

**CHEMICAL HAZARDS AND HEALTH RISKS OF CONSUMING RICE
GROWN AROUND MINING AREAS IN TANZANIA: A CASE OF GEITA
DISTRICT**

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**A DISSERTATION SUBMITTED IN PARTIAL FULFILMENT OF THE
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ABSTRACT

The main objective of this study was to assess health risks that may be associated with consumption of rice grown in three selected Wards of Rwamgasa, Magenge and Kaseme in Geita District. The agricultural soils of these wards are thought to have been contaminated with Mercury (Hg) used in amalgamation of Gold, and Arsenic (As) from anthropogenic sources (Arsenopyrites). Rice samples were collected from 15 villages of the selected Wards after which the concentrations of Hg and As were determined. The Association of Official Agricultural Chemists (AOAC 2015.01) was used for quantification of heavy metals. Macro Plasma Emission Spectroscopy (MP-AES 4210) equipped with Auto sampler SPS4 was used in this case to quantify Hg and As. Concentrations (ppm) for each metal was significantly different ($p < 0.05$) among sampling locations. The concentration of Hg and As per kg of rice in the sampled villages ranged from 0.78 mg/kg to 3.58 mg/kg at Rwamgasa and Mnekezi villages and 0.64 mg/kg to 1.01 mg/kg at Nyamalulu and Bingwa villages, respectively. Exposure analysis was established in line with health risks characterization for adults of 65 kg and children with 30 kg average body weight in which the calculated exposure dietary intake (EDI) was calculated. These results revealed that, concentration of Mercury exceeded the Maximum Allowable Concentration (MAC) of 0.02 mg/kgbw/day as per Ministry of Health of Health of the Republic of People of China (MHRPC). Further more, the health risk index (HRI) was computed as per USEPA/IRIS (2013) in which, for Mercury the Health Risk Index (HRI) exceeded 1 for children in villages of Rwamgasa, Iseni, Nyakayenze, Magenge, Nyamtondo, Nyamalimbe and Msasa implying health risks to children but there was no risks for adults. In the case of Arsenic it was observed that it did not pose health risks in both age groups since the concentration did not exceed the MAC of 0.35 mg/kgbw/day approved by FAO/WHO (2016).

DECLARATION

I, MAKAYA WILLIAM NSONDA, do hereby declare to the Senate of Sokoine University of Agriculture that, this thesis is my own original work done within the period of registration and that it has neither been submitted nor concurrently being submitted in any other institution.

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May the Lord richly reward and bless you.

DEDICATION

This work is dedicated to my parents Mr and Mrs Nsonda.

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LIST OF ABBREVIATIONS

AOAC	Association of Official Agricultural Chemists
ASTDRs	Agency for Toxic Substances and Disease Registry
As	Arsenic
BW	Body Weight
C	Concentration
DC	District Council
DMA	Dimethylarsonic acid
EDI	Estimated Dietary Intake
EFSA	European Food Safety Agency
FAO/WHO	Food and Agriculture Organization/World Health Organization
GEP	Global Environmental Protection
HACCP	Hazard Analysis Critical Control Point
HRI	Health Risk Index
Hg	Mercury
HQ	Health Quotient
IARC	International Agency for Research on Cancer
IRi	Variation in intake rate
JECFA	Joint Food and Agriculture Organization/World Health Organization Expert Committee of Food Additives
Kg	Kilogram
LOD	Limit of Detection
LQD	Limit of Quantification
TDI	Tolerable Dietary Intakes
TFDA	Tanzania Food and Drugs Authority

THRI	Total Health Risk Index
MeHg	Methyl Mercury
Mg	Milligram
MHPRC	Ministry of Health of People of Republic of China
MMA	Monomethylarsonic acid
MP-AES	Macro Plasma Atomic Emissions Spectroscopy
MSIS	Multimode Sample Introductive System
NBS	National Bureau of Statistics
NEMC	National Environmental Management Committee
PTWI	Provisional Total Weekly Intake
ppm	Parts Per Million
RfDo	Oral Reference Dose
R ²	Coefficient of Correlation
STAMICO	State Mining Corporation
UNDP	United Nation Development Programme
UNEP	United Nation Environmental Programme
UNIDO	United Nation Industrial Development Organization
USEPA	United States Environmental Protection Agency

CHAPTER ONE

1.0 INTRODUCTION

1.1 Background Information

There is a considerable evidence that human foods are frequently subjected to some form of contamination by heavy metals such as Lead, Arsenic, Mercury as a result of environmental pollution and anthropogenic activities (Berka,2009). Heavy metals such as mercury are important potential harmful environmental pollutants (Khanna, 2011; Orina, 2012) and the widespread of contamination with heavy metals in last decades has raised public and scientific interest due to their dangerous effects on human health (Ibrahim, 2011).

In Tanzania, artisanal and small scale mining activities are conducted in areas where rice production is also high, such areas are Kahama and Geita in the Lake Victoria zone. This suggests that, there are high chances of the rice produced from these areas to be contaminated with heavy metals at levels that exceed regulatory limits. 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 (Mathews *et al.*,2012). Due to this reason, protecting the agricultural soils in Tanzania 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 produced in Tanzania are limited and scattered.

Small scale miners in Geita use amalgamation process to recover gold from gold ores. The ore is initially finely ground then mixed with water to form a pulp. The pulp is then mixed

with liquid mercury allowing the gold to amalgamate onto mercury to form spongy amalgam. After a press or filtration process to remove excessive mercury from amalgam, gold is recovered from the amalgam by burning it over a fire, which releases mercury vapour to the atmosphere. The pressing process releases substantial amounts of mercury in to the environment through seepage and leakage (Appleton *et al.*,2004)

Rice (*Oryza Sativa*) is the third most important cereal crop in the world. In Tanzania it is consumed as the second major staple food after maize. Recently, the concern has been raised about possible contamination by heavy metals such as Mercury (Hg), Arsenic (As), Lead (Pb) and Cadmium (Cd) in soil, water and air due to industrialization. The contaminants can be accumulated, transferred and up taken by rice. *Fu et al.* (2008) found Pb with the mean level of 0.69 mg/kg in polished rice in a typical electronic waste recycling area from the Southeast China. Also studies of rice grown in highly contaminated area in China found mean levels of 0.080 mg/kg (As), 0.037 mg/kg (Cd) 0.005 mg/kg (Hg) and 0.060 mg/kg (Pb) (Huang *et al.*, 2013).

In the artisanal mining sites, there is a likelihood of heavy metal contamination of water from tailings and waste water from mining processes to contaminate crops grown in this area. In nature, Arsenic occurs primarily in its sulfide form in complex minerals containing silver, lead, copper, nickel, antimony, cobalt, and iron. As is present in more than 200 mineral species, the most common of which is arsenopyrite. Terrestrial abundance of arsenic is approximately 5 mg/kg, although higher concentrations are associated with sulfide deposits. Sedimentary iron and manganese ores as well as phosphate-rock deposits occasionally contain levels of arsenic up to 2900 mg/kg (WHO, 2001). All types of arsenopyrite contain gold (1250–3000 ppm) and other trace elements such as Cu

(200–1300 ppm), Sb (up to 9000 ppm) and Tl (3000–4500 ppm) as showed in the study by Cepedal *et al.* (2008).

1.2 Problem Statement and Justification of the Study

It has been mostly reported that the extent of heavy metals contamination in agricultural soils is influenced by their closeness to mining or industrial areas (Appleton *et al.*, 2005).

For instance, the increased mining, agricultural, industrial activities and domestic discharges around farming land if not properly managed, its likely to cause heavy metal contamination (Owa, 2013). Different studies have revealed health effects of heavy metals in humans, however, the toxicity of metal contamination depends on several factors. These factors include the dose, the route of exposure and nutritional status of the exposed individuals (Tchouwou *et al.*, 2012). High toxicity levels of Hg, Arsenic and Lead rank highly among the priority metals with public health significance (Khillare *et al.*, 2015).

Mercury is a toxic pollutant, which is distributed worldwide from both natural and anthropogenic sources, and cannot be broken down in the environment (Jaeger *et al.*, 2009). Hg has a half life of 1–2 years in the atmosphere, which allows it to be transported over long distances via oceanic and atmospheric processes (Li *et al.*, 2009b; Selin, 2009). There has been increasing attention on the neurotoxicity and neurodevelopmental risk among people exposed to low or moderate levels as once released into the environment. Mercury is quickly transformed into organic compounds by aquatic microorganisms mainly as methyl mercury (MeHg) which is more toxic than elemental and inorganic forms of Hg (Davidson *et al.*, 2004).

