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Investigation of uranium derived from phosphate fertilizers on plants uptakes and bacterial diversity in selected agricultural soils of east africa

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INVESTIGATION OF URANIUM DERIVED FROM PHOSPHATE FERTILIZERS ON PLANTS UPTAKES AND BACTERIAL DIVERSITY IN SELECTED AGRICULTURAL SOILS OF EAST AFRICA

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A Thesis Submitted in Fulfilment of the Requirements for the Degree of Doctor of Philosophy in Environmental Science and Engineering of the Nelson Mandela African Institution of Science and Technology

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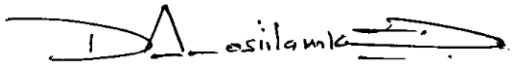
ABSTRACT

Different types of phosphate fertilizers (PFs) used in agriculture to increase soil fertility contain uranium (U) as an accompanying element. Uranium is both toxic and slightly radioactive. This study investigated the concentrations of U in phosphate rocks (PRs) and PFs used in growing tobacco (*Nicotiana tabacum* L.) and maize (*Zea Mays* L) plants from Burundi, Kenya, Rwanda, Tanzania, and Uganda. The results showed that the concentrations of U in phosphates from Burundi, Kenya, Tanzania and Uganda were statistically significant ($P \leq 0.01$). The lowest U concentration was 10.7 mg kg^{-1} found at the Mrima Hill deposit in Kenya and the highest was 631.6 mg kg^{-1} found at the Matongo deposit in Burundi. Uranium concentrations for local and imported PFs was also statistically significant ($P \leq 0.01$). Uranium concentration from PFs ranged from 107.88 ± 9.60 and 281.57 ± 15.82 mg kg^{-1}. The study also assessed the influence of U from PFs on the radioactivity of agricultural soils and uptake by tobacco plants. The results showed that applications of PFs in agricultural soils increased radioactivity in agricultural soil and tobacco crops. Tobacco smoking and snuffing behaviors resulted in an annual effective dose that was 2.41-6.53 and 1.14-2.45 times greater than annual recommended effective doses for snuffers and smokers. This work also investigated the influence of fertilizer derived U on maize plant uptake and bacterial diversity in soil after application of fertilizers with varying U concentrations. The pilot field experiments showed that application of PFs with different U concentrations influenced bacteria abundance and diversity in maize crops. Applications of Nafaka plus (NP) (3.93 mg kg^{-1}) and Minjingu Powder (MP) (3.06 mg kg^{-1}) PFs in soil increased bacteria abundance and diversity. Some bacteria were abundant on NP treated soil (high U content) because of their ability to tolerate higher U concentrations. Uptake from soil to crop for maize after applications of PFs of varying U concentrations was investigated by amending soil with *Eucalyptus globulus ssp maideii* bark and kaolin clay. The soil amendments in reduced U uptake from soil to plant in pot experiments. It is believed that the reduced U uptake is associated with *eucalyptus globulus ssp maideni* carboxyl groups that can reduce mobile hexa-uranyl ions to immobile tetra uranyl ions that can be absorbed by the kaolin clay. The study recommends further work in understanding the mechanisms of *Eucalyptus ssp maidenii* bark powder and kaolin U uptake reduction in soil complex matrix.

DECLARATION

I, Dennis Amos Mwalongo, do here declare before the Senate of the Nelson Mandela African Institution of Science and Technology that this Thesis is my own original work and it has neither been submitted nor being concomitantly submitted for degree award in any other institution.



16th August, 2024

Dennis Amos Mwalongo — Date

The declaration is hereby confirmed by:

16th August, 2024

Prof. Kelvin M. Mtei — Date

16th August, 2024

Prof. Thomas T. Kivevele — Date

16th August, 2024

Dr. Nils H. Haneklaus — Date

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CERTIFICATION

The undersigned certify that have read and hereby recommend for acceptance by the Senate of the Nelson Mandela African Institution of Science and Technology the thesis titled, "*investigation of uranium derived from phosphate fertilizers on plants uptakes and bacterial diversity in selected agricultural soils of East Africa*" in fulfilment of the requirements for the award of the Degree of Doctor of Philosophy in Environmental Science and Engineering of the Nelson Mandela African Institution of Science and Technology.

Prof. Kelvin M. Mtei Date

Prof. Thomas T. Kivevele Date

Dr. Nils H. Haneklaus Date

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DEDICATION

This work is dedicated to my lovely father, the late Mr. Amos Msanga Mwalongo and my lovely mother, the late Ms. Rhoda Msango Mtewele. Also, to my beloved wife, Ms. Joyce David Kivambe, and sons David Mwalongo and Danley Mwalongo.

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LIST OF ABBREVIATIONS AND SYMBOLS

ANOVA	Analysis of Variance
ASV	Amplicon sequence variants
Ca	Calcium
CEC	cation exchange capacity
CRM	Certified Reference Materials
DAP	Di-ammonium phosphate
DNA	Deoxyribonucleic Acid
EDXRF	energy dispersive X ray fluorescent
EU	European Union
GPS	Global Positioning System
ha	Hectare
IAEA	International Atomic Energy Agency
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
ICRP	International Commission on Radiological Protection
K	Potassium
LSD	Least Significant Difference (LSD)
mg kg^{-1}	Milligram per kilogram
MOHP	Minjingu hyper organic phosphate
NPK	Nitrogen, phosphorous and potassium
NEA	Nuclear Energy Agency
NIST	National Institute of Standards and Technology, United States of America
NM-AIST	Nelson Mandela African Institution of Science and Technology
NPS	Nafaka Plus
OM	Organic Matter
PFs	phosphate fertilizers
PG	Phospho-gypsum
PRs	Phosphate rocks
PRISMA	Preferred Reporting Items for Systematic Reviews and Meta Analyses
PT	Proficiency Testing
Ra	Radium 226
TAEC	Tanzania Atomic Energy Commission

