ASSESSMENT OF URANIUM LEVELS IN WATERAND SOILS AND ITS UPTAKE BY SORGHUM AND SUNFLOWER IN SINGIDA URBAN DISTRICT, TANZANIA

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A DISSERTATION SUBMITTED IN PARTIAL FULFILMENT OF THE REQUIREMENTS FOR THE DEGREE OF MASTER OF SCIENCE IN SOIL SCIENCE AND LAND MANAGEMENT OFSOKOINE UNIVERSITY OF AGRICULTURE. MOROGORO, TANZANIA

2016

ABSTRACT

In Tanzania, uranium deposits have recently been found in Bahi and ManyoniDistricts. Due to suspected high uranium levels in water sources as reported by District water authoritiesin Singida District, this study was carried out to determine levels of uranium in soils, and water and its uptake by commonly grown food cropsfor enhancing public awareness and safety to the residents of Singida Urban District. Rock, soil and plant samples were collected randomly from farmer's field.Water samples were collected from various water sources in Singida Urban District. Laboratory analyses of soil, rock and plant samples werecarried out at the Geological Survey of Tanzania in Dodoma and Soil Science laboratory at Sokoine University of Agriculture while water samples were analysed atthe Government Chief Chemist Laboratory in Dar es Salaam. Results foundin soil, water, and rocks. The highest U level in soil of 15.4mg $kg^{-1}$was obtained at Burudani area and the lowest level of<0.01mg $kg^{-1}$was obtained at Kwa Askofu area. The concentration of uranium in rocks ranged from 20.1 to 31.5 mg kg^{-1}, which in some places washigher than the critical level of 23 mg kg^{-1} recommended for agricultural land use. Uranium concentrations in sorghum and sunflower grainswere found torange from 1.4 to 5.2 mgkg $^{-1}$, while in leaves it was less than 2.3 mgkg $^{-1}$. Uranium concentrations in water ranged from 0.058 to 1.097 mgL^{-1}, the values which were higher than 0.03 mgL^{-1}tolerable limit set by WHO for human and animal consumption. It was concluded that uranium contents found in soils are within the tolerable limit set for agricultural use and consequently sorghumand sunflower grown in the District are safe for human and animal consumption. Most water sources in the District had higher levels of Uranium and thus, should not be used for safety reasonsunless they are treated to reduce uranium concentration. Further studies are recommended to assess uranium concentration in fish, and soda ash,which are produced from lakes and in Singida Urban District.

DECLARATION

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

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ACKNOWLEDGEMENTS

Finally, I must express my very profound gratitude to my parents and to my husband Fredy O. James for providing me with unfailing support and continuous encouragement throughout my years of study and through the process of doing research and writing this dissertation. This accomplishment would not have been possible without him. Thank you.

DEDICATION

This work is strictly dedicated to my lovely Parents, my late father Charles Kaishwa and my mother Veronica Omary for their role as parents, my husband Fredy O. Oreku, my sisters Pendo, Shuku, and Mariam, and my brothers, Yesse and Omary for their love, support and patience during my studies.

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LIST OF SYMBOLS AND ABBREVIATIONS

AGRA	Alliance for Green Revolution in Africa
ANOVA	Analysis Of Variance
ASL	Above Sea Levels
ATSDR	Agency for Toxic Substances and Disease Registry
Ca	Calcium
CCME	Canadian Council of Ministers of Environment
CEC	Cation Exchange Capacity
cm	centimetres
cmol (+) kg^{-1}	Centimole per kilogram
CO_2	Carbon dioxide
CV	Coefficient of Variation
DTPA	Diethylene Triamine Pentaacetic Acid
EN ISO	European International Standard Organization
FAO	Food and Agriculture Organisation
GMT	Greenwich Mean Time
H_2O_2	Hydrogen Peroxide
HNO_3	Nitric Acid
ICP-OES	Inductively Coupled Plasma-Optical Emission Spectrometer
ISO	International Standardization Organization
K	Potassium
LOAEL	Lowest-Observed Adverse-Effect-Level
LSD	Least Significance Difference
MAC	Maximum Acceptable Concentration
Mg	Magnesium

mg kg^{-1}	Milligram per kilogram
Mn	Manganese
N	Nitrogen
Na	Sodium
NDMRT	New Duncan's Multiple Range Test
^{O}C	Celsius
P	Phosphorus
pH	power of hydrogen
SOC	Soil Organic Carbon
SOM	Soil Organic Matter
SUA	Sokoine University of Agriculture
TDI	Tolerable Daily Intake
TEB	Total Exchangeable Base
U	Uranium
U$_3$O$_8$	Triuranium octaoxide
UO$_2$	Uranium oxide
UO$_2$$^{2+}$	Uranyl
USDA	United States Department of Agriculture
USEPA	United State Environmental Protection Agency
WHO	World Health Organisation 2
Zn	Zinc
m asl	Metre above sea level

CHAPTER ONE

1.0 INTRODUCTION

1.1 Background Information

Uranium is a metallic element that occurs at high concentration in granites, shales and carbonates (Bleise*et al.*,2003). It occurs in three natural isotopes: ^{238}U, ^{235}U and ^{234}U, all of which are radionuclides. Uranium has a low specific activity. Its chemical toxicity is of greater concern than its radiotoxicity. Uranium (IV) is the dominant species of uranium present in primary minerals (Smedley*et al*., 2006). This specie is associated with hydroxides, phosphates and fluorides (Keith-Roach*et al.,* 2010) and is relatively insoluble but can readily oxidise to Uranium (VI), which is stable. Uranium can be enriched in soil via natural paedogenesis processes, with uranium being released from parent materials to soil and soil solution, and via human activities. Mining phosphate ores can carry uranium to the soil and river water resources surrounding the mines. Phosphate rocks may contain considerable amounts of uranium (Romero *et al*., 1995).

Uranium in soils is enriched by weathering of parent rocks, mainly granites (Sasmaz and Yaman, 2008). Weathering of granites releases the element to the soil and makes it available for plant uptake. Furthermore, migration and accumulation of U in plants depend on factors such as soil characterises, climatic conditions, types of plants, parts of plant concerned, physical-chemical form of the uranium and the interfering elements (Tome *et al.,* 2003). For example, the availability of calcium and potassium in soil affects uranium uptake by plant. This is due to calcium and potassium which tend to replace uranium in soil solution by letting uranium to be adsorbedon the soil colloids leaving calcium and potassium in soil solution and makes them easily available to plants as compared to uranium (Kohler *et al.,* 2014).

Uranium and radioactive mineralization in Tanzania were identified in the pre-independence era and a number of occurrences are known in Tanzania (Kreuser*et al.,* 1990). Far back to 1950s uranium occurrences were identified in the Bahi swamp (Cygan, and Tazaki, 2014). In early post-independence era (1961– 1970s) the country was subjected to a systematic airborne survey for radioactive occurrences. In this period, it was evident that four geological areas in Tanzania contain potentially significant uranium deposits viable for commercial mining activities (Knivsland, 2012).These uranium occurrences were found in sandstones mainly of the Karoo and Bukoban Super groups and Carbonatite complexes of Mesozoic to Recent age (Ngulimi and Ishiga,2016). Some uranium occurrences were found in calcrete relatedsecondary environments in Quaternary deposits at Ndala, Itigi and Iseke and at the unconformity between the Karagwe-Ankolean and Bukoban Super group as vein-like type of uranium mineralisation.

Uranium VI (U^{6+}), like other heavy metals, is associated with various health hazards such as kidney toxicity which is caused by breathing air containing uranium dusts or by eating substances containing uranium, which enter the bloodstream. Very high uranium intakes ranging from about 20 - 30 mgL^{-1} depending on the individual can cause acute kidney failure and death (Zamora *et al.,* 1998). At lower uranium intake levels of around 15 to 20 mgL^{-1}, damage can be detected by the presence of protein and dead cells in the urine. However, at lower intake levels, the kidney repairs itself over a period of several weeks after the uranium exposure has stopped(Sheppard*et al*., 2005).

High level of uranium in agricultural soils enhances the chance of its assimilation and translocation to various parts of the plants (Ankle *et al.,* 2009). Once released from rocks U exists as U^{4+} in reducing state and as U^{6+} in oxidizing state.

According to Meinrath*et al.* (2003), U is soluble and mobile in oxidizing environments and it is generally insoluble in reducing condition. Antunes*et al.* (2007) reported that U-acetates, U-sulphates, U-carbonates, U-chlorides and U-nitrates can easily dissolve in groundwater and surface water.

