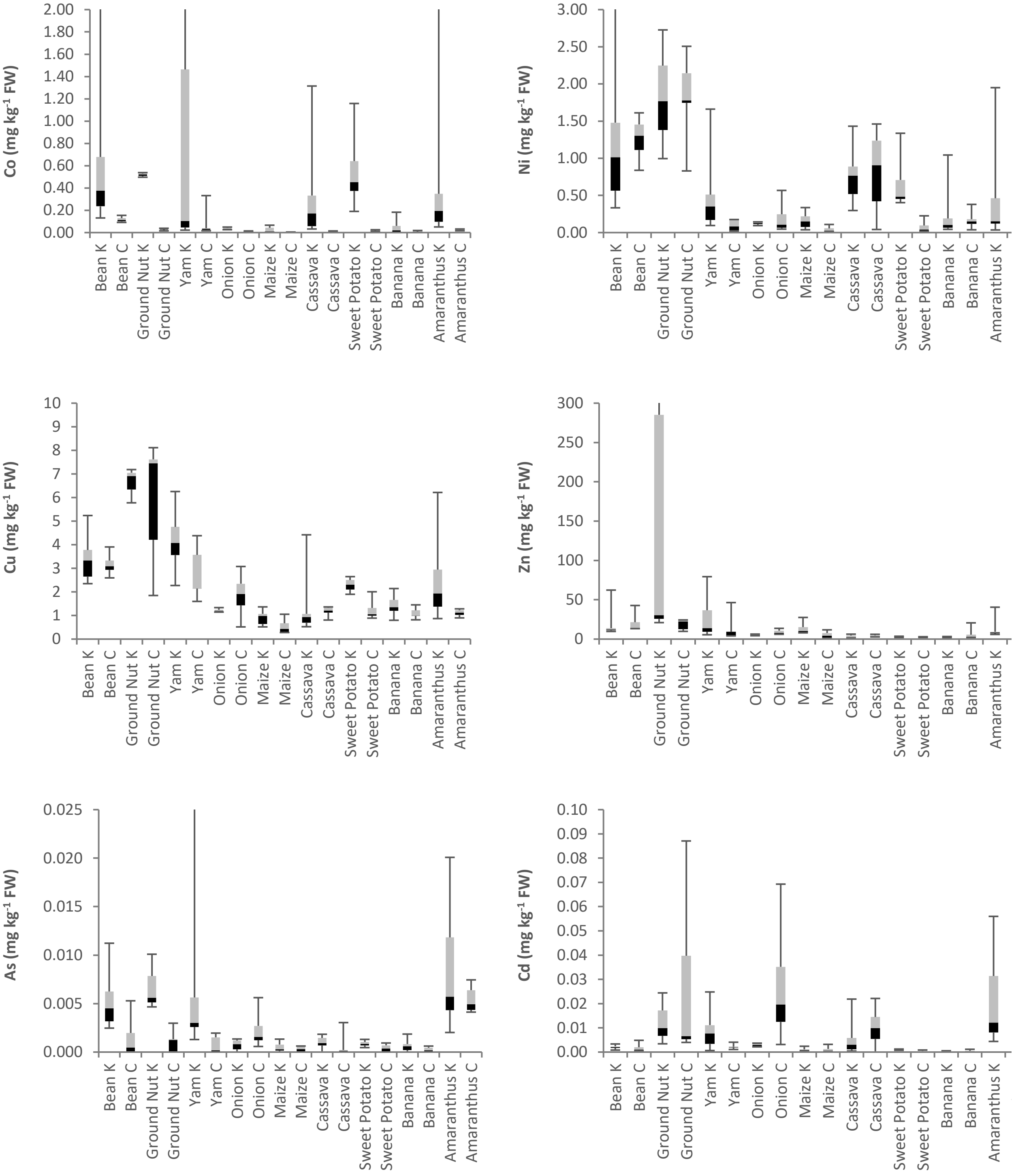
For bananas (*Musa species*) in the Kilembe catchment, Cu (P=0.04) and Co (P=0.045) were significantly higher than in controls. Much of the Pb in bananas also appeared to originate from extraneous soil dust rather being systemically taken up by the plant during growth. Nesta et al. (2015) also found that concentrations of Cu in *Musa* species planted in contaminated soils in Ghana were higher than in plants grown in non-contaminated soils.

