2016-04

Risk of heavy metals exposure through consumption of rice from Kahama and Geita districts

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https://doi.org/10.58694/20.500.12479/92

RISK OF HEAVY METALS EXPOSURE THROUGH CONSUMPTION OF RICE FROM KAHAMA AND GEITA DISTRICTS

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A Dissertation Submitted in Partial Fulfilment of the Requirements for the Degree of Master's in Environmental Science and Engineering of the Nelson Mandela African Institution of Science and Technology

Arusha, Tanzania

April, 2016

ABSTRACT

Heavy metals exposure is associated with various human health problems. This research aimed at determining the levels of heavy metals in paddy soils and in polished rice from villages around the artisanal gold mining areas in Kahama and Geita districts. It also intended to assess the risk of human exposures of heavy metals through rice consumption around the artisanal gold mining areas in Kahama and Geita districts, Tanzania. Twenty soil samples were collected from paddy fields and 20 polished rice samples grown from those fields were taken from the farmers. An additional 20 polished rice samples were collected from farmers in other villages from the same districts. Information about rice cultivation practices and rice consumption were collected from 40 farmers that were randomly selected, 20 from each of the two districts; Kahama and Geita districts, in which the mining areas are located. Chromium (Cr), copper (Cu), zinc (Zn), Mercury (Hg), cadmium (Cd), nickel (Ni), lead (Pb) and arsenic (As) were determined in the soil and rice samples using Energy Dispersive X–ray Florescence spectrometer. Heavy metals exposure through rice consumption, for each of the forty farmers, was determined using deterministic approach. In all the soil samples, concentrations of Cd and Cr were found to be above the maximum limits of 1 mg/kg and 100 mg/kg, respectively, as set by Tanzania Bureau of Standards (TBS). Concentrations of Pb and Cd in all the rice samples were above the maximum limits set by Codex Alimentarius Commission. Forty percent of the farmers consumed rice at least once daily with per capita consumption of 66.8 g per day for Geita district and 74.0 g per day for Kahama district. According to Joint FAO/WHO Expert Committee on Food Additives (JECFA), the estimated daily intake (EDI) for Cd was found to be above tolerable daily intake (TDI) for 95% of all population in both Kahama and Geita districts while the estimated daily intakes (EDIs) for As, Zn and Cu in both Kahama and Geita population were below the TDIs. The results also showed that sites which are closer to the mining activities had higher heavy metal concentration in both rice and soils compared to the sites further away from the mining activity.

DECLARATION

I, **FIDES SIMON** do hereby declare to the Senate of Nelson Mandela African Institution of Science and Technology that this dissertation is my own original work and that it has neither been submitted nor being concurrently submitted for degree award in any other institution.

Name and signature of candidate	**Date**

The above declaration is confirmed

Name and signature of supervisor	**Date**

Name and signature of supervisor	**Date**

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CERTIFICATION

The undersigned certify that they have read and found that the dissertation conform to the standard and format acceptable for submission, therefore do hereby recommends for acceptance of dissertation entitled "**Risk of heavy metals exposure through consumption of rice from Kahama and Geita districts"**, in fulfilment of the requirements for the degree of Master of Science in Environmental Science and Engineering at Nelson Mandela African Institution of Science and Technology.

Name and signature of supervisor **Date**

Name and signature of supervisor **Date**

ACKNOWLEDGEMENT

Special thanks to my Almighty God who is the source of entire knowledge and wisdom. Thank you God for protection, guidance and strength you provide to me throughout my life.

I express my thankfulness to my supervisors Dr. Kelvin M. Mtei and Dr. Martin Kimanya for their guidance, patient and cooperation. Their contribution, suggestions and constructive criticism ensured the consistency and good shape of this research work.

I would like to thank the government of Tanzania through Nelson Mandela African Institution of Science and Technology (NM-AIST) for funding my studies; it could not be possible to accomplish my studies at NM-AIST without its assistance. I am also indebted to thank the Ilemela municipality for granting me a study leave of two years.

I acknowledge the role played by Miss Tune Kaijage and Ms Furaha Chuma of Tanzania Atomic Energy Commission Laboratory for their support in laboratory analysis of the samples.

Special gratitude and appreciation is owed to my beloved husband Francisco Olambo for his love, care, encouragement and support throughout my studies. I also acknowledge the patience showed by my lovely son Frank Francisco for staying far away from his mother for the entire period of studies.

Finally, I thank everybody who contributed in one way or the other in accomplishing this work successful.

DEDICATION

I dedicate this work to my lovely parents Mr. and Mrs. Simon Mpanduji, my husband Francisco Olambo and my son Frank Francisco. Their prayers, care, inspiration and guidance have become a good support to my life.

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LIST OF ABBREVIATIONS AND SYMBOLS

Abbreviation/symbol	Description
%	Percent
µ/kg Bw/day	Microgram per kilogram body weight per day
ANOVA	Analysis of variance
As	Arsenic
BDL	Below detection limit
Bw	Body weight
Cd	Cadmium
Cons	Average daily per capita
Cr	Chromium
Cu	Copper
EC	European Commission
EDI	Estimated daily intake
EDXRF	Energy Dispersive X-ray Fluorescence Spectrometer
EFSA	European Food Safety Authority
FAO	Food and Agriculture Organization of United Nations
g	Gram
Hg	Mercury
IARC	International Agency for Research Cancer
ILO	International Labour Organization
JECFA	Joint FAO/WHO Expert Committee on Food Additives
Km	Kilometer
mg/Kg	Milligram per kilogram
N	Number of samples
Ni	Nickel
NM-AIST	Nelson Mandela African Institution of Science and Technology
p	P – value
Pb	Lead
PTMI	Provisional tolerable monthly intake
PTWI	Provisional tolerable daily intake
RLDC	Rural Livelihood Development Company

SPSS	Statistical Package for social sciences
TBS	Tanzania Bureau of Standards
TDI	Tolerable daily intake
WHO	World Health Organization of United Nations
Zn	Zinc

CHAPTER ONE
GENERAL INTRODUCTION

This chapter describes the general introduction of the study. It mainly focuses on the background information of the study, the justification of the research problem, research objectives, research questions as well as significance of the study.

1.1 Background information

Rice is the second most important food crop in Tanzania, after maize. It provides the bulk of calories for more than half of the Tanzanian population (RLDC, 2009). In Tanzania rice is normally consumed in the polished (white) grain form. Rice is cultivated in various regions of Tanzania mainly Morogoro, Mbeya, Geita, Shinyanga, Mwanza, Tabora, Rukwa and Manyara (Barreiro-Hurle, 2012). Payus and Talip (2014) report that, rice may be contaminated with heavy metals through the heavy metals present in soil. Heavy metals may be translocated to the rice grains through the roots of rice plants grown in the heavy metals contaminated soils (Deka and Sarma, 2012). Rice can also be contaminated by absorbing heavy metals from the heavy metals polluted air (Lugwisha and Othman, 2014).

Heavy metal contamination in agricultural soil refers to the excessive introduction of toxic heavy metals contained materials in the agricultural soils (Su *et al*., 2014). Agricultural soil is said to be contaminated with heavy metals when its natural condition has been changed due to the presence of these heavy metals beyond the maximum limit (Monitha *et al*., 2012). It has been reported by Vanita *et al*. (2014) that heavy metals contamination in agricultural soils is associated with mining activities and agricultural activities such as the utilization of pesticides, herbicides, fertilizers and other agrochemicals rich in heavy metals. Other sources of heavy metals in the agricultural soils are industrial emission as well as disposal of industrial waste (Deka and Sarma, 2012). Heavy metal contamination in soil may affect plant growth and lead to the decline of crop yield which results to food insecurity (Chibuike and Obiora, 2014; Nnaji and Igwe, 2014). In Tanzania, TBS has set the maximum limits of As, Cd, Cr, Pb, Hg, Ni, Cu and Zn in agricultural soils as; 1 mg/kg, 1 mg/kg, 100 mg/kg, 200 mg/kg, 2 mg/kg, 100 mg/kg, 200 mg/kg and 150 mg/kg, respectively (TBS, 2007).

Heavy metals availability for uptake by plants is reported to be influenced by some soil properties especially pH (Lugwisha and Othman, 2014). As the pH of the soil increases, the heavy metals concentration in plants becomes low (Lugwisha and Othman, 2014).

Globally, the contamination of rice by heavy metals has become a serious problem from year to year. The consumption of rice with heavy metals exceeding the maximum limit may create several health risks to consumers, including cancer which can occur as a result of excessive uptake of dietary heavy metals (Orisakwe *et al*., 2012). Various organs in the world have set standards of heavy metals in rice. For instance Codex Alimentarius Commission (2014) recommended that the level of Pb and Cd in rice should not exceed 0.2 mg/kg and 0.4 mg/kg, respectively. The maximum limit of As in polished rice is 0.2 mg/kg as set by Codex Alimentarius Commission (2014).

In Tanzania, information about heavy metals contamination in agricultural soils and rice as well as risk assessment for heavy metals through rice consumption is still inadequate. Therefore there is a need to research on heavy metals contamination in agricultural soil and rice grown near the mining areas as well as assessing the risk of exposure of heavy metals in rice around artisanal mining areas in Kahama and Geita districts.

1.2 Justification of the Research Problem

Heavy metals are toxic elements which are non – biodegradable and may persist in the environment for a long period of time; they affect both plants and human health (Obasohan, 2008). Heavy metals may enter into human body through air, water and food consumption (Obasohan, 2008). Human exposure to heavy metals may lead to health problems like renal dysfunction, neurological disorders, diarrhea, paralysis and total damage to the brain (Duruibe *et al*., 2007). Chronic exposure to heavy metals may cause cancer of different parts of human body (Vanita *et al*., 2014).

Heavy metals contaminations in soils affect crop production as low concentrations of heavy metals hinder seed germination whereas high concentrations of heavy metals cause over growth of the crops (Pourrut *et al*., 2011). Rice crops are more affected by heavy metals since they are grown under flooded conditions. Flooded fields have high capacity of accumulating nutrients as well as toxic materials (Machiwa, 2010).

As it has been reported by Guan *et al*. (2014) that mining operations near the agricultural sites may influence the increase of heavy metals in soils as well as in food crops, rice grown near mining areas in Tanzania may be at risk of being contaminated with heavy metals. Geita and Kahama districts lie in the Lake Victoria basin. The largest gold producing mines in Tanzania (Bulyanhulu, Buzwagi and Geita gold mines) are located in these districts (Makene *et al*., 2012). In these areas, the artisanal mining is also growing. Moreover, Geita and Kahama are among the main rice producing districts in Tanzania (Barreiro-Hurle, 2012). It is very likely that rice grown in Kahama and Geita districts may be contaminated with heavy metals from these mining sites. Continuous consumption of heavy metals contaminated rice as food may lead to potential hazard to human health. Several studies have reported rice to be a source of heavy metal exposure in China, Indonesia and Japan (Fu *et al*., 2008). In Tanzania only dietary mercury intake of people at Mugusu gold mine village in Geita has been studied (Tungaraza *et al*., 2011). It is, therefore, important to undertake studies on the risk of heavy metals exposure through rice consumption around the mining areas in Kahama and Geita districts.

1.3 Objective of the Study

The general objective of this study was to assess the risk of exposure to heavy metals from rice grown around artisanal mining areas in Kahama and Geita districts.

The specific objectives were;

i. To determine the concentrations of heavy metals namely chromium, copper, zinc, mercury, cadmium, nickel, lead and arsenic in paddy soils.

ii. To determine the relationship between heavy metals content in soils and polished rice for chromium, copper, zinc, mercury, cadmium, nickel, lead and arsenic.

iii. To estimate the risk of human exposures to heavy metals namely copper, zinc, mercury, cadmium, lead and arsenic through rice consumption.

1.4 Research Questions

The study was guided by the following research questions:

(i) What are the concentrations of heavy metals namely chromium, copper, zinc, mercury, cadmium, nickel, lead and arsenic in selected agricultural soils around mining areas in Kahama and Geita districts?

(ii) How is the heavy metals namely chromium, copper, zinc, mercury, cadmium, nickel, lead and arsenic content in soils relate to the same heavy metals concentration in rice grown in these soils?

(iii) What is the risk of heavy metals namely copper, zinc, mercury, cadmium, lead and arsenic exposure to human life through rice consumption?

1.5 Significance of the Study

Heavy metals may cause different health risk to human body. It is important to know the extent of heavy metals contamination in soils as well as in food consumed by majority of people as well as the estimated daily intakes of heavy metals through food consumption. . This study has revealed the extent of heavy metals contamination in paddy soils and rice, and the risk of heavy metals exposure through rice consumption in Kahama and Geita districts. The information from this study might be used by the government and the organs responsible for food safety to monitor and control heavy metals contaminations in soils and rice grown in Kahama, Geita districts and elsewhere. This will reduce human health risk due to heavy metals exposure.

CHAPTER TWO

HEAVY METALS CONTAMINATION IN AGRICULTURAL SOIL AND RICE IN TANZANIA: A REVIEW[1]

Abstract

Heavy metals contamination in agricultural soil is a potential environmental threat to the safety of agricultural food crops such as rice which is consumed by majority of people in Tanzania. The aim of this review was to put together available information on heavy metals contamination in agricultural soils, sources of the heavy metals and the extent of their contamination in rice. The information on risk of heavy metals exposure through rice consumption in Tanzania as well as all the other parts of the world will also be put together. It has been mostly reported that the extent of heavy metals contamination in agricultural soils is influenced by their closeness to the mining and industries areas. The use of wastewater contaminated with heavy metals from industries for irrigation has been reported to increase levels of heavy metals in agricultural soils. The elevated level of heavy metals in agricultural soils led to their accumulation in crops especially rice which upon consumption poses health effects to human and the ecosystem at large. This review suggests the need for determining the extent of heavy metals contamination in agricultural soils around potential areas such as mining and to link this with exposure assessment on heavy metals through rice consumption. This information is necessary to establish the extent at which rice consumers are at risk of heavy metals ingestion.

2.1 Introduction

Heavy metals contamination in the environment should be given much attention by the public and the government since it may cause detrimental effects to plant and human health. Whereas food crops may be exposed to heavy metals through contaminated soil or atmospheric dispersal of heavy metals from industrial areas, human beings may be exposed to heavy metals through consumption of contaminated foods such as rice (Machiwa, 2010).

Heavy metals such as copper (Cu), iron (Fe), manganese (Mn) and zinc (Zn) are considered to be useful micro-nutrients to plants when used in the proper needed amounts which facilitate the physical growth and development of the plants (Aziz *et al*., 2015). When they

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[1] International Journal of Environmental Protection and Policy (2016); 4(1): 16-23

exceed maximum limits they become toxic to plants (Vanita *et al*., 2014). The normal plant growth can be inhibited by high levels of heavy metal in the soil, leading to reduction of crop yield (Chibuike and Obiora, 2014; Nnaji and Igwe, 2014). The heavy metals cadmium (Cd), arsenic (As) and chromium (Cr) have been reported to be more toxic to plants and have no any reported biological functions to plants (Ji *et al*., 2012). Other heavy metals with no reported benefits to plants are lead (Pb), nickel (Ni) and mercury (Hg) (Chibuike and Obiora, 2014). Crops such as rice that are grown in submerged conditions are even more exposed to heavy metal sources both from the soil and the water. The accumulation of heavy metals in the rice fields is of more concern since the metals may enter rice grains through the soil - rice plant system (Zhao *et al*., 2015). Rice being the second most consumed crop in the world may in turn expose the majority of its consumers to the heavy metals (Emumejaye, 2014).

Just like in plants - Fe, Cu, Mn and Zn are useful in humans when used in trace amounts. When they exceed the maximum limits in the body, they pose health problems to human (Fernández-Luqueño *et al*., 2013). Some of these health problems are cancer, hypertension, fever, kidney disorder and DNA damage in living cells (Vanita *et al*., 2014). Heavy metals can also cause modifications in DNA methylations resulting to epigenetic silencing of gene expression (Vanita *et al*., 2014). Just like in plants, some heavy metals namely Cd, As, Cr, Pb and Hg have no any reported biological functions in humans (Ji *et al*., 2012).

The risk of human exposure to heavy metals through food increases when the food is grown on the heavy metals contaminated soils. Agricultural soils may be contaminated by heavy metals through practices such as irrigation and use of heavy metals containing agrochemicals such as pesticides, herbicides and fertilizers (Vanita *et al*., 2014). The highest risk of contamination has been reported for soils around mining operations (Zhuang *et al*., 2009). In Tanzania for instance, artisanal and small scale mining activities are conducted in areas where rice production is also high. Of such areas are Kahama and Geita in the Lake Victoria zone. This suggests that there are high chances for the rice produced from these areas to be contaminated with heavy metals at levels that exceed regulatory limits. Observations made in China by a study conducted around Dabaoshan mine in China reported high levels of Pb and Cd in rice to be 1.44 mg/kg and 0.82 mg/kg, respectively. The study also reported that in paddy soils, high concentration of Cu (276 mg/kg – 703 mg/kg), Zn (181 mg/kg – 1100 mg/kg) and Cd (3.0 mg/kg – 5.5 mg/kg) in agricultural soils of China (Zhuang *et al*., 2009).

Heavy metals from the mining sites may reach agricultural soils through leaching. Also during the rainy season large quantities of tailing and waste containing heavy metals are carried by runoff to the agricultural fields near the mining sites which lead to the elevated levels of heavy metals in the soils (Matthews – Amune and Kakulu, 2012). Due to this reason, protecting the agricultural soils from the heavy metals contamination is an urgent need. However, strategies to protect those areas from heavy metal contamination cannot be formulated when data on heavy metal contamination of agricultural soils in Tanzania are limited and scattered.

2.2 Heavy metals contamination in agricultural soil

Extent of contamination

In soil science perspective, heavy metals contamination in agricultural soil refers to the presence of heavy metals of significant toxicity (Su *et al*., 2014). In Tanzania there are some few available data of heavy metal contamination in agricultural soils. Kibassa *et al*. (2013) reported average concentrations of Zn, Pb, Cr, Cd and Cu in agricultural soils collected from six sites in Dar es Salaam to be 33.18 mg/kg, 14.32 mg/kg, 7.68 mg/kg, 0.22 mg/kg and 5.62 mg/kg, respectively. These sites were chosen based on their proximity to the steel, iron, food and detergents manufacturing industries which drain wastewater into the rivers and streams from which residents draw water for irrigation purposes. The mean concentrations of Cr (0.35 mg/kg), Pb (8.45 mg/kg), Cu (1.69 mg/kg) and Zn (13.9 mg/kg) were also reported for agricultural soil of Lushoto district (Lugwisha and Othman, 2014). A study by Machiwa (2010) in the lake Victoria basin reported mean concentrations of Cd (8.70 mg/kg), Hg (19.99 mg/kg), Pb (19.38 mg/kg), Cr (20.98 mg/kg), Zn (65.46 mg/kg) and Cu (14.58 mg/kg) in paddy soils collected from 18 sites within the basin. The Lake Victoria basin was chosen for the heavy metals investigation based on increasing mining activities, urbanization, industrial and agricultural development. However, all these studies did not assess and report as to whether the levels of heavy metals contamination constitute a significant risk of toxicity in the soils or not. One of the ways to assess the risk of heavy metal toxicity in soils is comparing the contamination levels with the maximum limits for the metals in soils.

As guidance to farmers and agricultural extension officers, Tanzania Bureau of Standards (TBS) sets the maximum limits for heavy metals in agricultural soils. The maximum limits set by the TBS (2007) for Cr, Cu, Zn, Pb, Ni, As and Cd are 100 mg/kg, 200 mg/kg, 150 mg/kg, 200 mg/kg, 100 mg/kg, 1 mg/kg and 1 mg/kg, respectively. There is an urgent need to determine the extent heavy metal contaminations in agricultural soils of Tanzania comply with the TBS set limits. Additionally, a need exist to determine the heavy metal contamination in as many farms as possible to avail sufficient data for risk assessment which will inform policy makers in formulation of strategies to prevent agricultural soils from contamination. In other countries data are available for a considerable part of their farm lands. For instance in Bulgaria 19 360 ha of agricultural soils are known to be contaminated with heavy metals, in Poland, 0.5% of the total agricultural areas is known to be contaminated with heavy metals, in France, approximately 800 000 agricultural sites are known to be contaminated with heavy metals, whereas in Western Europe, up to 1 200 000 agricultural sites are reported to be potentially contaminated with heavy metals (Puschenreiter *et al*., 2005).

2.3 Sources of Contamination

There are several sources of heavy metals contamination in agricultural soil. These include the parent material from which the soil is derived as well as anthropogenic activities (Chen *et al*., 2002). Although most farms in Tanzania are located in contamination prone areas, there are very limited reports on influence of the sources in heavy metal contamination status. Machiwa (2003) reported that many agricultural fields located near mining site in Geita and Tarime districts are contaminated with heavy metals. In Tanzania, major gold mines are located in Geita, Musoma, Kahama, Tarime, Chunya and Mpanda (Tungaraza *et al*., 2011) which are also main food crops producing areas. Therefore, food crops cultivated near these gold mines are vulnerable of contamination with heavy metals. For instance it has been reported that the level of Hg in agricultural soils in Mugusu gold mining site in Geita district, Tanzania is high due to the gold mining activities taking place near the agricultural soils (Kitula, 2006).