In Tanzania, uranium deposits haverecently been found in Bahi and ManyoniDistricts(Mbogoro andMwakipesile, 2010). These deposits occur in the lowland areas surrounding isolated hills of Precambrian granites. The source of this uranium is yet to be known. Presence of high levels of uranium in drilled water wells has also been reported in Singida Urban District (Personal communication with Basin Water Officer for the Internal Drainage Basin for central regions, 2014). The sources and distribution of this uranium in water wells in Singida are also not known. Singida is among the dry regions in central Tanzania with insufficient surface water for domestic uses.Drilledwater wells are used to supplement surface water for human and animal consumption.

The presence of uranium in drilled water wells in Singida is something that needs to be carefully studied for the safety of people because uranium and its compounds are carcinogenic and highly toxic (Sasmaz and Yaman, 2008).

It is not known whether or not agricultural soils in Singida Urban District are also contaminated with this uranium. Uranium in agricultural soils is enriched by weathering of parent rocks, particularly granites (Sasmaz and Yaman, 2008), which are the dominant rocks in the study area. Studies indicate that high level of uranium in agricultural soils enhances the chance of its assimilation and translocation to various parts of food crops (Ankle*et al*., 2009; Singh, 1997). Thus, it is possible for crops like sunflower and finger

millet that are dominantly grown in Singida to accumulate high contents of U. Through the food chain, this uranium could be transferred to humans and other living species.

This study was aimed to generate information on the levels of U contamination in water, agricultural soils and selected food crops in Singida Urban District. This information is vital in order to create public awareness on the potential hazards posed by U to the vulnerable communities in the District. In addition, the study focusedon improving scientific knowledge on U soil/plant interactions, its mobility and contamination levels in soils and water resources as well as better understanding on the ability of food crops to accumulate U in their conductive tissues.

Thus, the main objective of this study was to assess the level and distribution of uranium in major soils and water sources and its uptake by finger millet and sunflower in Singida District to insure human and animal safety.

The specific objectives of this study were to:

i) Characterize the soils of Singida Urban District for their physical and chemical properties.

ii) Determine the concentration and distribution of uranium in major soils and rocks in Singida Urban District.

iii) Determine uranium levels in surface and underground waters in Singida Urban District.

iv) Assess the uptake of uranium by finger millet and sunflower in Singida UrbanDistrict.

CHAPTER TWO

2.0 LITERATURE REVIEW

2.1 Physical and Chemical Properties of Uranium

Uranium is the heaviest metal in nature. It has 14 natural isotopes but the most abundant are ^{238}U (99.28 %) and ^{235}U (0.71 %). All of them are radioactive; this means that their nuclei emit radiation particles to achieve a stable configuration. Uranium is member of two decay series, the uranium and the actinium series, being a stable isotope of lead, the last element of them. The energy released during the disintegration of these nuclei warms the interior of the earth and determines the process that moves the continents and the elements cycles in the earth's crust (Cowart and Burnett, 1994). Radioactivity produced by U mainly consists of alpha particles formed by two protons and two neutrons, a helium nucleus with a net charge of 2+; they have a high energy normally but they do not penetrate more than 1 mm of tissues (Ribera *et al*., 1996). Besides, the ^{235}U isotope is fissile; this means its nucleus can be split spontaneously releasing an enormous amount of energy in chain reactions.

The ^{235}U and ^{238}U isotopes have similar chemical properties. The metal U is chemically very active and can react with most elements except the rare gases. It forms oxides with air, producing either UO_2 or U_3O_8. At room temperature, humidity is the principal cause of oxidation. In fragmented state (chips, powder or turnings) the metal U becomes pyrophoric (Ribera *et al*., 1996).

Uranium IV and VI are the oxidation states typically observed in the environment. Under strongly reducing conditions, U occurs in the tetravalent oxidation state (U (IV)). It

hydrolyzes in solution to form monomolecular hydroxo-complexes, such as $U(OH)_n^{4-n}$. In reduced ground waters condition, U (IV) is complex bounded by sulphate, chloride and phosphate, by fluoride at pH values less than 4 and also by organic humic and fulvic acids. Still under low oxygen contents the U is quickly oxidized to the hexavalent state (VI) and the uranyl ion ($UO2^{+2}$) is formed in aqueous solution. U forms mono- and poly-nuclear hydrolysis products.

At pH 5 and higher the major species in solution is $(UO_2)_3(OH)_5^{+}$. Aqueous uranyl forms complexes with halogens and with most oxo-anions, such as NO_3^-, SO_4^{-2}, ClO_4^-, PO_4^{3-}, HPO_4^- and CO_3^{2-}, and carboxylic acids as well. The formation of uranyl carbonate complexes is favoured by high pH, high uranyl ion activities and high partial CO_2 pressures. Uranyl phosphates may be important in systems with pH from 6 to 9, when the ratio of P/C is larger than 10. Sulphates, F^- and possibly Cl^- complexes are important when their concentrations are high. Organic acids, which contain several functional groups per molecule, also form strong complexes with U (Fellow*et al*., 1998; Mortvedt, 1994).

2.2 Uranium in Soil

Uranium occurs naturally in granites and other minerals deposits. Uranium enters the soil system through leaching from natural deposits, mill tailings, and use of phosphate fertilizers containing uranium (WHO, 2004). During the weathering process, U can be leached or accumulated in some horizons of the soil profile. In tropical environments, U is retained in the red soils, because it has a great affinity for iron minerals (Evans *et al*., 2006; Yoshida *et al*., 1998). U is mainly accumulated in the A horizons of soils. High U contents have been described in very rich organic matter soils coming from the sequestration and reducing of U (Fellow*et al*., 1998; Tipping, 1996).

Anthropogenic U contamination of soils is caused by the different steps of mining, uses and disposal of U containing products or by-products. Tekeda *et al.* (2005) reported that the uranium concentration in surface soils cultivated with applied phosphate fertilizer accumulated relatively high U levels than those of neighbouring non-agricultural areas, probably because of successive application of P fertilizer. Average concentrations of U in such fertilizers are by a factor of 100 higher than that of soils, mainly depending on the origin of the rock phosphates (Kratz andSchnug, 2006; Schnug*et al.,* 1996). In soils, uranium occurs in various states. Soluble uranium occurs as hydrated ions and uranyl complexes and exchangeable uranium that can be retained as cation forms by soil colloids (mineral and organic forms). Sometimes, uranium occurs as precipitated uranium, uranium minerals, and accumulated uranium in tissues of soil micro-organisms (Hooda, 2010).

High levels of uranium in soil enhance the chance of its assimilation and translocation in various parts of plants (Ankle *et al.*, 2009). Uranium in soils can reach concentration as high as hundreds of mgkg^{-1} (Rufykiri*et al.,* 2004).

2.3 Oxidation States of Uranium

Uranium occurs in 5 oxidation states (+2, +3, +4, +5 and +6).Only two oxidation states (+4, and +6) are considered stable enough to be of practical importance (ATSDR, 1999). Uranium is generally found as oxides, such as uranium dioxide (UO_2) or triuranium octaoxide (U_3O_8)(McManus *et al.*, 2005).

Uranium in soil is pH-dependent. Under acidic reducing conditions; UO_2^{2+} is the predominant uranium species in the soil while under oxidizing conditions, uranylUO_2^{+} form highly mobile compounds (Ebbs *et al.*, 1998).Under neutral conditions, hydroxide

complexes such as UO_2OH^+, $(UO_2)_2(OH)_2^{2+}$, $(UO_2)_3(OH)_5^+$ and $(UO_2)_3(OH)_7^-$ are formed. Phosphate complexes such as UO_2HPO_4 and $UO_2(HPO_4)_2^{2+}$ are formed under acidic conditions. Under alkaline conditions, carbonate complexes such as UO_2CO_3, $UO_2(CO_3)_2^{2-}$ and $UO_2(CO_3)^{4-}$ predominate (Ebbs *et al*., 1998).

2.4 Solubility and Mobility of Uranium in Soils

Solubility and mobility of U vary with soil characteristics and are influenced by pH, redox potential, soil temperature, soil texture, organic and inorganic compounds, soil moisture and microbial activities (Rivas, 2006). Uranium transport generally occurs in oxidizing surface water and groundwater as the uranyl ion U^{6+}, or as uranyl fluoride or carbonate complexes. The U^{6+} and uranyl fluoridecomplexes dominate in oxidizing acidic waters whereas the carbonate complexes dominate in near- neutral and alkaline oxidizing waters, respectively (Gavrilescu*et al*., 2009). An important point in considering U migration in soil is that when $U^{6+}$is reduced to U^{4+} by humus, peat, or other organic matter, it is essentially immobilized. It should also be noted that phosphate and sulphide usually precipitate U and hence, stop migration, a behaviour that can be exploited in remedial operations (Fellow *et al.,* 1998).