Th	Thorium-232
U	Uranium-238
UNSCEAR	United Nations Scientific Committee on the Effects of Atomic Radiation
US GS	United States of America Geological Survey
WPA	Wet Phosphoric Acid
YB	YaraMila blended
YC	YaraMila Cereal

CHAPTER ONE

INTRODUCTION

1.1 Background of the problem

Most of phosphate fertilizers processed from phosphate rocks contain high concentrations of naturally occurring Uranium (U). These high concentrations of U could be transferred to the phosphate fertilizer streams during wet phosphoric acid (WPA) production, which is the dominant process (> 90%) of all phosphate fertilizers production globally (Haneklaus *et al.,* 2017). The concentration of U in phosphate rocks and the resulting phosphate fertilizers thus depends largely on the type of the phosphate ores used. For instance, Bunus (2000) estimated that U in phosphate fertilizers processed from sedimentary ore contain about 80 –100 mg kg^{-1} while the higher-end concentrations occasionally reaching 160–180 mg kg^{-1} (Hellal *et al.,* 2019; Mogollón *et al.,* 2018).

Phosphate rocks can also be directly applied to agricultural soils without processing it into chemical fertilizers. This is done with some highly reactive phosphate rock components that is agronomically suitable for direct application on more acidic soils for releasing the nutrients into the soils, such as the ones found in East Africa (Casanova, 1995; Rajan *et al.,* 1996; Szilas, 2002). However, direct application of phosphate rocks into the agricultural soils means that, all U contained in the phosphate rocks is transferred to the agricultural soils as well.

The concentration of U in phosphate rocks and phosphate fertilizers is thus of concern and needs to be better understood. Systematic and timely data on the U content in most common phosphate fertilizers used for soil fertility in agricultural soils across East African countries is not available today.

Tobacco is a cash crop extensively grown in Kenya, Tanzania, and Uganda. Tanzania is one of the leading tobacco producer in the East African region, whereby it is the 3rd in Africa and the 8th in the world (Ndomba, 2018). Uganda is the 2nd in Africa, followed by Kenya, Rwanda and Burundi (James, 2019). In these countries, tobacco plantations intensively use nitrogen (N), phosphorous (P), and potassium (K) fertilizers from different brands (Lisuma *et al.,* 2022). Furthermore, Kenya, Tanzania, and Uganda are rural countries and more than 80% of the population depends on agriculture by cultivating various crops, including tobacco, for their livelihood (Lokuruka, 2021; Lokuruka, 2020). The fact that a large population in these countries are directly involved in agricultural production means that a large share of the

population could be exposed to radioactivity beyond natural background levels through the intensive application of phosphate fertilizers (Purnama & Damayanti, 2020; UNSCEAR, 2000). However, systematic studies on natural radionuclides levels associated with fertilizers use in tobacco production in Burundi, Kenya, Rwanda, Tanzania, and Uganda have to the best of our knowledge, not been performed yet, although different studies have determined the elevated natural radioactivity (Banzi *et al.,* 2000; Makweba & Holm, 1993; Meza *et al.,* 2015).

Recently, new fertilizers aimed to suit better maize crops, such as the YaraMila Cereal (YC) and the Nafaka Plus (NP) have been developed and introduced for use in Tanzania. YaraMila Cereal (YC) is developed by the Yara Fertilizer Company and is imported to Tanzania, while Nafaka plus (NP) is produced locally by the Minjingu Mines and Fertilizer Limited company located in northern Tanzania. In addition, some maize farmers have been using Minjingu Powder (MP) which is natural phosphate rock milled and ground into powder form so that it can be used for cereal crops.

The influence of U on bacterial activity has thus far only been investigated for mine, mine waste or U processing sites. Although the topic of environmental risks from fertilizer-derived U is gaining considerable momentum, the influence on bacterial diversity associated with different U concentrations in fertilizers has not been investigated in Tanzania or elsewhere yet. Therefore, it is important to investigate and contribute to the body of knowledge on the changes in soil bacterial community induced by the application of U derived from application of phosphate fertilizers and to identify the cereal fertilizers with the lowest effect on the soil bacterial community. This knowledge contributes to determination of the relationship between U impurities from phosphate rocks and phosphate fertilizers on maize crops in sandy loam soil that has to the best of our knowledge not been studied yet.

1.2 Statement of the problem

Phosphate fertilizers contain U which is a contaminant in agricultural soil. Removing U during phosphate fertilizers production, though commonly done in the past at industrial scale in Florida in the USA in the 1980s–1990s (Steiner *et al.,* 2020) is currently not a preferred option as it increases manufacturing costs. In addition, removing U from phosphate rocks destined for direct application is even more challenging since some phosphate rock types contain high organic matter and high concentration of Calcite ($CaCO_3$) which consume acid (Ruan *et al.,* 2019; Zafar *et al.,* 1995). Uranium can relatively easy be recovered from liquid wet phosphoric acid, an intermediate product in fertilizer production. In addition it could be directly leached

from the phosphate ore prior to processing as suggested by Al Khaledi *et al.* (2019) and Guzmán *et al*. (1995).