In *Amaranthus* species, with the exception of Ni, the concentrations of all trace elements were higher in Kilembe contaminated soils than in control vegetable samples. Over 26% of the vegetable samples exceeded consumption thresholds of 20 mg kg^{-1} of Cu recommended by the European Community (2006). The vegetables also showed a strong correlation between Co and Cd (r=0.574, P= 0.013). Chunilall et al. (2005) also found that Amaranthus species appeared to take up Pb from contaminated soils in large quantities. Kachenko and Balwant (2005) also found that vegetables grown in contaminated soils in Australia accumulated large quantities of Zn, Cd and Cu.

Maize *(Zea maize)* grown in Kilembe mine soils contained higher concentrations of Co, Cu and Zn but only Co (P=0.046) and Ni (P=0.04) concentrations were significantly higher than in control maize plants. None of the elemental concentrations in maize exceeded thresholds recommended for human consumption. Maize also appeared to show strong and positive correlations between Cr and Zn (r= 0.862, r=0.013).

Groundnuts (*Arachis hypogaea*) grown in the Kilembe mine catchment soils contained significantly higher concentrations of Co than in controls (P=0.001). However none of the elements in ground nuts exceeded thresholds for human consumption. It should be noted that the number of ground nut samples were too limited (n=3) to provide meaningful inferences and extensive surveys of the crop are necessary.

Figure 1 shows the trace element concentration profiles (FW) for the range of food crops included in the survey as box and whisker plots. We can draw only limited conclusions from what is still a limited reconnaissance survey of the area. However, crops grown within Kilembe were frequently more enriched with trace metals than controls. Furthermore, the high frequency of skewed distributions indicate instances of relatively high concentrations on both a crop-specific and element-specific basis. Examples were almost exclusively from the Kilembe (K) area and included: (i) Co in beans, yams, cassava, sweet potato and amaranthus, (ii) Ni in amaranthus, (iii) Cu in amaranthus, (iv) Zn in ground nuts, (v) As in beans, groundnuts, yams and amaranthus, and (vi) Cd in groundnuts, onions and amaranthus. Cadmium concentrations were generally low across all crops and there were also higher outliers in control cassava and groundnuts. Similarly, (vii) Pb concentrations were also low and there were high control outliers in cassava and amaranthus.



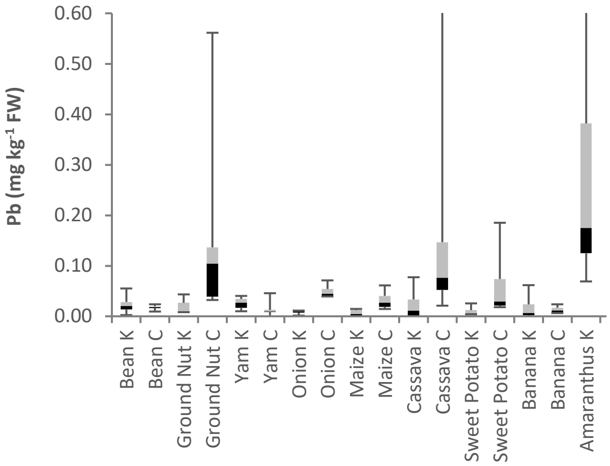


Figure 2: Box and whisker plots comparing the range of trace element concentrations in food crops and vegetables grown in the Kilembe area with those grown at control sites or procured from markets.Yams (n=13), Maize (n=7), Cassava (n=13), Sweet potatoes (n=4), bananas (n=13), Amaranth (n=18), Beans (n=18_), Ground nuts (n=3) and Onions (n-4)

2.2 Elemental contributions from soil dust

The estimated elemental contributions to washed food samples from residual soil dust contamination (Eq. 1) were very low for Cu, Co, Zn and Ni, lying within the range of 0.07-1.4%. However a significant proportion of Pb appeared to originate from extraneous soil dust, ranging between 26% and 45%. This implies that, with the exception of Pb, most of the elemental concentrations measured in the washed vegetables and foods resulted from plant root uptake during growth. It should also be noted that this study used average soil elemental concentrations from a previous study in the area (Mwesigye et al., 2016) and so the soil concentrations used may differ from actual elemental concentrations in a given plot or the wind-blown dust affecting individual crops.