Mobility and solubility of U can be affected by aeration (water saturation, high biological or chemical oxygen demand), carbonate content (organic material content, pH, parent material, and weathering) and cation exchange capacity (texture, clay, organic matter and pH) (Gavrilescu *et al.,* 2009). Heavy metals become more water soluble under acid conditions and can move downward with water through the soil, and in some cases they move to aquifers, surface streams, or lakes (Chuan *et al*., 1996). In aerated aqueous solutions at pH 2.5, the uranyl ion is very stable. Near pH 7, uranyl ion forms stable complexes with phosphate and carbonate.

Soil redox status varies temporally and spatially. In a surface soil, it is influenced by rainfall, bioactivity, and changes in land use (Boulet*et al*., 2003). Decrease in redox potential may cause changes in metal oxidation state, formation of new low-soluble minerals, and reduction of Fe, resulting in release of associated metals (Amrhein*et al*., 1994; Chuan*et al*., 1996; Masscheleyn*et al*., 1991).

The formation of soluble uranyl carbonate complexes gives rise to a strong positive correlation with U concentrations in soil solutions. This is particularly clear under anaerobic conditions and also at high temperatures which encourage microbial activity and high CO_2 partial pressures **(Gu*et al.,* 2005)**.

2.5 Uptake of Uranium by Plants

Uranium distribution in plants follows a pattern similar to Ca, which is accumulating more in older than in younger leaves (Mortvedt, 1994). Uranium absorbed by plant as hexavalent U^{6+} and that oxidation state does not change when in plant parts and remainhexavalent U^{6+} (Duquene *et al.,* 2006). Normally, U absorbed by plant tend to concentrate in there tissue in seedling stage than in the flowering stage (Larache *et al.,* 2005).

UO_2^{2+} is the species mostly readily taken up and transported to various plant parts(Ebbs *et al*., 1998). Uranium accumulates in the roots of plants (Kloke*et al*., 2002), which result in different uranium concentrations in different plant parts. Variation in uranium uptake and translocation between different plant types has also been reported. According to Kloke*et al*.(2002), dicotyledonous plant species tend to accumulate more uranium than monocotyledonous plant species.

Furthermore, U uptake and accumulation in plant is influenced by soil properties such as pH, cation exchange capacity (CEC), organic matter and aeration. High soil pH, CEC and good soil aeration favour U uptake while soil with high organic matter reduces uptake of U by plants (Meinrath *et al.,* 2003). Some species have developed the ability to tolerate high concentrations of heavy metals. Such plants can be used as bio-indicators in geo-chemical explorations or, more practically, to extract those contaminants from polluted soils. Many members of the *Brassica* species are classified as hyper-accumulators, and hydroponically grown sunflowers (*Helianthus annuus*) have been reported as a means to remove U from contaminated waters (Salt *et al.*, 1995).

In studying soil-plant transfer factors, AL-Kharouf*et al.* (2008) found that the green parts (leaves, stems and roots) of most crops tend to accumulate U at about two orders of magnitude higher than fruits. Plants also possess the potential to take up U present in soil and associated water bodies (Neves*et al.,* 2009).

2.6Uranium in Water

Water is the main transport vector for U. In confined aquifers, U occurs in the tetravalent state, whereas in unconfined or surface water its state is hexavalent (Rosing*et al*., 2006).The soluble products of rock weathering are transported into the groundwater and eventually into the rivers and then to the ocean. The actual U content of sea water is about $3gL^{-1}$ (Cowart and Burnett, 1994; Pais and Jones, 1997). Reductive precipitation of U(IV) in anoxic marine sediments is globally the most significant sink for dissolved U.

Uranium in surface water can disperse over large distances to ponds, rivers, and oceans. The transport and dispersion of uranium in surface water and groundwater are affected by adsorption and desorption of uranium on aquatic sediments. As with soil factors, that

control mobility of uranium in water include oxidation-reduction potential, pH, and sorbing characteristics of sediments and the suspended solids in the water (Katsoyiannis*et al.*, 2007; Phan*et al.,*2015). In groundwater, the weathering of uranium-bearing rocks and minerals is the source of dissolved uranium. The concentration of uranium in groundwater is usually in the range 0.1-50 μgL^{-1} (Rivas, 2005), and in contaminated groundwater plumes, concentration of uranium is much more, e.g., uranium concentration of groundwater in some uranium mill tailing in United States varies from $0.07 - 3.05$ mg L^{-1}(Baumgartner *et al.*, 2000).

The current guideline for uranium in drinking water is a Maximum Acceptable Concentration (MAC) of 0.02 milligrams/litre (mgL^{-1}). This means that no adverse health effects would occur from the ingestion of 1.5 L of drinking water per day containing uranium at this level over a lifetime of 70 years (Zamora*et al.*, 1998).

2.7 Hazardous Effects of Uranium

There are two different health hazards produced by U. The first is the short-time chemical toxicity of soluble compounds like UO_2^{2+}, by influencing directly the function of internal organs, especially the kidneys. The second is in the long time influence, because of the effect of the short distance alpha radiation of the U staying in the body, which could cause the development of cancer and genetic defects by deformation of chromosomes(Kumar*et al.*, 2003).

Uranium penetrates into an organism by different paths: pulmonary (inhalation), ingestion (gastro-intestinal system) or trans-cutaneous (skin and wounds) (Ribera*et al.*, 1996; WHO, 2004). Only a minor part (10 %) of the uranium that enters an animal or human organism by the respiratory route will be retained in the bronchial tree, the highest

proportion (up to 65 %) will reach the gastro-intestinal tract and the rest will be exhaled. Insoluble compounds that remain in the lungs will affect the alveolar tissue by radiation, and soluble compounds will be transferred to the extra-cellular fluids and this will lead to diffusion of U throughout the organism.

In the organism, blood is the principal carrier of U to target organs. From the U absorbed in blood, approximately 67 % is filtered by kidneys and excreted in urine during the next 24 hours (WHO, 2004). Insoluble U oxides do not seem to be a significant toxic risk when applied through the skin (WHO, 2004). Long term ingestion of U by humans leads to progressive or irreversible kidney injury. Both structural and functional kidney damages are known. $UO2^{2+}$ ions depress glomerular function, tubular secretion of organic anions and re-absorption of filtered glucose in the proximal tubule. In addition, damaging U effects on liver and the whole nervous system areknown. U can cross the blood-brain barrier, placenta, and foetus and can also be in milk (Ribera *et al*., 1996). The U.S.EPA proposed a drinking water standard for U of $0.02 mg \, L^{-1}$ based on kidney damage (Pontius, 1990). Limit for U in water has been established at $0.02 mg \, L^{-1}$ by WHO (2004).

CHAPTER THREE

3.0 MATERIALS AND METHODS

3.1 Description of the Study Area

3.1.1 Location

Soil, water, rock and plant samples were collected in Singida Urban District. Singida UrbanDistrict is located between 4° 40'East and 4° 53' South of the Equator and longitude 34° 30' and 34° 53' East of GMT. It covers an area of 754 km^2.The District is bordered to the south and west by the Ikungi District and to the north and east by the Singida Rural District. It is situated at an elevationof 1547masl. The sites where the studied samples were collected are as indicated in Figure 1.