Essentially, during phosphate fertilizers production U is neither recovered as a mineral resource nor removed as a contaminant as no country has legislations and regulations in place that restrict U concentrations in mineral fertilizers (Haneklaus *et al.,* 2017; Kratz *et al.,* 2016) and this practice is also not economically viable given the relatively low value of natural U (Haneklaus, 2021; López *et al.,* 2019; Shang *et al.,* 2021). It is suspected though, and part of an active scientific debate, that prolonged application of phosphate rocks and phosphate fertilizers can cause U accumulation in agricultural soils (Bigalke *et al.,* 2017; Campos *et al.,* 2021; Ratnikov *et al.,* 2020; Schipper *et al.,* 2011; Takeda *et al.,* 2006; Yamaguchi *et al.,* 2009). It is further speculated that a high concentration of U in agricultural soils could influence its uptake by plants through the root system in a similar way that other essential elements such as nitrogen, phosphorous, potassium, calcium and magnesium, and other supplemented micronutrients are absorbed (Baumann *et al.,* 2014; Harguindeguy *et al.,* 2019; Semioshkina & Voigt, 2021; Sheppard, 2011; Shtangeeva, 2010; Velasco *et al.,* 2009). Saleh *et al.* (2018) reported for instance that, the uptake of U from soils to plants behaves chemically similar to that of Calcium (Ca).

The concentration of U in phosphate rocks and phosphate fertilizers is thus of concern and needs to be better understood. Systematic and timely data on the U content in the most common phosphate fertilizers used for soil fertility in agricultural soils across the East African countries is not available today. Recently, new fertilizers aimed to suit better maize crops, such as the YaraMila Cereal (YC) and the Nafaka Plus (NP) have been developed and introduced for use in Tanzania. In addition, some maize farmers have been using Minjingu Powder (MP) which is natural phosphate rock milled and ground into powder form so that it can be used for cereal crops. The information on the U concentration in these PFs is to the best of our knowledge not available. Moreover. The influence of U concentration on bacterial activity has thus far only been investigated for uranium mine effluents, mine wastes or active and former U processing sites. Although the topic of environmental risks from PFs-derived U is gaining considerable momentum, the influence on bacterial diversity associated with different U concentrations in fertilizers has not been investigated in Tanzania or elsewhere yet. Reducing U uptake from soil to the most consumed cereals (such as maize) using natural materials such as *Eucalyptus globulus ssp maidenii* and kaolin in the wake of reluctance in setting regulatory limit of U in PFs needs to be understood.

1.3 Rationale of the study

Phosphate rocks used as source of elemental phosphorous contain U which pose significant concerns to humans and the environments. It is known that East African phosphate rocks can contain relatively high concentrations of this heavy metal if compared to the concentrations found in phosphate rocks globally (Banzi *et al.,* 2000; Makweba & Holm, 1993; Meza *et al.,* 2015). For example, Minjingu phosphate ores from Northern Tanzania are directly applied to agricultural soils, distributing all the U with it. Since U is highly soluble in oxidizing environments, radioactive U may leach into groundwater depending on the pH and redox potential of the soil or even be absorbed by plants such as tobacco and maize.

There is a current scientific debate regarding the fate of U in agricultural soil, which is a challenging issue as some scholars urge that U uptake by plants depends on the type of plant grown in P fertilizer contaminated soil as well as the soil pH. Even though using locally produced fertilizers in East Africa may pose environmental risks, and some East Africans have already voiced these concerns. Local P fertilizers are significantly less expensive than imported fertilizers and they are therefore usually preferred. Also, the U content in the imported fertilizers which may contain significant concentration of U as well is still unknown. Consequently, U can progressively accumulate in agricultural soils over longer periods of fertilizer application. Although U is recognized to cause agricultural soil contamination and pose chemical and radiological risks to humans and the environment in East Africa, there is currently no country in the region with regulation to limit the U content in the fertilizers. Globally such a regulation does also not exist. The availability of information on the magnitude of the problem may improve and suggest for policy formulation. Thus, this thesis clarified the situation based on information gathered about the amount of U in East African phosphate rocks and mineral fertilizers used, as well as the possible effects of fertilizer application on soil contamination in agriculture and the potential for plant uptake of U, which could then be limited.

A systematic information on the U content in phosphate rocks of East African origin and common fertilizers used in five East African countries remains unknown. This study investigated U levels in East African phosphate rocks and mineral phosphate fertilizers, as well as the possible effects of P fertilizers applications on soil pollution, tobacco and maize uptake and ultimately explored the potential material that can limit U uptake by maize plant.

1.4 Research objectives

1.4.1 General objective

Investigate uranium concentration in phosphate fertilizers, its potential uptake by tobacco and maize plants and assess bacteria diversity in selected agricultural soils in East Africa.

1.4.2 Specific objectives

The specific objectives of this study were:

(i) To investigate the concentration of U in phosphate rocks and phosphate fertilizers applied in agricultural soils in selected East African countries.

(ii) To investigate impact of phosphate fertilizers application on the U concentration in agricultural soils and tobacco plant.

(iii) To investigate the change in agricultural soil bacterial diversity following the application of phosphate fertilizers with varying U concentration.

(iv) To evaluate the effect of kaolin and *Eucalyptus globulus ssp maidenii* on U uptake from soil to maize plant.

1.5 Research questions

(i) What is the concentrations U in phosphate rocks and phosphate fertilizers applied in selected countries in East Africa?

(ii) What is the extent of phosphate fertilizers application on the U concentration in agricultural soils and uptake by tobacco plant?

(iii) To what extent does U variations in phosphate fertilizers influence bacterial diversity in agricultural soils?

(iv) What is the effectiveness of reducing U uptake from soil to maize plant compartments after treatment with kaolin and *Eucalyptus globulus ssp maidenii*?

1.6 Significance of the study

The findings from this study improve understanding of Uranium content in phosphate rocks, phosphate fertilizers, bacterial diversity and use of natural materials to reduce its uptake by crops. This study is of significant importance as it addresses a critical issue at the intersection of agriculture, environmental science, and public health. By shedding light on the uranium content in phosphate rocks, mineral fertilizers and its impact on soil, crops uptakes, bacterial diversity and human health and potential of reducing uranium uptake. The study provides valuable insights that can inform agricultural practices, public health policies, and environmental protection efforts. The findings underscore the need for a balanced approach to fertilizer use, ensuring that the benefits of increased soil fertility do not come at the cost of environmental contamination and health risks. The study's recommendations for future research also pave the way for the development of innovative solutions to recover U from phosphate fertilizers and hence, mitigate the risks associated with uranium in agriculture.