Concentrations of heavy metals in areas close to mining operations can exceed maximum limits. It has been recorded that the average concentrations of Cu (502 mg/kg), Zn (498 mg/kg), Pb (278 mg/kg) and Cd (3.92 mg/kg) in Dabaoshan mines in South China were above grade II of environmental quality standards values for agricultural soils in China (Zhuang *et al*., 2009). According to Escarré *et al*. (2011) and Duruibe *et al*. (2007), the levels of heavy metals in the agricultural soils depend on the distance from the source of contamination. Escarré *et al*. (2011) reported that levels of heavy metals in agricultural soils decrease as the distance from the mining sites increase, and proved that high levels of heavy metals were concentrated within 0 – 1.5 km from the mining sites. In Tanzania, data on the extent at which distances from certain mining operations to agricultural sites affect heavy metal contamination are not known. These data are indeed necessary to provide advice to farmers on where to locate their farms in order to prevent heavy metals contamination in the soils.

Another source of heavy metals contamination of the environment is industries. Industries emit heavy metals to the atmosphere which may be deposited on the agricultural soils. The heavy metals originate from uses of different heavy metals as raw materials during the industrial processes (Hu *et al*., 2004). The disposal of industrial waste on or near the agricultural land may lead to the heavy metals concentration on the agricultural soil beyond the maximum limits (Anyakora *et al*., 2013; Deka and Sarma, 2012; Machiwa, 2010). The study by Machiwa (2010) reported that Lake Victoria basin wetlands are highly contaminated with heavy metals through runoff from the industrial waste disposals. Similar observations were made in China, where heavy metals such as Cu, Pb and Zn in the agricultural soil of Xuzhou, Guangzhou and Wuxi were reported to be due to industrial activities such as electroplating plants, spring factory, band steel factory, leather factory and petrochemical complex (Wei and Yang, 2010).

The use of waste water from industries for irrigation may lead to introduction and accumulation of heavy metals in the agricultural soil (Emumejaye, 2014). For examples, the Msimbazi valley in Da es Salaam-Tanzania is highly contaminated with heavy metals from industrial wastewater draining into the valley. Small farmers use this contaminated water from the valley and use it to irrigate vegetable gardens. This may influence the elevated levels of heavy metals in the soils (Kibassa *et al*., 2013). To reduce the effects of heavy metals in agricultural soils, industries and mines operators should adhere to the environment protection standards set by bodies such as TBS in the case of Tanzania.

It has also been found that the sources of Cd, Hg and As in agricultural soil of many locations in China are influenced by excessive use of pesticides and fertilizers (Ji *et al*., 2012; Su *et al*., 2014; Wei and Yang, 2010). In Tanzania, phosphate fertilizers originating from Minjingu phosphate rock which are widely used in agricultural production in Tanzania are reported to be rich in Hg, Cd, As, Pb, Cu and Ni (Lema *et al*., 2014). The use of this kind of fertilizer on agricultural land may act as another source of heavy metals in the soils. Malidareh *et al*.

(2014) investigated the levels of As, Cd and Pb on paddy soils in North of Iran before and after phosphate fertilizer application. The author reported the ranges of these heavy metals in paddy fields before application of chemical fertilizers to be from 0.001 mg/kg – 0.007 mg/kg (As) and 0.066 mg/kg – 0.103 mg/kg (Pb). After applying chemical fertilizers, the level of As ranged from 0.10 mg/kg – 0.30 mg/kg and the level of Pb increased at the range of 0.201 mg/kg – 0.447 mg/kg. Before application of chemical fertilizers, the level of Cd in paddy soils was recorded below the detection limit whereas after application of chemical fertilizers the level of Cd was detected at the range of 0.045 mg/kg to 0.052 mg/kg. The study done by these authors proved that the excessive use of fertilizers may increase the levels of heavy metals on agricultural soils.

2.4 Heavy Metals Contaminations in Rice Grains

In Tanzania, rice is considered as the seventh most important agricultural crop contributing to about 5% of total crop production and is consumed by large population. Rice production in Tanzania has increased from 781 538 tons to 1 341 846 tons from year 2000 to 2007 (Barreiro-Hurle, 2012). Rice is a very important crop in Tanzania as it is considered as most Tanzanian’s food energy supply (Smith and Subandoro, 2007). Rice provides about 8% of the Tanzanian’s calories intake (Barreiro-Hurle, 2012). Although rice is the third energy supplier crop after maize (24.3%) and cassava (10.5%) (FAO, 2013), but it is mostly consumed by large number of people in Tanzania, especially urban areas than other foods (Smith and Subandoro, 2007). Shabbir *et al*. (2013) also report that rice is considered as one of the important agricultural crop in different parts of the world.

Rice can be contaminated by heavy metals when grown in contaminated paddy soils or through irrigating using groundwater and municipal wastewater containing heavy metals (Zhao *et al*., 2015). Heavy metals from the contaminated paddy soils may be taken-up by the rice plant and accumulate in the grains (Nagarajan and Ganesh, 2014; Payus and Talip, 2014).

The heavy metals contaminated atmosphere can also be the source of heavy metals contamination in rice. Heavy metals e.g. Pb, Cr, Mn and Cd from the industries such as Hg from paper industries and vehicle exhaust can pollute air which then can be absorbed directly by the rice grains (Jia *et al*., 2010; Li *et al*., 2008; Lugwisha and Othman, 2014). Just like in the case of agricultural soils, the levels of heavy metals in rice may be influenced by factors such as distance from the source of metals e.g. mining operations, use of pesticides and fertilizers in rice cultivation and distances to the farm from heavy metal waste discharging industries (Lee *et al*., 2001).

Rice is widely grown in wetlands. Heavy metals from mining activities areas and industries may be taken by runoff to the wetlands. Wetlands have high capacity of accumulating heavy metals (Machiwa, 2010). Hence rice crop is more susceptible to heavy metals contamination than other food crops since it is grown in wetlands.

Buzwagi and Bulyanhulu gold mines in Kahama district and Geita gold mine in Geita district are the largest gold producing mines in Tanzania (Makene *et al*., 2012). Geita and Kahama are among the main rice producing districts in Tanzania (Barreiro-Hurle, 2012). As it has been reported that mining operations near the agricultural sites may influence the increase of heavy metals in soils as well as in food crops (Guan *et al*., 2014), rice grown near mining areas in Tanzania may be at risk of being contaminated with heavy metals. Therefore there is a need to research on heavy metals contamination in rice grown near the mining areas.

According to the Codex Alimentarius Commission (2014) heavy metal contamination in rice should not exceed the maximum limits which are; 0.4 mg/kg and 0.2 mg/kg for Cd and Pb, respectively. Furthermore, it is recommended that the levels of As in polished rice should not exceed 0.2 mg/kg (Codex Alimentarius Commission, 2012).

In Tanzania, Machiwa (2010) reported mean concentration of Cu and Zn in rice collected from four different locations of Mwanza, Geita, Bunda and Magu within the Lake Victoria Basin to be 3.7 mg/kg and 21.7 mg/kg, respectively. These levels were associated with the effects of industrialization in that basin. Tungaraza *et al*. (2011) reported average level of Hg (0.026 mg/kg) in rice grown in Mugusu mining area in Tanzania. Data of heavy metal contamination in rice is essential for food safety control organs to be able to perform risk assessments which inform food safety managers on strategies to prevent contamination in rice. However, the available data are inadequate for such risk assessments. This implies that more studies on heavy metals contamination in rice grown in different regions in Tanzania are needed.

2.5 Exposure Assessment for Heavy Metals from Rice

The consumption of heavy metal contaminated rice has detrimental effects to human health (Lugwisha and Othman, 2014). The risk of exposures in humans and the associated health effects depend on the extent of heavy metal contamination in rice and the amount of the rice consumed by an individual. In Tanzania, rice is increasingly becoming a staple food for majority of the people. It has been reported that from the year 2000 to 2007, annual rice consumption per capita has increased from 28 kg to 29 kg, respectively (Barreiro-Hurle, 2012). Similar consumption rates were reported by Mghase *et al*. (2010) who showed that the average daily per capita rice consumption is about 68.5 – 82.2 g/person.

Based on the Joint FAO/WHO Expert Committee on Food Additives (JECFA) (2010), the TDI for Cd, Hg and As are recommended to be 1.0 µg/kg body weight/day, 0.57 µg/kg body Bw/day and (2 - 7 µg/kg body Bw/day), respectively. Tungaraza *et al*. (2011) reported the estimated daily intake (EDI) for Hg in rice grown around Mugusu artisanal gold mining areas in Tanzania to be 4.0 µg/day / person of 68 kg weight which is equivalent to 0.06 µg/ kg body weights/day. Although this value is lower when compared with TDI for Hg set by the JECFA, but the frequency of rice consumption can lead to the increase of EDI to the above TDI. This information is not enough to conclude that Tanzania is not at high risk of heavy metal contamination through rice consumption; more studies should be conducted on risk assessment for heavy metals in rice in Tanzania especially around the mining sites.

2.6 The Influence of Processing Rice on Heavy Metal Contamination

Rinsing and cooking rice using uncontaminated water may decrease the levels of heavy metals in the polished rice. Naseri *et al*. (2014) studied the effects of rice cooking process on the concentrations of heavy metals and found that the raw polished rice had the mean concentrations of Cd (0.33 mg/kg), Pb (1.75 mg/kg), Cr (0.38 mg/kg), Ni (0.89 mg/kg) and Co (0.2 mg/kg) which were higher compared to the levels of heavy metals Cd (0.1 mg/kg), Pb (1 mg/kg), Cr (0.29 mg/kg), Ni (0.19 mg/kg) and Co (0.03 mg/kg) in cooked rice. Therefore, it is advisable that in order to minimize exposure of heavy metals, one should not consume uncooked rice. However, it has been observed that polished rice can accidentally be contaminated with heavy metals during the polishing and cooking process. In the study done by Ziarati and Azizi (2014), arsenic was detected in raw and cooked polished rice while it was not detected in rice husk. This might be due to the unintentionally use of arsenic contaminated water in cooking or the leakage of arsenic from the equipment used during the whole polishing process. In Tanzania, there is no any available information about the extent traditional rice processing and cooking practices can reduce heavy metal contamination. Data on the influence of the practices in heavy metal contamination of rice are important to advise organs responsible for water treatment on whether or not there is need to improve the treatment procedures to ensure that heavy metals in potable water do not exceed the maximum limits.

2.7 Relationship between Heavy Metals Contamination in Agricultural Soils and Rice

Elevated levels of heavy metals in agricultural soils may influence the uptake of heavy metals in rice plant system (Ziarati and Azizi, 2014); although there are factors such as high pH of the soil may hinder availability of heavy metals in soils to be absorbed by plant roots (Lugwsha and Othman, 2014). While Moradi *et al*. (2013) reported a positive correlation ($p < 0.01$) between the levels of Zn in agricultural soil and in rice collected in industrial sites in Iran,Chanda *et al.* (2011) reported that the elevated level of heavy metals contamination in agricultural soil may not necessary lead to the elevated levels of heavy metals in rice. The authors further reported the mean levels of heavy metals in agricultural soils to be 72.03 mg/kg, 38.7 mg/kg and 3.0 mg/kg for Cr, Pb and Hg, respectively, while the concentration of Pb and Cr in rice grown in the same soil were found to be below the detection limits. In Tanzania, there is no adequate information which shows correlation of heavy metals between agricultural soil and rice. There is a need to investigate how heavy metals levels in agricultural soils may be correlated with levels in rice.

2.8 Conclusion

Heavy metal contamination in the environment is of more concern worldwide. The knowledge on the sources of heavy metals contamination in agricultural soils and rice is necessary in order to minimize the possibility of food insecurity as well as human health problems. It is explained that the increase of heavy metals in agricultural soils and rice may be influenced by mining activities done close to the agricultural fields. Heavy metals emitted from industries may be directly absorbed by rice grain, also improper disposal of industrial waste rich in heavy metals on the agricultural land. The use of phosphate fertilizers may increase the levels of heavy metals in the soils and rice as they consist of heavy metals such as Hg, Cd, As, Pb, Cu and Ni. High levels of heavy metals in rice lead to high value of EDI for heavy metals which may pose human health problems, therefore the levels of heavy metals in rice should be monitored as rice is considered as a source of energy for the large population in Tanzania. This review work concluded that in Tanzania, only little information on the heavy metals contamination in agricultural soils and rice is available. Hence, more researches need to be conducted in areas around the probable sources of heavy metals contamination. This will facilitate the investigation of the extent of heavy metals contamination in agricultural soils and rice as well as conducting the exposure assessment for heavy metals through rice consumption.

CHAPTER THREE

HEAVY METAL CONTAMINATIONS IN PADDY SOILS AND POLISHED RICE AND THEIR RELATIONSHIP WITH DISTANCE FROM THE GOLD MINING SITES

Summary

Rice grown in farms that are located near mining operations is at a high risk of contamination with heavy metals from the mining processes. This study investigated the extent of heavy metals (Cr, Cu, Zn, Hg, Cd, Ni, Pb and As) contamination in 20 samples of paddy soils and 20 samples of polished rice. Soil samples were collected at a depth of 0 – 15 cm from paddy fields and rice samples from farmers' residences in Kahama and Geita districts. The concentrations of heavy metals in soil and rice samples were determined using Energy Dispersive X- ray Florescence Spectrometer. The results showed that in paddy soils, the mean concentrations of Cd, Cr, Ni, Cu, Zn, Pb and Hg were 9.81 mg/kg, 150.92 mg/kg, 25.27 mg/kg, 55.23 mg/kg, 57.75 mg/kg, 17.97 mg/kg and 1.57 mg/kg, respectively, in Kahama district, and 11.95 mg/kg, 124.38 mg/kg, 12.66 mg/kg, 55.23 mg/kg, 25.99 mg/kg, 23.46 mg/kg and 1.89 mg/kg, respectively, in Geita district. In Kahama the concentrations of As in paddy soils were found to be below the detection limit at Segese, Malito and Shilela villages. While at Mwendakulima (1.01 mg/kg), Bulyanhulu (2.30 mg/kg), Busulwangili (3.02 mg/kg) and Kakola (58.64 mg/kg) sites the concentrations of As in paddy soils exceeded the maximum limit. In Geita district, whereas the concentrations of As in paddy soils at Nzera, Nyamboge, Bugogo, Lwezera, Igate, Kaduda and Kasesa sites, were below the detection limit, the concentrations of As in paddy soils at Katoma (2.52 mg/kg) and Katoro (5.50 mg/kg) sites exceeded the maximum limit. The concentrations of Cd and Cr in all soil samples were above the maximum limits suggested by Tanzania Bureau of Standards (TBS). In rice samples, the mean concentrations of Cd, Cr, Ni, Cu, Zn, and Pb were 4.60 mg/kg, 20.43 mg/kg, 1.17 mg/kg, 9.33 mg/kg, 13.62 mg/kg and 1.97 mg/kg, respectively, in Kahama and 2.83 mg/kg, 19.39 mg/kg, 1.77 mg/kg, 2.77 mg/kg, 9.45 mg/kg and 2.39 mg/kg, respectively, in Geita. In all rice samples, the concentrations of Cd and Pb were recorded to be above the maximum limits set by Codex Alimentarius Commission. In Kahama the concentrations of As in rice at Isaka, Isagehe, Mwime, Mwendakulima, Segese, Malito and Shilela villages were below the detection limit, while arsenic concentrations at Bulyanhulu (0.10 mg/kg) and Busulwangili (0.20 mg/kg) villages were found to be below the maximum limit set by Codex Alimentarius Commission. The mean concentrations of Cr, Ni, Cu, Zn, As, Pb, Hg and Cd in rice samples are 20.43 mg/kg, 1.17 mg/kg, 9.33 mg/kg, 13.62 mg/kg,

0.59 mg/kg, 1.97 mg/kg, 0.54 mg/kg and 4.60 mg/kg, respectively in Kahama district and 19.37 mg/kg, 1.77 mg/kg, 2.77 mg/kg, 9.47 mg/kg, 0.05 mg/kg, 2.39 mg/kg, 0.34 mg/kg and 2.83 mg/kg, respectively, in Geita district. Among all heavy metals detected in rice samples from Geita and Kahama, only Hg showed significant difference ($p < 0.05$) between the two districts. Generally, sampling sites within 0 – 1.5 km from the mining sites were found to have higher concentrations of heavy metals than sites far away from the mining sites. This suggests the mining activities to be the likely potential source of heavy metals contaminations in agricultural soils from the sampled sites in Kahama and Geita districts.

3.1 Introduction

Heavy metals contamination in rice is a potential threat to human health. Rice is used as staple food in various countries in the world (Chamannejadian *et al*., 2013; da Silva *et al*., 2013; Mehrnia, 2013). Consumption of heavy metals contaminated rice may result in health problems such as neurotoxic effects, destruction of genes and cardiovascular diseases (Batista *et al*., 2012). Heavy metals can also cause kidney damage, diabetes, anaemia, diarrhea, headache and reduction of hearing capacity (Lokeshappa *et al*., 2012). Some heavy metals such as cadmium, arsenic and copper are categorized in carcinogenic group 2A, meaning that there is inadequate evidence of their carcinogenicity (IARC, 2006).

Heavy metals from paddy soils may translocate to the rice grains through soil – rice plant systems. Several studies have revealed the significant relationship between heavy metals contamination in soil and rice systems (Aziz *et al*., 2015; Chanda *et al*., 2011; Zhao *et al*., 2015; Moradi *et al*., 2013).

Heavy metals contamination in agricultural soil may be due to both natural conditions and anthropogenic activities (Chopra *et al*., 2009; Singh, 2015; Dobran and Zagury, 2006). Natural conditions include; biological activity, weathering, decomposition of parent material and volcanic activities (Rahman *et al*., 2014) whereas anthropogenic sources include industrial activities and the application of fertilizers and pesticides rich in heavy metals during agricultural activities as well as mining activities (Sherene, 2010). Heavy metals such as Cu, Cr, As, Hg, Pb, Zn, Ni and Cd may reach paddy soils through emissions from rapidly expanding mine tailings, disposal of metal wastes as well as atmospheric deposition of heavy metals from the mines (Lee *et al*., 2001). Lin (1998) reported that mining activity is one of the factors for Pb pollution in a downtown central Sweden whereby fine Pb particles are spread by wind from mining waste heap to the surrounding areas.

In Tanzania, the information on heavy metals contamination in paddy soils or rice in the vicinity of mining operations is still inadequate. This chapter reports investigation on heavy metals contents in paddy soils and rice around mining sites in Kahama and Geita districts in Tanzania. The heavy metals investigated include chromium (Cr), nickel (Ni), zinc (Zn), copper (Cu), arsenic (As), lead (Pb), cadmium (Cd) and mercury (Hg) which were selected due to their toxicity to human being. The information in this chapter aims at helping organs responsible for environmental protection and food safety to establish means of reducing heavy metals contamination in rice and other food crops, to reduce human health problems which may be posed by the elevated levels of heavy metals in the environment.

3.2 Materials and Methods

3.2.1 Study area

The study was conducted in areas around Geita gold mine in Geita district and around Bulyanhulu and Buzwagi gold mines in Kahama district. Geita and Bulyanhulu are the two largest gold producing mines in Tanzania while Buzwagi is the fourth largest gold mine (Makene *et al.*, 2012). These areas are also among the main rice producing districts in Tanzania (Barreiro-Hurle, 2012). Geita district is located at latitude -3°12’0” and longitude 31°54’0”. Kahama district is located at latitude -3.6667° and longitude 33.5000°. Farms were selected from villages which were located near distance from the mining sites. Farms selected from villages around Geita gold mining site (with distance from the mining site in brackets) were; Katoma (5.7 km), Busanda (15.4 km), Igate (26.9 km), Katoro (30.1 km), Kasesa (32.3 km), Kaduda (33.3 km), Nzera (34.1 km), Lwezera (35.4 km), Nyamboge (36.0 km) and Bugogo (38.6 km) (Figure 3.1). In Kahama district the villages selected with the distances from Bulyanhulu or Buzwagi gold mine in brackets were Bulyanhulu (0.8 km), Mwendakulima (1.3 km), Kakola (1.6 km), Busulwangili (2.6 km), Mwime (4.7 km), Isagehe (12.1 km), Shilela (26.4 km), Malito (31.0 km), Segese (32.4 km) and Isaka (33.3 km) (Figure 3.2). The farms in Geita are further from the mining site as compared to Kahama because of the large area fencing. The distances were obtained using ArcGIS 10.2 software through point distance analysis tool. The tool determines the distances from input point features to all points in the near features within a specified search radius.