3.1.2 Climate and economic activities

The mean annual rainfall in Singida Urban District ranges from 600 to 800 mm per year and temperature ranges from 15^{0}C to 31^{0}C. The main economic activities of the people include business, agriculture (corn, finger millet and sunflower) and livestock keeping. Sampling was carried out after finger millet and sunflower had matured and ready for harvesting.
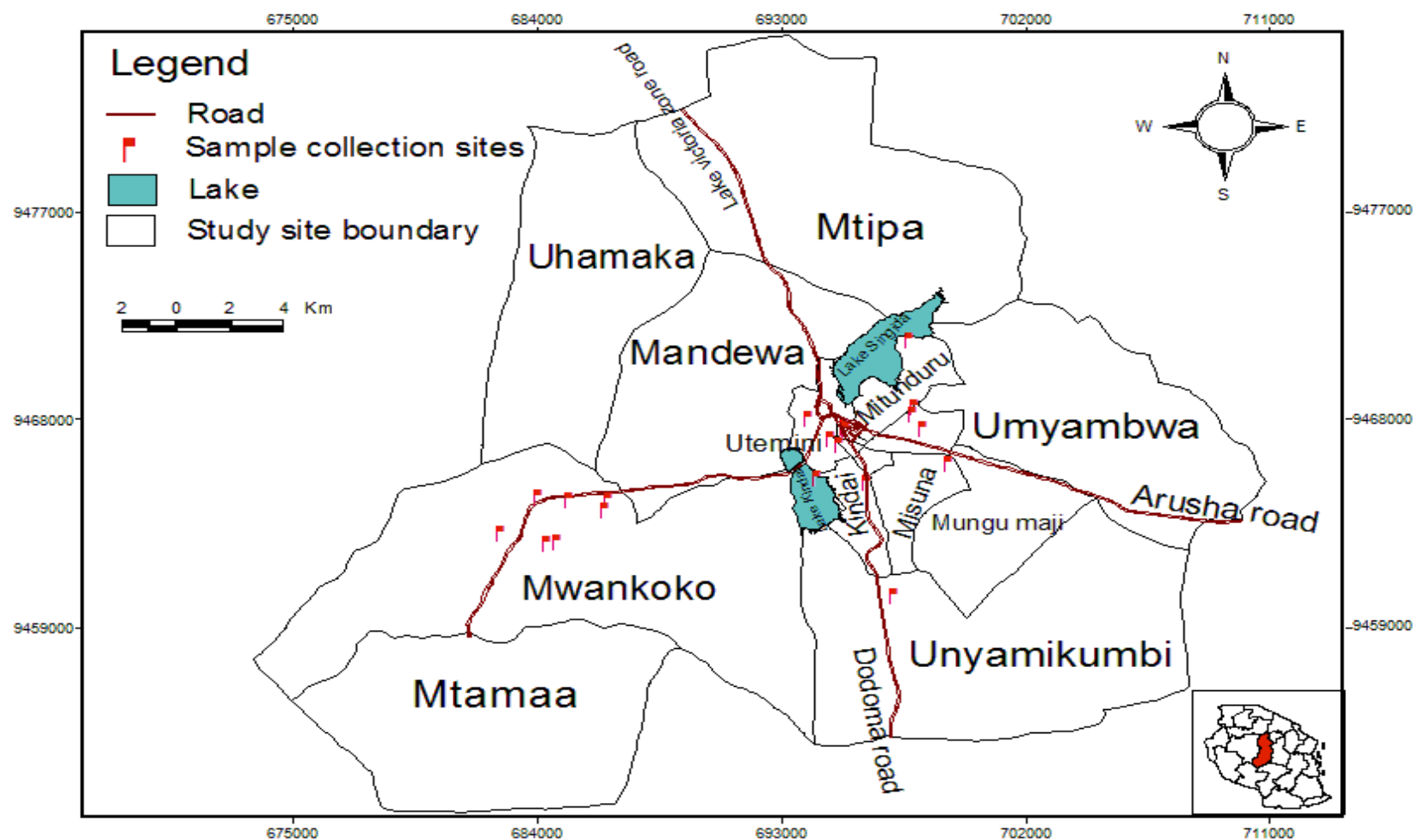
Figure 1:The siteswhere the studied samples were collected

3.1.3 Geology of Singida

The geology of Singida is dominated by the Tanzanian granite-gneiss craton (Kabete*et al*., 2012). The volcanic rocks encountered are predominantly basaltic lava and localised hypabyssal diorite/microdiorite intrusives. Meta-sedimentary rocks consist mainly of metaquartzitic lithologies, with subordinate siltstones, mudstones and banded ferruginous chert. The Nyanzian lithologies are generally metamorphosed to greenschist- and lower-amphibolite facies with localised occurrences of upper amphibolite grade. Post tectonic granite intrusives are common throughout the area. Gold mineralisation is associated with penetrative brittle-ductile, steeply dipping, quartz veined shear zones that generally approximate the strike of lithological contacts.

The area south of Singida isroughly flat with little relief except for the granite intrusives that form prominent tors. Weathered overburden profiles are dominated by Neogene transported pedogenic gravels and related soils. Black cotton soils (Mbuga) occur locally. It is more likelythat the graniterocks which are found in Singida Urban District are the source of uranium in soils and waterwhen they undergo weathering.

3.1.4 Soils

Singida Urban District is largely covered by granite rocks. Weathering of these granite rocks has resulted into the formation of sandy soils dominated by sandy particles, and are classified as CambisolsKnicker*et al.,*(2005). The soil dominated by sandy particles usually has low fertility status and low organic matter. The soil is greyish brown in colour. Sand washes are found on the soil surface in lowlands, which is believed to be brought from uplands through sheet erosion. Salt content in soil is generally high in some areas and during dry seasons salt pans crystallize on the surface.Other soils found in the studied area include Leptosols, Acrisols and Solonetz.

3.2Characterization of Surface Soils

3.2.1Soil sampling

Reconnaissance survey was carried out in order to determine the physical and chemical properties of the soils in the study area. Soil samples were taken randomly at the depth of 0- 30cm by using an auger. Three sub-samples were obtained from a sampling unit to get one composite sample as representative soil for laboratory analysis.

3.2.2 Soil processing

Bulk soils from the field were air- dried, crushed and sieved to obtain <2 mm fraction that was used for laboratory analysis. Analysis for the physical and chemical properties was carried out in the Soil Science Laboratory at Sokoine University of Agriculture (SUA).

3.2.3 Analysis forchemical and physical properties of soils

The pH H_2O values were measured by mixing soil with deionised water at the ratio of 1:2.5 (soil: water) followed by shaking the suspension for 30minutes and pH determined using a Corning glass electrode pH-meter (Okalebo *et al.,* 2002). Organic carbon was determined by the wet oxidation method of Walkley-Black (Nelson and Sommers, 1982). Particle size distribution of the soils was determined by the hydrometer method (Day, 1965) and textural class was determined using the USDA textural class triangle.The exchangeable bases (K^+, Ca^{2+}, Mg^{2+} and Na^+) were extracted by neutral ammonium acetate solution and determined by Atomic Absorption Spectrophotometer for Ca^{2+} and Mg^{2+}, while a flame photometer was used for K^+ and Na^+ (Sparks,1996). The cation exchange capacity (CEC) was determined by the neutral ammonium acetate (Buffered at pH 7.0) saturation method (Sparks *et al.,* 1996).

3.3 Determination of the Concentration and Distribution of Uranium in Soils and

    Rocks in Singida Urban District

3.3.1 Soil and rock sampling

Along the dominant slope, the study site was partitioned into three zones, designated as upper, middle and lower slopes. In each segment,soil sampling was carried out by using an auger at 0-30cm, 30-60cm and 60-100cm depth. The main objective of this sampling was to determine vertical and horizontal U distribution along the slopes.Samples of the exposed rock around the area were taken in order to analyse and determine levels of uranium in those rocks.

3.3.2 Soil and rock processing and analysis

Rock samples were crushed and sieved to pass through a < 2 mm sieve for laboratory analysis. Similarly, the soil samples were sieved through the same mesh. Analyses of rock and soil samples were carried out at Sokoine University of Agriculture in the Department of Soil and Geological Sciences.A Total X-ray fluorescence analyser (XL3 ANALYSER)was used to determine the levels of uranium in soils and rocks as described by Hunter andBertsch(1998).Fifty grams of soil and rock samples were weighed in a test tube by ausing analytical balance (EN ISO 20483) and placed in transparent plastic bags. After that the plastic bags were placed in the screen of the XL3 ANALYSER equipment. Uranium concentration in soil and rock samples was read directly using the XL3 ANALYSER.

3.4 Determination ofUranium Levels in Surface and Underground Waters

3.4.1 Water sampling

Underground and surface water samples were obtained from 16 different sources of drinking water in Singida Urban District. Underground water sources werepredominant among the samples (14 samples) and are the major sources used as compared to only two surface watersources. Underground water samples were collected from tapwater, whereby tap waterwas left to run for about 3 minutes to empty standing water before collection. Surface water samples were collected from the two lakes such as Lake Kindai and Lake Singidani in SingidaUrban District.   Each water sample had a volume of half a litre and was sampled by using clean and sterile bottles.

3.4.2 Analysis of water samples

Eightml of water samples weremixed with 1ml of HNO_3.Thisvolume was put into vials and U concentration was directly determined using an Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES) (Hunter and Bertsch 1988). The laboratory analysis of uranium in water samples was carried out in Dar es Salaam at the Government Chief Chemist Laboratory.