1.7 Delineation of the study

Uranium concentrations in phosphate rocks, common phosphate fertilizers and the influence of phosphate fertilizers on activity concentrations in soil and tobacco plant uptake were studied and are reported in this study. Furthermore, effects of U on bacterial diversity and use of natural materials such as kaolin and *eucalyptus globulus ssp maidenii* bark powder in reducing U uptake by maize plant are as well presented in this work. However, the following were not covered in this study:

(i) The current study did not investigate U from all phosphate rocks and phosphate fertilizers used in East African region because of lack of financial resources as well as phosphate ores import restrictions.

(ii) This study found relatively high U concentration in phosphate rocks and phosphate fertilizers but was unable to estimate the U resources presently available in these deposits. as more geological information would be required to do so.

(iii) This study determined the concentration of gamma emitting radionuclides from 232Th, 238U and 40K in tobacco leaves and assessed radiological risk for smokers and snuffers. However, the study did not determine the concentration of polonium- 210 (210Po) which is an alpha emitter and more damaging to smokers and suffers was not assessed.

(iv) Detailed study on bacteria abundance and diversity involving different ecological zones and different physical-chemical properties of soil should be studied.

(v) The study did not investigate the influence fertilizer derived U on crop yield and its impact on nutrients uptake by plants.

CHAPTER TWO

LITERATURE REVIEW

2.1 Rationale of the literature review

Phosphate fertilizer is a source of elemental phosphorus (P), which is one of the three essential nutrients, together with nitrogen (N) and potassium (K), that plants require in large amounts for reproduction and growth. It is crucial since it is required by plants and its deficiencies impede plant development and yields. The world population growth has intensified world initiatives to increase the use of P fertilizers in the agriculture sector to increase food production (Searchinger *et al.,* 2018).

The U contained in P fertilizers is preferentially transferred to mineral fertilizers during fertilizer production with the commonly used WPA process (Al Khaledi *et al.,* 2019) and if recovered U would most likely be recovered from the WPA (Reitsma *et al.,* 2018; Ye *et al.,* 2019). The presence of U in phosphate fertilizers, as well as their continuing application on agricultural soils, causes U dissemination in the agricultural soils (De Souza Braz *et al.,* 2021; Sun *et al.,* 2017; Kratz *et al.,* 2016).

There is no global consensus on the regulatory limit for U in P fertilizers today. A growing number of studies have confirmed global U buildup in agricultural soils and potential increase of U in surrounding ground and surface waters as a result of prolonged P fertilizer applications (Bigalke *et al.,* 2018; Takeda *et al.,* 2006). Depending on the origin of the PR, U concentrations are usually in the order of 80- 200 mg kg^{-1} for sedimentary PR, which account for approximately 80% of the PR mined today, and much lower (30 mg kg^{-1}) for PR of igneous origin, as they are mainly found in Russia and South Africa. U concentrations in PR in East Africa are known to show higher U concentrations than the world average (Haneklaus, 2021; IAEA, 2023). Although U has proven to be an environmental and a health concern there are no legal limits to the level of U in fertilizers today (Bigalke *et al.,* 2018; Takeda *et al.,* 2006). Therefore, the aim of this review is to examine the extent of U concentration in P fertilizers in the world and investigate reported U accumulations in agricultural soils in East African countries as a result of P fertilizer application.

2.2 The focus of the literature review

The focus of this literature review is based on the effect of the dissemination of U with P fertilizers on agricultural soils. The U is present as an impurity in PR, which is the raw material

used for P fertilizer production. The U in P fertilizers is then disseminated on agricultural soil with fertilizers leading to potential accumulation in the soil. The U loads in agricultural soil may cause plant uptake from the soil so that this element could in trace concentrations even enter the food chain. The availability and uptake of non-essential substances like U are influenced by the availability and uptake of important macro and micronutrients like nitrogen (N), phosphorous (P), potassium (K), and zinc (Zn), among others, by plants (Duhan *et al.,* 2023). The uptake of U by plants may pose both chemical and radiological damage to human and animal tissues (Semioshkina & Voigt, 2021; Uchida *et al.,* 2007). It is imperative to review the current state of U in P fertilizers, its dissemination in agricultural soils, its potential uptake by plants, and its possible entry into the food chain. This is very important, particularly in areas with the highest average U concentration in phosphate fertilizers, such as East Africa agricultural soils.

2.3 Countries included in this literature review

The review was conducted in 54 countries on U in P fertilizers and its reported impact on agricultural soil. The countries include 27 countries of the European Union (EU) and United Kingdom, 6 East African countries (DRC, Tanzania, Kenya, Uganda, Burundi, and Rwanda), and 21 other countries from North Africa (Algeria and Egypt), Oceania (Australia and New Zealand), Asia (Japan, Kazakhstan, Serbia, Thailand, Vietnam, Saudi Arabia, Pakistan, India, and Bangladesh), and the Americas (Mexico and Brazil). The list of countries is presented in Fig. 1.