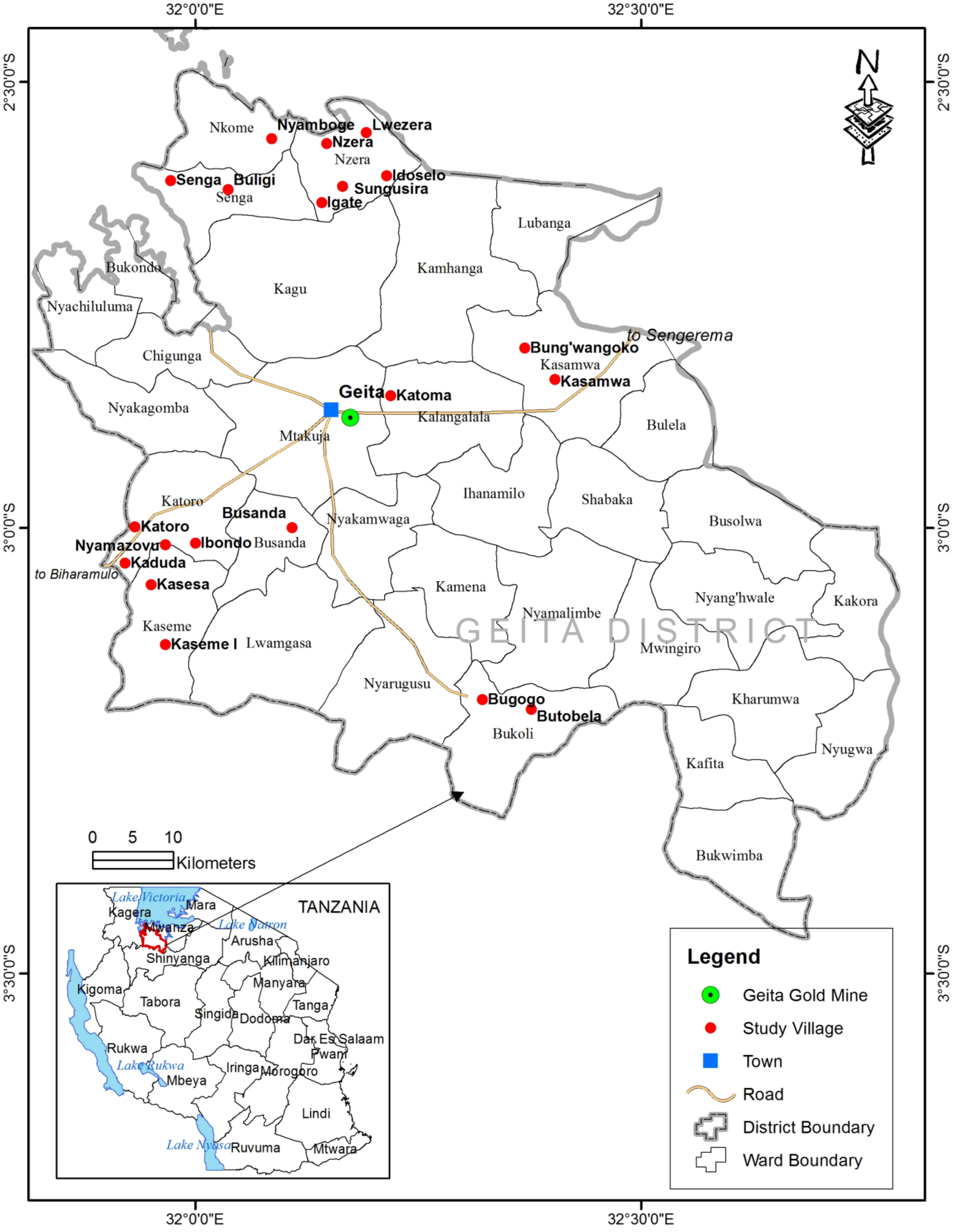


Figure 3.1: Map showing sampling sites around Geita gold mine in Geita district.

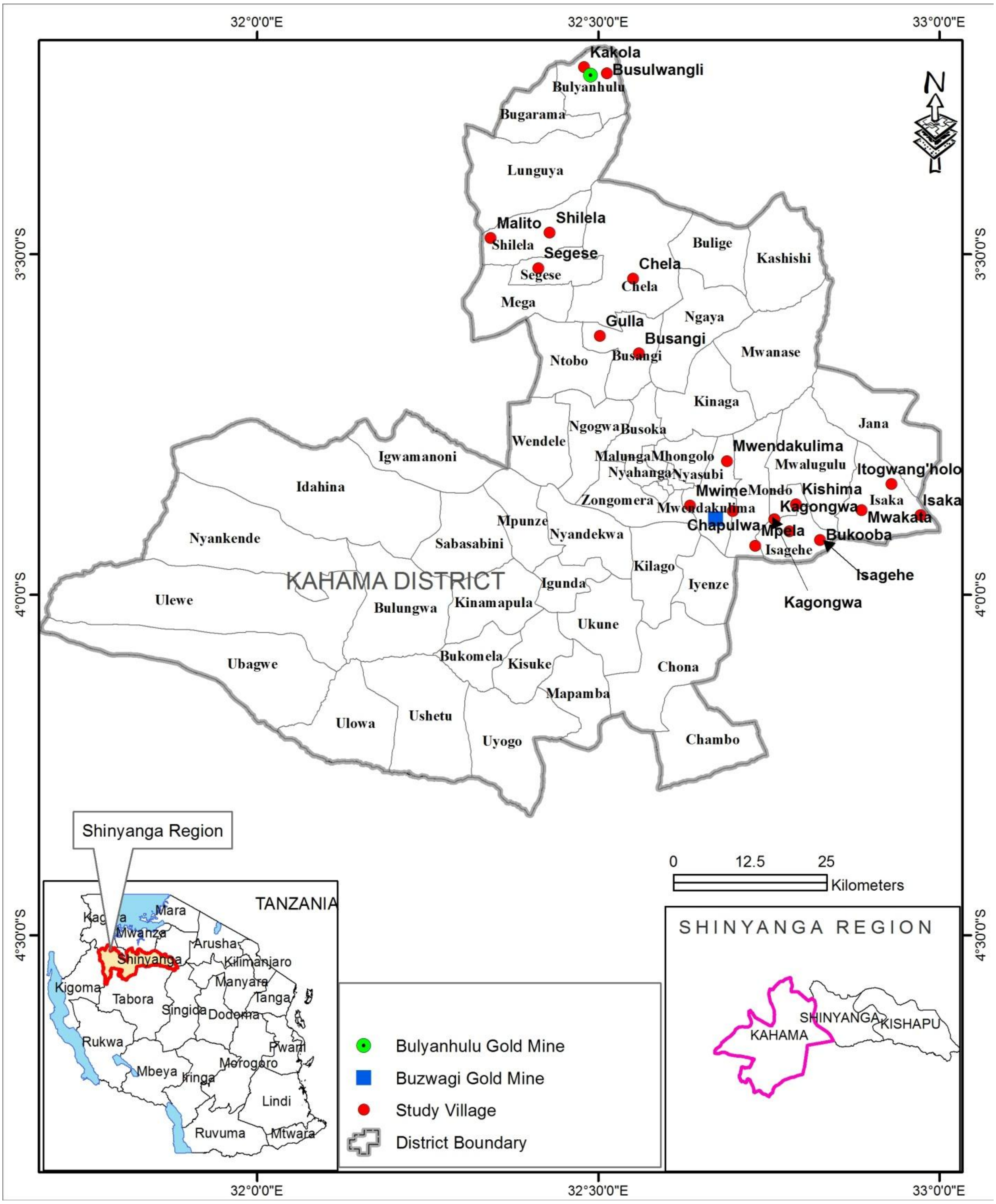


Figure 3.2: Map showing sampling sites around Bulyanhulu and Buzwagi gold mines in Kahama district.

3.2.2 Sampling and sample preparation

Twenty composites paddy soil samples and twenty polished rice samples were collected from the selected farms/villages around Geita, Buzwagi and Bulyanhulu gold mines. Paddy soils were collected at a depth of 0-15 cm. Composite soil samples were formed after mixing soils collected from five different points in each paddy field. Vegetation and other non-soil materials were removed. Soil samples were immediately sundried and stored in polyethylene bags (Santos - Santos *et al.*, 2010). Polished rice samples were collected from farmers' residences in same villages as soil samples. Sand and other non-rice materials were removed and the rice was kept in polyethylene bags.

3.2.3 Laboratory analysis

Soil and rice samples analysis was done at the Tanzania Atomic Energy commission laboratory using the method of Energy Dispersive X-ray Florescence (EDXRF). Before analysis, soils or rice samples were dried at a temperature of 60 °C for 72 hours in an oven (Heraeus Thermo Scientific, model: DIN12880). The samples were then ground using a porcelain mortar and pestle and sieved by 150 µm diameter sieves. Dry weight of 6 g was then measured using beam balance (Sartorius, model: cp 3245) then mixed with 1.35 g cellulose binder. The mixture was placed in a grinding bowl with four spherical balls of 3 mm radius and fixed to Pulverisette 6 Planetary mono-mill for further grinding and homogenization at a speed of 180 revolutions per minutes for 15 minutes. Homogenized sample was pressed into a pellet by using Hydraulic Press (Retsch, model: pp 25) at a pressure of 15 tons. The pellets were placed in a sample holder in EDXRF spectrometer (Bench top Specto Xepos, model XEP01). The pellets were energized for 15 minutes with 50 Kilo volts from inbuilt x-tube in the spectrometer. Detection limits of spectrometer for As, Cu, Ni, Hg, Cd, Cr, Pb and Zn were 0.02 mg/kg, 0.04 mg/kg, 0.01 mg/kg, 0.03 mg/kg, 0.07 mg/kg, 0.09 mg/kg, 0.05 mg/kg and 0.06 mg/kg, respectively.

3.2.4 Quality assurance and control

Quality assurance for the analysis was carried out by determination of elemental composition in certified standard reference materials, namely 2711a Montana Soil II and1568b Rice Flour. The deviation from actual values and measured values of standard reference materials were used in computing measured values of the samples in study to obtain the true elemental concentration of the samples.

3.2.5 Statistical Analysis

The data were analyzed by excel software for mean and standard deviation of heavy metals concentrations as well as for single factor analysis of variance heavy metals contents between Kahama rice and Geita rice. Pearson correlation between heavy metal concentrations in the paddy soil and rice samples was determined using the SPSS-software statistics version 21.

3.3 Results and discussion

3.3.1 Heavy metals concentration in paddy soil and polished rice

3.3.1.1 Contamination of heavy metals in paddy soil and polished rice

In both Kahama and Geita districts, the ranges of heavy metals in soils samples were 104.58 mg/kg – 250.29 mg/kg (Cr), 0.18 mg/kg – 73.45 mg/kg (Ni), 7.93 mg/kg – 297.84 mg/kg (Cu), 6.60 mg/kg – 176.66 mg/kg (Zn), BDL – 58.64 mg/kg (As), 9.69 mg/kg – 31.65 mg/kg (Pb), 1.12 mg/kg – 2.86 mg/kg (Hg) and 1.04 mg/kg – 17.67 mg/kg (Cd) as shown in Table 3.1 and 3.2.While the ranges of heavy metals in rice samples were 12.73 mg/kg – 47.37 mg/kg, 0.17 mg/kg – 10.13 mg/kg, 0.59 mg/kg – 62.4 mg/kg, 0.68 mg/kg – 19.66 mg/kg, BDL – 5.60 mg/kg, 0.27 mg/kg – 5.07 mg/kg, BDL – 1.13 mg/kg, 1.24 mg/kg – 17.97 mg/kg for Cr, Ni, Cu, Zn, As, Pb, Hg and Cd, respectively as shown in Tables 3.3 and 3.4. The analysis of variance results revealed that only the concentration of Hg in Kahama rice showed significant difference ($p < 0.05$) with the concentration of Hg in Geita rice. Table 3.6 shows the percentage of the soil and rice samples exceeding the maximum limit of heavy metals.

Table 3. 1: Heavy metals concentrations in paddy soils collected from villages around Bulyanhulu and Buzwagi gold mine in Kahama district

Village	**Cr**	**Ni**	**Cu**	**Zn**	**As**	**Pb**	**Hg**	**Cd**
				(mg/kg)				
Isaka	116.28	11.73	15.08	46.43	0.83	18.94	1.41	10.48
Bulyanhulu	226.61	31.67	43.27	176.66	2.30	25.09	1.99	12.64
Isagehe	115.30	17.08	19.48	39.43	0.76	11.32	1.74	3.36
Mwime	161.79	37.57	28.79	21.10	0.29	18.52	1.48	12.07
Mwendakulima	241.59	73.49	76.83	54.55	1.01	25.93	1.69	11.14
Segese	118.45	12.57	16.69	43.48	BDL	14.30	1.25	6.61
Malito	120.35	12.34	11.61	19.36	BDL	9.69	1.25	10.87
Shilela	112.58	7.31	9.51	13.25	BDL	10.76	1.38	1.04
Kakola	164.47	24.74	297.84	134.87	58.64	31.65	2.12	12.18
Busulwangili	131.73	24.16	33.24	28.41	3.02	13.47	1.58	17.67
TBS Limit	100	100	200	150	1	200	2	1

BDL: Below detection limit

Table 3. 2: Heavy metals concentrations in paddy soils collected from villages around Geita gold mine in Geita region.

Village	**Cr**	**Ni**	**Cu**	**Zn**	**As**	**Pb**	**Hg**	**Cd**
					(mg/kg)			
Nzera	113.96	9.89	17.15	46.25	BDL	19.25	1.97	11.16
Nyamboge	109.19	5.28	9.34	15.07	BDL	27.88	1.79	11.97
Bugogo	109.39	5.57	10.08	16.59	BDL	18.19	2.07	10.56
Busanda	110.88	10.76	17.44	29.12	0.50	27.44	2.40	14.26
Lwezera	110.84	8.31	11.67	21.68	BDL	24.56	1.30	7.73
Igate	110.53	0.18	7.93	18.06	BDL	26.15	1.87	12.49
Katoro	250.29	61.83	86.20	45.80	5.50	29.73	2.35	14.21
Katoma	116.99	13.11	38.99	21.59	2.52	26.57	2.86	13.55
Kaduda	104.58	8.89	16.09	39.11	BDL	20.95	1.20	12.10
Kasesa	107.18	2.80	8.53	6.60	BDL	13.83	1.12	11.50
TBS Limit	100	100	200	150	1	200	2	1

BDL: Below detection limit

Table 3.3: Heavy metals concentrations in rice collected from villages around Bulyanhulu and Buzwagi gold mine of Kahama district.

Village	**Cr**	**Ni**	**Cu**	**Zn**	**As**	**Pb**	**Hg**	**Cd**
					(mg/kg)			
Isaka	17.57	0.70	2.89	16.42	BDL	4.8	0.53	1.53
Bulyanhulu	36.53	0.87	3.32	6.01	0.10	0.73	1.00	4.01
Isagehe	18.23	0.87	4.41	15.70	BDL	1.6	0.87	3.68
Mwime	14.53	1.00	3.20	9.66	BDL	1.4	BDL	1.41
Mwendakulima	18.10	0.80	3.59	17.68	BDL	0.8	0.60	1.83
Segese	24.93	2.13	3.59	7.54	BDL	2.73	0.27	2.27
Malito	15.87	1.10	4.33	16.42	BDL	0.93	0.67	4.36
Shilela	12.73	0.47	1.64	4.23	BDL	0.8	0.40	6.60
Kakola	27.67	2.60	62.40	27.00	5.60	5.07	0.54	2.33
Busulwangili	18.10	1.17	3.90	15.50	0.20	0.8	0.48	17.97
Codex limit	-	1.5	20	50	0.2	0.2	-	0.4

BDL: Below detection limit

Table 3.4: Heavy metals concentrations in rice collected from villages around Geita gold mine

Village	**Cr**	**Ni**	**Cu**	**Zn**	**As**	**Pb**	**Hg**	**Cd**
					(mg/kg)			
Nzera	14.9	0.33	1.72	16.8	BDL	1.13	1.13	3.68
Nyamboge	14.93	0.37	1.01	3.31	BDL	0.27	0.47	5.07
Bugogo	19.73	0.23	1.6	7.17	BDL	2.53	0.27	3.86
Busanda	12.8	1.63	1.91	9.76	BDL	3.6	0.27	4.21
Lwezera	16.07	1.43	2.42	5.29	BDL	2.47	BDL	2.21
Igate	15.77	0.17	0.59	9.53	BDL	3.93	0.47	4.15
Katoro	47.37	10.13	10.18	19.66	0.38	4.2	0.33	1.38
Katoma	18.53	1.8	5.62	10.07	0.14	2.4	0.47	1.27
Kaduda	17.63	1.27	2.03	12.22	BDL	2.47	BDL	1.27
Kasesa	16.17	0.37	0.62	0.68	BDL	0.87	BDL	1.24
Codex limit	-	1.5	20	50	0.2	0.2	-	0.4

BDL: Below detection limit

Table 3.5: Correlations of heavy metals in paddy soils and rice in Geita and Kahama districts

	Pearson correlation (r - value)	
Heavy metals	Geita	Kahama
Cr	0.977**	0.499
Ni	0.990**	-0.107
Cu	0.984**	0.974**
Zn	0.929**	0.16
As	0.987**	0.993**
Pb	0.645*	0.651*
Hg	0.338	0.362
Cd	-0.032	0.371

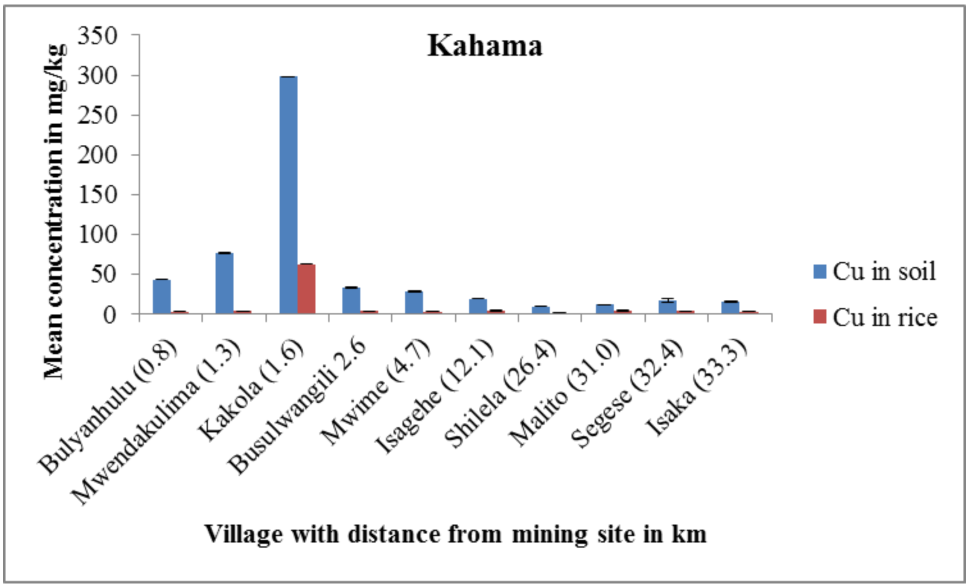
**Correlation is very significant ($p \leq 0.01$)

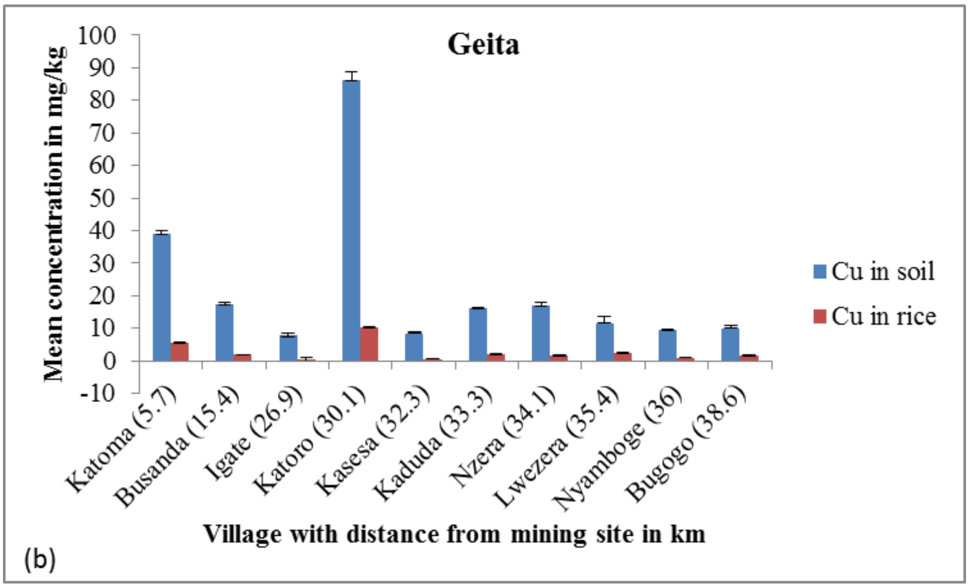
*Correlation is significant ($p \leq 0.05$)