Comparing a range of crops (beans, yams, cassava, banana and amaranthus), it was clear that the amaranthus showed the greatest influence of residual dust contamination after washing. Figure 3 shows a very strong correlation between Fe and V for amaranthus, implying significant contributions of other elements from soil dust (Joy et al., 2015). Other food crops showed a poor correlation between Fe and V, at much lower concentrations, implying that contribution of the elements from soil dust was minimal. This general pattern is consistent with the relatively exposed situation of leafy vegetables, and their ability to retain sub-micron sized particulates in leaf cuticles. By contrast, the ‘protected’ nature of the edible components in beans, maize and bananas and the removal of peel required in preparation of yams and cassava suggests that the influence of dust on food prepared from those crops would be very limited.

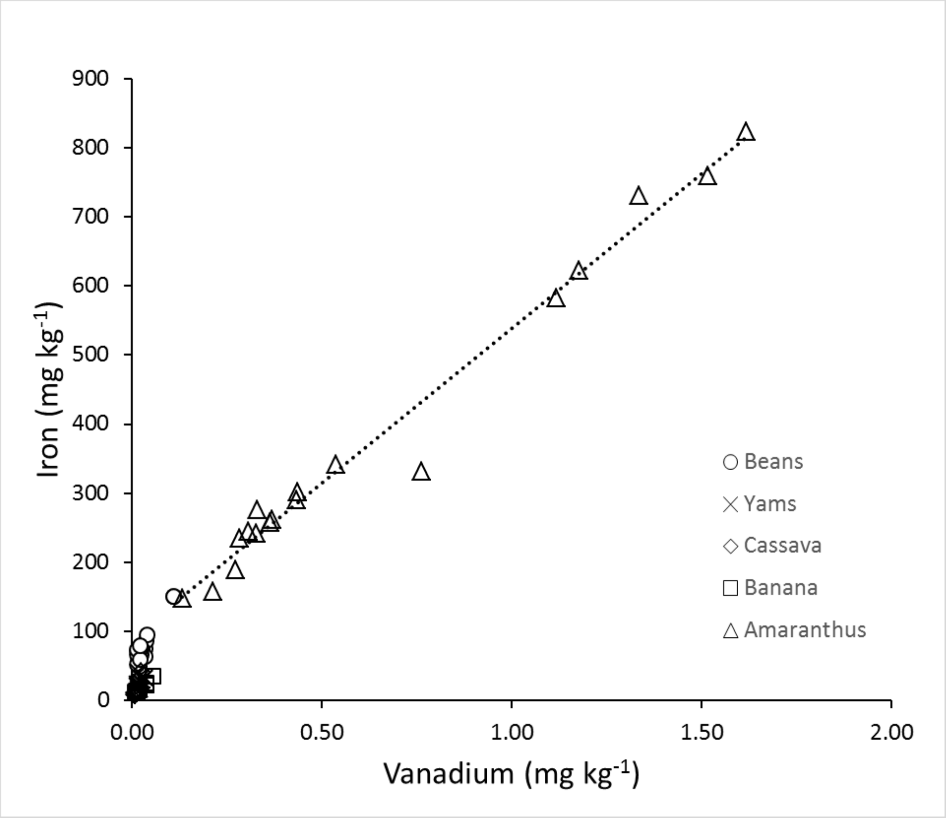


Figure 3: Soil V and Fe concentrations (mg kg^{-1}) in all food samples collected from locations within the Kilembe mining area.

2.3. Provisional risk assessment (hazard quotients)

Table 2 shows hazard quotients calculated for key crops and assuming, as a comparative exercise, that the crops are consumed at rates equivalent to the parameter values given in Section 1.1.3. Yams appeared to present a possible risk to children through excessive Co and Zn consumption while adults were exposed to high Zn concentrations. Maize consumption presented a hazard quotient that was of risk to children through excessive Zn intake while cassava consumption presented risks to children through excessive Ni and Cu consumption. Consumption of Kilembe catchment sweet potatoes could present risks for children due to excessive Ni intake. Amaranthus also appeared to present a risk to children from Co and Ni and this could be attributed to root uptake during growth but also aerial deposition of the trace elements from tailings and soil dust which may not be washed off completely during food preparation. Of all the foods considered, only bananas revealed no potential hazards to consumers from trace elements for all the Kilembe samples, possibly because of limited trace element uptake or because of reduced contamination of the edible part from extraneous dust. The risks found for all the foods exceeded the earlier levels reported by Mwesigye et al. (2016) partly because the estimated food consumption in the earlier study was less than measured values in this study.