However, organic Hg compounds are more easily absorbed via ingestion than inorganic Hg compounds (WHO, 2003). More than 90% of MeHg could be absorbed and accumulated in the body posing a risk of damage to the neurological system, cardiovascular and reproductive systems (Mergler *et al.*, 2007). Some specific pediatric health effects are subtle neuro developmental abnormalities, such as visual spatial errors (Chevrier *et al.*, 2009), decrements in motor speed and attention have been reported (Debes *et al.*, 2006).

Lack of capital to purchase the requisite crushing and milling equipment to facilitate the process compels artisanal miners to use rudimentary manual methods of gold extraction which involves “pounding” (crushing and grinding) of ores using locally designed metal mortars and pestles. The resultant powder is mixed with water and sluiced to obtain a gold concentrate, which is later amalgamated with Hg (Benjamin *et al.*, 2002). These processes expose miners to heavy metals as well as soil and water pollution. Health of miners and other people living within the areas may be affected through inhalation of Hg vapour or contaminated dusts, direct contact with Hg, eating fish, cereals and other foods also through ingestion of water affected by Hg and As contamination. Although numerous studies on Hg pollution have been done in this area limited studies have been conducted on chemical hazard exposure assessment on Hg for consumers of rice in the study area. A number of studies indicate that in countries not suffering from high level of As in drinking water, rice is a major contributor to inorganic As in human (Meacher *et al.*, 2002).

Rice on other hand has high proportions of inorganic As (Ackerman *et al.*, 2005) and it is particularly susceptible to assimilating arsenic into its grain (Williams *et al.*, 2007). Preliminary data released by United States Food and Drugs Administration (U.S.FDA)

revealed that rice had higher levels of inorganic As than any other food, because as the rice plant grows tend to absorb Arsenic more readily than other food crops. Rice is a staple in the US which is widely consumed by the population including by infants. Additionally rice intake primarily through infant rice cereal is about three times greater for infants than adults in relation to body weight (U.S.FDA report, 2011). The U.S.FDA report has prompted similar study to be conducted similarly in Tanzania. Rice is a staple food grown in areas thought to be contaminated with these heavy metals. The European Food Safety Authority (EFSA) has assessed along with many other international authorities, that As is a substance that should be avoided as much as possible (EFSA, 2015). Therefore, the findings of this study will provide necessary information to various institution that will guide them on measures in minimizing damages caused by heavy metals in question to the health of individuals in the research areas.

1.3 Study Objectives

1.3.1 General objective

To assess chemical hazards and health risks of consumers of rice grown around mining areas in Geita district.

1.3.2 Specific objectives

Specific objects were to :-

- i. Determine levels of contamination of heavy metals (Hg and As) in rice grown around selected gold mining areas of Geita District.
- ii. Assess the health risk to consumers of rice contaminated with Hg and As.

1.4 Hypothesis

H₀: Chemical hazards and health risks of consumers of rice grown around mining areas contains safe levels of Mercury and Arsenic.

H₁: Chemical hazards and health risks of consumers of rice grown around mining areas contains unsafe levels of Mercury and Arsenic.

CHAPTER TWO

2.0 LITERATURE REVIEW

2.1 Overview

This chapter summarizes literature relating to heavy metals pollution, toxicity of heavy metals, methylation of Hg and As, human exposure to Hg and As, risk based approach to food safety, tolerable dietary intakes (TDI) of heavy metals, estimated dietary intakes (EDI) of heavy metals and risk assessment to heavy metals via rice consumption.

2.2 Heavy Metals

Heavy metals can be defined as a group of elements with a density higher than 5 g/cm³. respect to their toxicity, they can be divided into two groups namely micronutrients like Fe, Mn, Mo, Cu, Cr, Ni and Zn that are essential in small amounts and the toxic like Ar, Cd, Hg and Pb without any known biological importance (Lottermoser, 2007). Heavy metals are extremely persistent in the environment because of their non-biodegradable nature, long biological half-lives, thermal stability and potential to accumulate to toxic levels in both plants and animals (Adah *et al.*, 2013). Even at low concentrations, heavy metals have been reported to produce damaging effects on human and animals (Adah *et al.*, 2013).

2.3 Heavy Metals Pollution

Heavy metals applications in industrial, domestic, agricultural sphere have led to their wide distribution in the environment. However, their potential effects on human health and environment have raised concerns among researchers. This is because these metals are commonly associated with pollution and toxicity problems (Ratan and Verma, 2014). Studies in Ghana raised a great concern of potential risk of exposure to gaseous Mercury

(Hg) during amalgamation process (Hogarh *et al.*, 2016). As, the important element in iron ores, have been implicated with soil contamination around mining areas in China where high levels of As have been also reported (Liao *et al.*, 2005). Mining methods employed by small-scale miners vary according to the type of deposit being exploited and its location. In view of the poor financial base of small-scale miners, a great majority rely solely on traditional/manual methods of mining, which are largely artisanal, featured by simple equipment like shovels, pick-axes, pans, chisels and hammers in which environmental pollution is not considered (Benjamin *et al.*, 2002).

The speciation of Hg in the soil environment is dynamic where it can be biotically and abiotically interconverted between the dominant solution phase inorganic species of Arsenate (As_v) and Arsenite (As^{III}), the oxidized and reduced form respectively (Abedin *et al.*, 2002). Inorganic As can also be methylated through microbial action to give monomethylarsonic acid (MMA) and dimethylarsinic acid (DMA). All four species are present in the solution phase of paddy soils (Abedin *et al.*, 2002). These four species can also be assimilated by rice roots.



Figure 1: Photo taken during mapping of study area contaminated swamp at Magenge 2016.

2.4 Mercury and Arsenic

Mercury (Hg) is a naturally occurring metal that is present in several forms. Metallic mercury is shiny, silver-white and odorless liquid. Hg, which has the lowest melting point (-39°C) of all the pure metals, is the only pure metal that is liquid at room temperature. However, due to its several physical and chemical advantages such as high boiling point (357°C) and easy vaporization. Hg is still an important material in many industrial products.

Mercury exists in several forms namely elemental or metallic mercury, inorganic mercury compounds, and organic mercury compounds. The most reduced is Hg metal with the other two forms being ionic of mercurous ion and mercuric ion (Hg^{2+}) in oxidizing conditions especially at low pH. Hg^{+} ions are not stable under environmental conditions since it dismutates into Hg and Hg^{2+} (Chang *et al.*, 2009).

Exposure routes for Mercury to the environment contamination due to Hg is caused by industrial activities, petrochemicals and also by agricultural sources such as fertilizer and fungicidal sprays. Some of the more common sources of Hg found throughout the environment include mining activities but may not be limited to the household bleach, acid, and caustic chemicals (e.g. battery acid, household lye, muriatic acid (hydrochloric acid), sodium hydroxide, and sulfuric acid), instrumentation containing Hg (e.g., medical instruments, thermometers, barometers and manometers), dental amalgam (fillings), latex paint (manufactured prior to 1990), batteries, electric lighting (fluorescent lamps, incandescent wire filaments, mercury vapor lamps, ultraviolet lamps), pesticides, pharmaceuticals (e.g. nasal sprays, cosmetics, contact lens products), household detergents and cleaners, laboratory chemicals, inks and paper coatings, lubrication oils, wiring devices and switches and textiles (Resae *et al.*, 2005). Until the 1970s organomercurials,

especially methyl and ethyl mercury, were widely used in agriculture as antifungal agents in seed grain. This practice was discontinued as a result of a number of mass poisonings both humans and in certain wildlife species; diuretic, and/or cathartic properties in Europe, North America, Australia, and elsewhere (WHO, 2003; Clarkson and Magos, 2006). The largest present intentional use of Hg is by artisanal and small-scale gold miners because many metals including gold dissolve in Hg to form amalgams (alloys) (Landis and Yu, 2004).

Studies conducted by Clarkson and Magos (2006) Hg compounds were used in skin creams to treat infections. The medical applications of Hg compounds were common during the 20th century. For example, the addition of calomel (Hg_2Cl_2) to teething powders. Mercuric chloride, mercuric oxide, mercuric iodide, mercurous acetate, and mercurous chloride are, or have been, used as antiseptic.

Arsenic is a chemical element with the symbol As, atomic number 33 and relative atomic mass 74.92. It has a specific gravity 5.73 g/cm^3 , melting point of 817°C (at 28 atmospheres). It boils at 613°C and a vapor pressure of 1 mmHg at 372°C . Arsenic is a semi metallic, odorless and tasteless element (Mohan *et al*, 2007.). As is number one on the ATSDR's toxic and hazardous substances "Top 20 List," and is the most common cause of acute heavy metal poisoning in adults. From a biological and toxicological perspective, there are three major groups of arsenic compounds namely;

- Inorganic arsenic compounds
- Organic arsenic compounds and
- Arsine gas.