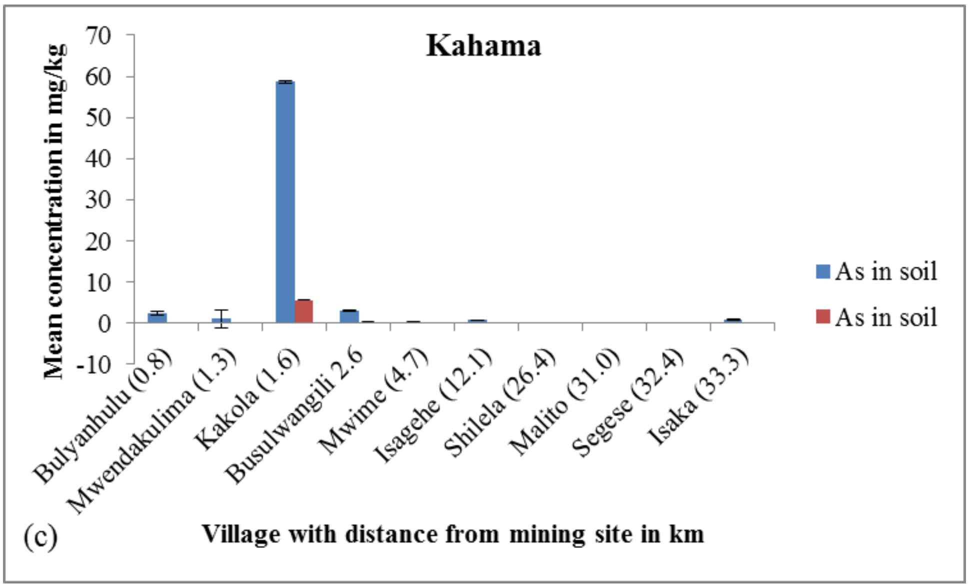
Table 3.6: Percentage of all soil and rice samples exceeding the maximum limit for stated metals

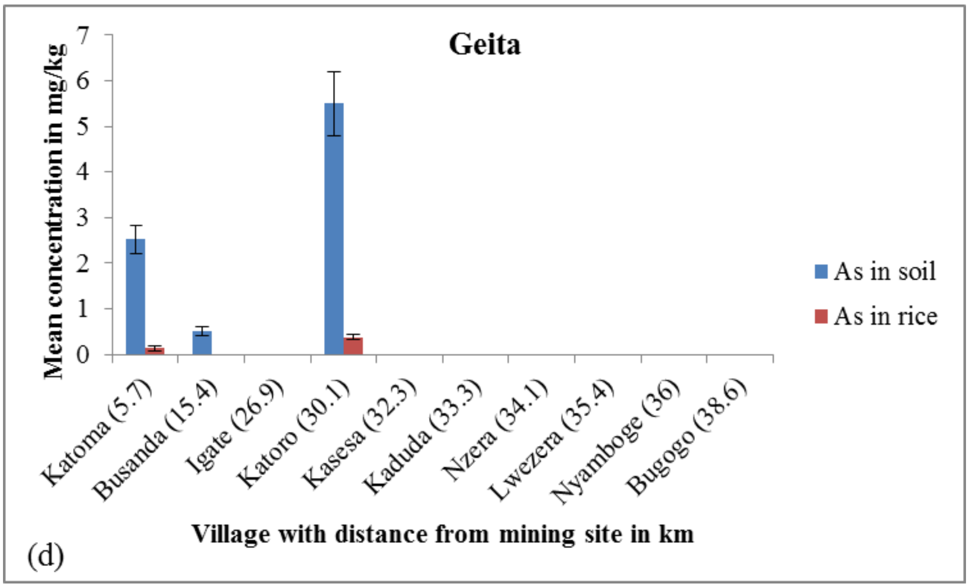
	Cu	As	Pb	Cd
Rice samples exceeding the Codex Alimentarius Commission maximum limit (%)	5	5	100	100
Soil samples exceeding Tanzania maximum limit (%)	0	30	0	100

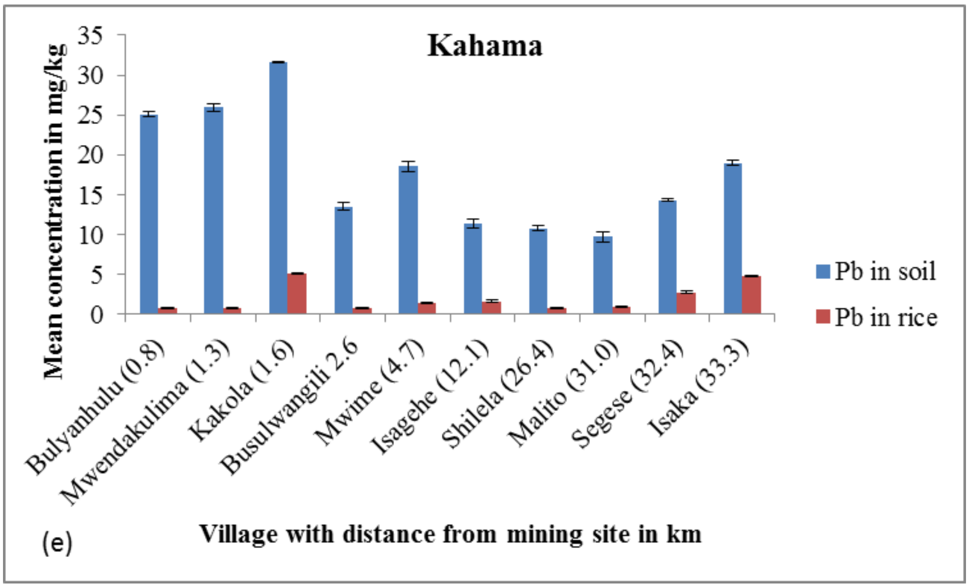
In Figure 3.3 (a-h) the relationship between the heavy metals concentration in soil and the heavy metals concentration in rice is shown. The graphs indicate that the higher the concentrations of heavy metal in the soil, the higher the concentration of that heavy metal in the rice. This suggests that the contamination of rice by most of the heavy metals studied is from soil.

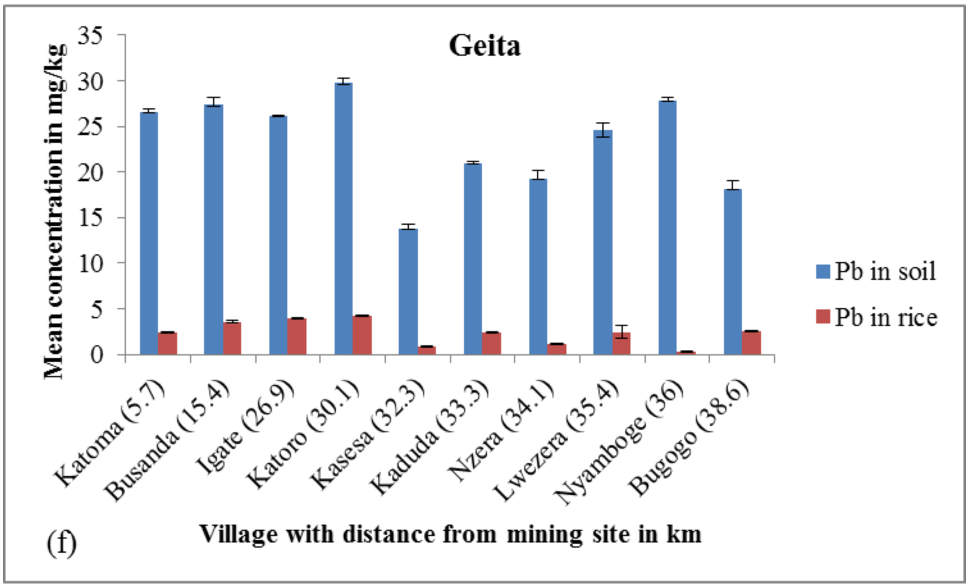

Kahama
Mean concentration in mg/kg
350
300
250
200
150
100
50
0
Bulyanhulu (0.8)
Mwendakulima (1.3)
Kakola (1.6)
Busulwangili 2.6
Mwime (4.7)
Isagehe (12.1)
Shilela (26.4)
Malito (31.0)
Segese (32.4)
Isaka (33.3)
Village with distance from mining site in km
Cu in soil
Cu in rice


Geita
Mean concentration in mg/kg
100
90
80
70
60
50
40
30
20
10
0
-10
Katoma (5.7)
Busanda (15.4)
Igate (26.9)
Katoro (30.1)
Kasesa (32.3)
Kaduda (33.3)
Nzera (34.1)
Lwezera (35.4)
Nyamboge (36)
Bugogo (38.6)
Village with distance from mining site in km
Cu in soil
Cu in rice
(b)


Kahama
Mean concentration in mg/kg
70
60
50
40
30
20
10
0
-10
Bulyanhulu (0.8)
Mwendakulima (1.3)
Kakola (1.6)
Busulwangili 2.6
Mwime (4.7)
Isagehe (12.1)
Shilela (26.4)
Malito (31.0)
Segese (32.4)
Isaka (33.3)
Village with distance from mining site in km
As in soil
As in soil
(c)


Geita
Mean concentration in mg/kg
0
1
2
3
4
5
6
7
As in soil
As in rice
Katoma (5.7)
Busanda (15.4)
Igate (26.9)
Katoro (30.1)
Kasesa (32.3)
Kaduda (33.3)
Nzera (34.1)
Lwezera (35.4)
Nyamboge (36)
Bugogo (38.6)
Village with distance from mining site in km
(d)


Kahama
Mean concentration in mg/kg
0
5
10
15
20
25
30
35
Pb in soil
Pb in rice
Bulyanhulu (0.8)
Mwendakulima (1.3)
Kakola (1.6)
Busulwangili 2.6
Mwime (4.7)
Isagehe (12.1)
Shilela (26.4)
Malito (31.0)
Segese (32.4)
Isaka (33.3)
Village with distance from mining site in km
(e)


Geita
Mean concentration in mg/kg
0
5
10
15
20
25
30
35
Pb in soil
Pb in rice
Katoma (5.7)
Busanda (15.4)
Igate (26.9)
Katoro (30.1)
Kasesa (32.3)
Kaduda (33.3)
Nzera (34.1)
Lwezera (35.4)
Nyamboge (36)
Bugogo (38.6)
Village with distance from mining site in km
(f)

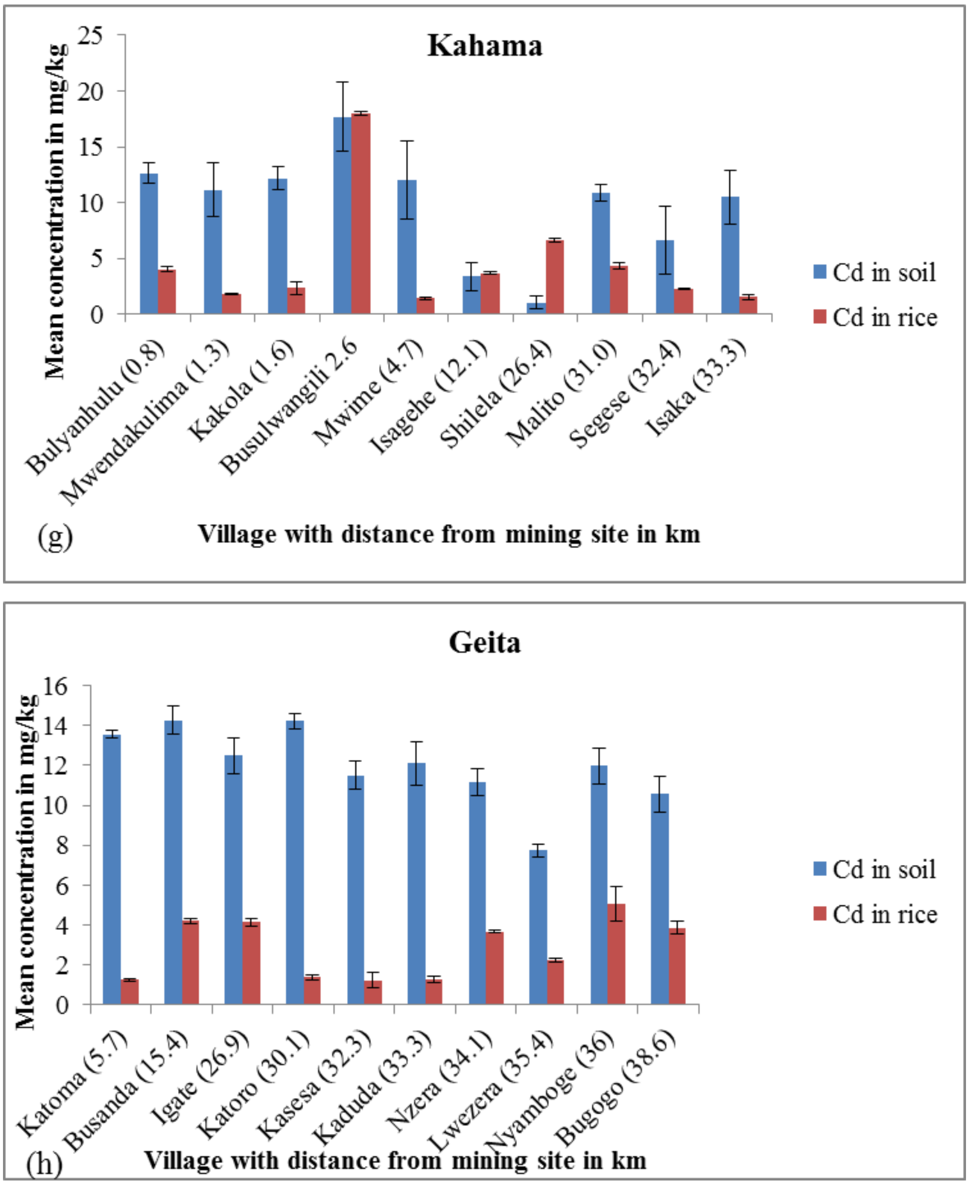


Figure 3.3 (a-h): Relationships between mean levels of heavy metals in soil and level of heavy metals in rice collected from villages in Kahama and Geita districts.

3.3.1.2 Metal specific information

Chromium

For paddy soils samples, the results for heavy metals concentration in Kahama district (Table 3.1) indicate that; the mean concentration of Cr was 150.92 ± 47.78 mg/kg. As shown in Figure 3.4, the maximum concentration of Cr (241.59 mg/kg) was recorded at Mwendakulima site which is located 1.3 km from Buzwagi gold mine and the minimum concentration (112.58 mg/kg) was found at Shilela which is located 26.4 km from

Bulyanhulu gold mine. The concentration of Cr in polished rice ranged from 12.73 mg/kg (Shilela) to 36.53 mg/kg (Bulyanhulu) with the mean concentration of 20.43 ± 7.23 mg/kg in Kahama districts (Table 3.3).

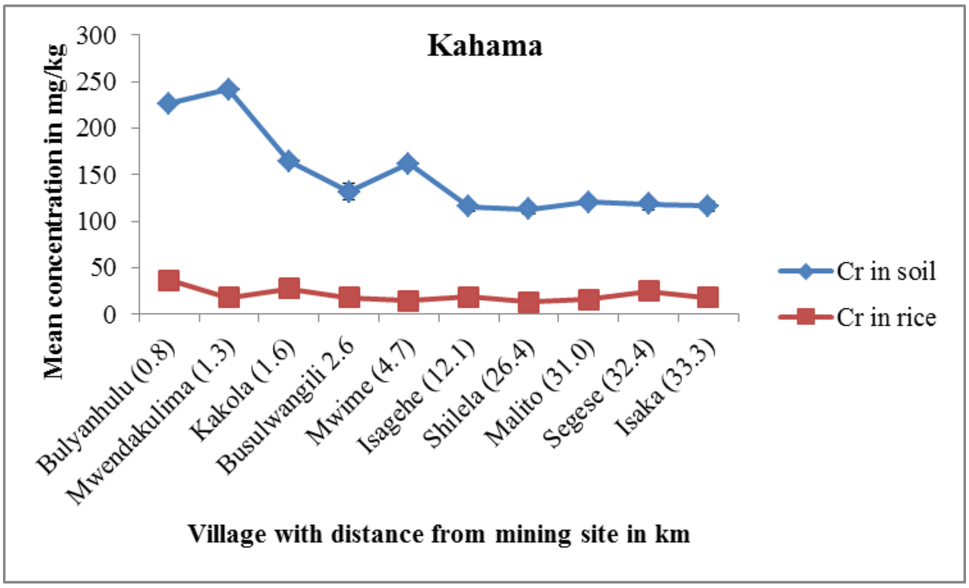


Figure 3.4: The effect of distance from mining site on Cr contamination to paddy soil and rice in Kahama district

The higher concentration of Cr paddy soil at Mwendakulima as compared to other villages in Kahama district may be caused by its closeness to Buzwagi gold mine. Mwendakulima is located 1.3 km from Buzwagi gold mine which might have led to leakage of heavy metals from the gold mine to Mwendakulima paddy soils. The higher contamination of Cr to rice from Bulyanhulu and Kakola as compared to other villages in Kahama district might be caused by the closeness to Bulyanhulu gold mine.

In Geita district results, the mean concentration of Cr in paddy soils samples was 124.38 ± 44.37 mg/kg. The maximum concentration of Cr (250.29 mg/kg) was recorded at Katoro, 30.1 km from Geita gold mine and the minimum concentration (104.58 mg/kg) was determined at Kaduda, 33.3 km from Geita gold mine. The level of Cr in rice ranged from 12.8 mg/kg (Busanda) to 47.37 mg/kg (Katoro) with the mean concentration of 19.39 ± 10.03 mg/kg as shown in Table 3.4. The level of Cr in paddy soils and rice from Geita district with the distances from the mine are as shown in Figure 3.3.

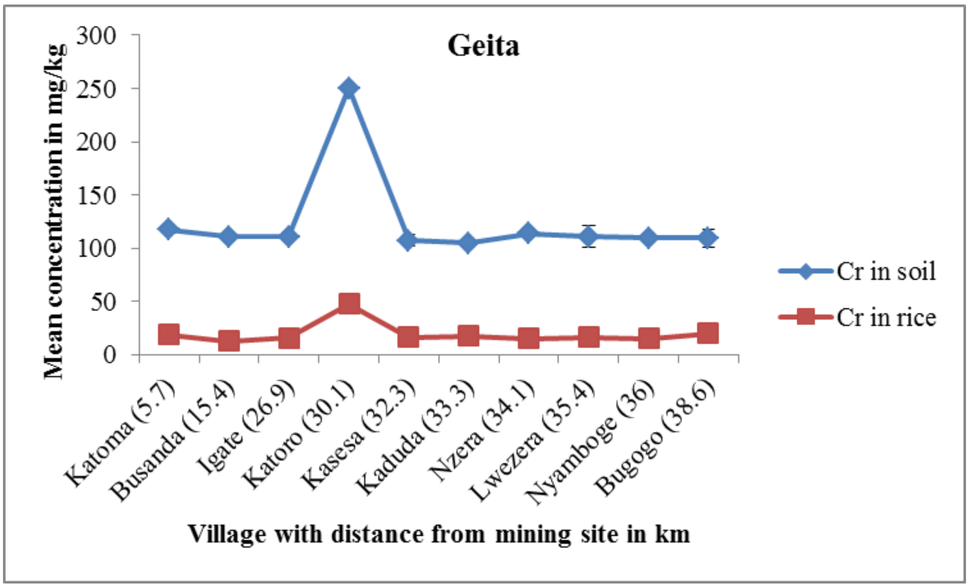


Figure 3.5: The effect of distance from mining site on Cr contamination to paddy soil and rice in Geita district

In Geita district, farms in Katoro found 30. 1 km from Geita gold mine was reported to have the highest level of Cr (250.29 mg/kg). The reasons for the high Cr concentration in soil and rice from Katoro village might be due to small scale mining activities which were taking place at the village (He *et al*., 2013). According to TBS (2007), the level of Cr in all soil samples collected from both Kahama and Geita districts exceeded the maximum limit which is 100 mg/kg in agricultural soil. The concentration of Cr in soils higher than the results found in this study was reported by Aziz *et al*. (2015) in paddy soils samples from contaminated field at Ranau valley, Sabah as (590.80 - 7272.24 mg/kg).

Pearson correlation analysis showed significant correlation at a level of 0.01 between the level of Cr in paddy soils and in rice in Geita as shown in Table 3.5. The levels of Cr in rice in Bulyanhulu, Segese and Kakola might not be influenced by high level of Cr in paddy soil since the concentration of Cr in paddy soils and in rice in Kahama were not significantly correlated. The reason for high level of Cr in rice in these sites might be due to the direct absorption of Cr from the contaminated atmosphere, as reported by Machiwa (2010) that heavy metals disperse in the atmosphere.