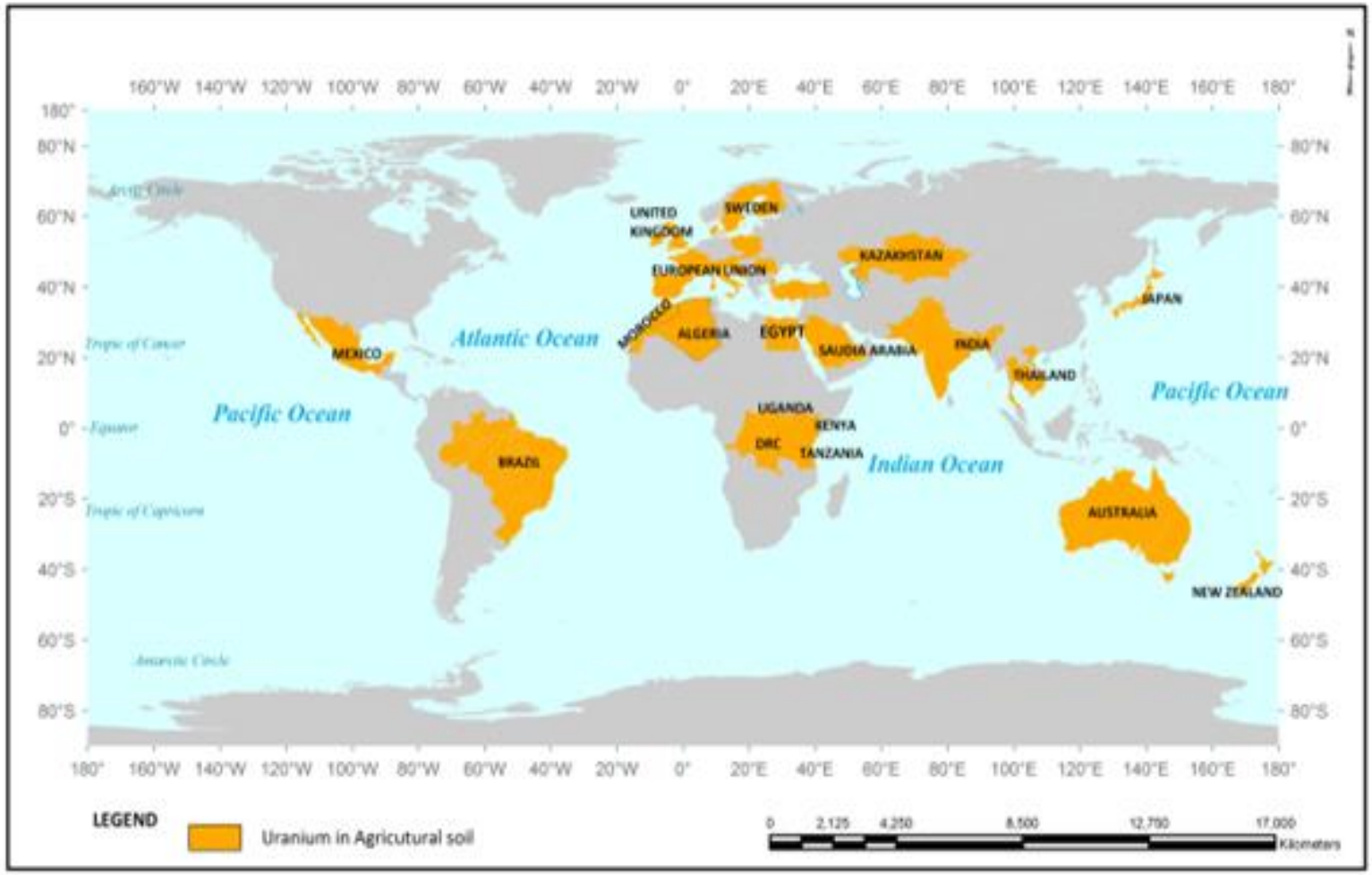

Figure 1: Countries/regions (orange) from where articles on U dissemination in agricultural soils as a result of the application of P-derived fertilizers were considered in this review

2.4 Descriptive Statistics of the Selected Articles

The distribution of articles per year relevant to the reviewed topic during the last 20 years is displayed in Fig. 3. According to descriptive statistical inferences, the number of articles published annually in 2003–2022 varied from 1 to 10 (average 2.7). The gradual change in the number of articles per year may be attributed to a lack of interest in or scientific relevance of the subject due to the lower U concentration in P fertilizers used in agricultural soils in many countries globally. Also in 2004, no relevant article was published in the selected countries for review.