Of the inorganic arsenic compounds, arsenic trioxide, sodium arsenite and arsenic trichloride are the most common trivalent compounds. Arsenic pentoxide, arsenic acid and arsenates (e.g. lead arsenate and calcium arsenate) are the most common pentavalent compounds. Common organic Arsenic compounds include arsanilic acid, methylarsonic acid, dimethylarsinic acid (cacodylic acid) and arsenobetaine (WHO, 2000).

At intermediate redox conditions, such as those found in paddy soils which continuously fluctuate between aerobic and anaerobic conditions, As is mobilized from both pyrites and oxy hydroxides as the relatively mobile Arsenite (Smedley *et al.*, 2002). Thus, for aerobically grown crops the relatively immobile arsenate is the dominant plant available form. But for anaerobically cultivated rice the more mobile Arsenite is the dominant plant available form. The inorganic forms Arsenate and Arsenite are the dominant species in soil solution, the former predominant in aerobic soils and the later in anaerobic soils (Abedin *et al.*, 2002). Methylated species are also widely found in soils, particularly monomethylarsonic acid (MMA) and dimethylarsinic acid (DMA), which are found at high proportions in paddy soils (Abedin *et al.*, 2002).

2.5 Toxicity of Mercury and Arsenic

The toxicity of these metals has two main aspects:

- (a) The fact that they have no known metabolic function, but when present in the body they disrupt normal cellular processes, leading to toxicity in a number of organs;
- (b) The potential particularly of the so-called heavy metals Hg and As, to accumulate in biological tissues, a process known as bioaccumulation.

This occurs because the metal, once taken up into the body, is stored in particular organs, for example liver or kidney and is excreted at a slow rate compared with its uptake. This process of bioaccumulation of metals occurs in all animals, including food animals such as fish and cattle as well as humans. Agency for Toxic Substances and Diseases Registry (ATSDR,1999) ranked Mercury to number three on the "Top 20 List" of toxic and hazardous substances. Mercury is generated naturally in the environment from degassing of the earth's crust, from volcanic emissions. It is therefore necessary to control the levels of these toxic metals in foodstuffs in order to protect human health (Ali andAl-Qahtani, 2012).

Mercury damages the developing brain, and causes a lifelong negative effect in exposed population (Bose-O'Reilly *et al.*, 2008). When exposed to high levels of mercury vapour, children exhibit a syndrome known as acrodynia (painful limbs) or pink disease (Davidson *et al.*, 2004). Garca-Fernandez *et al.*, (1996) reported that, the health problems caused by Mercury toxicity include headache, metabolic abnormalities, respiratory disorders, nausea and vomiting. Inorganic Hg occurs as salts of its divalent and monovalent cationic forms (WHO, 2003). Mercury salts affect primarily the gastrointestinal tract and the kidneys, and can cause severe kidney damage. Both inorganic and organic Mercury compounds are absorbed through the gastro intestinal tract and affect other systems via this route (WHO, 2003).

Despite the wide range of metal toxicity and toxic properties, there are a number of toxicological features that are common to many metals. Metals toxicity depends on several factors including the dose, route of exposure, and chemical species, as well as the age, gender, genetics, and nutritional status of exposed individuals (Tsuzuki *et al.*, 1994).

Because of their high degree of toxicity, heavy metals like Arsenic, Cadmium, Chromium, Lead, and Mercury rank among the priority metals that are of public health significance (Tsuzuki *et al.*, 1994). Concern over such incidents of toxicity, has prompted numerous investigations into the metabolism and toxic effects of these elements (Ali and Al-Qahtani, 2012).

One of the earliest, and the most comprehensively documented case of Hg poisoning in man and other animals occurred in the 1950's at Minamata Bay and 1965 in Niigata, in Japan especially among fishermen and their families. The source of the Hg was discharged wastes from an acetaldehyde plant that used inorganic Hg as a catalyst (Hodgson, 2004). The illness became known as the "Minamata Disease" (Lourie, 2003).

Minamata disease reached epidemic proportions during 1956-1965 where 111 cases of poisoning were reported and 41 deaths. By 1982, there were 1800 verified human victims of Hg poisoning in a total regional population of 200,000 (Hodgson, 2004; Eisler, 2007; Selin, 2009), however, the total number of victims remained unconfirmed (Hodgson, 2004; Selin, 2009). Abnormal Hg content of more than 30 mg/kg fresh weight was measured in fish, shellfish, and mud from the Bay and in organs of necropsied humans and cats that succumbed to the disease (Hodgson, 2004).

Another sentinel outbreak occurred in rural Iraq in 1971-1972 from seed grain treated with organo-mercurials as fungicides. Farmers received around 95,000 tonnes of wheat and barley seeds, the grain was intended for planting, (Lourie, 2003; Selin, 2009), but ended up being consumed (Lourie, 2003). Tragically, more than 6500 individuals were hospitalized and 459 died from consumption of Hg contaminated bread. The Iraqi

government broadcasted warnings to avoid eating the grain. However, people in rural areas did not have radios, did not believe the warnings or chose to ignore them because they lacked other types of food. When it faced the initial disaster of the Hg treated seed being distributed among the farmers, the government announced that any farmer possessing these seeds was liable to prosecution involving death. The peasants then disposed the hazardous seeds in nearby rivers and lakes, spreading the contamination to even more remote regions (Veiga and Baker, 2007).

In general, inorganic compounds of Arsenic are regarded as more highly toxic than most organic forms which are less toxic (Andrianisa *et al.*, 2008). The first symptoms of long-term exposure to high levels of inorganic As (e.g. through drinking-water and food) are usually observed in the skin, and include pigmentation changes, lesions and hard patches on the palms and soles of the feet (hyperkeratosis) (Ikeda *et al.*, 2000). These metallic elements are considered as systemic toxicants which are known to induce multiple damages to human organs, even at lower levels exposure (Jan *et al.*, 2015). It also seems to have a negative impact on reproductive processes (infant mortality and weight of new born babies) (Hopenhayn, 2006). As is classified as an established human carcinogen by the International Agency for Research on Cancer (IARC). Epidemiologic studies have provided substantial evidence for the association of As in drinking water with skin cancers (non-melanoma), lung and bladder. Limited epidemiologic evidence also suggests a possible association of As in drinking water with liver, kidney, and prostate cancers (International Agency for Research on Cancer, 1987).

2.6 Mercury Mode of Action in Human Body

Direct effects usually occur when the biochemistry and physiology of an organism are altered by abnormal chemical action. Several mechanisms of chemical toxicity are

recognized (Landis and Yu, 2004). They include interruption, blocking, or interference of specific cellular functions, such as the increased production of highly reactive molecules such as hydrogen peroxide (H_2O_2), macrophage disruption and Nucleic acids may be broken or bound to reactive molecules. Cellular responses to exposure to contamination may include stimulation of protection mechanisms, such as lysosomes and stress proteins. Tissue damage may occur as a result of these cellular processes, and may result in developmental abnormalities (Thompson *et al.*, 2007).

Brain pyruvate metabolism is known to be inhibited by Hg, as are lactate dehydrogenase and fatty acid synthetase (Timbrell, 2004). Mercury inactivates the Na^+/K^+ -ATPase, which leads to membrane depolarization, calcium entry, and eventual cell death (Landis and Yu, 2004; Huang *et al.*, 2008). Further more, it decreases α - and γ -globulins while increasing β -globulin, causing liver dysfunction, decreases DNA content in cells by affecting the DNA polymerase activity and adversely affects chromosomes and mitosis by disrupting the microtubule formation (Landis and Yu, 2004; Gupta, 2007), leading to mutagenesis (Landis and Yu, 2004).

2.7 Health risk Assessment Approach

Risk assessment is a process of estimating the probability that a chemical substance to cause adverse effects to a given population. Under particular conditions of exposure, risk assessment provides the logical basis for public health decisions and actions aimed at reducing or eliminating the risk involved. Health risk assessments are used to determine if a particular chemical poses a significant risk to human health and, if so, under what circumstances. It is important to assess the daily intake of metals from rice and compare it with the Tolerable Daily Intake (TDI) values set by international organizations for health safety (Hajeb *et al.*, 2009).

The common approaches for food safety risk analysis; include Codex Alimentarius Commission framework. The risk analyses applied in the food safety context are the Codex Alimentarius Commission risk analysis and HACCP frameworks.

The present study used the Codex Alimentary Commission framework for heavy metals analyses. Risk assessment consists of the following steps:

- i) Hazard identification,
- ii) Hazard characterization,
- iii) Exposure assessment, and
- iv) Risk characterization.

The risk assessment definitions used in this study are similar to that of Potter (1996):

Hazard: A biological, chemical or physical agent in or property of food with the potential to cause an adverse effect.

Hazard identification: Identification of known or potential health effects associated with a particular agent in food

Exposure assessment: The evaluation of degree of intake likely to occur.

Risk characterization: The estimation of the adverse effects likely to occur in a given population, and a summary of assumptions and sources of uncertainty.

Dose response assessment: Determination of relationship between the magnitude of exposure and magnitude of frequency of adverse effect.