3.5Uranium Uptakeby Sorghum and Sunflower

3.5.1 Plant and soil sampling

Plantsamples (sorghum and sunflower) and soil samples were sampled in June 2015. Soil and plant samples were collected from three farms of sunflower and four farms of sorghum within the Singida Urban area. Soil samples were collected at depth 0-30 cm. Plant samples consisting ofshoots and grains were collected from the same field where soil samples were collected. In each farm, triplicate soil and plant samples were collected randomly at varying distances depending on the size of the farm. After collection, the soil and plant samples were placed into plastic sample bags and labelled for analysis.

3.5.2 Preparation and analysis of plant samples

Plant samples were oven-dried at 70-80°C to constant weight. After drying the plant samples were ground to obtain the required fine particles for analysis of uranium level in plant. Uranium in soil and plants were analysed using ICP-OESas described by Hunter and Bertsch(1998). Analysis was carried out at the Geological Survey Laboratory in Dodoma. Plant samples (sorghum and sunflower) of 0.5g were weighed and then transferred into the digestion tubes. Digestion procedure as recommended by ISO217 (2007) was used for plant analysis. After digestion, 100ml of the digest were placed in volumetric flask and topped with distilled water. Ten ml of the solution was poured into vials and uranium concentration was read directly using Inductively Coupled Plasma Optical Emission Spectrometry.

3.6 Statistical Data Analysis

Results were described using descriptive statistics and tables were generated using MSExcel. The analysis of variance (ANOVA) was employed and the Duncan's New Multiple Range Test (DNMR) was applied for means separation where high and low levels of uranium in soil, rock, leaves and grains were identified.

CHAPTER FOUR

4.0 RESULTS AND DISCUSSIONS

4.1 Characterization of Surface Soils in Singida Urban

The results for general chemical and physical properties of the soil are presented in Tables 1 and 2.

Table 1: **Chemical properties of the studied surface soils**

Location	pH	CEC	Na	K	Ca	Mg	P	Organic carbon
				$(cmol(+)\ kg^{-1})$				$(mg\ kg^{-1})$	(%)
Kwa Askofu	4.9	12.0	0.06	0.11	0.10	0.13	7.67	0.10
Unyankindi	7.3	33.4	0.60	0.76	0.63	0.72	3.47	1.50
Mwankonko	6.4	18.4	0.11	0.30	0.65	1.22	1.84	0.84
Ichungukia	7.3	16.0	0.24	0.28	0.54	0.81	13.12	0.24
Burudani	6.3	26.0	0.08	0.10	0.23	0.22	10.17	0.30
Kindai	5.2	10.2	0.12	0.19	0.53	0.05	1.86	3.59
Singidani	6.3	6.0	0.15	0.50	0.32	0.63	19.27	0.08

CEC = Cation exchange capacity; Na = sodium; K = potassium; Ca = calcium;Mg = magnesium; P = phosphorus;

Table 2: **Physical properties of the studied surface soils**

Location	Particle size (%)			Textural class
	Silt	clay	Sand	
Kwa Askofu	1.2	5.8	93	S
Unyankindi	11.3	55.7	33	SC
Mwankonko	1.3	24.7	74	SCL
Ichungukia	1.3	14.7	84	LS
Burudani	10.7	1.3	88	LS
Kindai	1.3	12.7	86	LS
Singidani	5.3	10.7	84	LS

S = Sandy; SC= sandy clay; SCL= sandy clay loamy; LS = loamy sandy

4.1.1   Soil pH

Soil pH in the studied area ranged from 5.2-7.3except for Kwa Askofu which was strongly acidicas indicated in Table 1. Theseresult imply that most of soil pH of the studied area ranged from slight acidity to neutral.Basically, metal toxicity occurs at soil pH lower than 5.0 when elements such as Al, Fe, Mn, and Cu have much greater solubility than crop nutrient requirements(Adamchuk and Mulliken, 2005).

The pH value is an important geochemical parameter and is anticipated to have a significant impact on uranium leaching, precipitation, and mobilization, because it can change uranium chemical forms in soil that have different mobility characteristics. Uranium is amphoteric, meaning that it can be mobilized at either high or low pH(Sutton and Burastero, 2004), as shown here under:

As an acid:$UO_3 + H_2O \rightarrow UO_2^{4-} + 2\ H^+$

Dissolving uranium oxide in a strong base like sodium hydroxide forms the doubly negatively charged uranate anion (UO_2^{4-}). Uranates tend to concentrate, forming diuranate, $U_2O_2^{7-}$, or other poly-uranates. Important diuranates include ammonium diuranate ($(NH_4)_2U_2O_7$), sodium diuranate ($Na_2U_2O_7$) and magnesium diuranate (MgU_2O_7), which form part of some yellowcakes. It is worth noting that uranates of the form M_2UO_4 do not contain $UO2^{4-}$ ions, but rather flattened UO_6octahedra, containing a uranyl group and bridging oxygen.

As a base: $UO_3 + H_2O \rightarrow UO_2^{2+} + 2\ OH^-$
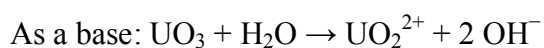
Dissolving uranium oxide in a strong acid like sulfuric or nitric acid forms the double positive charged uranyl cation. The uranyl nitrate formed ($UO_2(NO_3)_2 \cdot 6H_2O$) is soluble in ethers, alcohols, ketones and esters, for example, tributylphosphate. This solubility is used

to separate uranium from other elements in nuclear reprocessing, which begins with the dissolution of nuclear fuel rods in nitric acid. The uranyl nitrate is then converted to uranium trioxide by heating.

In acidic environments, speciation is dominated by UO_2^{2+}, the highest soluble uranium form. At pH > 4, cationic uranyl hydroxide and uranyl carbonate complexes form, of which the latter complexes are anionic above pH 9 (Amrhein andLosi, 1994; Davis *et al.,* 1994). In alkaline conditions, the U distribution ratio (Rd) isless than 20 when the solution pH isless than 8. Johnson *et al.* (2004) revealed that U sorption increases drastically as the soil pH increases. This means that the mobility of U decreases as the pH increases above 8 in alkaline soils.

However, most of the surface soils in this studied area are acid soils. Acid soils are dominated by the uranyl cation (UO_2^{2+}) and soluble carbonate complexes.Uranyl cation and uranyl carbonate complexes together with anionic P complex are probably the species mostly readily taken up by plant roots and transferred to the shoot.

4.1.2 Cation exchange capacity

The results from the studied area indicated that cation exchange capacity (CEC)mostly ranged from very low to low except for two areas of Unyankindi and Burudani which showed high cation exchange capacity as shown inTable 1.TheCEC gives an idea of the potential fertility status of the soil and the capacity of the soil to retain nutrients and other cations against leaching (Alam*et al.,*1993). The CEC levels observed in these studied soils indicate that the soils have low nutrient retention capacity (Landon, 1991).

Uranium can be adsorbed on a variety of negatively charged soil constituents including clay minerals, oxides and silicates, and organic materials. Johnson *et al.,* (2004) found a positive correlation between U and the cation exchange capacity of the soils.This phenomenon suggested that the adsorption of uranium through weak electrostatic bonds by the clay is an important mechanism in controlling uranium mobility. Soils which have high CEC values exhibit higher uranium sorption and low uranium concentrations in the soil solution compared to those with low CEC values (Vandenhove *et al.,* 2011).

However, most of the soils in this studied area have low cation exchange capacity.This impliesthat the soil cannot retain very highamount of uranium because cation exchange capacity is low, thus able to hold low levels of positively charged ions (cations) by electrostatic force.

4.1.3Available Phosphorus

The available phosphorus in the studied area ranged from very low (1.84) to low (10.17)mgPkg^{-1} soil except for two areas in Ichungukia and Singidani, which indicated medium levels of phosphorus (Table 1).Low available phosphorus in the soil may also be attributed to low soil pH (<5.8).Phosphorus could react with iron (Fe) and aluminium (Al) to produce insoluble Fe and Al phosphates that are not readily available for plant uptake (Hodges*et al.,* 2007). Phosphorus is an important plant nutrient necessary for root development, nodulation which is important for nitrogen fixation process, pod formation and filling in legumes (Marschner*et al.,* 1995).