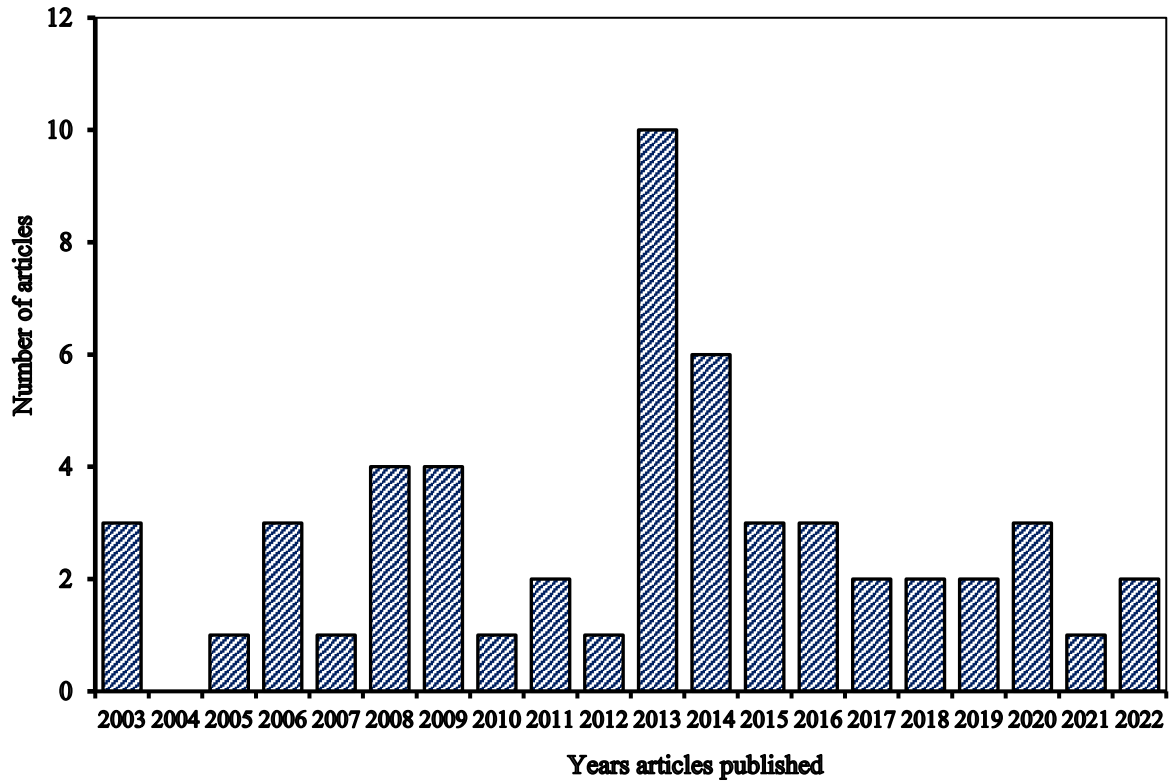

Figure 2: Articles published from 2002–2022 on U contamination in agricultural soils

From the reviewed articles, examined the trend of individual country/region article contributions were examined. The region with the most significant article contributions was Europe which is not surprising given that the EU is actively discussing the implementation of legal U limits in fertilizers. In Europe, Germany had the highest number of articles (10) followed by Switzerland (3), and Turkey (3). Also, two studies were done for all countries in the EU. Indian researchers published the second highest number of articles (10) after the EU (Bergen *et al.,* 2022; Verbeeck *et al.,* 2020).

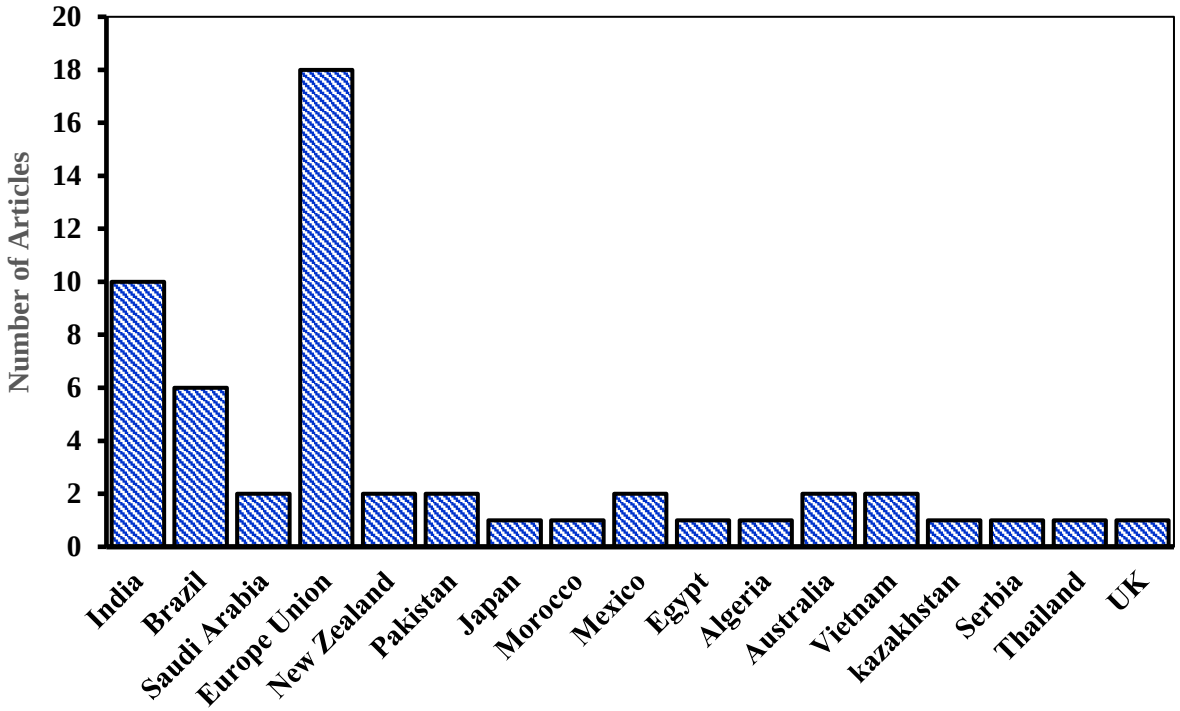

Figure 3: Number of articles published on U in agricultural soils in research in the 10 most featured countries and regions

2.5 Uranium dissemination with phosphate rocks and mineral fertilizers

Natural U (U) is present as an impurity in phosphate rocks (PR), the ore that is used for mineral fertilizer production. It is estimated that during P fertilizer production over 80% of the U contained in the PR ends up in the fertilizers (Beltrami *et al.,* 2014; Komar & Carlson, 2013; Reyes *et al.,* 2019; Wu *et al.,* 2018), with the rest transferring to the phosphogypsum mine waste tailings (Bilal *et al.,* 2023; Diwa *et al.,* 2023; Hakkar *et al.,* 2021). Most P fertilizers contain at least low U concentrations. Thus, long term applications of P fertilizers can increase U loads in agricultural soils. The concentration of U in P fertilizers depends on the type and origin of the PR used in fertilizer production and also, but to a lesser degree, the production process that decides how much of this U transfers to the final fertilizer product (Arhouni *et al.,* 2022). Besides, impurities in the used industrial sulfuric acid can introduce additional impurities. For instance, according to Kouzbour *et al.* (2019) and Kouzbour *et al.* (2019), the wet phosphoric acid (WPA) process that is used in the production of P fertilizers frequently results in a product which contains more than 80% of the U impurities originally present in the PR (Hoffmann *et al.,* 2016). In sedimentary PR, for instance, U concentrations are in the range of 80–200 mg kg^{-1} (Haneklaus, 2021). Sedimentary PR accounts for about 80% of the PR mined globally, while in igneous PR the U concentrations are typically much lower (about 30 mg kg^{-1}) (Al Khaledi *et al.,* 2019; Diwa *et al.,* 2021; Haneklaus, 2021; Ulrich *et al.,* 2014). The

application of P fertilizers can disseminate considerable amounts of U on agricultural soils that may leach from the soil to ground- and surface waters (Campos *et al.,* 2021; Haneklaus, 2021).