CHAPTER THREE

3.0 RESEARCH METHODOLOGY

3.1 Overview

This chapter is divided into five main sections;- study area, rice samples collection and preparation, methods and procedures for analysis including digestion of samples, optimization of Agilent technologies 4210 MP-AES and quantification of Hg and As.

3.2 Study Area

This study was conducted in Geita district which is one of the major gold mining areas in Tanzania. Geita district is located at latitude -2.8667 and longitude 32.2500. According to the 2012 census given by NBS, the population of the district was 807,619. The district is bordered to the East by Mwanza region and Nyang'hwale district, to the South by Shinyanga region and Mbogwe district, and to the West by Chato district. Geita district is characterized by a large number of small scale mines including Rwamgasa, Tembomine, Iseni, Buziba, Mgusu mines and others.

3.3 Study Design and Sample Size

Cross-sectional research design was conducted in this study. A household survey independent structured questionnaire (with closed ended questions) was used to gather information on household rice consumption patterns. Purposively sampling was used in which rice fields were sampled within the wards in the villages with mining activities and rice cultivation. The sample size was estimated using the formula for infinite population as proposed by Kothari (2004).

$$n = \frac{Z^2 P(1 - P)}{e^2}$$

Where by n = size of sample, P = Sample proportion, assuming 5% (0.05) for this study
 e = acceptable error (the precision), set at 5% (0.05) for this study and z = standard variate
 at a given confidence level, for this study 95%, confidence level= 1.96 (Kothari, 2004).

$$\begin{aligned} \text{Thus, } & 1.96^2 \times 0.05(1 - 0.05) / 0.05^2 \\ & = 72.99 \approx 73 \end{aligned}$$

3.4 Method of Investigation

Data was collected in two main ways; primary and secondary.

3.4.1 Primary data collection

The primary data was collected through different quantitative and qualitative methods. The data was obtained from the field through surveys of the project through administration of questionnaires and personal interviews. Samples of rice for laboratory analysis was collected from each village as per sampling plan.

3.4.2 Questionnaires administration

A structured questionnaire, consisted of both open-ended and closed-ended questions which were administered in each of the selected areas.

3.4.2 Secondary data

Secondary data were obtained through review of published books and journals.

3.5 Sample Collection

The District has thirty five (35) Wards but only three wards of Rwamgasa, Magenge, and Kaseme were studied. The combined population of the wards is about 180,500 and average house hold of six (6) individuals (NBS, 2012). The choice of the research area was based on the presence of rice cultivation and mining activities. Samples were collected from farmers during harvesting in which a total of seventy five (75) samples were collected in villages from the three wards of Magenge, Kaseme and Rwamgasa where mining activities have been conducted for long period of time. Five (5) samples from five (5) villages from each of the three (3) wards were collected and placed in plastic bags.

Table 1: Samples collected

Locations	Number of Samples
Rwamgasa Ward	
Rwamgasa	5
Bingwa	5
Iseni	5
Imalanguzu	5
Nyakayenze	5
Maagenge Ward	
Maagenge	5
Nyamalulu	5
Nyamtondo	5
Nyamalimbe	5
Msasa	5
Kaseme Ward	
Sobola	5
Kaseme	5
Mnekezi	5
Nyamisebehi	5
Tembomine	5
Total	75

3.6 Sample Preparation

The samples were dehusked separately placed in plastic bags, stored in a cool dry place. The dehusked rice samples were grounded with a help of a blender to obtain fine flour, and placed in plastic bags and stored in a dry cool place for further analysis.

3.6.1 Preparation of 1000ppm stock AAS standard solutions for Mercury

A Mercuric (Hg) standard stock solution of 10ppm was prepared from 1000ppm standard using 2% Nitric acid. This stock solution was used to prepare solutions for calibration curve (0.00, 0.50, 1.00, 2.00 and 3.00ppm) at the intervals of 0.50ppm. The standard was used for injection into Agilent Technologies MP-AES 4210 for quantification (AOAC, 2015.01)

3.6.2 Preparation of 1000ppm stock AAS standard solutions for Arsenic

An Arsenic (As) standard stock solution of 10ppm was prepared from 1000ppm standard using 2% Nitric acid. This stock solution was used to prepare solutions for calibration curve (0.00, 0.10, 0.50, 1.00 and 2.00ppm) at the intervals of 0.50ppm. The standards were ready for injection into MP-AES 4210 for heavy metal quantification (AOAC, 2015.01).

3.6.3 Sample digestion for heavy metals

The official method AOAC (2015.01) was used to quantify Hg and As in which MP-AES 4210 equipped with Auto Sampler S4 and Nitrogen Generator.

3.6.3.1 Ashing by drying

About 5g of finely grounded rice was placed in a Platinum desiccator. The sample was gently heated on a hot plate until enough water was driven off for partial carbonization to

occur, then placed in an electrical furnace and increased the heat at a rate of 100 °C per hour. Then the sample was heated at 500°C for about 8hours. 2ml of Nitric acid was added. After drying, 2ml of water was added followed by addition of hydrochloric acid to dissolve the salts. Deionized water was used to prepare fixed volumes of measurement solutions. The sample was injected into MP-AES 4210 for quantification. Digestion of blank reagent blank was also performed in parallel with rice samples keeping all digestion parameters the same.

3.6.3.2 Wetashing

About 0.5g of finery grounded rice was added into digestion flask where 4ml of 32% Nitric acid was added followed by 1ml of 30% Hydrogen peroxide. Sample was digested by Ethos Easy Microwave digester at 150°C for 45minutes. The extract was filtered through whatman filter paper No 1 and transferred to 50ml volumetric flask; the flask was rinsed with Ultra-pure distilled water then filled the volumetric flask to the mark. The sample was injected into MP-AES 4210 for quantification. Digestion of blank reagent blank was also performed in parallel with rice samples keeping all digestion parameters the same. (AOAC, 2015.01).

3.7 Method and Procedures

AOAC First Action Method 2015.01 was used for quantification of heavy metals. MP-AES 4210 equipped with Auto sampler SPS4 was used.

3.8 MP-AES Optimization

After sample preparation, wavelength optimization was performed in order to select the optional spectral wavelengths for measurement. Multi element standard solutions from

Agilent technologies were analyzed by MP-AES 4210 over the wavelength range of interest to monitor spectral interferences. In this work, a standard reference solution was used to optimize parameters such as nebulizer pressure, number of replicates, read-time and stabilization time (Agilent Technologies MP-AES 4210 Manual).

3.9 Analysis of Metal Contents of Digests

Agilent Technologies MP-AES 4210 equipped with Auto sampler SP4 was used to determine the concentration of digested samples after treatment through different conditions of parameters. The analysis was done at working conditions of MP-AES 4210 for As and Hg as indicated in Table 2 below;

Table 2: Operating conditions for Hg and As byMP-AES

Parameter	Condition employed
Nebulizer	Concentric
Spray chamber	Multimode Sample Introdutive System (MSIS)
Nitrogen consumption (L/min)	20
Read-time (s)	10
Number of replicates	3
Rinse time (s)	30
Sample uptake delay (s)	45
Stabilisation time (s)	15
Pump speed (rpm)	15
Wavelength (nm)	253.652 for Hg and 228.812 for As

3.10 Statistical Data Analysis

Data obtained from the survey were analyzed using IBMSPSS software version 16, where as laboratory data were analyzed usingMicrosoftoffice excel and SAS system version 9.1.3. Analysis ofvariance was used to determine the significant ($p<0.05$) variations in intake rate (IRi). Means were separated by Duncan’s Multiple Range Test.

3.11 Healthy Risk Assessment

3.11.1 Estimated Daily Intake (EDI) of Hg and As through Rice Consumption

The daily intake of metals depends on both the metal concentration in food and the daily food consumption. In addition, the body weight of the human can influence the tolerance of pollutants. The estimated daily intakes (EDI) are the concept introduced to take into account of these factors. Based on the dietary nutrition intake level survey by Zhong *et al.* (2006), the EDI was calculated as follows:

$$EDI = C \times FF/BW$$

Where EDI was the estimated daily intakes, C was the concentration of the heavy metals in contaminated rice, FF-Food Factor for the daily average consumption of rice and BW the body weight.

3.11.2 The health risk index (HRI)

The health risk index (HRI) described by the percentage of the safe value was used for the risk assessment. HRI is calculated by dividing estimated daily intake of heavy metal (EDI) by their Oral Reference Doses (RfDo) for a particular heavy metal. Estimate of risk to human health (HRI) through consumption of rice grown in metal contaminated soil is calculated by the following equation:

$$HRI = EDI/RfDo$$

Whereby,

HRI = Estimate of risk to human health

EDI = Estimated daily intakes

RfDo = Oral Reference Dose is an estimate of daily exposure to the human population that is likely to be without an appreciable risk of deleterious effects during a life time.