Table 2. Hazard quotients of foods grown within contaminated soils around Kilembe mine

	Co		**Ni**		**Cu**		**Zn**		**Pb**	
RfD (mg kg^{-1} day^{-1})	0.02c		0.02b		0.4a		0.3b		0.0035a	
	children	**adults**	**children**	**adults**	**children**	**adults**	**children**	**adults**	**children**	**adults**
Yams (n=13) cf=0.5	**1.3**	0.62	0.74	0.43	0.28	0.16	**2.2**	**1.2**	0.12	0.04
Maize (n=7) cf=0.43	0.24	0.04	0.20	0.04	0.06	0.04	**1.2**	0.63	0.12	0.06
Cassava (n=13) cf=0.4	0.44	0.24	**1.2**	0.62	**1.60**	0.84	0.28	0.14	0.062	0.04
Potatoes (n=4) cf=0.22	0.82	0.44	**1.03**	0.42	0.14	0.08	0.24	0.14	0.02	0.014
Bananas (n=13) cf=0.33	0.06	0.030	0.26	0.14	0.14	0.04	0.22	0.12	0.04	0.02
Amaranthus (n=18) cf=0.15	**1.6**	0.84	0.46	0.26	0.16	0.08	**1.1**	0.56	0.54	0.28
Beans	0.54	0.32	**1.5**	0.93	0.22	0.13	**1.1**	0.62	0.07	0.04

(n=21) cf= 0.47										
Ground nuts (n=3) cf= 0.4	0.3	0.18	**1.1**	0.6	0.02	0.013	**1.2**	0.68	0.03	0.16

cf = dry weight to fresh weight conversion factor; RfD = reference dose.

a. Hough et al. (2004).

b. US EPA Iris Database (2015).

c. New Jersey Department of Environmental Protection (2008).

Bold figures represent HQs that exceed 1

Locally, most households prepare one main meal comprising 80% carbohydrate foods, such as bananas, cassava, yams, maize and sweet potatoes. The main meal is consumed with a vegetable sauce, made of either beans, ground nuts or amaranthus, which makes up approximately 20% of the meal. Calculations have been developed to establish HQ values for typical meal combinations consumed by locals and the results are shown in Table 3.

Table 3: HQ calculation based on a typical balance of dietary components.

Trace element	**Co**		**Ni**		**Cu**		**Zn**		**Pb**	
Food combinations	**CHL**	**ADL**	**CHL**	**ADL**	**CHL**	**ADL**	**CHL**	**ADL**	**CHL**	**ADL**
Bananas + Beans	0.16	0.08	0.43	0.23	0.15	0.09	0.32	0.24	0.05	0.03
Bananas + Gnuts	0.11	0.06	0.51	0.31	0.12	0.06	0.40	0.26	0.04	0.003
Bananas + Amaranth	0.37	0.21	0.31	0.16	0.14	0.12	0.40	0.21	0.14	0.06
Yams +Beans	**1.15**	0.56	0.82	0.46	0.26	0.16	**1.9**	**1.1**	0.12	0.06
Yams + Gnuts	**1.1**	0.54	0.9	0.54	0.23	0.13	**1.98**	**1.01**	0.11	0.03
Yams +Amaranth	**1.36**	0.67	0.7	0.39	0.25	0.19	**1.98**	**1.1**	0.21	0.1
Cassava + Beans	0.46	0.25	**1.18**	0.62	**1.32**	0.7	0.36	0.25	0.07	0.04
Cassava + Gnuts	0.41	0.23	**1.26**	0.7	**1.29**	0.67	0.44	0.16	0.06	0.03
Cassava + Amaranth	0.67	0.36	**1.06**	0.55	**1.31**	0.73	0.44	0.25	0.16	0.1
Maize + Beans	0.3	0.1	0.38	0.42	0.09	0.06	**1.1**	0.64	0.12	0.08
Maize + Gnuts	0.25	0.07	0.46	0.23	0.06	0.03	**1.18**	0.55	0.11	0.05
Maize +Amaranth	0.51	0.2	0.26	0.08	0.08	0.09	**1.18**	0.61	0.21	0.11
Sweet potatoes + Beans	0.77	0.41	**1.04**	0.46	0.15	0.09	0.34	0.25	0.04	0.02
Sweet Potatoes + Gnuts	0.72	0.39	**1.12**	0.54	0.12	0.06	0.42	0.16	0.03	0.01