The applications of P fertilizers have been reported to enhances U concentrations in agricultural soils in different continents around the world. For instance in Asia (Chauhan & Chauhan, 2014; Chauhan & Kumar, 2015; Nain *et al.,* 2008; Pantelica *et al.,* 1997; Porntepkasemsan *et al.,* 2018; Pulhani *et al.,* 2005; Shang *et al.,* 2021; Yamaguchi *et al.,* 2009; Yamazaki & Geraldo, 2003; Ye *et al.,* 2019; Bergen *et al.,* 2022; Bigalke *et al.,* 2020; Sun *et al.,* 2022; Sun *et al.,* 2020; Tulsidas *et al.,* 2019), in Australia (Abraham *et al.,* 2018; Wu *et al.,* 2021), in North America (Hore-Lacy, 2016; Zielinski *et al.,* 2000), and in Africa (Ahmed & El-Arabi, 2005; Boukhenfouf & Boucenna, 2011). The concentration of radioactivity from Minjingu phosphate fertilizers from Tanzania has been reported (Banzi *et al.,* 2000; Makweba & Holm, 1993; Meza *et al.,* 2015). Minjingu P fertilizers have been reported to be used in neighboring East African countries such as Kenya, Uganda, the Democratic Republic of Congo (DRC), Burundi, Rwanda and Zambia as well so that this issue can hardly be described as a local phenomenon, but does indeed affect millions (Abuli *et al.,* 2012; Kifuko *et al.,* 2007; Ndeleko-Barasa *et al.,* 2021; Nziguheba, 2007).

Phosphate fertilizer application is regarded as a major cause of U dissemination in agricultural soils (Schnug & Lottermoser, 2013). It is no secret that radiotoxic of U is hazardous to human health and the environment (Bigalke *et al.,* 2020; De Souza Braz *et al.,* 2021; Van Dung *et al.,* 2022; Zlobina *et al.,* 2022). Yamaguchi *et al.* (2009) and (Yamazaki & Geraldo, 2003) reported for instance that agricultural soils with a long history of P fertilizer application showed elevated U concentrations if compared with unfertilized soils. Phosphate fertilizers with high U concentrations are prone to soil contamination and may influence soil-to-plant uptake (Hu *et al.,* 2020; Kratz & Schnug, 2006). Besides, P fertilizers have also been identified as a major risk for U contamination of drinking water (Kaishwa *et al.,* 2018; Ren *et al.,* 2022; Schnug & Lottermoser, 2013).

Therefore, the objective of this work is to present a comprehensive overview of current knowledge regarding the U concentration in different P fertilizers in the world and its reported distribution in agricultural soils and potential U uptake by plants. Additionally, we performed a comparison of the reported U concentrations from different countries and concludes by providing a sustainable policy recommendation.

2.5.1 Uranium in phosphate fertilizers and in agricultural soils

In this literature review, U concentrations in P fertilizers used in arable soils in different countries around the world were extensively examined and reported in comparison to the countries of East Africa. The reviewed literatures involved short and long-term field experiments where fertilized and unfertilized (control or forest) agricultural fields were compared. The comparison between U concentrations in fertilized and unfertilized soils was conducted to determine if there is a measurable statistically significant difference. Out of the 54 articles selected here, 10 data points from 35 countries (27 EU and 8 others) were plotted in box and whiskers for easy data visualization (Fig. 4), while the remaining articles data were presented using range and central tendency (mean, median, maximum and minimum). The discussion was done for continents. Almost all articles (52 out of 54 or 96%) reported that the application of P fertilizers significantly ($p < 0.05$) increases U concentrations presented as specific activity (Bq kg^{-1}) or mass fraction (mg kg^{-1} or parts per million, ppm) in countries outside of East Africa. However, two articles (about 4%) found a nonlinear relationship between U in P fertilizers and U enrichment in agricultural soils of Germany, Denmark and Greece (Servitzoglou *et al.,* 2018; Sun *et al.,* 2020). The results show that different countries have reported varying U concentrations in P fertilizers and in agricultural soils.

(i) Europe

The trace metal accumulation, in agricultural soils including U from mineral phosphate fertilizers applications in long-term field studies in 27 European union countries (Austria, Belgium, Bulgaria, Croatia, Republic of Cyprus, Czech Republic, Denmark, Estonia, Finland, France, Germany, Greece, Hungary, Ireland, Italy, Latvia, Lithuania, Luxembourg, Malta, Netherlands, Poland, Portugal, Romania, Slovakia, Slovenia, Spain and Sweden) (Bergen *et al.,* 2022). The results showed that the average U concentration in unfertilized soils ranged between 0.18–0.95 mg kg^{-1} and 0.29–4.88 mg kg^{-1} for fertilized soil. The findings showed that long-term use of mineral P fertilizer enriches agricultural soils with U. The concentration of U was attributed by application of P fertilizer which has no limitation in EU policy. There is for instance a legal limit on cadmium (Cd) concentrations in fertilizers used in the EU (Verbeeck *et al.,* 2020) and it was recommended to consider similar regulations for U (Bergen *et al.,* 2022; Suciu *et al.,* 2022; Ulrich, 2019). In Greece, Servitzoglou *et al.* (2018) investigated the U content in common phosphate fertilizers. The results showed that U ranged from 3.3–57.1 mg kg^{-1} (average 32.1 mg kg^{-1}). Also, U contained in P fertilizers did not change its concentration

in fertilized soil compared with unfertilized soils and hence did not enhance the natural radioactivity of wheat grain grown on this soil.