Health Risk Index (HRI) is below one, no health risk may occur as a result of ingestion of the rice. The greater the value HRI above 1, the greater is the level of risk associated with the rice consumption. Hence, HRI ranging from 0.0 to 1, means no hazard. The HRI ranging from 0.1 to 1.0 means moderate hazard while HRI greater than 1 means that there is high risk for long time consumption (USEPA/IRIS, 2013).

CHAPTER FOUR

4.0 RESULTS AND DISCUSSION

4.1 Respondents Demographic Characteristics (n=75)

In this study, 72% of respondents were male and 28% were female in which 44% were aged between 38-47 years, 24% aged between 28-27 years, 18.7% were above 45 years and 13.3% were between 18-27 years. Most of the respondents (48%) had attained the primary school education level, 14.7% of the respondents had attained secondary education level, 12.3% had attained vocation training and 25% had informal education. About 51% of respondent had a household size of about 7 individuals, 30% had a household of about 5 individuals, 19% had a household of about 10 individuals. In average these results are moreless the same with results by National Bureau of Statistics (NBS),2012 shown that,the household in the study area was 6individuals. All of the respondents practiced rice cultivation, 52% practiced crop production only, 32% practiced crop production and livestock keeping and 16% practiced both crop production and gold mining.

More than half of the respondents (53.3%) obtained all food from their own farms, 47.7% bought some food from the markets and non of them depended on relief food. Regarding to components of breakfast, 30% their breakfast comprised of rice left overs, bans and scones and tea, 28% stiff porridge served with vegetables/beans/meat and tea, 25% did not get formal breakfast and 17% had vitumbua made from rice flour, sweet potatoes/cassava. About 70% of respondents claimed to own rice farms, 20% of hired rice farms each year and 10% were business individuals who bought crops during harvesting. Most of the rice plots owned by the respondents were located away (about 3km) from homestead (16%) and 78% had their rice plots to a distance less than 1km.

Most of the rice plots (84%) were located near by gold amalgamation activities carried out by the use of chemicals (Mercury). Regarding portion size, the respondents about 60% claimed to prepare 1kg of rice/maize flour for 4 people, 30% prepared 1kg of rice for three individuals. In gold amalgamation process almost 97.3% handle Hg without any protective gear. Equipment used included Hammer, slugbox, slantedbox, stoneclusher, rope, basins, waremesh, sisal bags and chisel. All of the respondents claimed to have no knowledge about the adverse health effect associated with Hg. Health problems recorded in the study area included malaria, HIV/AIDS, diarrhoea, chestpain, pneumonia and cancers (skin and urinary systems).

Table 3: Mean Concentration of Hg and As (mg/kg) in rice from different location

Location	Mercury (Hg)	Arsenic (As)
Rwamgasa ward		
Rwamgasa	3.58 ± 0.09 ^a	0.74 ± 0.03 ^b
Bingwa	1.56 ± 0.04 ^d	0.64 ± 0.04 ^c
Iseni	2.57 ± 0.06 ^b	0.94 ± 0.06 ^a
Imalanguzu	1.45 ± 0.01 ^d	0.74 ± 0.02 ^b
Nyakayenze	2.16 ± 0.02 ^c	0.98 ± 0.04 ^a
Magenge ward		
Magenge	2.89 ± 0.01 ^a	0.84 ± 0.08 ^b
Nyamalulu	1.92 ± 0.02 ^e	1.01 ± 0.05 ^a
Nyamtondo	2.02 ± 0.14 ^d	0.78 ± 0.04 ^{bc}
Nyamalimbe	2.35 ± 0.01 ^b	0.73 ± 0.05 ^{cd}
Msasa	2.13 ± 0.05 ^c	0.66 ± 0.08 ^d
Kaseme ward		
Sobola	1.25 ± 0.03 ^c	0.68 ± 0.05 ^c
Kaseme	0.99 ± 0.03 ^d	0.83 ± 0.07 ^{ab}
Mnekezi	0.78 ± 0.02 ^e	0.73 ± 0.05 ^{bc}
Nyamisebhehi	1.64 ± 0.01 ^b	0.84 ± 0.09 ^a
Tembomine	1.92 ± 0.01 ^a	0.83 ± 0.07 ^{ab}

*Mean concentration with the same superscript on the same ward are not significant different at $p < 0.05$

4.2.1 Mercury (Hg) and Arsenic (As) concentration in rice from Rwamgasaward

The results in Table 3, shows the mean concentrations of Hg in rice samples drawn from Rwamgasa ward starting with the highest mean of 3.58 ± 0.09 mg/kg for Rwamgasa,

followed by the mean of 2.57 ± 0.06 mg/kg for Iseni, 2.16 ± 0.02 mg/kg for Nyakayenze, 1.56 ± 0.04 mg/kg for Bingwa and 1.45 ± 0.01 mg/kg for Imalanguzu village. The mean concentration of Arsenic was detected in the following trend starting with the highest mean of 0.98 ± 0.04 mg/kg (Nyakayenze), 0.94 ± 0.06 mg/kg (Iseni), 0.74 ± 0.02 mg/kg (Imalanguzu), 0.74 ± 0.02 mg/kg (Rwamgasa) and 0.64 ± 0.04 mg/kg (Bingwa). There was a significant difference in mean concentration of mercury in rice samples ($P < 0.05$).

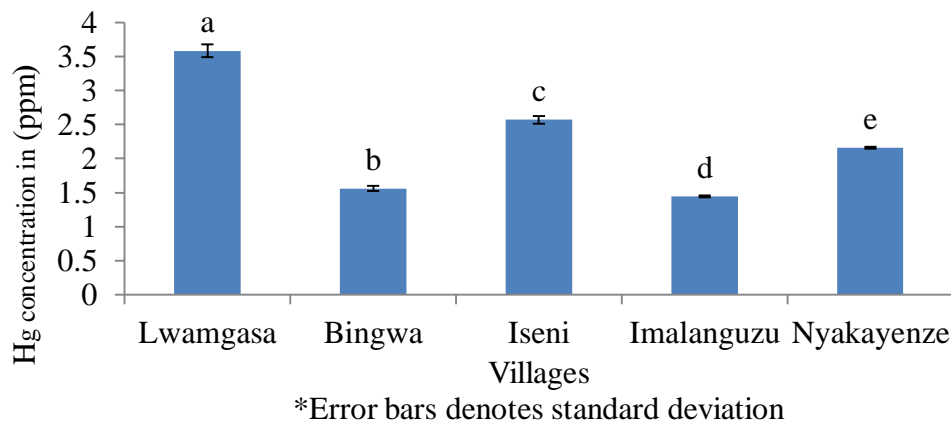


Figure 1: Variation of Hg in rice from Rwamgasa ward

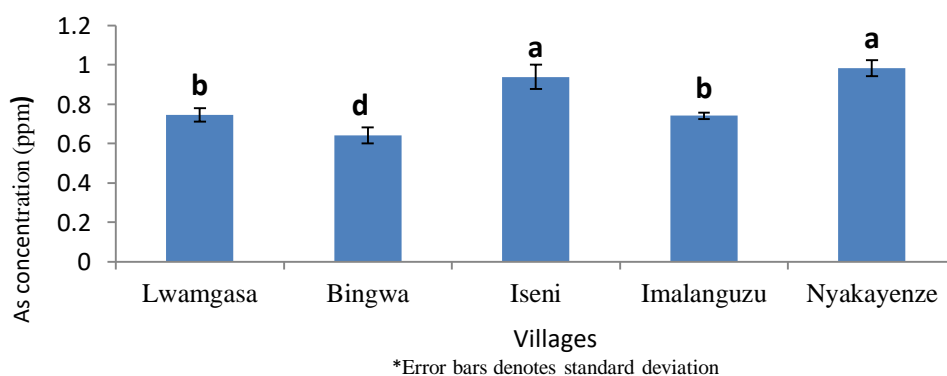


Figure 2: Variation of As in rice from Rwamgasa ward

Studies by Taylor *et al.* (2004) in Rwamgasa village in his finding on Mercury contamination found 0.01 mg/kg and 0.013 mg/kg in dehusked rice, further more samples of pond tailings from Rwamgasa was found to have mercury concentration in a range of

28.5 mg/kg to 193 mg/kg. In this study the range in mercury concentration was found to 1.45 mg/kg to 3.58 mg/kg and the rice plants are grown mostly depending water from those ponds suggesting that they absorbed the same from the tailings. Arsenic concentration in Rwamgasa ward ranged from 0.64 mg/kg to 0.98 mg/kg which is inline with finding by Jiang *et al.* (2015) in Suxian country Human province in Southern China found concentration of Arsenic in rice ranging from 0.02mg/kg to 1.48 mg/kg of rice.