Chromium is considered as toxic element in the environment. In plants, it leads to the decline of seed germinations and affects the growth of plant roots which results to poor plant growth and hence low yields (Iyaka, 2009). Through soil-plant system, Cr may enter food chain and cause health impairment to human being (Nagarajan and Ganesh, 2014; Barouchas *et al*., 2014). Consumption of Cr contaminated rice will certainly lead to health effects as Cr is considered to be carcinogenic (Barouchas *et al*., 2014). Emumejaye (2014) reported the concentration of Cr in brands of rice imported from different countries and consumed in Delta State, Nigeria to be 1.15 mg/kg (Thailand) and 0.12 mg/kg (Nigeria).

Nickel

In soil samples, the concentration of Ni in Kahama ranged from 7.31 mg/kg at Shilela (26.4 km) to 73.45 mg/kg at Mwendakulima (1.3 km) with the mean concentration of 25.27 ± 19.45 mg/kg. The mean concentration of Ni in rice in Kahama district was 1.17 ± 0.67 mg/kg. The maximum concentration of Ni was recorded at Kakola (2.60 mg/kg) while the minimum concentration of Ni in rice in Kahama district was recorded at Shilela (0.47 mg/kg). The Ni concentration in rice from Kakola (2.60 mg/kg) and Segese (2.13 mg/kg) were above the maximum limit suggested by Codex Alimentarius Commission. Figure 3.6 shows the effect of distance from the mining site to the contamination of Ni in paddy soil and rice.

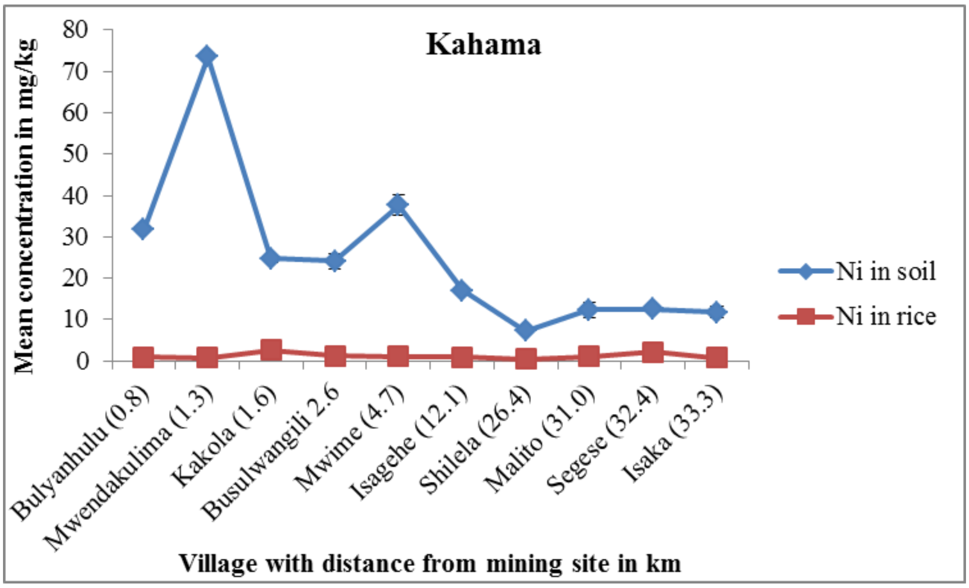


Figure 3.6: The effect of distance from mining site on Ni contamination to paddy soil and rice in Kahama district

While in Geita district the level of Ni ranged from 0.18 mg/kg at Igate, 26.9 km from Geita gold mine to 61.83 mg/kg at Katoro, 30.1 km from Geita gold mine with the mean concentration of 12.66 ± 17.70 mg/Kg. The level of Ni in Geita district with the distances from the mine is shown in Figure 3.7. In Geita district, the concentration of Ni in polished rice ranged from 0.17 mg/ kg (Igate) to 10.13 mg/kg (Katoro) with the mean concentration of 1.77 ± 3.00 mg/kg as shown in Table 3.4. The Ni concentration in rice from Katoro (10.13 mg/kg), Katoma (1.8 mg/kg) and Busanda (1.63 mg/kg) were above the maximum limit of 1.5 mg/kg as suggested by Codex Alimentarius Commission.

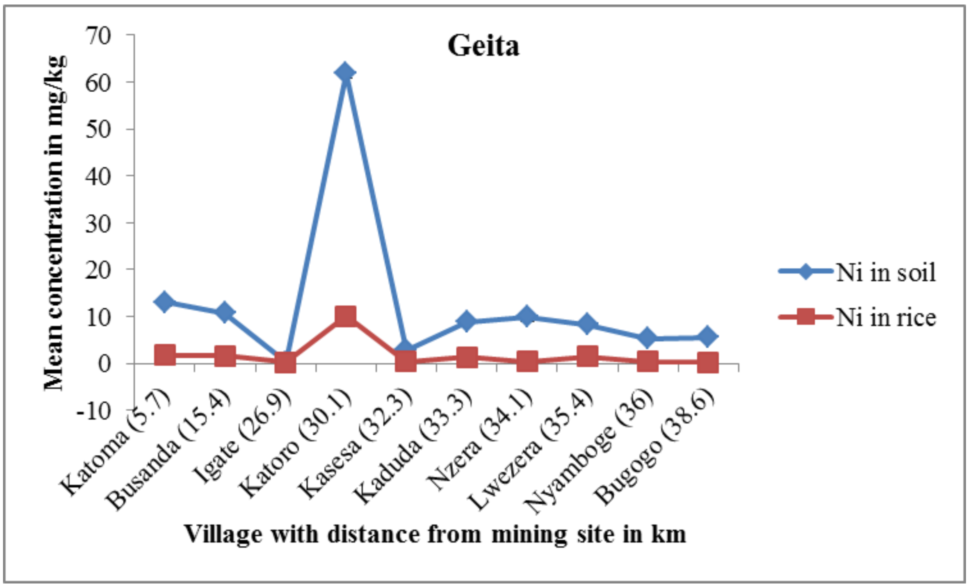


Figure 3.7: The effect of distance from mining site on Ni contamination to paddy soil and rice in Geita district

The probable reason for high levels of Ni in soil and rice sample at Katoro might be due to the artisanal mining activities done near the sampling site. All soil samples studied were found to have less Ni concentration than maximum limit of 100 mg/kg (TBS, 2007). The results suggest that the concentration of Ni in paddy fields in Kahama and Geita districts should be controlled to avoid the increase of the concentration of Ni above the maximum limit. In other study (Emumejaye, 2014), the concentration of Ni in brands of rice imported from Thailand and consumed in Delta State, Nigeria was reported to be 2.11 mg/kg, 2.37 mg/kg, 1.84 mg/kg, 1.05 mg/kg and 6.05 mg/kg. Pearson correlation analysis showed significant correlation at a level of 0.01 between the level of Ni in paddy soils and in rice in Geita whereas in Kahama the correlation was not significant as shown in Table 3.5.

Nickel is considered as carcinogenic element. High level exposure of Ni to human being may cause depression, heart attacks, haemorrhages, kidney damage, decrease in blood pressure, nausea as well as paralysis (Das *et al*., 2008).

Copper

Figure 3.8 shows that the level of Cu in the paddy soils ranged between 9.51 mg/kg at Shilela, 26.4 km from Bulyanhulu gold mine to 297.84 mg/kg at Kakola, 1.6 km from Bulyanhulu gold mine in Kahama district. The concentration of Cu in rice from Kahama district ranged from 1.64 mg/kg (Shilela) to 62.4 mg/kg (kakola) with mean concentration of 9.33 ± 18.66 mg/kg.

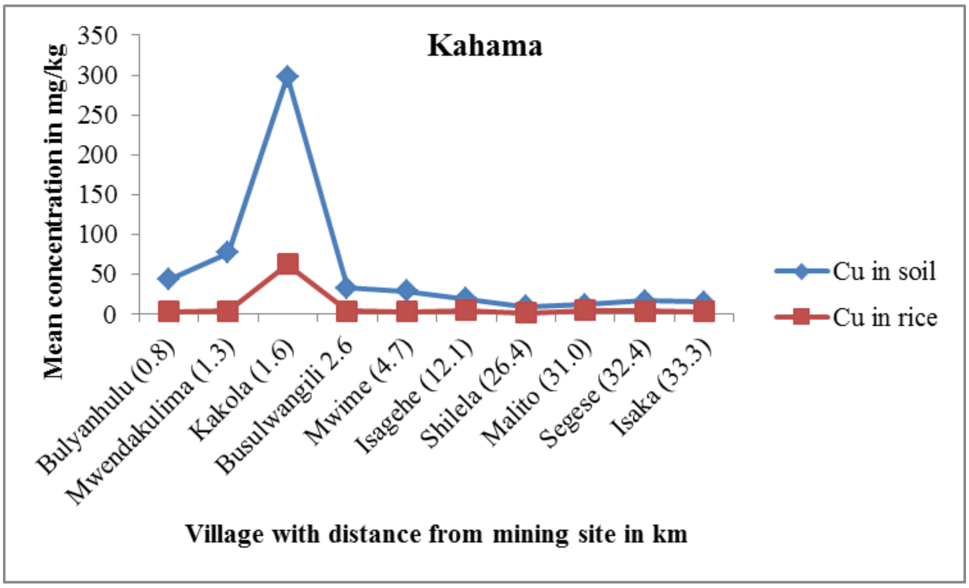


Figure 3.8: The effect of distance from mining site on Cu contamination to paddy soil and rice in Kahama district

These results indicated that paddy soil from Kakola has Cu concentration above the maximum limit of 200 mg /kg (TBS, 2007). Apart from being close to the Bulyanhlu gold mine, some small scale mining activities are conducted at Kakola village. Small scale miners used to wash their working instuments and the sand assumed to contain gold using paddy water as well as stagnant water near the paddy fields. Therefore the eleveted level of Cu in soil samples at Kakola might be due to the effects of mining (Zhuang *et al*., 2009).

All rice samples in Kahama recorded Cu level below the maximum limit of 20 mg/kg set by Codex Alimentarius Commission except at Kakola (62.4 mg/kg). The elevated level of Cu at Kakola might be due mining effects. This is similar to the information given by Zhuag *et al*. (2009) that the elevated level of heavy metals (Cu, Zn, Pb and Cd) in soils, vegetables and rice grown in Quangdong, South China was the effect of mining activities at Dabaoshan mine. Consumption of rice from Kakola may result to copper related health problems such as anaemia, acne, adrenal hyperactivity, depression, diabetes, headache, hypertension as well as panic attack (Lokeshappa *et al.*, 2012). Exposure to high level of Cu may result to Wilson's disease to human being (Vanita *et al.*, 2014)

Figure 3.9 shows that in Geita the high level of Cu (86.20 mg/kg) were found at Katoro, 30.1 km from Geita gold mine and low level of Cu 7.93 mg/kg was found at Igate, 26.9 km from Geita gold mine. In Geita district the concentration of Cu in rice ranged from 0.59 mg/kg (Igate) to 10.18 mg/kg (Katoro).

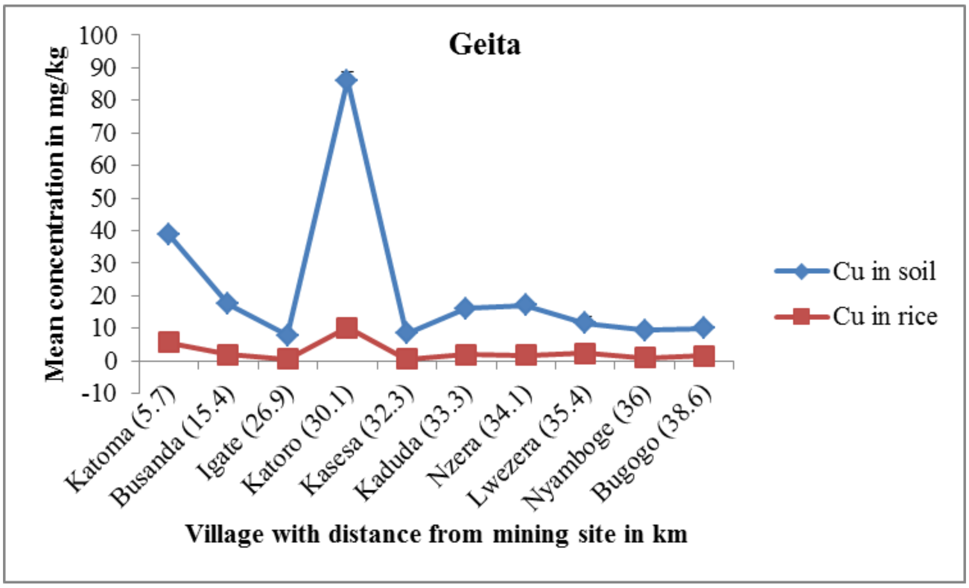


Figure 3.9: The effect of distance from mining site on Cu contamination to paddy soil and rice in Geita district

The concentration of Cu in all soil samples of Geita district were below the maximum limit of 200 mg kg (TBS, 2007). All rice samples in Geita also recorded Cu level below the Codex Alimentarius Commission maximum limit of 20 mg/kg (Machiwa, 2010). The level of Cu in the paddy soils in this study were higher compared to that reported by Machiwa (2010) which ranged from 6.4 mg/kg to 22.8 mg/kg.

In Pearson correlation results, it has been observed that in both Kahama and Geita districts the concentration of Cu in paddy soils was strongly correlated with the concentration of Cu in the rice samples at significant level of 0.01 as shown in Table 3.5. This suggested that high level of Cu in rice sample at Kakola was influenced by eleveted level of Cu in paddy soils.

Similar results were reported by Fong *et al*. (2015) in Japan that the availability of micronutrient in soil influences their concentration in rice grain.

Zinc

Figure 3.10 indicate that the concentration of Zn in soil samples from Kahama district ranged from 13.25 mg/kg at Shilela, 26.4 km from Bulyanhulu gold mine to 176.66 mg/kg at Bulyanhulu, 0.8 km from Bulyanhulu gold mine. The concentration of Zn in rice ranged from Shilela (4.23 mg/kg) to Kakola (27.00 mg/kg) with the mean concentration of 13.62 ± 6.18 mg/kg (Table 3.3).

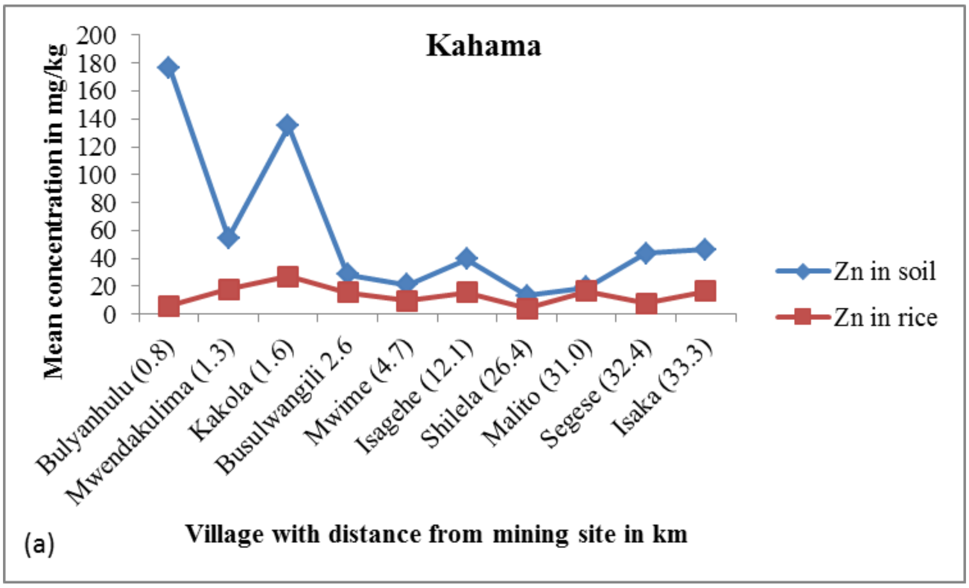


Figure 3.10: The effect of distance from mining site on Zn contamination to paddy soil and rice in Kahama district

Only soil sample from Bulyanhulu had above acceptable limit of Zn in agricultural soils of 150 mg/kg as stated by TBS (2007). The probable cause of high concentration of Zn in this site might be due to drainage from mining operations in Bulyanhulu gold mine which is just 0.8 km from the sampling site. This value is lower compared to the study done by Zhuang *et al*. (2009) around the Dabaoshan mine in China which ranged from181 mg/kg to 1100 mg/kg. Yadav *et al.* (2014) reported the mean level of Zn in agricultural soils irrigated with wastewater from natural drainage to be 1.62 mg/kg. The level of Zn above the maximum limit indicates that the soil is highly contaminated with Zn (Moradi *et al*., 2013).

The concentration of Zn in soil samples from Geita ranged from 6.60 mg/kg at Kasesa, 32.3 km from Geita gold mine to 46.25 mg/kg at Nzera, 34.1 km from Geita gold mine with the mean concentration of 25.99 ± 13.62 mg/kg (Table 3.2 and Figure 3.11). The concentration of Zn in rice (Geita district) ranged from 0.68 mg/kg (Kasesa) to 19.66 mg/kg (Katoro) with the mean concentration of 9.45 ± 5.81 mg/kg (Table 3.4 and Figure 3.11).

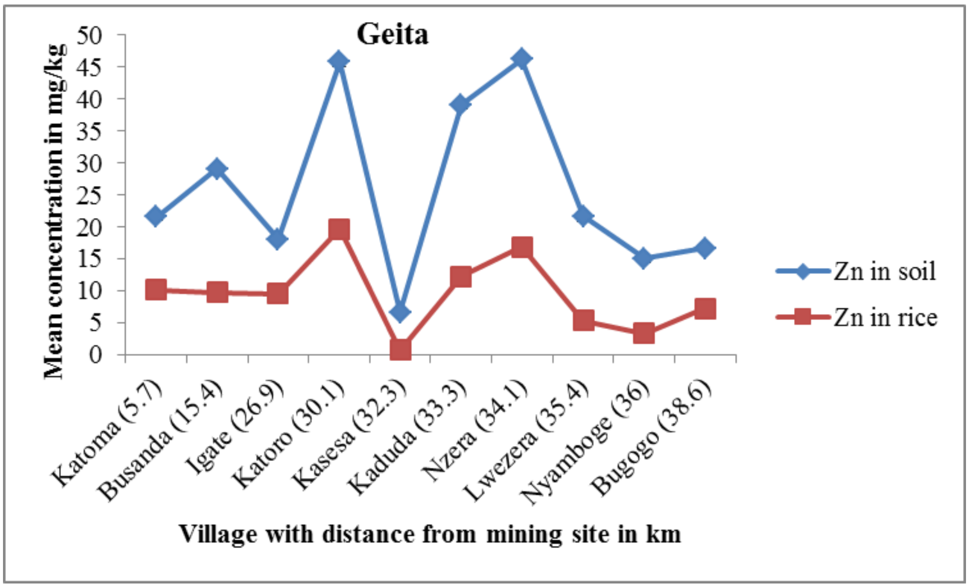


Figure 3.11: The effect of distance from mining site on Zn contamination to paddy soil and rice in Geita district

The levels of Zn in all soil samples from Geita were below the maximum limit stated by TBS (2007). According to Codex Alimentarius Commission, the maximum limit of Zn in rice is 50 mg/kg (Machiwa, 2010). All rice samples in both Kahama and Geita districts were found below the maximum limit. Consumption of rice from these sampled sites may not result to the Zn related health problems since the concentrations of Zinc range within maximum values. Yadav *et al.* (2014) reported the mean level of Zn in rice (0.84 mg/kg) irrigated with wastewater from natural drainage. Zinc is considered as essential heavy metal when present in trace amount. When it exceeds the maximum limit it becomes toxic to living organisms (Vanita *et al.*, 2014). Pearson correlation analysis showed significant correlation at a level of 0.01 between the level of Zn in paddy soils and in rice in Geita as shown in Table 3.5.

Arsenic

From results given in Table 3.1, in Kahama district, the mean concentration of As in paddy soils was 6.69 ± 18.28 mg/kg. The maximum concentration of As (58.64 mg/kg) was recorded at Kakola, 1.6 km from Bulyanhulu gold mine while the concentration of As at Malito (31.0 km), Segese (32.4 km) and Shilela (26.4 km) were below the detection limit.

Concentration of As in Kahama district in the sampling sites with the distance from the mines were as shown in the Figure 3.12.