Phosphates (PO_4^{3-})are among the most insoluble uranium (VI) compounds, and when phosphate is present in the soil at appreciable concentrations, uranyl phosphate compounds can control dissolved uranium concentrations. The influence of phosphorus

ligands in soil solution on the mechanisms of U sorption remains thus far unclear, meaning that a lot of research is still needed to understand the relation between the PO_4^{3-} and U mobility. The results from the literature revealed that maximum dissolved U in soil solutionis observed in different soils with low PO_4^{3-} level compared to that with a higher level of PO_4^{3-} (Van den Hove*et al.,* 2011).

4.1.5 Organic carbon

The results in the Table 1 indicate that organic carbon in the studied area range from very low to low accept atKindai area which showshigh organic carbon.Organic carbon isthe basis for soil fertility evaluation. Organic matter releases nutrients for plant growth, promotes soil structure, biological and physical health of soil, and is a buffer against harmful substances(Eswaran*et al.,* 2012).

UranylUO_2^{2+} has been reported to combine with organic ligands such as soil humic substances as well as acetate,fumerate, oxalate and citrate (Ganesh *et al*., 1997; Huang *et al*., 2003). Furthermore, U has been found in the soils assorbed, chelated or complexed with soil organic matter. Zhou and Gu (2005) found a strong linear relationship between uranium (VI) and total organic carbon released from the soil.The presence of humic substances could also enhance the mobility of uranium (VI) as a result of its complexation reactions with humic substances.However, most organic carbon in the studied area is low and this implies that the soil does not retain much uranium in the form of complexes with organic ligands.

4.1.6 Exchangeable bases

Table 1indicates that exchangeable bases (Ca, Mg and Na) in the study area were very low to low whereas K was very low to medium based on ratings compiled by Landon (1991). There is very little information in the literature on the effect of Ca on U(VI) sorption. Zheng*et al.* (2014) found that the presence of calcium carbonate in soils decreased U(VI) sorption and attributed this effect to the presence of calcium uranyl carbonate complexes. Hsi and Langmuir (1985) saw no effect of Ca or Mg on U(VI) sorption by iron oxides in carbonate-free systems, while Losiand Amrhein (1994) observed lower U(VI) sorption on goethite in synthetic drainage waters containing elevated levels of Ca and Mg. They attributed this effect to competition between Ca and Mg ions and positivelycharged U(VI) species for sorption surface sites.

The availability of calcium and potassium in soil affects uranium uptake in plants. This is due to calcium and potassium tending to replace uranium in soil solution by letting uranium to be absorbed on the soil colloids, leaving calcium and potassium in soil solution and thus making makes them easily available to plants as compared to uranium (Kohler*et al.,* 2014).

4.1.7 Particle size

In general, the soils in the study area were mainly composed of sand (Table 2). Soils at Kwa Askofu area were sandy composing of 93.0% sand, 1.2% silt and 5.8% clay whereas Unyankindi were sandy-clay and at Mwankonko were sandy-clay loam. Other areas were as indicated in Table 2.

The particle size is an important geochemical parameter for uranium distribution in soils. For instance, there is a strong positive correlation between U and clay content in the soil. Clay minerals are negatively charged hence they have ability to retain uranium cations

which are positive. Uranium content increases with decrease in soil particles. However, sand and silt did not show any particular significant correlationwith U (Kumar*et al*., 2015).

4.2 Levels of Uraniumin Soils and Rocks in Singida Urban District

4.2.1Concentration and distribution of uranium in soil

Table 3 shows levels of uranium in different soils at Darajani, Irao, Misuna, and Burudani areas.Elevation and soil sampling depths had no significant (P = 0.217) difference in uranium levels in the studied soils as shown in Appendix 1.These results indicated that ignoring other possible attributes the largest amount of uranium (8.75 mg kg^{-1}) was obtained in the soil collected atBurudani site. This was followed by the uranium in soils atMisuna (5.42 mg kg^{-1}) and Irao (5.35 mg kg^{-1}) which did not differ significantly between them. The significantly lower uranium level (3.74mg kg^{-1}) was obtained in soil at Darajani area. The variation in uranium contents in soils could be related with the concentrations of U in host rocks which are the granites. It is well known that granites are the primary source of U and release U during weathering (Sasmaz and Yaman 2008).Therefore, high level of U in granites implies on weathering release a lot of U in soils.

Table 3: Mean values of uranium levels in soils

Location	Mean Uranium (mg kg^{-1})
Darajani	3.74a[*]
Irao	5.35b
Misuna	5.42b
Burudani	8.75c
SE±	0.85
LSD$_{0.05}$	1.72
CV (%)	25.3
P- Value	<0.001

[*] *Means along the same columnsharing similar letter(s) do not differ significantly at 5% based on Duncan's' New Multiple Range Test (DNMRT).*

Based on the critical level of uranium (23 mg kg^{-1}) for agricultural land use, none of the soils in the present study indicatedfatal effect to crop life (Gupta and Gupta 1998). According to the Canadian Council of Ministers of the Environment (CCME, 2005), both environmental and human health soil quality guidelines have been developed for four land uses: agricultural, residential/parkland,commercial and industrial. Based on the recommendation given by CCME basing on Canadian Soil Quality Guidelines for the protection of environmental and human health, the acceptable levels of U are 23 mg kg^{-1} for agricultural land use and for residential/parkland land use, 33 mg kg^{-1} for commercial land use, and 300 mg kg^{-1} for industrial land use.Therefore, the level of U in agricultural soil in the study areas was low and more likely may not cause environmental health hazards to plant, livestock and human beings.

Table 4 shows variation of uranium with elevation and depth of sampling. The mean uranium elevation value in all areas indicated that uranium level increases with decreasing elevation down slope as shown in Table 4. High level of uranium at lower elevation could be due to uranium concentrated from parent material (granite) by weathering and erosion and eventually washed down the slope.

Usually water velocity on a slope affects deposition of materials in suspension as sand drops out of suspension first, while clay size particles can be carried further away from the base of the slope before they are deposited. This kind of geological sorting brings about variation in soil in relation to landscape(Wang*et al.*, 2001).

Table 4:Variation of uranium with elevation and depth of sampling

Location	Elevation	Depth (cm)	Uranium level (mg kg^{-1})	Mean U elevation value (mg kg^{-1})	Textural class
Irao	Lower	0–30	8.15		CL
		30–60	7.43	7.06	SCL
		60–100	5.61		SC
	Middle	0–30	7.61		SCL
		30–60	3.65	4.97	SL
		60–100	3.65		LS
	Top	0–30	5.42		SC
		30–60	4.00	4.03	SCL
		60–100	2.68		LS
Darajani	Lower	0–30	5.24		SCL
		30–60	2.55	4.23	LS
		60–100	2.33		S
	Middle	0–30	4.05		CL
		30–60	3.89	3.76	SL
		60–100	3.35		SC
	Top	0–30	4.82		SCL
		30–60	4.68	4.23	CL
		60–100	3.19		SL
Burudani	Lower	0–30	7.83		S
		30–60	8.66	9.95	SL
		60–100	12.65		SCL
	Middle	0–30	7.86		SL
		30–60	10.44	8.72	SCL
		60–100	11.06		SC
	Top	0–30	5.55		LS
		30–60	8.55	7.59	SL
		60–100	9.38		SC
Misuna	Lower	0–30	2.55		S
		30–60	7.52	6.93	SL
		60–100	7.72		SCL
	Middle	0–30	4.00		SL
		30–60	4.60	5.72	SCL
		60–100	5.23		SG
	Top	0–30	5.52		LS
		30–60	6.14	6.18	SCL
		60–100	6.88		SC

Textural class: SL= Sandy loamy, CL= Clay Loamy, SCL= Sandy Clay Loamy, SC= Sandy Clay, S= Sandy, LS= Loamy Sandy

The results indicated that the levelof U atIrao and Darajani areas decreases with depth along the soil as presented in Table 4. There is high U content in upper surface and it decreases with increasing soil depth. The higher amount of U in upper surface could be due organic substances including acetate,fumerate, oxalate and citrate whichoccurs in the upper soil surface.These organic substances cause uranium retention through complexationand cause high uranium level in upper surface.

High amount of finer particles in the form of clays and low sand content in the upper surface also can cause high uranium level in upper surface, This occurs in Irao and Darajani as shown in the Table 4where uranium level decreases as clay content decreases. This is because clay minerals are negatively charged hence they have ability to retain uranium cations which are positive. This could be the case because clay content in the Irao and Darajani decreased with soil depth.