The highest concentration of Hg in Rwamgasa ward (Table 4) in rice samples was obtained from Rwamgasa village 3.58 ± 0.09 mg/kg and the lowest concentration in rice samples from Imalanguzu village 1.45 ± 0.01 mg/kg. The concentration of Hg in rice samples in Rwamgasa ward was significantly high ($P < 0.05$) compared to levels detected from other wards. The highest levels of Hg concentration in rice samples from Rwamgasa might be due to intensive mining activities that have been carried in Rwamgasa greenstone belt since colonial era which was very active in the 1930's and 1950's with main mines in the area like Mawemeru, El Dorado and STAMICO according to study by Appleton *et al.*, (2004). Also artisanal miners have been working at Rwamgasa since 1974. It has been estimated by BGS (British Geological Survey) that, in average, around 30 kg of Mercury is released into the environment from Rwamgasa village annually (Appleton *et al.* 2004). Also, the high levels of Hg concentration in rice samples were found at Iseni and Nyakayenze villages. This might be attributed by the reason that, there was an ongoing processing of gold ores and reprocessing of tailing. The concentration of Hg in rice samples grown in Rwamgasa is higher compared to those of rice samples grown on the highly Hg contaminated Philippines, which had a mean of 0.016 mg soils of Naboc irrigation system ranged from 0.008 mg/kg to 0.05 mg/kg Hg wet weight as reported by Appleton (2000). The findings of this study are similar to those reported by Taylor *et al.*

(2004) who found Hg concentration range of 0.005 mg/kg to 5.1 mg/kg of rice in the same area. However the values observed in this study are higher than those reported by Machiwa *et al.* (2003) in Saragurwa, Nyarugusu and Nungwe in Geita district, which had lower concentrations of 0.04 mg/kg, 0.09 mg/kg and 0.13 mg/kg of rice respectively. However, in agricultural soils, elevated concentrations of Hg and As are mostly found in the immediate surroundings of gold processing sites.

The elevated Hg concentrations in the soils are probably a result of Hg physically lost by the small-scale miners during the gold amalgamation process. Elevated As concentrations in the soils are most probably related to As content of the inherent mineralogy of the ores which contain the mineral arsenopyrite FeAsS_2 . These results of high As and Hg concentration in soils close to amalgamation sites are consistent with findings conducted in South America by Lacerda and Salomons (1998). Studies of As concentration by William *et al.* (2007) focused on the Mississippi delta/flood plain and Texas, which has long been the practice to grow rice on soils previously used for cotton production when cotton was treated against *Anthonomus grandis* (boll weevil) infection with arsenical pesticides. It was suspected that this past arsenical pesticide usage was a reason why South Central US rice has an average arsenic content almost double than that of California rice, this report can apply to the research area due to long term cultivation of cotton.

There was no variation in rice samples from Rwamgasa ward in this study for As except for villages of Nyakayenze and Iseni. The concentration in the ward ranged from 0.64 mg/kg to 0.98 mg/kg at Bingwa and Nyakayenze, respectively (Table 3). Although As concentration may be due to the inherent mineralogy of the ores which contain the mineral arsenopyrite FeAsS_2 . Also application of phosphate fertilizers produced in Tanzania which

consist of Hg, Cd, As, Pb, Cu and Ni may increase the levels of these metals as reported by Lema *et al.* (2014). This study is similar to that of Taiwan by Lin *et al.* (2004) who found the concentration of 0.80 mg/kg for As in rice.

4.2.2 Health risk for adult and children due to consumption of rice from Rwamgasa

The Estimated Daily Intake (EDI) of Hg and As indicated higher values at Rwamgasa village in comparison to other villages in the ward and the entire research area, and were recorded at 1.65×10^{-2} mg/kg bw/day (0.0165 mg/kgbw/day) and 3.58×10^{-2} mg/kg bw/day (0.0358 mg/kgbw/day) for adult and children, respectively (Table 4). The approved safe value in rice for Hg by MHPRC (Ministry of Health of republic of People of China, 2005) is 0.02 mg/kg bw/day. Therefore the EDI detected for children exceeded the safe value in accordance to Chinese standard. This indicated that the long term large consumption of rice from this area will result in the higher exposure of Hg. Since the EDI for Hg at Rwamgasa, Iseni and Nyakayenze exceeded the safe value. The approved safe limit for As is 0.035 mg/kgbw/day (FAO/WHO, 2016). Moreover, the total hazard indice of As greater than or closeto the safety threshold of 1. Long-term As exposure through the regular consumption of rice in the investigated area poses potential health problems to residents in the vicinity of the mining industry.

Table 4: Calculation of estimated Daily Intake (EDI) And Health Risk Index (HRI) due to consumption of rice from the Research Area

Location	Mercury							Arsenic							Total	
	C Mg/kg	FF kg	EDI a Mg/kgbw/day	EDIC Mg/kgbw/day	RfDo Mg/kg bw/day	HRIa -	HRIc -	C Mg/kg	FF kg	EDI a Mg/kgbw/day	EDIC Mg/kgbw/day	RfDo Mg/kg bw/day	HRIa -	HRIc -	THRIa	THRIc
Lwamgasa	3.58	0.30	0.016	0.036	0.02	0.826	1.79	0.771	0.30	0.004	0.008	0.35	0.010	0.022	0.836	1.81
Bingwa	1.56	0.30	0.007	0.016	0.02	0.360	0.78	0.642	0.30	0.003	0.006	0.35	0.008	0.018	0.368	0.79
Iseni	2.57	0.30	0.011	0.025	0.02	0.593	1.29	0.938	0.30	0.004	0.009	0.35	0.012	0.027	0.605	1.31
Imalanguzu	1.45	0.30	0.007	0.015	0.02	0.333	0.72	0.741	0.30	0.003	0.007	0.35	0.010	0.021	0.343	0.70
Nyakayenze	2.16	0.30	0.010	0.022	0.02	0.498	1.08	0.918	0.30	0.004	0.009	0.35	0.012	0.026	0.511	1.10
Magenge	2.89	0.30	0.013	0.029	0.02	0.666	1.45	0.841	0.30	0.004	0.008	0.35	0.011	0.024	0.678	1.46
Nyamalulu	1.92	0.30	0.009	0.019	0.02	0.443	0.96	1.012	0.30	0.004	0.010	0.35	0.013	0.029	0.456	0.98
Nyamtondo	2.02	0.30	0.009	0.020	0.02	0.466	1.01	0.784	0.30	0.004	0.008	0.35	0.010	0.022	0.477	1.03
Nyamalimbe	2.35	0.30	0.011	0.024	0.02	0.541	1.18	0.734	0.30	0.003	0.007	0.35	0.010	0.021	0.551	1.19
Msasa	2.13	0.30	0.010	0.0213	0.02	0.492	1.07	0.658	0.30	0.003	0.007	0.35	0.009	0.019	0.501	1.08
Sobola	1.25	0.30	0.006	0.0125	0.02	0.288	0.62	0.678	0.30	0.003	0.007	0.35	0.009	0.019	0.297	0.64
Kaseme	0.99	0.30	0.005	0.0099	0.02	0.229	0.50	0.827	0.30	0.004	0.008	0.35	0.011	0.024	0.240	0.52
Mnekezi	0.78	0.30	0.004	0.0078	0.02	0.1800	0.39	0.727	0.30	0.003	0.007	0.35	0.010	0.021	0.190	0.41
Nyamisebhehi	1.64	0.30	0.008	0.0164	0.02	0.3792	0.82	0.835	0.30	0.003	0.008	0.35	0.011	0.024	0.390	0.84
Tembomine	1.93	0.30	0.009	0.0193	0.02	0.4461	0.96	0.829	0.30	0.004	0.008	0.35	0.011	0.024	0.457	0.99

4.2.3 Mercury (Hg) and Arsenic (As) concentration in rice from Magenge ward

The concentrations of Hg for the five sampling villages showed variations. The mean concentrations of Hg in rice samples drawn from Magenge ward were 2.89 ± 0.01 mg/kg, 1.92 ± 0.02 mg/kg, 2.02 ± 0.14 mg/kg, 2.35 ± 0.01 mg/kg and 2.13 ± 0.05 mg/kg of rice samples from Magenge, Nyamalulu, Nyamtondo, Nyamalimbe and Msasa respectively (Table3). The findings in the five villages of Magenge ward showed unpredictable variations. The concentration of As in rice samples drawn from this ward were 0.841 ± 0.08 mg/kg, 1.01 ± 0.05 mg/kg, 0.78 ± 0.04 mg/kg, 0.73 ± 0.05 mg/kg and 0.66 ± 0.08 mg/kg of rice samples from Magembe, Nyamalulu, Nyamtondo, Nyamalimbe and Msasa, respectively (Table 3). The highest concentration was found at Magenge village and the lowest was at Nyamalulu village. The results of this study in these five villages are higher to those established in Cisu Indonesia whereby the prevalence of Hg contamination was found to be (1.186 mg/kg) Bose-O'Reilly *et al.* (2016). The concentration of Hg is mostly high at Magenge village and Nyamalimbe due to the ongoing mining activities and reprocessing of tailing, also there has been the intensive mining activities at Nyamalimbe village since 1980's.