Sweet potatoes + Amaranth	**1.1**	0.52	0.92	0.39	0.14	0.12	0.42	0.22	0.13	0.07

Bold figures represent HQs that exceed 1

The combination of Yams with amaranth, ground nuts (Gnuts) and beans presented possible health risks to children through excessive consumption of Co and Zn while adults could also get excessive Zn intakes. A combination of cassava with either beans, amaranth and ground nuts also posed health risks to children due to excessive Ni and Cu intakes. A combination of maize with either beans, amaranth or ground nuts posed health risks to children due to excessive zinc contents in the diets while sweet potatoes consumed with beans, amaranth or ground nuts posed risks to children due to high amounts of Co, Ni and Cu.

3. Conclusions

The findings of this study revealed that most food crops grown in Kilembe contaminated catchment soils accumulated the trace elements Cu, Co, Ni, Pb and Zn to concentrations greater than equivalent crops from control sites. The elevated trace elements found in food crops grown in the Kilembe catchment are known contaminants within the Kilembe area associated with past copper mining and processing activities. Some leafy vegetables of *Amaranthus* species had concentrations of Cu which exceeded recommended human consumption thresholds (20 mg kg^{-1}). In some instances, concentrations of Co, Zn, Cd and Pb in *Amaranthus vegetables* also exceeding human consumption thresholds, suggesting possible exposure of consumers but also suggesting the presence of soil dust despite washing of samples.

Hazard quotients for yams, maize, sweet potatoes and amaranthus suggest that further investigation of the food crops grown in contaminated Kilembe soils is needed with more focus on cultivation approaches and the suitability of specific crops for specific types of location. Leafy vegetables, especially Amaranthus, may pose a particularly strong risk to consumers because of retention of extraneous soil dust which cannot be eliminated completely through washing. This is potentially very dangerous on an ex-mining site with poor vegetative cover on more contaminated areas. Permitting cultivation of selected crops in the right places and avoiding minespoil patches could allow local people to produce less contaminated food. However, community sensitization is needed so as to be able to identify hotspots to avoid during cultivation but also taking measures against soil dust contaminations during food harvesting and preparations. Additional studies are needed in the area so as to design appropriate remediation and phytostabilisation programs on contaminated areas to prevent tailing erosion into agricultural soils and water systems.

Acknowledgements

This study was funded by the UNESCO Abandoned Mines Project in Sub Sahara Africa, supported by the Swedish International Development Cooperation Agency (Sida). Laboratory analysis was sponsored by The University of Nottingham (UK), School of Biosciences.

References

Alloway B.J., 1994. Toxic metals in soil–plant systems. Chichester UK, John Wiley and Sons.

Antonious, G., Kochhar, T., 2009. Mobility of heavy metals from soil into hot pepper fruits: a field study. Bulletin of Environmental Contamination and Toxicology 82, 59-63

Chunilall, V.A., Kindness, and Jonnalagadda S.B.,2005. Heavy Metal Uptake by Two Edible Amaranthus Herbs Grown on Soils Contaminated with Lead, Mercury, Cadmium, and Nickel. Journal of Environmental Science and Health 40, 375–384

Clark, R. B., 1992. Marine Pollution pp. 61-79. Oxford, UK Cleavendo Press.

Datta, S.P., Young, S.D., 2005. Predicting metal uptake and risk to the human food chain from leaf vegetables grown on soils amended by the long-term application of sewage sludge. Water Air Soil Pollution 163, 119–136.

E. C. Commission Regulation No 1881., 2006. Setting maximum levels for certain contaminants in foodstuffs. Official Journal of European Union 346, 18–20.

FAO/WHO., 2001. Joint FAO/WHO Food Standards Programme–Codex Alimentarius Commission.