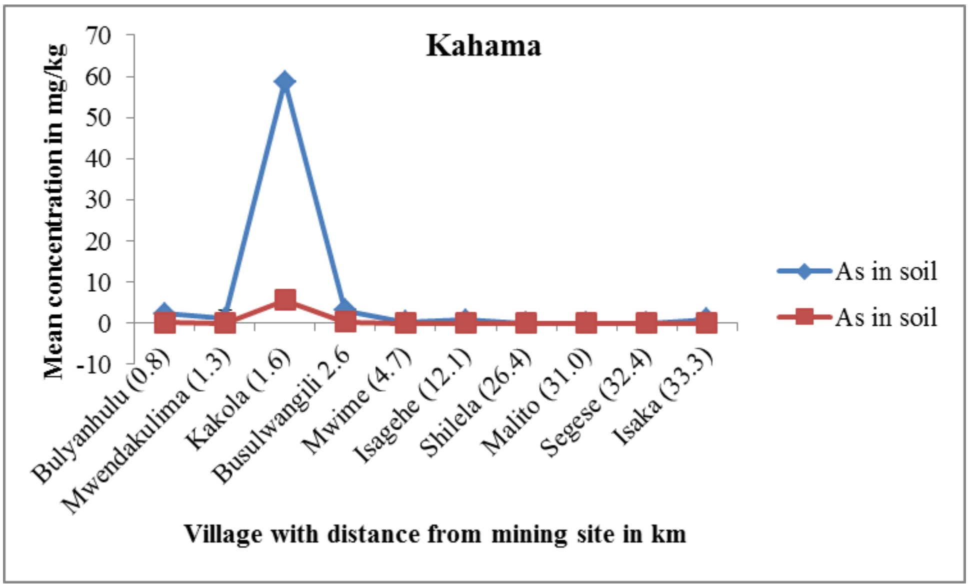


Figure 3.12: The effect of distance from mining site on As contamination to paddy soil and rice in Kahama district

The highest arsenic concentration in soil at Kakola (58.64 mg/kg) in Kahama district is of more concern in soil as well as in human life. According to TBS (2007), the maximum limit of As in agricultural soils is 1 mg/kg. The probable cause for high level of As in Kakola might be due to the leakage and drainage from mining activities taking place at Bulyanhulu gold mine. Kakola is found closer to the Bulyanhulu gold mine at a distance of 1.6 km. Other studies have reported several effects of elevated arsenic levels in soil. For instance; Santos-santos *et al*. (2006) reported high level of As in agricultural soil (182.41 mg/kg) in Zacateas-Mexico which was due to the mining activities. Cao *et al*. (2003) reported that many sites in different countries are highly contaminated with arsenic concentration greater than 26.5 mg/kg. Banejad and Olyaie, (2011) and Srivastava *et al*. (2013) reported that high level of As in paddy soil may result to reduction of rice production. Brammer and Ravenscroft (2009) reported that at a range of 26.3 to 57.5 mg/kg As level in paddy soil in Bangladesh lead to the decrease of rice production from 8.92 t/ha to 2.99 t/ha.

In Kahama district the concentration of As in rice was detected at Kakola (5.60 mg/kg), Busulwangili (0.20 mg/kg) and Bulyanhulu (0.10 mg/kg), while concentration of As in 70%

of all rice samples in Kahama district were recorded to be below the detection limit. (Figure 3.12).

As shown in Table 3.2 and Figure 3.12, the mean concentration of As in soil was 0.85 ± 1.81 mg/kg in Geita district. The maximum concentration of As (5.50 mg/kg) was recorded at Katoro, 30.1 km from Geita gold mine while the concentration of As at Nzera, Nyamboge, Bugogo, Lwezera, Igate, Kaduda and Kasesa were below the detection limit.

In Geita district the concentration of As in rice was detected at Katoro (0.38 mg/kg) and Katoma (0.14 mg/kg) only, while the concentration of As in 80% of all rice samples were recorded below the detection limit of the instrument (Figure 3.13).

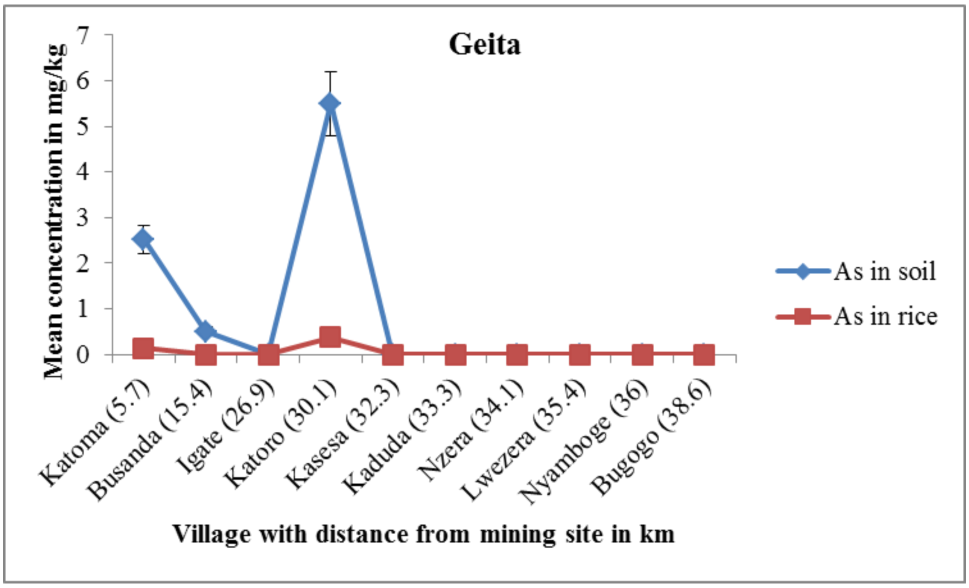


Figure 3.13: The effect of distance from mining site on As contamination to paddy soil and rice in Geita district

Rice is considered to have high capacity of arsenic accumulation especially under the flooded conditions (Bhattacharya *et al*., 2010). It is reported that rice is a potentially important route of human exposure to arsenic especially in populations with rice based diets (Bhattacharya *et al*., 2010). According to Codex Alimentarius Commission (2012) only rice samples at Kakola (5.62 mg/ kg) in Kahama district and Katoro (0.38 mg/kg) in Geita district were found to be above the maximum limit of 0.2 mg/kg. The concentration of As in rice ranged from 0.06 to 0.78 mg/kg was reported in West Bengal by Bhattacharya *et al*. (2010). Even Huang *et al*. (2013) reported high level of As in rice of Zheiang, China. Arsenic exposure to human being at high or low level may cause serious health problems such as kidney and liver failure, weakness and peripheral neutitis. Arsenic is also considered as carcinogenic

(Lokeshappa *et al*., 2012; Morton and Mason, 2006; Navas-Acien and Guuallar, 2008; Rauf *et al*., 2011; Reimann *et al*., 2009; Singh, 2015).

Pearson correlation results indicated that, in both Geita and Kahama districts, the levels of As in paddy soils were strongly correlated with the levels of As in rice samples at significance level of 0.01 as shown in Table 3.5. This suggests that, the elevated level of As in rice at Kakola might be influenced by the elevated levels of As in paddy soils. Similarly it was reported by Zhao *et al*. (2015) that the accumulation of heavy metals in the soil leads to high distribution of heavy metals to the rice plants especially in the rice grains.

Lead

In paddy soils, the levels of Pb ranged from 9.69 mg/kg at Malito, 31.0 km from Bulyanhulu gold mine to 31.65 mg/kg at Kakola, 1.6 km from Bulyanhulu gold mine with the mean concentration of 17.97 ± 7.46 mg/kg in Kahama district as shown in Table 3.1. The level of Pb with the distances from the mine is as shown in the Figure 3.14.

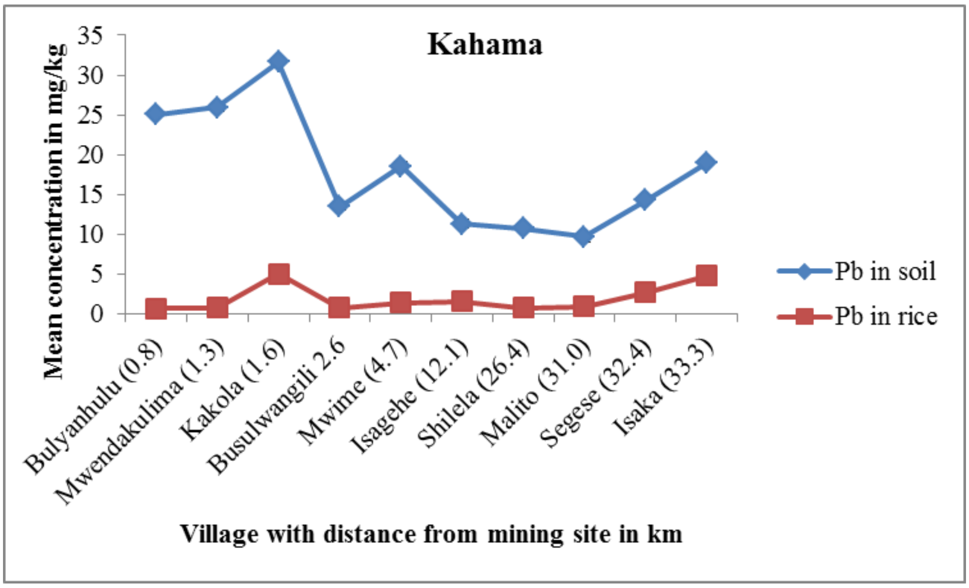


Figure 3.14: The effect of distance from mining site on Pb contamination to paddy soil and rice in Kahama district

The level of Pb ranged from 13.83 mg/kg at Kasesa, 32.3 km from Geita gold mine to 29.73 mg/kg at Nzera, 34.1 km from Geita gold mine with the mean concentration of 23.46 ± 5.14 mg/kg in Geita district (Table 3.2). The concentrations of Pb with the distances from Geita gold mine is shown in the Figure 3.15.

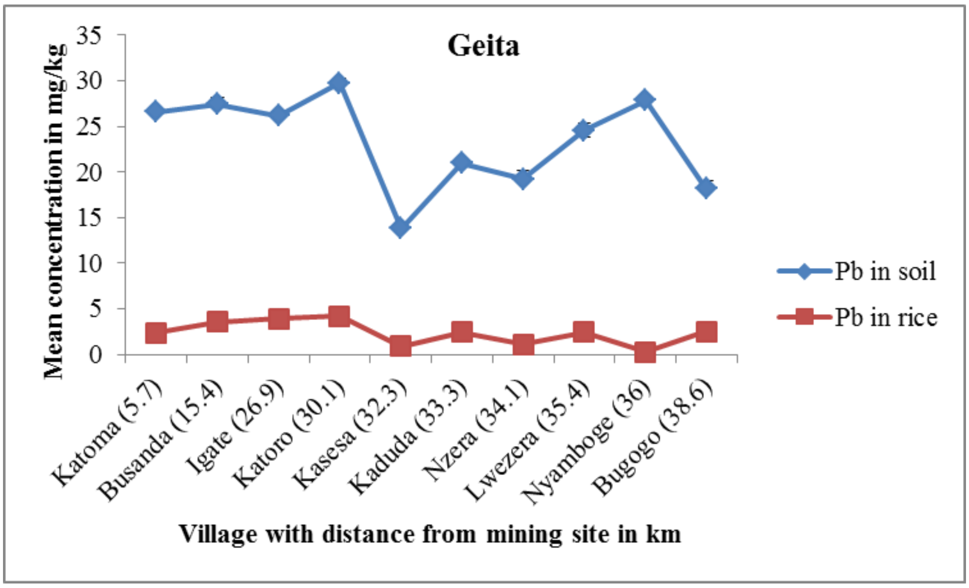


Figure 3.15: The effect of distance from mining site on Pb contamination to paddy soil and rice in Geita district

Lead is a metallic element which occurs by nature in the earth's crust and normally remains naturally below 50 mg/kg (Pourrut *et al*., 2011). In this study, the concentrations of lead in all soil samples were found to be below the maximum limit of 200 mg/kg (TBS, 2007). The highest level of Pb in Kahama district was at Kakola (31.65 mg/kg) and in Geita district was at Katoro (29.73 mg/kg). The probable reason for the existence of this concentration of Pb at Katoro might be due to the intentionally or unintentionally introduction of compounds rich in lead materials in paddy fields (Vanita *et al*., 2014). The maximum concentration of Pb (34.5 mg/kg) was reported by Micó *et al*. (2006) in agricultural soil in Spain. In Tanzania, Machiwa (2010) reported the maximum concentration of Pb to be 28.5 mg/kg for paddy soils within Lake Victoria basin in Tanzania. In India, high level of Pb (79.7 mg/kg) was reported by Chanda *et al*. (2011). Pb pollution in the agricultural soil is of more concern since it has no any biological benefit to both plants and animals even in low concentration. Low level of Pb retards seed germination whereas high level of Pb leads to over growth of plants which affects crop production (Pourrut *et al*., 2011).

The level of Pb in rice samples from Geita district ranged from 4.20 mg/kg (Katoro) to 0.27 mg/kg (Nyamboge) with the mean concentration of 2.39 mg/kg (Table 3.4). The level of Pb in rice samples from Kahama district ranged from 0.73 mg/kg (Bulyanhulu) to 5.07 mg/kg (Kakola) with the mean concentration of 1.97 mg/kg (Table 3.3). In all rice samples the concentration of Pb was recorded to be above the Codex Alimentarius Commission (2014) limit of 0.2 mg/kg. Despite being closer to the mining activities, Katoro site in Geita districts and Kakola site in Kahama district are business centers and there is so much automotive transportation around the sites which may lead to pollution of the atmosphere with lead. Therefore, the elevated level of Pb in rice might be due to direct absorption of Pb contaminated air from the automobile exhaust (Su *et al*., 2014). The level of Pb in this study is alarming situation as Pb exposure to human body may lead to death (Shabbir *et al*., 2013). The results in this study were lower as compared to that reported by Orisakwe *et al*. (2012) in South Eastern Nigeria which was 61.17 mg/kg of lead in rice concentration of Pb (1.44 mg/kg) in rice collected around Dabaoshan mine in China was reported by Zhuang *et al*. (2009). Shabbir *et al*. (2013) reported high level of Pb in rice in Pakistan to be 0.27 mg/kg. Emumejaye (2014) found the high concentration of Pb in rice consumed from Delta state in Nigeria which was imported from Thailand and India to be 5.45 mg/kg (Thailand), 14.55 mg/kg (Thailand), 17.64 (India) mg/kg and 22.73 mg/kg (Thailand).

Pearson correlation showed that in both Geita and Kahama districts, the concentration of Pb was significantly correlated at the level of 0.05 (Table 3.5). This indicated that the elevated level of Pb in rice might be influenced by the level Pb in agricultural soils as reported by Zhao *et al*. (2015).

Mercury

In paddy soils, high level of Hg (2.12 mg/kg) was found at Kakola, 1.6 km from Bulyanhulu gold mine and low level of Hg (1.25mg/kg) was found at Segese,32.4 km from Bulyanhulu gold mine in Kahama district (Table 3.1 and Figure 3.16).

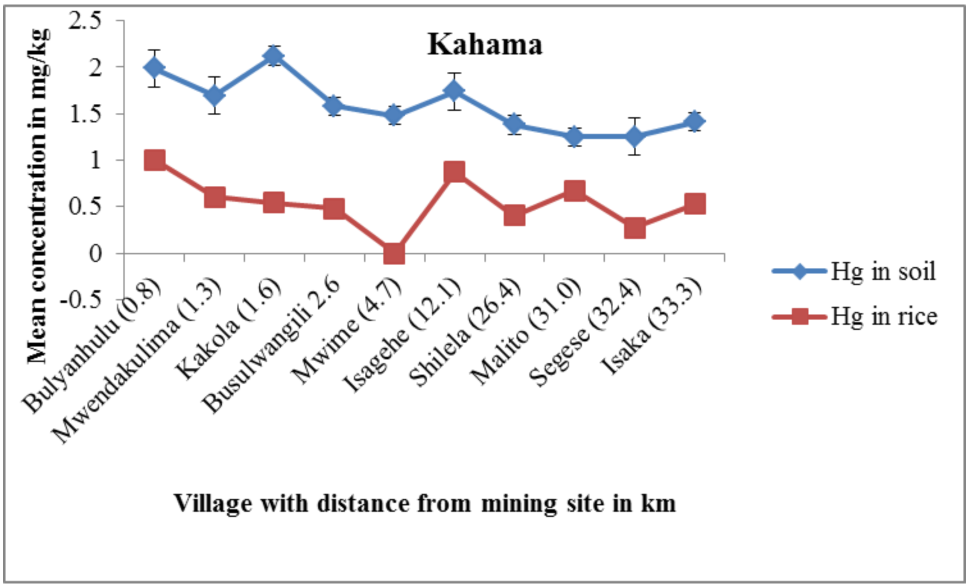


Figure 3.16: The effect of distance from mining site on Hg contamination to paddy soil and rice in Kahama district

As seen in Figure 3.17, In Geita district the high level of Hg (2.86 mg/kg) was found at Katoma 5.7 km from Geita gold mine and low level (1.12 mg/kg) of Hg was found at Kasesa, 32.3 km from Geita gold mine.

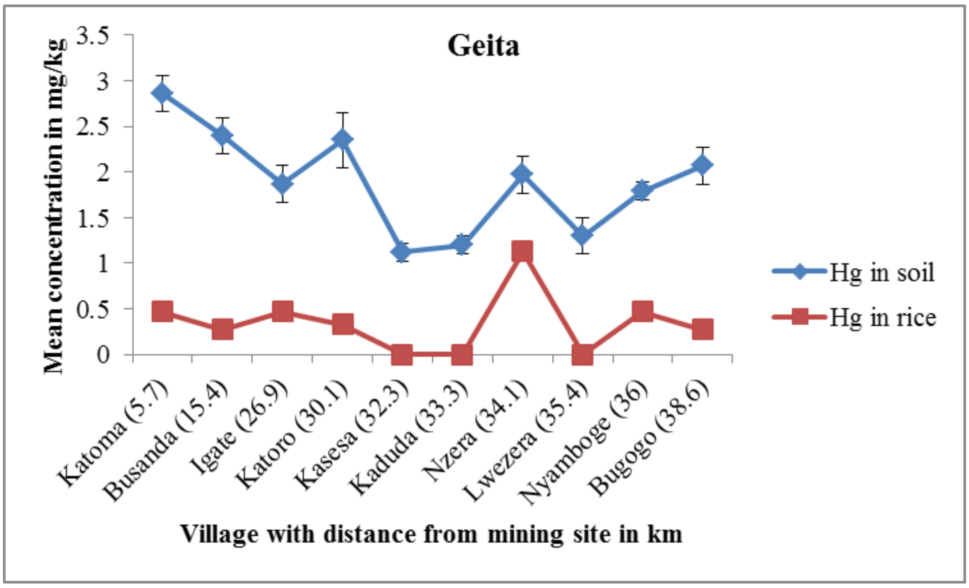


Figure 3.17: The effect of distance from mining site on Hg contamination to paddy soil and rice in Geita district

In this study, only soil samples at Kakola in Kahama district, Katoma, Busanda, Katoro and Bugogo in Geita district were found with Hg concentration above the maximum limit of 2 mg/kg (TBS, 2007). Kakola is located near Bulyanhulu gold mine at a distance of 1.6 km while Katoma is found near Geita gold mine at a distance of 5.7 km. The probable cause for high level of Hg above the maximum limit might be due to the effects of mining activities in the two districts (Santos-santos *et al*., 2006).

In Geita district the maximum concentration of Hg in rice samples was recorded to be at Nzera (1.13 mg/kg) while the concentration of Hg at Lwezera, Kaduda and Kasesa were below the detection limit. The mean concentration of Hg in all samples was 0.34 mg/kg (Table 3.4). In Kahama the high level of Hg was determined from rice samples obtained at Bulyanhulu (1.00 mg/kg) while the concentration of Hg at Mwime was below the detection limit of the instrument. The mean concentration of Hg in all samples was 0.54 mg/kg (Table 3.3).

Mercury is a toxic element to human being. It leads to potential different health problems in human being such as neurotoxic effects, destruction of genes and may affect cardiovascular system (Batista *et al*., 2012). It has also been considered as carcinogenic to human being as reported by International Agency for Research Cancer (IARC) (Batista *et al*., 2012). Rice has been reported to be a cause of Hg exposure to human being in different countries (Batista *et al*., 2012). The consumers of rice with the high concentration of Hg are at the risk of having health problems, since even low level of Hg exposure may lead to health problem such as high blood pressure, diarrhea, headache as well as reduction of hearing capacity (Lokeshappa *et al*., 2012). The detectable levels of Hg in rice samples were higher compared to that reported by He *et al*. (2013) which was (0.059 mg/kg) in Yangtse River Delt, China. Shabbir *et al*. (2013) reported the concentration of Hg (0.03 mg/kg) in Pakistan.

Pearson correlation results showed no significant correlation between the level of Hg in paddy soils and in rice in both Kahama ad Geita districts (Table 3.5). Therefore, the level of Hg in rice might not be due to high level of Hg in paddy soil. The probable reason for the high level of Hg in rice samples under the study might be due to the direct absorption of Hg from the contaminated atmosphere (He *et al*., 2013; Jia *et al*., 2010; Li *et al*., 2008).