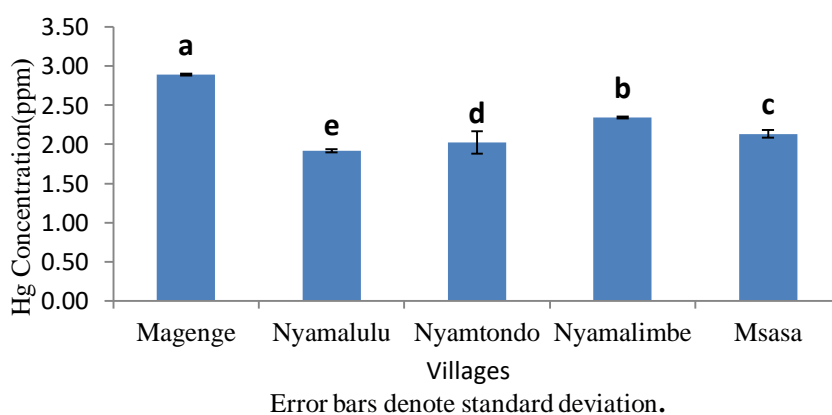


Figure 3: Variation of Hg in rice from Magenge ward

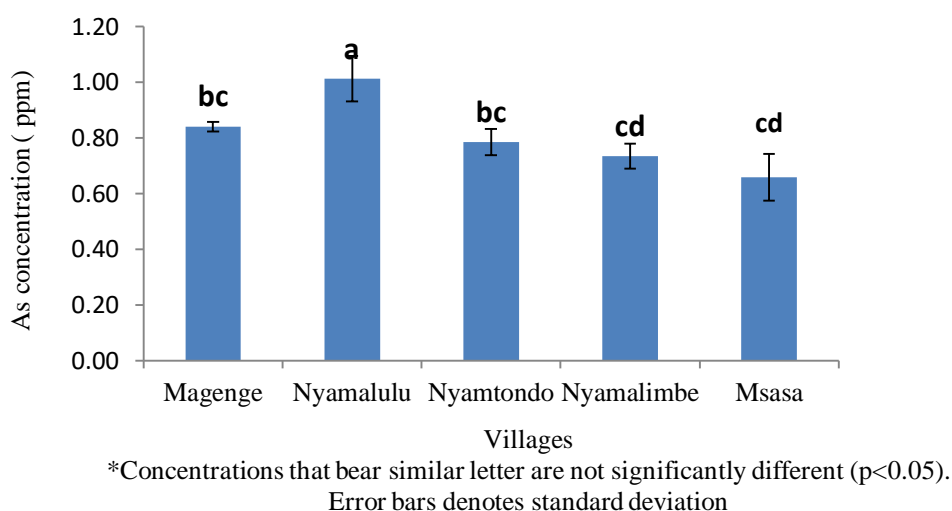


Figure 4: Variation of Hg in rice from Magenge ward

The findings in this study also showed that, the concentration of As was very high at Nyamalulu (1.012 mg/kg) compared to other areas studied in Geita district (Table3), the lowest concentration was recorded at Msasa (0.66±0.08) mg/kg. In general with exception of Nyamalulu village, there were no significant variations in As concentration among the villages ($P < 0.05$). This is probably because cotton cultivation is highly practiced in the all wards of Rwamgasa, Magenge and Kaseme. The finding of Magenge is in line with that of Turkey by Gunduz *et al.* (2013) who found the concentration of 0.98 mg/kg for As in rice samples.

4.3.1 Health risk for adult and children due to consumption of rice from Magenge

The estimated daily intakes (EDI) for Hg through consumption of rice from Magenge ward was characterized by a very slight variation among villages with values not exceeding 0.029 mg/kg bw/day and 0.013 mg/kg bw/day for children and adult respectively who consume rice from Magenge village. With exception of Nyamalulu

village which had EDI for children below the maximum allowable limit of 0.02 mg/kg bw/day the other four villages of Magenge, Nyamtondo, Nyamalimbe and Msasa villages had concentration above the maximum allowable limit. HRI for adult did not exceed 1 for all villages and for children HRI exceeded 1 in all villages in the Magenge ward with exception of Nyamalulu. Therefore, there is no health risk for adult due to Hg but there is a moderate risk to children consuming rice grown in Magenge ward.

The general population at Magenge ward had EDI of As minimum value at Msasa village for adult and children of 3.00×10^{-3} mg/kgbw/day and 6.60×10^{-3} mg/kg bw/day respectively. The highest As exposure was at Nyamalulu 1.01 mg/kg due to high concentration of As in the rice grown. The exposure estimates in all five villages of Magenge, Nyamalulu, Nyamtondo, Nyamalimbe and Msasa was below the value approved by FAO/WHO (2016) which is 0.35 mg/kgbw/day. However, the HRI computed for the ward was far below 1, implying that there is no health risk to the general population associated with consumption of rice grown around Villages of Magenge ward (Table 4).

4.3.2 Mercury (Hg) and Arsenic (As) concentration in rice from Kaseme ward

The concentration of Hg in rice samples drawn from Kaseme ward in this study were 1.25 ± 0.03 mg/kg, 0.99 ± 0.03 mg/kg, 0.78 ± 0.02 mg/kg, $1.6401 \text{ mg/kg} \pm 0.02$ and 1.92 ± 0.01 mg/kg of rice samples from Sobola, Kaseme, Mnekezi, Nyamisebehi and Tembomine respectively.

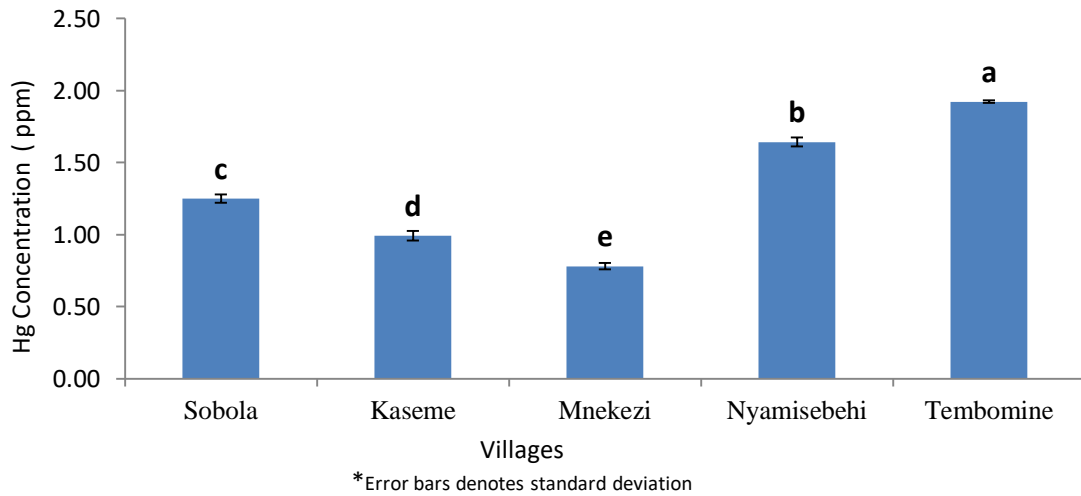


Figure 5: Variation of Hg in rice from Kaseme ward

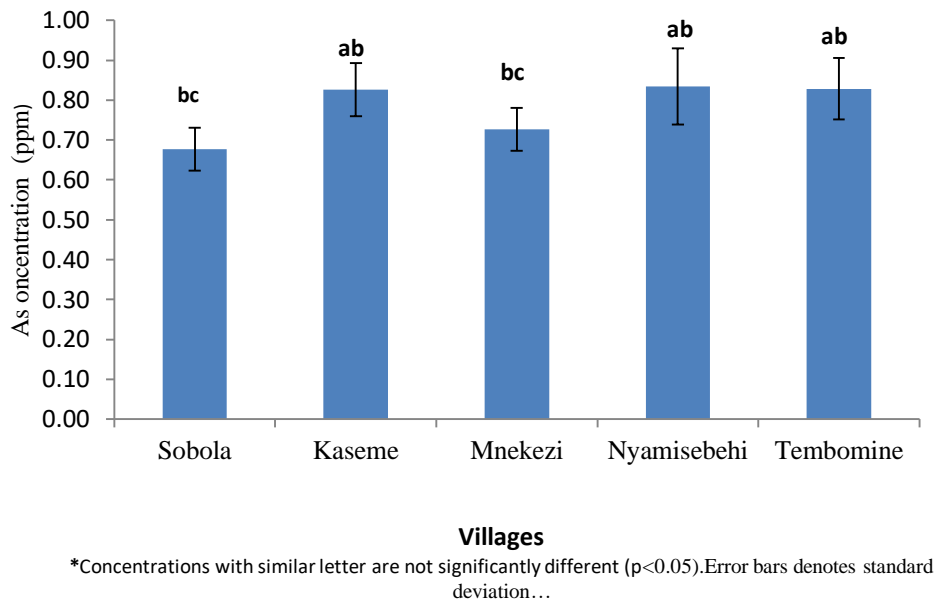


Figure 6: Variation of As in rice from Kaseme ward

The results show that, Kaseme ward had the lowest concentration of Hg among the wards studied (fig. 6). The concentration of Hg at Kaseme ward ranged from 0.78 mg/kg to 1.19 mg/kg at Mnekezi and Tembomine villages respectively. The concentration of Hg among villages of Kaseme wards showed a significant different ($P < 0.05$). The low level of Hg