Furthermore, the resultsin Burudani and Misuna areas indicated that the uranium levelincreasedwith soil depth along the slope(Table 4). This could bedue to increase in clay content with soil depth and the retention of uranium by clay.According to De Craen*et al.* (2004), uranium content increases with decrease in soil particles size.Mostly clay minerals are negatively charged hence they have ability to retain uranium cations which are positive. This could be the case because clay content in the Misuna and Burudaniincreases with soil depth.

Soil with higher cation-exchange-capacity will retain more uranium, while carbonate in the soil increases the mobility of uranium through the formation of anionic U and CO_3 complexes (Vandenhove*et al.,* 2007).Uranium does not migrate substantially in loam compared to sandy soils (Sheppard *et al.,*2005). Uranium migration in soil occurs over the

period of a few months, depending on sorption, and may be upwards when there is a net water deficit or downwards as a result of net leaching (Sheppard *et al.,*2005).

However, high level of uranium at Burudani areas could be due to high level of uranium ingranite rocks around the area as shown in Table 5which on weathering releases substantial amounts of U in the soil.

Plants are surface feeders; therefore, they are capable of absorbing high amount of uranium. In the short period of time the accumulation level may be below the critical levels. However, with time heavy metals tend accumulate and may reach to unacceptable levels especially for perennial crops, and even for annual like cereals (sorghum and sunflower). The amount might be low butthroughthe food chain they can accumulate in organism to high levels.

4.2.2 Concentration and distribution of uranium in rocks

Table 5 shows that levels of uranium in granite rocks at different sites. The highest uranium level (31.57 ± 2.474 mgkg^{-1}) wasrecorded in Burudani followed by Misuna rocks (28.98 ± 2.474 mg kg^{-1}). On the other hand, the lowest uranium amount (20.01mgkg^{-1}) was recorded from Darajani rocks. The highest soil uranium levels (Table 4) was also recorded at Burudani area. The trend in U soil level was the same in the rocks suggesting that the main source of uranium in soil was granite rocks. Weathering of granites might have released equivalent quantity of U as those found in the soil.

Table 5: Levels of uranium in granite rocks

Location	Mean (mg kg^{-1})
Darajani	20.01a[*]
Misuna	28.98ab
Burudani	31.57b
LSD$_{(0.05)}$	10.64
CV (%)	9.2
SE±	2.47
P- value	0.077

[*]*Means along the same columnsharing similar letter(s) do not differ significantly at 5% based on Duncan's' New Multiple Range Test (DNMRT).*

4.3 Uranium Levels in Surface and Underground Water

Table 6 shows uranium levels in underground and surface waters. Results from underground water samples indicated higher uranium concentrations as compared to critical level given for water by WHO (2011). The concentration of U ranged from <0.01to 0.46mgL^{-1} with a mean value of 0.09mgL^{-1} (Table 6). The current provisional recommended WHO guideline value of U in drinking water is 0.03mgL^{-1} (WHO, 2011). This means that the levels of U in Singida underground water is higher than the recommended level. This is an indication of high potential health hazards associated with drinking water contaminated with high uranium levels.

Uranium concentrations from the two surface water samples in this study werefound to be very high at 0.087mgL^{-1} and 1.097mgL^{-1} (Table 6)as compared with the WHO drinking water guidelines value of 0.03mgL^{-1} (WHO, 2011). The two lakes are not used and should not be used as sources of drinking water for humans although they are used for fishing activities.Livestock are using this water for drinking. Thus, there is possibility, through the food chain, for uranium to enter living organism including human beings.

Furthermore,the variation of U in water sources among the study areas couldbe contributed by two factors such as pH of water where the sample was taken and concentration of uranium in surrounding rocks. Waterspassing through uranium rich soils or rocks are likely to have a high level of uranium compared to the water flowing through soils or rocks with lowlevel of uranium (Ankle *et al.,* 2009).Whenthe pH is high, likewise U level is high too.Thisis because at high pH uranium form soluble uranium carbonate complex (Waite *et al.,* 1994). U-carbonate complexes are predominant at neutral to alkaline pH value, and this could be expected in most areas where water was sampled (Table 6).

Table 6: Uranium levels in underground and surface waters

Location	Coordinates	Elevation (m)	Source	Uranium levels (mgL^{-1})	H$_2$O pH value
Mwankonko A1	4^{0}50'36.2668"E&34^{0}50'1.9047"S	1493	Underground	0.059	7.7
Mwankonko A2	4^{0}50'35.2621"E&34^{0}38'38.5507"S	1479	Underground	0.058	5.99
Darajani A	4^{0}50'35.5341"E&34^{0}39'15.6236"S	1464	underground	0.062	5.43
Mwankonko B1	4^{0}51'36.3799"E&34^{0}38'48.447"S	1480	Underground	0.201	7.25
Mwankonko B2	4^{0}51'36.7607"E&34^{0}39'2.0418"S	1478	Underground	0.193	6.62
Kindai lake	4^{0}50'4.5871"E&34^{0}44'9.0929"S	1481	Surface	1.097	7.81
Sido area	4^{0}49'7.4125"E&34^{0}44'27.0478"S	1508	underground	0.464	7.49
Kititimo area	4^{0}49'43.2446"E&34^{0}46'48.4006"S	1516	Underground	0.018	6.81
Busta A	4^{0}48'54.8008"E&34^{0}44'45.0585"S	1500	Underground	0.011	6.25
Busta B	4^{0}48'51.5511"E&34^{0}44'42.0876"S	1491	Underground	0.023	6.70
Sema office	4^{0}49'16.1764"E&34^{0}44'36.9678"S	1516	Underground	<0.01	6.73
Sema residential	4^{0}48'39.6505"E&34^{0}43'59.2315"S	1506	Underground	<0.01	6.67
Singidani lake	4^{0}46'51.2954"E&34^{0}46'0.9357"S	1484	Surface	0.087	7.34
Irao	4^{0}52'59.3864"E&34^{0}45'44.0135"S	1503	Underground	<0.01	6.64
Kibaoni	4^{0}50'10.7811"E&34^{0}45'9.6568"S	1503	Underground	0.24	6.70

4.4 Uptake of Uranium by Finger Millet and Sunflower

4.4.1 Uranium concentration in selected farms

Table 7 shows uranium levels in agricultural soil in sorghum and sunflower farms. Uranium concentrations in surface soils taken at different points in selected farms of sorghum and sunflower (Table7) indicated that the highest U concentration from this study was at Burudani (15.9mgkg[-1]) and the lowest concentration was at Kwa Askofu (< 0.01mgkg[-1]).

There is a significance relationship between soil acidity and U concentration such that the increase in soil acidity reduces the U concentration. It is well depicted to results from Darajani and Unyankindi which have about neutralpH and higher U concentrations than other sites with acidic soil. The lowest soil pH (acidic) at Kwa Askofu has the lowest U concentration. This Uranium-pH relationship relates to the findings of the study done by Chuan*et al.* (1996), which highlighted high uranyl ion solubility in acidic conditions resulting in its transfer further below the surface soil.

Levels of U found in sorghum and sunflower farms are low and may not cause toxicity to plants. Plants can survive in the area where the uranium level is less than 23mgkg[-1] (Singh, 1997). Therefore, the levels of U in agricultural soils in the study area werelow and are more likely not to causeenvironmental hazards to plant, livestock and human beings in the short term. However, with time heavy metals tend to accumulate and may reach to unacceptable levels that can be harmful to human beings.

Furthermore, there wereother potentially toxic metals that have been found to occur in association with uranium. Such metals were Zr, Sr, Pb, Ti, S, Ba and Cr as shown in Appendices 2 and 3. These metals similarly to uranium might be taken up by plants and

through food chain may have negative impacts to humans and livestock. A follow up study on these metals is recommended to ensure the safety of Singida urban residents.

Table 7:Uranium levels in agricultural soils in sorghum and sunflower farms

Site name	Coordinates	Elevation (m)	Uranium levels (mgkg^{-1})	Soil pH value
Kwa askofu	$04^0$48'073S&$034^0$46'091"E	4931	<0.01	4.9
Unyankindi	$04^0$48'426"S&$034^0$45'028"E	4928	13.1	7.3
Mwankonko	$04^0$50'338"S&$034^0$39'644"E	4803	7.0	6.4
Darajani	$04^0$48'873"S&$034^0$44'728"E	4945	15.9	7.3
Ichungukia	$04^0$48'800"S&$034^0$45'028"E	4928	1.5	5.0
Singidani	$04^0$47'733"S&$034^0$45'857"E	4869	2.3	5.2
Kindai	$04^0$45'723"S&$034^0$46'634"E	4942	6.3	6.9

4.4.2   Uranium levels in different plants parts

Table 8 shows uranium levels in plants samples. Although U is not essential or beneficial to plants, many species will absorb U and incorporate it into their biomass along with other metals(Dushenkov*et al.,* 1997).