Cadmium

In paddy soils, the concentration of Cd ranged from 1.04 mg/kg at Shilela, 26.4 km from Bulyanhulu gold mine to 17.67 mg/kg at Busulwangili, 12.6 km from Bulyanhulu gold mine with the mean concentration of 9.81 ± 4.86 mg/Kg in Kahama (Table 3.1). The concentrations of Cd with the distances from the mine is shown in the Figure 3.18.

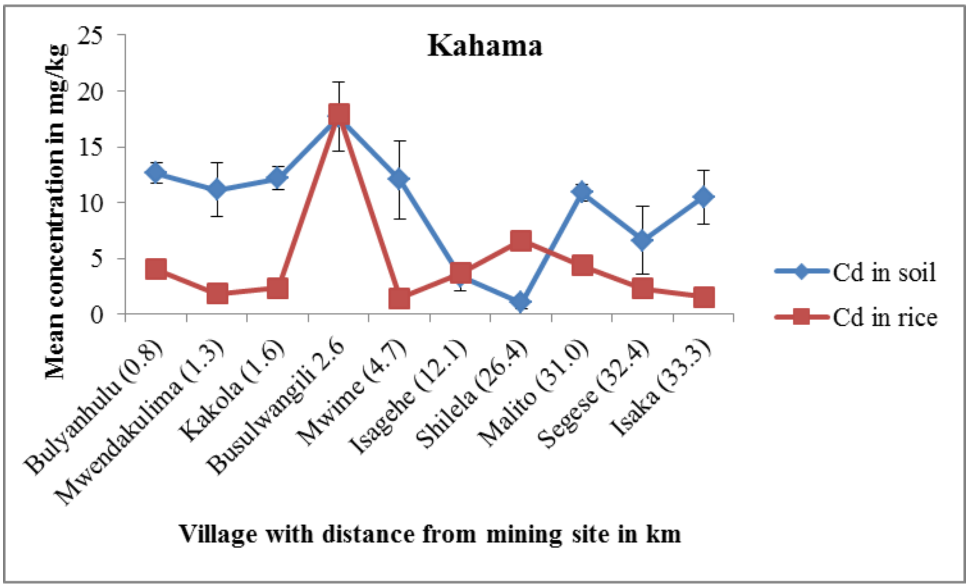


Figure 3.18: The effect of distance from mining site on Cd contamination to paddy soil and rice in Kahama district

The concentration of Cd ranged from 7.73 mg/kg at Lwezera, 35.4 km from Geita gold mine to 14.26 mg/kg at Busanda, 15.4 km from Geita gold mine with the mean concentration of 11.95 ± 1.94 mg/kg in Geita (Table 3.2). The concentrations of Cd with the distances from the mine is shown in the Figure 3.19.

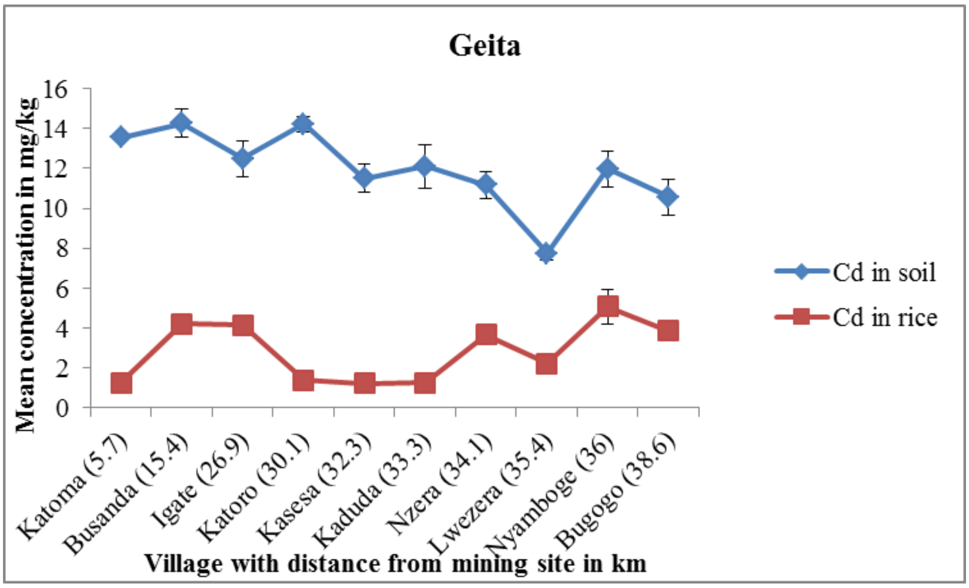


Figure 3.19: The effect of distance from mining site on Cd contamination to paddy soil and rice in Geita district

In this study, Cd concentrations in all soil samples in both Geita and Kahama districts, were recorded to be above the TBS (2007) maximum limit of 1 mg/kg in all sampled sites

Machiwa *et al*. (2010) reported that the concentration of Cd in paddy soil beyond 20 mg/kg reduces soil fertility and crop yield by weakening the roots of the rice plants. He also reported high level of Cd (29.2 mg/kg) in paddy soil around lake Victora basin in Tanzania. In this study, high level of Cd in soil samples at contaminated locations in Geita district were probably due to excessive utilization of livestock manure which was applied in order to increase soil fertility as well as crop yield (Kairiah *et al*., 2012). While in Kahama district, high level of Cd in paddy soils might be influenced by effects of mining activities near the contaminated areas (Zhuang *et al*., 2009).

The concentration of Cd in rice (Geita district) ranged from 1.24 mg/kg (Kasesa) to 5.07 mg/Kg (Nyamboge) with the mean concentration of 2.83 ± 1.50 mg/kg (Table 3.4). In Kahama district the concentration of Cd ranged from 1.41mg/kg (Mwime) to 17.97 mg/kg (Busulwangili) with the mean concentration of 4.60 ± 4.97 mg/kg. The concentrations of Cd in rice in all samples were recorded above the Codex Alimentarius Commission(2014) maximum limit of 0.4 mg/kg. In human being Cd may cause serious health problems even in trace amount (Vanita *et al*., 2014). Cd is considered as carcinogenic (Shabbir *et al*., 2013). It can also cause kidney damage, hypertention, diabetes anaemia, vascular disease and stroke to human being (Lokeshappa *et al*., 2012).

The results in this study indicated that levels of Cd in rice were very higher when compared with the high concentration of Cd (0.521 mg/kg) reported in South of Iran (Mehrnia, 2013). In Pakistan, Shabbir *et al*. (2013) found the concentration of Cd in different rice samples with the average of 0.55 mg/kg, 0.47 mg/kg and 0.41 mg/kg. High concentration of Cd (0.24 mg/kg) was reported in Southern Eastern, Nigeria (Orisakwe *et al*., 2012). Also, the high Cd level ranged from 2.1 mg/kg to 6.5 mg/kg in rice samples collected from Cd contaminated paddy soils as reported in South China (Wang *et al*., 2011).

In this study it was observed that low concentrations of heavy metals were recorded in the rice as compared with levels of heavy metals in the soils except for Cd at Isagehe and Shilela in Kahama district, by which more concentration of Cd was recorded to be higher in rice than in soils (Figure 3.3). This high level of Cd might be due to rice absorbing heavy metals directly from the contaminated air (Lugwisha and Othman, 2014).

According to pearson correlation analysis, in both Kahama and Geita districs, the concentration of Cd in paddy soils showed no significant relationship with the concentration of Cd in rice (Table 3.5). This suggested that the levels of Cd in rice were not influenced by the levels of Cd in paddy soils, it might be due to other causes such as direct absorption of Cd from the Cd contaminated air (Su *et al*., 2014).

3.4 Conclusions

This study aimed at determining the extent of heavy metals (Cr, Cu, Zn, Hg, Cd, Ni, Pb and As) contamination in paddy soils and polished rice around mining areas in Kahama and Geita districts. The results indicated that in all soil samples the concentrations of Cr and Cd exceeded the maximum limits set by TBS while in all rice samples, the concentrations of Pb and Cd exceeded the Codex Alimentarius Commission maximum limits. The results further indicated that there is strong relationship between the levels of heavy metals in paddy soils with the levels of heavy metals in rice. This means that as the levels of heavy metals in paddy soils become higher, the levels of heavy metals in rice increase. This has also been shown by the Pearson positive correlation of As, Pb and Cu in all soil and rice samples which indicated that the levels of these heavy metals in soils were the reason for high levels of these heavy metals in rice. It has been observed that paddy soils and rice from villages which are very close to the mining operations are highly contaminated with heavy metals. This suggested that mining activities near paddy soils might be the main source of heavy metals in rice grown in those areas. Therefore, this study recommends for periodical monitoring of paddy soils contaminations with heavy metals to assess the trend of contamination and establish remediation techniques of contaminated paddy soils.

CHAPTER FOUR

THE RISK OF HEAVY METALS EXPOSURE THROUGH CONSUMPTION OF RICE GROWN IN GEITA AND KAHAMA DISTRICTS

Summary

In chapter three it has been indicated that rice grown near the artisanal mining areas in Kahama and Geita districts may be contaminated with the heavy metals. The consumption of heavy metals contaminated rice from the sites indicated in chapter three may pose different health problems to human being. Chapter four aimed at estimating the risk of human exposures to heavy metals namely Cu, Zn, Hg, Cd, Pb and As through consumption of rice produced in Kahama and Geita districts. A standardized questionnaire was administered to 40 respondents from whom rice samples were collected. The heavy metals in the rice samples determined and reported in Chapter three were used in the assessment. The human exposure to heavy metal was estimated using the deterministic approach. The average daily per capita rice consumption used was 66.8 g/person/day for Geita district and 74.0 g/person/day for Kahama district. The estimated exposures were compared with tolerable daily intakes (TDIs) set by the Joint FAO/WHO Expert Committee on Food Additives to determine the health risk of exposure. The results indicated that 38 adult individuals out of 40 respondents were exposed to Cd above the TDI of 1 µg/kg bw/day. The results further showed that only 15% of all sampled population was exposed to Pb above the TDI (3.0 µg/kg Bw/day). The estimated daily intake (EDI) for As in all population were below TDI range (2 - 7 µg/kg Bw/day) for total As. The EDI for Hg exceeded TDI (0.57 µg/kg Bw/day) in 40% of the sampled population. The EDIs for Cu were recorded to be below the TDI (500 µg/kg Bw/day) in both Kahama and Geita population. The EDI for Zn in all samples were below the TDI (1000 µg/kg Bw/day). The highest EDIs for heavy metals through rice consumption were recorded from local population living at the villages located very close to the mining operations. This suggests the main cause of unacceptable human exposure to heavy metals is the proximity to artisanal mining and more studies need to be conducted to estimate the risk of heavy metals exposure through rice consumption in areas near the mining sites in Tanzania.

4.1 Introduction

Rice (*Oryza sativa*) is the third most important agricultural crop in Tanzania after maize and cassava (FAO, 2015). It is considered as a major component of human diet in Tanzania

(Machiwa, 2010). As reported by Mghase *et al*. (2010), the Tanzanian daily per capita consumption of rice ranged from 68.5 g/person to 82.2 g/person.

The intake of heavy metal contaminated rice may pose potential risk to human health (Machiwa, 2010). Pan *et al*. (2013) reported that human exposure to Pb and Hg may result to dysfunction of central nervous system. Human exposure to Cd may lead to destruction of kidney and liver, As may cause cancer of different organs in the human body, whereas Hg poses neurotoxicity and teratogenicity (Fu *et al*., 2008). Cu and Zn are considered as essential elements to many organisms when exposed at low level, but when exceeds the maximum limits they lead to serious health problems to human being (Nazir *et al*., 2015).

Mining activities near the paddy fields have been identified as contributors to the rapid increase of heavy metals contaminations in rice (Zhuang *et al*., 2009). Kahama district located in Shinyanga region, and Geita district located in Geita region have the largest gold mining sites in Tanzania which are Buzwagi and Bulyanhulu gold mines found in Kahama district and Geita gold mine found in Geita district (Makene *et al*., 2012). Apart from artisanal mining activities, small scale gold mining activities are also conducted in different areas in Kahama and Geita districts (ILO, 2002; Kitula, 2006). Local consumers of rice cultivated in Kahama and Geita districts may be exposed to heavy metals contaminations which may pose different human health impairment. This research aimed at assessing human exposure to heavy metals mainly Cd, Hg, As, Pb, Cu and Zn through rice consumption around mining sites in Kahama and Geita districts, Tanzania.

4.2 Materials and Methods

4.2.1 Study sites

The study was conducted in 40 households around mining areas in Geita and Kahama districts in Tanzania. Sites were selected based on the fact that they are among the main rice producing areas and they are located around the gold mining areas. Geita district sites were Nzera, Nyamboge, Bugogo, Busanda, Lwezera, Igate, Katoro, Katoma, Kaduda, Kasesa, Butobela, Idoselo, Bung’wangoko, Nyamazovu, Sungusira, Ibondo, Buligi, Kasamwa, Senga and Kaseme (Figure. 3.1). In Kahama district the sites selected were Isaka, Malito, Isagehe, Busulwangili, Mwendakulima, Mwime, Bulyanhulu, Shilela, Kakola, Segese, Chapulwa, Itogwan’gholo, Mwakata, Mpela, Kagongwa, Bukooba, Kishima, Gulla, Busangu and Chela (Figure.3.2).

4.2.2 Agricultural practice and consumption survey

Information about rice cultivation practices and rice consumption as well as other food consumption data was collected from 40 households that were randomly selected, 20 from each of the two districts; Kahama and Geita districts in which the mining areas are located. A close ended questionnaire was designed to understand sources of heavy metals exposure to human being. The questionnaire was initially designed in English and was then translated to Kiswahili to suit the common language of respondents. The questionnaire consisted of three sections; demographics, rice growing practices as well as rice and other food intake questions. The answered questionnaire was then translated back to English language to allow easy data interpretation.

4.2.3 Rice sampling and preparation

Rice sampling and preparation was done as explained in section 3.2.2 of Chapter three. Additional 20 polished rice samples were collected from other villages in Geita and Kahama districts to meet the number of samples required for risk estimation of heavy metals to rice consumers in Geita and Kahama districts.

4.2.4 Chemical analysis of rice

Rice sample analysis was done at Tanzania Atomic Energy commission laboratory as explained in section 3.2.3 of Chapter three.

4.2.5 Quality assurance and quality control

Quality assurance and quality control was performed as explained in section 3.2.4 of Chapter three.

4.2.6 Estimated daily intake (EDI) of Cu, Zn, As, Pb, Hg and Cd through rice consumption

Heavy metals intake through rice consumption was determined using deterministic exposure assessment method (Chamannejadian *et al*., 2013; Fu *et al*., 2008; Kimanya *et al*., 2008; Mehrnia, 2013; Zazoli *et al*., 2006) as shown below (Equation 1).

$$\mathrm{EDI} = \frac{\mathrm{C \ x \ Cons}}{\mathrm{Bw}} \qquad \text{Equation 1}$$

Where EDI is estimated daily intake; C means concentration of the heavy metals in contaminated rice; Bw is 60 kg body weight similar with that used by Fu *et al*. (2008); Cons is average daily per capita rice consumption which is about 66.8 g/person/day for Geita district and 74.0 g/person/day for Kahama district (Smith and Subandoro, 2007). The EDI calculated were then compared with the daily body tolerable intake to estimate the risk of heavy metals exposure through rice consumption (Chamannejadian *et al*., 2013; Fu *et al*., 2008; Mehrnia, 2013; Zazoli *et al*., 2006).

4.2.7 Statistical analysis

Statistical analysis was done using Microsoft Excels Spread Sheet in calculating EDI and finding the mean at a confidence level of 95%. To evaluate statistical differences in the elemental EDI between Kahama and Geita districts, the single factor ANOVA test was used.

4.3 Results and discussion

4.3.1 Heavy Metals Contents in Polished Rice

Tables 4.1 and 4.2 show the summaries of heavy metals contents in Kahama district and Geita district, respectively.

Table 4.1: Summary of heavy metals contents (mg/kg) in rice from Kahama district sites

Variable	Cu	Zn	As	Pb	Hg	Cd
Minimum	1.64	4.23	BDL	0.40	BDL	0.47
Maximum	62.40	27.00	5.60	5.07	2.00	19.53

BDL: Below detection limit

Table 4.2: Summary of heavy metals contents (mg/kg) in rice from Geita district sites

Variable	Cu	Zn	As	Pb	Hg	Cd
Minimum	0.59	0.68	BDL	0.27	BDL	0.65
Maximum	10.18	19.66	0.38	4.20	1.13	7.10

BDL: Below detection limit

Rice grown on contaminated soils is highly contaminated with heavy metals which may lead to several human health effects (Li *et al*., 2014). In Kahama district, the mean concentration of Cu, Zn, As, Pb, Hg and Cd was found to be 6.23 mg/kg, 14.49 mg/kg, 0.30 mg/kg, 1.50

mg/kg, 0.60 mg/kg and 5.58 mg/kg, respectively. The concentration of As was determined to be below the detection limit of the instrument of 0.02 mg/kg in 85% of all Kahama rice samples. In Geita district, the mean concentration Cu, Zn, As, Pb, Hg and Cd was determined to be 2.91 mg/kg, 12.32 mg/kg, 0.03 mg/kg, 1.50 mg/kg, 0.43 mg/kg and 2.87 mg/kg, respectively. The concentration of As was determined to be below the detection limit of instrument of 0.02 mg/kg in 85% of all Geita rice samples.

In this study, the maximum levels of Cu, Zn, As and Pb were reported at Kakola in Kahama district and at Katoro in Geita district. Kakola is located very close (1.6 km) to the Bhulyanhulu gold mine while small scale mining activities are performed at Katoro site. This suggested that the elevated levels of Cu, Zn, As and Pb might be influenced by the mining activities taking place near the study areas (Zhuang *et al.*, 2009). The elevated levels of Hg at Chela and Nzera as well as the high level of Cd at Bukooba and Sungusira might be due to the application of fertilizers in the paddy fields since all respondents in the study were reported to use different types of fertilizers to their paddy fields in order to increase crop yields (Payus and Talip, 2014). Both inorganic as well as livestock manure have been reported to increase the levels of heavy metals in agricultural soils (Khairiah *et al.*, 2012; Roy *et al.*, 2013).

In this section, the data of concentrations of heavy metals in polished rice were mainly for determination of risk of heavy metals exposure through rice to consumers of rice cultivated in Kahama and Geita districts. For the purpose of risk assessment of heavy metals exposure, samples found to be below the detection limit of the instrument were assigned half value of the detection limits similar to Tungaraza *et al*. (2011).

4.3.2 Socio-demographic characteristics of respondents

Questionnaire results indicated that 50% (n = 20) of all respondents were males and 50% were female. Among them 80% (n = 32) were married while 20% (n = 8) were not married. The age of respondents ranged from 20 years to 60 years. However most of them were between 30 years and 50 years of age (n = 24). In the study areas, the participants who had not received primary education were 22.5% (n = 9), 57.7% (n = 23) had primary education while 20% (n = 8) had secondary education. Majority of respondents were farmers (n = 30, 75%), while others (n = 7, 17.5%) dealt with other economic activities such as mining. A few of respondents work in public services (n = 3, 7.5%). All respondents (n = 40, 100%) depend on solar energy to dry rice after harvesting.

4.3.3 Dietary intake of heavy metals

The estimated dietary intake of heavy metals in Kahama and Geita districts are presented in Tables 4.3 and 4.4.