FAO/WHO Codex Alimentarius Commission., 2011. Working Document for Information and Use in Discussions Related to Contaminants and Toxins in GSCTFF Food Standards Programme ,Rome, Italy

General Standardization Organization (GSO)., 2013. General Standard for contaminants & toxins in food, Gulf standard.

Hough, R.L., Breward, N., Young, S.D., Crout, N.M.J., Tye, A.M., Moir, A.M., Thornton, I., 2004.Assessing potential risk of heavy metal exposure from consumption of homeproduced vegetables by urban populations. Environmental Health Perspectives 112, 215–221

Joy, J.M., Broadley, M.R., Young, S.D., Black, C.R., Chilimba, A.D.C., Ander, E.L., Barlow, S.T., Watts, J.M., 2015.Soil type influences crop mineral composition in Malawi. Science of the Total Environment 505, 587–595.

Kachenko A.G and Singh B., 2005. Heavy metals contamination in vegetables grown in urban and metal smelter contaminated sites in Australia. Water, Air, and Soil Pollution 169, 101–123.

Kim I.S., Kang H.K., Johnson G.P., Lee E.J., 2003. Investigation of heavy metal accumulation in Polygonum thunbergii for phytoextraction. Environmental Pollution 126, 235–43.

Kríbek, B.; Majer, V.; Knésl, I.; Nyambe, I.; Mihaljevic, M.; Ettler, V.; Sracek, O., 2014. Concentrations of arsenic, copper, cobalt, lead and zinc in cassava (Manihot esculenta Crantz) growing on uncontaminated and contaminated soils of the Zambian Copperbelt. Journal of African Earth Science 99, 713–723.

McLaughlin, M. J., Parker, D. R. and Clarke, J. M., 1999. ‘Metals and micronutrients – food safety issues’, Field Crops Research 60, 143–163, 1999.

Mwesigye, A.R., Young, S.D., Bailey, E.H., Tumwebaze, S.B., 2016. Population exposure to trace elements in the Kilembe copper mine area, Western Uganda: a pilot study. Science of the Total Environment 573, 366-375.

Mwesigye A.R and Tumwebaze S.B., 2017. Water contamination with heavy metals and trace elements from Kilembe copper mine and tailing sites in Western Uganda; implications for domestic water quality. Chemosphere 169 281-287.

Nabulo G, Young S.D., Black; C.R., 2010. Assessing risk to human health from tropical leafy vegetables grown on contaminated urban soils. Science of the Total Environment 408, 5338–5351.

Nagajyoti, P.C., Lee K.D, and. Sreekanth S.V.M., 2010. “Heavy metals, occurrence and toxicity for plants: a review”, Environment, Chemical Letters 8, 199-216.

Nesta B.S., Shouta M. M., Nakayama, Osei A., Yoshinori I., Julius N., Fobil B.B., Hazuki M. and Mayumi I. (2015), Accumulation of Heavy Metals and Metalloid in Foodstuffs from Agricultural Soils around Tarkwa Area in Ghana, and Associated Human Health Risks. Int. J. Environ. Res. Public Health, 12, 8811-8827.

New Jersey Department of Environmental Protection (2008. Health Effects Subcommittee Report.

Sawidis, T., Chettri, M. K., Papaionnou, A., Zachariadis, G., & Stratis. J., 2001. A study of Metal Distribution from Lignite Fuels Using Trees as Biological Monitors. Ecotoxicology and Environmental Safety 48, 27-35.

Singh, B., 2001. ‘Heavy metals in soils: sources, chemical reactions and forms’, in: D. Smith, S. Fityus and M. Allman (eds.), Geotechnics: Proceedings of the 2nd Australia and New ZealandConference on Environmental Geotechnics, Newcastle, NSW, Australia, November, pp. 77–93.

US EPA Iris Database, 2015. http://cfpub.epa.gov/ncea/iris/.

Wierzbicka M., 1999. Comparison of lead tolerance in Allium cepa with other plant species. Environmental Pollution104, 41–52.

Wilson, B., Pyatt, F.B., 2007 Heavy metal dispersion, persistence, and bioaccumulation around an ancient copper mine situated in Anglesey, UK. Ecotoxicology and Environmental Safety 66, 224-231.