The concentration of U in plants has been shown to be dependent on the degree of U contamination in the soil (Huang*et al.,* 1998) as well as soil pH. Values of U in plant tissues are shown in Table 8**.** The results indicated that uranium levels assimilated in sunflower and sorghum (grains and shoot) in the study area were low. The sorghum accumulated U in grains ranged from 1.4 to 2.8 mgkg^{-1}and from <0.01 to 2.2 mgkg^{-1} in shoot. Whileuranium levels accumulated in sunflower grains ranged from 3.0 to5.2 mgkg^{-1}and inshoot ranged from 1.3-2.3 mgkg^{-1}.The highest quantity of U was obtained in grains because grains are important storage parts of the plants in forms of starch for sorghum and oil for sunflower.

The level of uranium found insorghum and sunflower was low and thus may notpose health problem to livestock and human beings.The World Health Organization (WHO) (2004) has established a Tolerable Daily Intake (TDI) for soluble uranium of 0.6 µg/kgbody weight per day, based on the lowest-observed-adverse-effect-level (LOAEL) for uranium nephrotoxicity of 0.06 mg/kg body weight per day. This suggested that when U concentration above TDI is consumed, a person is at risk of getting cancer and kidney failure due to consuming high levels of U, as could happen upon long-term consumption of some of crops shown in Table 8.

Table 8: Uranium levels in plants samples

Location	Type of plant parts	Uranium levels (mgKg^{-1})
Kwa askofu	Sorghum shoot	<0.01
	Sorghum grain	2.2
Unyankindi	Sorghum shoot	<0.01
	Sorghum grain	1.4
Singidani	Sorghum shoot	2.2
	Sorghum grain	2.8
Darajani	Sunflower shoot	1.3
	Sunflower grain	3.0
Ichungukia	Sunflower shoot	2.2
	Sunflower grain	3.4
Kindai	Sunflower shoot	2.3
	Sunflower grain	5.2
Mwankonko	Sorghum shoot	1.2
	Sorghum grain	2.1

Furthermore, sunflower and sorghum were grown in similar type of soil.The difference in theability to accumulate Uisdue to the difference in their root systems. Sunflowers have a single taproot and smaller, hairy secondary roots. Sunflower roots usually grow 30.5 to 91.4 cm deep. Inthe United States Department of Agriculture soil scientists have

measured sunflower roots to more than 152.4 cm long (Mukhtar, 2010). The roots system of sunflower gives higher ability to extract soluble nutrients and other materials including U in the soil than sorghum because sorghum has a fibrous root system that is similar to corn. But sorghum and sunflower can grow in dry areas. In other words uranium uptake by plants may be limited to the outer membrane of the root system and may not occur on the interior of the root at all and this raises the selectivity of the root to U.

CHAPTER FIVE

5.0 CONCLUSIONS AND RECOMMENDATIONS

5.1 Conclusions

From the results of this study, it is concluded that:

i.   Water from Mwankonko A1, Mwankonko A2, Darajani A, Mwankonko B1, Mwankonko B2, Kindai Lake, SIDO area and Singidani Lake showed high uranium levels ranging from 0.058 to 1.097mgL^{-1}, the values which are higher than the recommended limit of 0.03mgL^{-1} set by WHO (2011). Thus, they are not safe for human and livestock consumption.

ii. Levels of uranium found in sunflower and sorghum grain and shootare within the tolerable limit to human and the environment. However,sunflower indicated slightly increased ability to accumulate high levels of uranium in grain as compared to sorghum

iii. Uranium levels in soil varied with elevation. Low levels of uranium occurred in the top elevation and high in the lowerelevation.

iv. Uranium levels in agricultural soilsare within the tolerable limit set by the WorldHealth Organisation.

v.   Low levels of uranium in sorghum and sunflower parts provided assurance that they are currently safe for livestock consumption.

5.2 Recommendations

From the results of this study, the following recommendations aremade:

i.   The data obtained on U levels in drinking water should be communicated to the community in the study area by relevant authorities in the District.

ii. The study recommends the use of ion exchange with anion resin and reverse osmosis (RO) techniques in order to treat U in water or non- use of the water for domestic purposes.Alternative sources of water that aresafe to human and livestock should be sought.

iii. Further studies are recommended to assess other potentially toxic metals like Zr, Sr, Pb, Ti, S, Ba and Cr whichoccur with uranium in the study area.

iv. It is recommended that an alternative source of good quality water be found for the people of Singida even if Lake Victoria is used as the source.

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APPENDICES

Appendix 1:**Analysis of variance (ANOVA) for the uranium levels for the soils collected from different sites in Singida**

Source of variation	d.f.	s.s.	m.s.	v.r.	F pr.
Replication	1	0.116	0.116	0.05	
Site (S)	3	239.251	79.75***	36.9	<.001
Elevation (E)	2	8.239	4.119 NS	1.91	0.164
Depth (D)	2	4.868	2.434 NS	1.13	0.336
Site.Elevation (S×E)	6	64.47	10.745***	4.97	<.001
Site.Depth (S×D)	6	29.124	4.854 NS	2.25	0.061
Elevation.Depth (E×D)	4	87.91	21.978***	10.17	<.001
Site.Elevation.Depth (S×E×D)	12	36.04	3.003 NS	1.39	0.217
Residual/Error	35	75.635	2.161		
Total	71	545.653			

Key: *** = very highly significant ($P < 0.001$); NS = Not Significant ($P > 0.05$)

Appendix 2:Potentially toxic metal occurring in soil with uranium

Location	Mo	Zr	Sr	U	Rb	Th	Pb	As	Zn	Cu	Ni	Co	Mn
Kwa askofu	7.20	139.65	26.05	<0.01	34.12	6.17	7.00	1.32	11.27	16.93	36.30	8.94	109.50
Unyankindi	3.31	378.76	116.18	13.05	112.36	32.61	34.42	3.42	63.82	42.53	46.46	53.22	427.05
Singidani	5.99	404.03	64.70	7.02	33.86	15.79	15.66	3.43	21.78	24.53	39.69	17.56	262.64
Darajani	3.62	275.60	113.17	15.89	73.02	14.17	30.56	2.54	1007.97	30.71	36.05	4.78	254.74
Ichungukia	3.85	150.99	19.00	1.52	31.97	6.17	9.15	<0.01	19.86	18.23	25.14	8.48	118.93
Kindai	5.79	207.32	23.72	2.26	24.82	10.09	7.95	1.31	11.58	11.81	33.70	3.69	110.32
Mwankonko	4.55	258.13	51.03	6.87	36.32	11.08	11.70	0.55	17.86	17.91	26.30	30.46	193.98
NIST2710a	7.76	186.97	253.42	11.6	62.84	69.16	5531.54	1675.88	4518.27	3575.59	65.47	<0.01	2368.84
Certified value			255.00	9.11			5520.00		4180.00	3420.00		5.99	2140.00

Appendix 3:Potentially toxic metal occurring in soil with uranium

Location	Cr	V	Ti	Sc	S	Ba	Cs	Te	Nb	Bi	P	Cl
Kwa askofu	255.52	25.86	645.12	<0.01	40.52	250.78	23.55	30.50	4.92	<0.01	344.22	280.65
Unyankindi	144.18	118.85	5779.98	22.52	275.42	352.26	7.09	<0.01	46.62	38.97	495.17	230.16
Singidani	130.27	69.61	3246.68	7.42	160.49	271.76	16.63	6.97	18.50	15.13	616.83	190.83
Darajani	121.49	47.72	2445.76	21.46	1839.63	373.24	19.85	7.31	17.56	12.75	732.01	348.78
Ichungukia	225.90	38.03	1067.867	0.39	199.58	186.49	17.57	10.81	6.01	<0.01	641.22	197.38
Kindai	173.34	48.16	1559.36	2.68	166.63	190.14	14.95	11.91	10.04	4.33	546.93	173.03
Mwankonko	254.15	44.95	1950.57	<0.01	232.19	211.66	8.19	<0.01	11.79	5.89	697.60	182.57
NIST2710a	45.81	101.03	3201.48	27.90	25207.9	802.72	48.12	69.64	14.83	34.20	1963.14	<0.01
Certified value						792.00						