Table 4.3: Estimated dietary intake of heavy metals (µg/kg Bw/day) in Kahama district

Village	Cu	Zn	As	Pb	Hg	Cd
Isaka	3.56	20.25	0.01	5.92	0.65	1.89
Malito	5.34	20.25	0.25	1.15	0.82	5.38
Isagehe	5.44	19.36	0.01	1.97	1.07	4.54
Busulwangili	4.81	19.12	0.01	0.99	0.57	22.16
Mwendakulima	4.43	21.81	0.01	0.99	0.74	2.26
Mwime	3.95	11.91	0.01	1.73	0.02	1.74
Bulyanhulu	4.09	7.41	0.12	0.91	1.23	4.95
Shilela	2.02	5.22	0.01	0.99	0.47	8.14
Kakola	76.96	33.30	6.93	6.25	0.67	2.87
Segese	4.43	9.30	0.01	3.37	0.33	2.80
Chapulwa	4.38	23.08	0.01	0.95	0.74	2.84
Itogwang’holo	4.61	20.00	0.01	0.83	0.62	2.04
Mwakata	3.42	18.28	0.01	0.99	0.02	7.88
Mpela	6.15	18.86	0.01	0.49	0.58	3.02
Kagongwa	3.95	13.10	0.01	0.62	0.27	0.58
Bukooba	2.60	17.22	0.01	2.13	1.64	24.09
Kishima	2.89	18.36	0.01	1.97	1.23	22.13
Gulla	2.74	17.51	0.01	1.60	0.02	5.28
Busangu	4.09	19.54	0.01	0.99	0.74	10.61
Chela	3.70	23.53	0.01	2.22	2.47	13.81
JECFA limit	500	1000	(2 -7)	3.0	0.57	1.0

Table 4.4: Estimated dietary intake of heavy metals (µg/kg Bw/day) in Geita district

Village	Cu	Zn	As	Pb	Hg	Cd
Nzera	1.91	18.7	0.01	1.26	1.26	4.10
Nyamboge	1.12	3.68	0.01	0.30	0.52	5.64
Bugogo	1.78	7.98	0.01	2.82	0.30	4.29
Busanda	2.13	10.87	0.01	4.01	0.30	4.69
Lwezera	2.69	5.89	0.01	2.67	0.02	2.46
Igate	0.66	10.61	0.01	4.38	0.52	4.62
Katoro	11.33	21.89	0.42	4.68	0.37	1.54
Katoma	6.26	11.21	0.16	2.67	0.52	1.41
Kaduda	2.26	13.60	0.01	2.75	0.02	1.41
Kasesa	0.69	0.76	0.01	0.97	0.02	1.38
Butobela	3.47	21.05	0.01	0.52	0.52	2.59
Idoselo	2.99	19.57	0.01	0.75	0.52	1.80
Bungwangoko	5.03	19.83	0.01	0.44	0.67	1.84
Nyamazovu	2.74	11.44	0.01	0.59	0.52	2.59
Sungusira	3.08	15.85	0.01	0.59	1.11	7.90
Ibondo	1.17	18.97	0.01	1.34	0.67	7.45
Buligi	4.08	15.85	0.01	0.89	0.37	4.69
Kasamwa	3.43	13.98	0.08	0.52	0.37	1.6
Senga	3.38	14.67	0.01	0.75	0.37	0.72
Kaseme	5.59	20.79	0.01	0.52	0.37	1.21
JECFA limit	500	1000	(2 -7)	3.0	0.57	1.0

These EDI values shown in Tables 4.3 and 4.4 indicate that consumers of rice cultivated in Kahama and Geita districts are exposed to heavy metals (As, Hg, Cd, Zn, Pb and Cu). When estimated dietary intakes for heavy metals in Kahama and Geita districts were compared, it was found that the EDI for Zn ($p = 0.048$), Cd ($p = 0.014$) and Hg ($p = 0.037$) were significantly different ($p < 0.05$). While the EDI for Cu ($p = 0.251$), As ($p = 0.344$) and Pb ($p = 0.906$) showed no significant difference ($p > 0.05$).

The TDI set for Cd by Joint FAO/WHO Committee on Food Additives and Contaminants (JEFCA) (2003) is 1 µg/Kg Bw/day. In this study, the EDI for Cd through rice consumption ranged from 0.58 µg/kg Bw/day (Kagongwa) to 24.90 µg/kg Bw/day (Bukooba) in Kahama district. In Geita district the lowest EDI was recorded at Senga (0.72 µg/kg Bw/day) while the highest EDI was recorded at Sungusira (7.90 µg/kg Bw/day). These results show that the dietary intake of Cd by all respondents exceeded TDI except for respondents at Senga (0.72 µg/kg Bw/day) in Geita district and at Kagongwa (0.58 µg/kg Bw/day) in Kahama district. This means that almost 95% of the whole population under this study exceeded the TDI, indicating high human exposure to Cd within the study areas. The dietary intake of Cd through consumption of Cd contaminated rice may lead to serious kidney and liver damage (Zhuang *et al*., 2009). Fu *et al*. (2008) reported that Cd exposure to human being through rice consumption caused the Itai – itai disease in Japan in 1960s. Local inhabitants around the Dabaoshan mine in Guangdong, China were /exposed to Cd (5.1 µg/kg Bw/day) via consumption of Cd contaminated rice as it was reported by Zhuang *et al*. (2009). For example in Southeast China, Fu *et al*. (2008) reported the maximum EDI for Cd (3.6 µg/kg Bw/day) which was 3.6 times higher than TDI set by Joint FAO/WHO Committee on Food Additives and Contaminants (JEFCA). Zazouli *et al*. (2008) determined the EDI of Cd through *Iranian* rice consumed in North of Iran to be at the range of 22.57µg/kg Bw/day to 25.43µg/kg Bw/day and Zazoli *et al*. (2006) estimated the Cd daily intake through *tarom* rice consumption in North of Iran at the range of 0.36 µg/kg Bw/day to 2.23 µg/kg Bw/day.

In this study, the estimated Pb intake through rice consumption ranged from 0.49 µg/kg Bw/day (Mpela) to 6.25 µg/kg Bw/day (Kakola) in Kahama district. In Geita district, the highest EDI for Pb was recorded at Katoro (4.48 µg/kg Bw/day) while the lowest EDI for Pb was reported at 0.30 µg/kg Bw/day. According to JECFA limits (3.0 µg/kg Bw/day), only 15% of all respondents were remarkably exposed to Pb above the maximum TDI for Pb intakes (Joint FAO/WHO, 2014). These included local population at Busanda (4.01 µg/kg Bw/day), Igate (4.38 µg/kg Bw/day) and Katoro (4.68 µg/kg Bw/day) in Geita district, also Isaka (5.92 µg/kg bw/day), Kakola (6.25 µg/kg Bw/day) and Segese (3.37 µg/kg Bw/day). Similarly, Fu *et al*. (2008) observed that Southeast China population was exposed at 14.0 µg Pb /kg Bw/day which was about 3.9 times higher than the TDI.

In this study, the maximum EDI of As was found to be in Kakola households (6.93 µg/Kg Bw/day) as calculated from the maximum level of As in rice which is within the TDI (2 – 7 µg/kg Bw/day) for total As suggested by JECFA (2010). The rest of the EDI for As were below the TDI range. Although the results showed that in all respondents the EDI for As were within the range of TDI, but As may accumulate in the body through rice consumption frequency. Some people in the study areas consumed more than the average quantity of rice in a single meal, this may cause the EDI of As to exceed the TDI which may lead to human health problems. According to EFSA (2009) and Joint FAO/WHO (2011) arsenic is a toxic metalloid as it is considered as carcinogenic to human being. Fu *et al*. (2008) reported the range of EDI for As through rice consumption as 0.5 µg/Kg Bw/day to 1.7 µg/kg Bw/day in Southeast China. The maximum EDI for As through rice in Nigeria was reported by Onwukeme *et al*. (2014) to be 16.62 µg/kg Bw/day which is approximately 2.4 times the maximum TDI. This indicated high risk of As exposure due to Southeast China population.

The maximum EDI of Hg in Kahama district was found in population at Chela (2.47 µg/kg Bw/day) and the lowest EDI of Hg was found in populations at Mwime, Gulla and Mwakata (0.02 µg/kg Bw/day) with the mean EDI of 0.75 µg/kg Bw/day. The EDI of Hg in Geita district population ranged from 0.02 µg/kg Bw/day at Kasesa, Kaduda and Lwezera, while the maximum EDI was recorded at Nzera (1.26 µg/kg Bw/day) with the mean EDI of 0.47 µg/kg Bw/day. The TDI for Hg recommended by JECFA (2010) is 0.57 µg/kg Bw/day. The results in this study indicated that only 40% of all respondents in both Kahama and Geita districts exceeded the TDI. These results indicating that consumers of rice cultivated around mining sites in Kahama and Geita districts are more exposed to Hg toxicity and thus exposure to Hg health effects such as birth defects and skin discoloration (Charles *et al*., 2013). Although birth defects and skin discoloration might be due to other causes apart from Hg exposure toxicity, Charles *et al*. (2013) reported that half of the studied population near artisanal mining areas in Rwamagasa village in Geita district in Tanzania experienced birth defects and observed to have skin discoloration. The maximum EDI intake of Hg in this study was higher compared to the maximum EDI of Hg (4 µg/day for an adult of 55 kg body weight) in Mugusu artisanal gold mining village, in Geita district using the similar approach reported by Tungaraza *et al*. (2011). Batista *et al*. (2012) reported that Brazil population was exposed at 0.22 µg Hg /kg Bw/day.

In Geita district, the minimum and maximum EDIs for Cu were 0.66 µg/kg Bw/day (Igate) and 11.33 µg/kg Bw/day (Katoro), respectively with the mean of 3.69 µg/kg Bw/day, whereas the EDIs for Cu in Kahama district ranged from 2.02 µg/kg Bw/day (Shilela) to 76.96 µg/kg Bw/day (Kakola), with the mean of 7.68 µg/kg Bw/day. The results in this study indicate that in all respondents, the EDI for Cu was below the maximum TDI (500 µg/kg

Bw/day) stated by JECFA (Joint FAO/WHO, 2014). This indicates that the local populations in the study areas were not significantly exposed to the effects of Cu contamination. However, the observed maximum EDI for Cu from the study areas was higher compared with results of other studies; 6.59µg/kg Bw/day (Lin *et al*., 2004) in Taiwan population, 1.66µg/kg Bw/day (Satpathy *et al*., 2014) in Indian population and 10.69 µg/kg Bw/day in Nigeria population as reported by Onwukeme *et al*. (2014). The levels of Cu in agricultural soil and in rice should be maintained in order to prevent the EDI of Cu exceeding the TDI. High exposure of Cu to human being through rice may lead to different health problems to such as neurological diseases and heart diseases (EC, 2003).

The Zn intake was estimated to be at the range of 5.22 µg/kg Bw/day (Shilela) to 33.30 µg/kg Bw/day (Kakola) with the mean value of 17.87 µg/kg Bw/day, in local population in Kahama district. In Geita district population, the maximum EDI for Zn was recorded at Katoro (21.89 µg/kg Bw/day) and the minimum EDI was recorded at Kasesa (0.76 µg/kg Bw/day) with the mean value of 13.84 µg/kg Bw/day. In this study the EDI for Zn in all populations in both Kahama and Geita districts were recorded to be below the maximum TDI for Zn which is 1000 µg/kg Bw/day recommended by JECFA (Joint FAO/WHO, 2014). This suggested that the levels of Zn in rice should be controlled since too much intake of Zn is lethal to human being, but also the Zn deficiency may lead to deficiency in immunity, poor growth as well as underdevelopment of neurons (Plum *et al*., 2010). Comparing human exposure to Zn in this study with others; 40.96 µg Zn/kg Bw/day (Onwukeme *et al*., 2014) and 43.57 µg Zn/kg bw/day (Lin *et al*., 2004), it can be observed that the EDI for Zn exposure to the local inhabitants in Kahama and Geita districts still seems to be much higher. This indicates that the accumulation of Zn in human bodies in Kahama and Geita districts through rice consumption may result to human health problems.

Large values of EDI of heavy metals above the TDI result to high levels of risk to human health due to exposure to high levels of heavy metals in the rice samples. Rice plants uptake heavy metals from the heavy metals contaminated agricultural soil and translocate to the rice grains (Muraoka *et al*., 2012). In this study, high levels of heavy metals exposure are due to the elevated levels of heavy metals in rice which might be caused by the effects of mining activities around the sample sites (Lee *et al*., 2001; Li *et al*., 2008; Zhuang *et al*., 2009). The results indicate that the EDIs for Cu, Zn, As and Pb were higher at Kakola in Kahama district and at Katoro in Geita district as compared with the other sites. This might have been contributed by mining activities. Kakola is located very close to the Bhulyanhulu gold mine while at Katoro, the small scale mining activities are conducted. Thus the rice consumers at Kakola and Katoro are highly exposed to heavy metals. Similar report was given by Zhuang *et al*. (2009). Contribution from other sources such as fertilizers which contain heavy metals is of potential concern. The application of such fertilizers on agricultural soil may increase the heavy metals contamination (Bhartiya and Singh, 2012; Zhang *et al*., 2012; Veeken and Hamelers, 2002; Zazouli *et al*., 2008). Khairiah *et al*. (2012) reported that the high level of anthropogenic Cu in paddy soils in Malaysia was due to the application of inorganic fertilizers as well as livestock manure. Livestock manure contributed to the increase of levels of heavy metals in agricultural fields in Northeast China as it was reported to consist of Cu (31.1 mg/kg), As (2.5 mg/kg) and Cd (0.5 mg/kg) Zhang *et al*., 2012). The median value of Pb (14.3 mg/kg) in phosphatic fertilizers used in the kingdom of Saudi Arabia was reported by Modaihsh *et al*. (2004). According to the study on investigation of heavy metals contents in commercial inorganic fertilizers in the Kingdom of Saudi Arabia conducted by Modaihsh *et al*. (2004), the highest values of heavy metals detected in phosphatic fertilizers were Pb (32.4 mg/kg) and Cd (36.8 mg/kg). In this study, all respondents reported to have used either inorganic fertilizers or livestock manure in their paddy fields to increase rice production. The study done by Alrawiq *et al*. (2014) reported that heavy metals from heavy metals contaminated agricultural soils may be translocate into rice plants and accumulate in different parts of the plant including grains which are potential route of heavy metals exposure to human being. This suggested that the use of fertilizers in Kahama and Geita districts might be another source of heavy metals contamination in rice and hence increase the EDI values to the population. However more studies need to be done to investigate the effect of fertilizers in heavy metals contamination in rice in Kahama and Geita districts.

4.4 Conclusion

The study indicates that 95% of all local populations in both Kahama and Geita districts is exposed to Cd above the tolerable daily intake. This suggests that most of people in Kahama and Geita districts are at high risk of being affected by Cd through rice consumption. The results indicated that there is no potential risk of Cu, Zn and As from rice consumption in both Geita and Kahama districts, their EDIs being lower than TDI. The EDIs for Pb and As were recorded to be higher at Kakola in Kahama district as compared with all other study areas in both Kahama and Geita district. Kakola being located very close to Bulyanhulu gold mine, the probable cause of Pb and As exposure through rice consumption might be mining operations. The application of fertilizers to the paddy soils may increase the levels of heavy metals in rice since fertilizers consist of a certain quantity of heavy metals. Further studies should be conducted to investigate other sources of heavy metals contamination in rice and estimate the risk of human exposures of heavy metals through other food such as maize, cassava and potatoes and suggest the measures to minimize such exposures in Kahama and Geita districts as well as in other areas in Tanzania.

CHAPTER FIVE

5.1 General Discussion

The present work was guided by three study questions which aimed at addressing the concentrations of selected heavy metals in paddy soils around artisanal mining areas in Kahama and Geita districts; the correlation between these heavy metals in soil and in rice as well as the risk of heavy metals exposure to human life through rice consumption.

The results of this study indicate that paddy soils and rice in Kahama and Geita districts have some levels of heavy metals contamination. The levels of Cd and Cr in all soil samples were above the maximum limits set by TBS. The result revealed that in both Kahama and Geita districts, the concentration of As was extremely high at Kakola site in Kahama district as it was 58.64 times higher than the TBS maximum limit of 1 mg/kg. This value is nearly equal to 57.50 mg/kg of As in paddy soils of Bangladesh reported by Brammer and Ravescroft (2009). High level of As in agricultural soils should be given much attention as it may affect human health through food chain as well as decrease crop production (Banejad and Olyaie, 2011).

According to Codex Alimentarius Commission the concentrations of Pb and Cd in all rice samples exceeded the maximum limit whereas the concentrations of Cu and As exceeded the maximum limit in only 5% of all rice samples. The results indicated that there is strong relationship between the levels of heavy metals in paddy soils and heavy metals in rice, this means that as the concentrations of heavy metals in paddy soils increases, the levels of heavy metals in rice become higher except for some reasons. This suggested that heavy metals contaminated paddy soils might be the main source of heavy metals in rice (Zhuang *et al.*, 2009). Moradi *et al.*, (2013) reported that high levels of heavy metals in contaminated soils may influence the increase in concentrations of heavy metals in food crops grown in that contaminated soils. Pearson correlation analysis, found that in both Kahama and Geita districts, the concentrations of Cu, As and Pb in paddy soils were positively correlated with the concentration of these heavy metals in rice at the significance level of 0.01, 0.01 and 0.05, respectively. This means that the elevated levels of Cu, As and Pb in paddy soils in the study sites lead to the increase of concentrations of these heavy metals in the rice grown in the same sites (Moradi *et al.*, 2013). The level of Zn in agricultural soils and rice collected from industrial sites in Iran was positively correlated at significance level of 0.01 as reported by Moradi *et al*. (2013).

In this study, the results showed that most of the villages located very close to the mining activities were observed to have high levels of heavy metals in both paddy soils and rice as compared with the villages located very far from the mining activities. Kakola (1.6 km) in Kahama district was recorded high levels of heavy metals as compared to other villages. At Katoro village (30.1 km) in Geita district where small scale mining activities are conducted was observed to be highly contaminated with heavy metals. Escarré *et al.* (2011) reported that the distance from mining sites has the influence on heavy metals contamination in soils. That means the small the distance from the mining sites the high the heavy metal contamination in the soils.

The risk of heavy metals exposure to human life was determined using deterministic methods (Fu *et al.*, 2008). The results indicated that 95% of households in both Kahama and Geita districts are exposed to Cd through rice consumption. The results further indicated that the EDIs for Zn and Cu were below the TDIs set by JECFA. This means the local population in Geita and Kahama districts are not at risk of Zn and Cu exposure through rice consumption sites. However, Kakola site in Kahama district was observed to have the highest values of EDIs for As (6.93 µg/kg Bw/day) and Pb (6.25 µg/kg Bw/day) suggesting high risk of being affected by As and Pb contamination through rice consumption. These highest values of EDIs were associated with the closeness of mining sites to this village (Kakola). High EDIs for Pb (8.90 µg/kg Bw/day) and Cd (5.10 µg/kg Bw/day) from rice collected around Dabaoshan mine in Guangdong, China was reported by Zhuang *et al.* (2009). Fu *et al.* (2008) reported the maximum EDI for Pb through rice collected from a typical e – waste recycling area in Southeast China to be 14.0 µg/kg Bw/day.

5.2 Conclusion

This study aimed at determining the concentrations of heavy metals in paddy soils and in rice from villages around the artisanal gold mining areas, as well as assessing the risk of human exposures of heavy metals through rice consumption around the artisanal gold mining areas in Kahama and Geita districts. The concentrations of Cu, As and Pb in paddy soils showed positive correlations with the concentration of Cu, As and Pb in polished rice in all villages in Kahama and Geita districts. This suggested that the levels of Cu, As and Pb in rice might be influenced by elevated levels of these heavy metals in paddy soils. Cd and Pb were observed to be above the maximum limits in the polished rice collected form the villages in Kahama and Geita. This indicates potential threat to the health of rice consumers in these areas. Generally levels of heavy metals in paddy soils and in rice were observed to be higher in the villages which are very close to the mining areas. EDIs analysis has shown the possibility for local population in Kakola village (Kahama district) being at high risk of effects of heavy metals Pb, As, Cd and Hg which were observed to be above the TDIs. Although the EDIs for Cu and Zn were below the TDIs in all study sites, but they were observed to be higher at Kakola site than in other study sites.

5.3 Recommendations

Based on the results, this study recommends that;

A. For further studies

- Studies should be conducted to pin point other sources of heavy metals contamination in soils and rice and provide the information on how to minimize the contamination in Kahama and Geita districts.
- Other studies should be conducted to determine the levels of heavy metals in other food crops such as maize, cassava and potatoes grown around artisanal and small scale mining areas in Kahama and Geita districts and estimating the risk of heavy metals exposure through consumption of such contaminated food.

B. For community or authorities

- The concentration of heavy metals in the agricultural soils and rice in Kahama and Geita districts should be constantly monitored to minimize heavy metals exposure through food.
- There is a need to ascertain the source of these heavy metals by the mine wastes.
- Government and environmental agencies should introduce immediate measures to prevent heavy metals exposures in Kahama and Geita districts.

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APPENDICES

QUESTIONNAIRE

THE NELSON MANDELA AFRICAN INSTITUTION OF SCIENCE AND TECHNOLOGY

SCHOOL OF MATERIAL, ENERGY, WATER AND ENVIRONMENTAL SCIENCE

QUESTIONNAIRE FOR THE RESPONDENTS OF THE STUDY AREA

Region............................... District.......................

Village……………………. Ward…………………

A. Population Information

1. Sex…… 4. Occupation…………………….

2. Age…… 5. Education level…………………

3. Marital status……

B. Rice growing practice

1. Do you grow rice using irrigation or rain fed farming system?

……………………….

2. Do you use fertilizer in rice growing?

YES/NO…………..

If YES which fertilizer do you use?

3. Which month do you harvest rice?

…………………………

4. Do you dry rice after harvesting?

YES/NO…………………

If YES, how do you dry? ……………………………………………………………

C. Rice meal intake questions

1. How often do you eat rice?

   A) Daily

   B) Weekly

   C) Monthly

   D) Others (explain)………………………………..

2. How many times per A) Day…….. , do you use to eat rice?

   B) Week…..

   C) Month….

   D) Others…..

3. Which amount of rice (in gram) do you take per meal?

   …………………………………………………………..

4. Which food do you consume when you do not eat rice as a meal?

………………………………………………………………………