concentration found at Mnekezi and Kaseme was because currently there are no mining activities carried out and is a place where artisanal miners are living and therefore the level of contamination of agricultural soil is decreasing. Higher levels at Tembomine, Sobola and Nyamisebehi is attributed by historical background as mining activities have been there for some years and currently small scale investments by Chinese is underway and also geographically is an area for run off for Hg contaminated water from Rwamgasa mining during heavy rain seasons. The Hg levels observed in Kaseme are still very high compared to the Hg level recorded in Mugusu artisanal mining area in the study by Tungaraza *et al.* (2011) who found concentration of 0.026 mg/kg of rice grown in Mugusu. In the other hand the result that, the concentration of Arsenic in Magenge ward was high at Nyamisebehi (0.84 mg/kg) and low at Sobola (0.68 mg/kg). There was no significant variations in As concentration among the five villages. The levels found at these five villages were higher than 0.08 mg/kg which was found in rice samples in China by Huang *et al.* (2013).

4.4.1 Health risk for adult and children due to consumption of rice from Kaseme

Among the three wards studied, rice grown in Kaseme ward was found with the minimum concentrations of Hg leading to low estimated daily intakes (EDI) in rice consumed. The minimum EDI for both adult and children were 3.600×10^{-3} mg/kgbw/day and 7.80×10^{-3} mg/kgbw/day respectively for rice grown at Mnekezi village. In general the EDI did not exceed the maximum allowable concentration (0.02 mg/kg bw/day) approved by Ministry of Health for the People of Republic of China (MHPRC, 2005). Since the health risk index for villages did not exceed 1 due to Hg for both adult and children therefore there is no health risk associated with consumption of rice grown in villages of Kaseme (Table 4).

The general population at Kaseme ward had EDI of As below the maximum allowable concentration of 0.35 mg/kg bw /day as approved by FAO/WHO (2016). Nyamisebehi had the highest and Sobola village had the lowest level of EDI. In all cases there was no health risk to the general population due to As associated with consumption of rice grown in the villages of Sobola, Kaseme, Mnekezi, Nyamisebehi and Tembomine in Kaseme ward (Table 4).

CHAPTER FIVE

5.0 CONCLUSION AND RECOMMENDATIONS

5.1 Overview

This chapter presents conclusion and recommendations for reduction of the health risks associated with contamination of rice from the study area.

5.2 Conclusion

Heavy metals contamination in the environment is of more concern worldwide. The knowledge of the source of heavy metals contamination in agricultural soil and rice is necessary in order to improve food safety by minimizing possibility of food insecurity as well as food human problems. The demographic data indicated that about 84% of the rice of the rice plots were located near by gold amalgamation activities which was carried out by the use of chemicals (Mercury). In gold amalgamation process almost 97.3% handle Hg without any protective gear. All of the respondents claimed to have no knowledge about the adverse health effect associated Hg. Based on the findings this study revealed that, there was a high concentration of Hg in the study areas with similar mining activities such as those in Chunya, Kahama, Ushirombo, Singida, Morogoro and Muheza due to amalgamation of gold by artisanal miners.

Individuals in the study area were exposed to Hg through ingestion in various food crops such as vegetables, yams and rice which impair their health status. The computed estimated daily intakes for Mercury (Hg) in children in most cases exceeded the MAC limit of 0.02 mg/kgbw/day in the research area. Through risk characterization of the results of sample analysis, health risk index was calculated whereby it was revealed that there is a health risk to children who consume rice over a long period of time from the

following locations;Lwamgasa, Iseni, Nyakayenze, Magenge, Nyamtondo, Nyamalimbe and Msasa. Non-carcinogenic risk due to Hg and As for adult was within the safe limit. This review work conclude that,in Tanzania there's insufficient information on heavy metals contaminationin agricultural soils and rice. Moreover it sugest that more research need to be conducted in areas around the problem sources of heavy metal contamination in agricultural soil and rice as well as conducting exposure assessment for heavy metas through rice consumption. Local government authorities near local mining areas in Geita district need to make sure that Environmental regulation and approved practices are followed. Improved technologies such as the use of retorts should be emphasized to reduce Hg vapour exposure to environment. Different study design should be used to assess possibility of Hg related to birth and growth defects, increased abortion/miscarriage rate learning difficulties in childhood, infertility problems and neuro-psychological related problems linked to Hg exposure.

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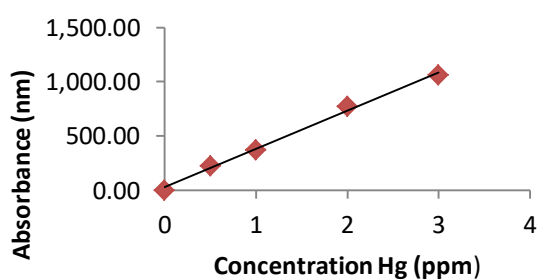
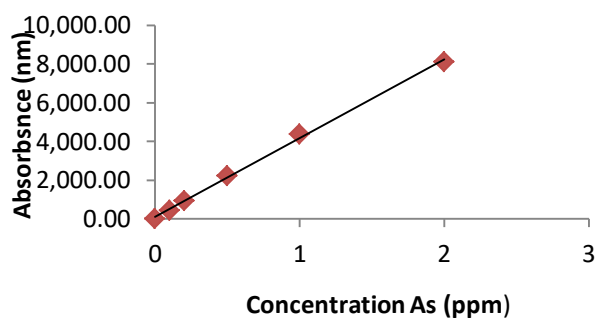
APPENDICES

Appendix 1: Results for method validation and calibration curves

Metal	Concentration of working standards (mg/l)	Correlation Coefficients of calibration curves (R^2)	Limit Of Detection (LOD)	Limit Of Quantification (LOQ)	Assay (%)	Calibration curve model
Hg	0.00, 0.50, 1.00, 2.00 and 3.00	0.99521	0.711ppm	2.155ppm	102.83 ± 0.095	Linear
As	0.00, 0.10, 0.20, 0.50, 1.00 and 2.00	0.99839	0.276ppm	0.835ppm	105.74 ± 0.039	Linear

$Intensity = 359.5461073 * concentration + 21.68145059$ for Hg, and

$intensity = 4061.37831797 * concentration + 110.6018986$ for As



Appendix 2: Structured questionnaire for assessing rice consumption pattern in the research area

Region District
Division.....Ward..... Village.....
Household Identification No.....
Name of Enumerator.....Date.....

Research Questionnaire

1. Name of respondent.....Mobile phone.....
2. Sex 1. Female 2. Male
3. Age in complete years.....
4. Household size.....
5. Level of education i.e. number of years spent in school.....
6. Religion. 1.Christian
2. Muslim
3. Others.
7. Main occupation 1.Agro-pastoralism
2. Farmer
8. Source of food consumed.
1. Own farms
2.Bought
3.Relief
9. What are the components for your breakfast?.....
10. What are the components of your dinner?.....
11. How many kilogram of food do you cook for your household?.....
12. When you intend to prepare rice for dinner what is the estimated portion size?.....
13. Do you own a plot for rice production? 1. Yes 2.No
14. If yes where is your plot located? 1. Homestead
2. Away from homestead
10. How many walking hours is your plot to gold mining ores?.....

11. Are gold amalgamation activities carried out within your rice plots? 1. Yes 2.No
12. What is the distance in meters from rice crops to the gold extraction plant?.....
13. Do you engage in gold mining activities? 1. Yes 2.No.
14. If yes how do you extract gold?.....
15. Specify the equipment mentioned above.1.....2.....3.....
16. Apart the above equipment started do you use chemicals in amalgamation of gold?
1. Yes 2.No Mention the chemical used in amalgamation of gold.....
17. Do you handle it with bare hands? 1. Yes 2.No.
18. What type of health problems encountered in this area?.....
Record the physical appearance of respondent in terms of health.....

End