The U in P fertilizers and their effects on agricultural soils have been extensively studied in Germany. Ten articles have reported examining U concentrations in P fertilizers and their impact on Germany agricultural soils. Kratz *et al.* (2016) investigated U concentrations in phosphate fertilizers commonly used on German agricultural soils and found that U concentrations ranged from 14.3 to 141 mg kg^{-1}. The results further showed that all phosphate fertilizers had higher U concentrations than the limit proposed by the German Commission for the Protection of Soils (Kratz *et al.,* 2016). The article further investigated the impact of long-term (from 3–78 years) application of P derived fertilizers on U concentration in Europe using 218 soil samples. The results showed that long-term application of mineral P fertilizers enriched soils with U. The results further suggested that the concentration of U in soils in all sites were directly proportional to applied P (increase of 1 ton P ha^{-1} linearly increased the U concentration in the surface soil (0–23 cm depth) by 0.11 mg U kg^{-1}).

Schnug and Haneklaus (2014) reported that P-derived fertilizers were found to be a major source of U pollution, that U would buildup in agricultural soils over time and that the prolonged use of P fertilizers with elevated U concentration could also lead to increased U concentrations in groundwater. The article proposes the recovery of U as an environmental contaminant as well as a raw material for nuclear power plants for the generation of electricity. The authors further argued that U recovery during the fertilizer manufacturing processes can reduce environment pollution since the recovered U resources do not only reduce heavy metal loads in soils but could be used to produce greenhouse gas lean electricity. The study was complemented by Liesch *et al.* (2015) who also reported that P fertilizers are a source of U in agricultural soils. The study did, however, mention that background concentrations of U are another important factor. In addition to U buildup in agricultural soils, Campos *et al.* (2021) investigated the possibility of U mobility in vineyard soils in Germany's Rhineland-Palatinate region. The findings demonstrated that U in fertilized soils varied from 0.48 to 1.26 mg kg^{-1}, which was slightly higher than nearby non-agricultural soils (0.50 mg kg^{-1}), very homogeneous along slope positions, and slightly higher in top soils.

Sun *et al.* (2022) complemented the study by investigating long-term P fertilizer application and P fertilizer-derived U accumulation in topsoil (0–23 cm) from 1876 to the 2010s. According to the findings, the total U accumulation rates, which ranged from 2.8 to 6.1 µg U kg^{-1} yr^{-1},

were comparable to those noted forty years prior. Additional investigations of the authors, showed that, in comparison to soil from forests (the control), soil obtained from fertilizers did not show significantly enhanced U levels (Sun *et al.,* 2020). Rogasik *et al.* (2008) examined, using data from long-term field trials conducted in Germany, the relationship between P fertilizer application rates, fertilizer products, and soil parameters and fertilizer-based U accumulation in soils. The overall long-term field results demonstrated that surface soils that receive continuous mineral phosphate fertilization had greater U contents than unfertilized soils.

(ii) Australia and New Zealand

In Australia, Lottermoser (2009) conducted a study in Northern Queensland, Australia, on the concentration of U in P fertilizers and its possible accumulation in P fertilized sugarcane soils after long-term P fertilizer application. The result found that U concentrations in P fertilizers and agricultural soils ranged from 6.0–183 mg kg^{-1} and 1.3–6.3 (average 3.7) mg kg^{-1}. The U concentrations in the control (forest) soil ranged from 1.3–2.3 mg kg^{-1} (average 1.83). The study indicated that, the average U concentration of the control soils (1.83 mg kg^{-1}) was half compared to the fertilized soils (3.7 mg kg^{-1}). The results suggested that the U accumulation in sugar cane soils was influenced by the application of P fertilizers.

New Zealand uses locally available, inexpensive, reactive P fertilizers after simple beneficiation and granulation (Haneklaus, 2021). This fertilizer contains slightly higher concentrations of radioactive radionuclides when compared to other P fertilizer (Hilton *et al.,* 2010). If PR is directly applied on agricultural soils, all impurities are also disseminated with the PR. The concentration of U in P fertilizers in New Zealand ranged from 12.1 to 129.5 mg kg^{-1} while in agricultural soils concentrations of 2.1–7.1 mg U kg^{-1} were reported (Pearson *et al.,* 2019). The concentrations of U in New Zealand agricultural soils was reported to display a linear correlation to the concentration of U in the applied P fertilizers (Pearson *et al.,* 2019).

Taylor (2007) reported, soil U accumulation after long term P fertilizer application in New Zealand. The study analyzed samples that were collected and preserved from four sampling sites between 36 and 43 years ago to determine whether there was a statistically significant variation in U accumulation in the same four sites in New Zealand. The results showed that for soils collected roughly 40 years ago showed mean levels of U increases of 1.30 mg kg^{-1}, or 0.033 mg kg^{-1} annually. Therefore, agricultural soil U increase was linked to the use of P fertilizers in Australia and New Zealand (Taylor, 2007).

(iii) Middle East

The world's greatest PR deposits are in the North African and Middle Eastern belts. The belt encompasses all of North Africa as well as several Middle Eastern nations, such as Jordan and the Kingdom of Saudi Arabia. In the Kingdom of Saudi Arabia, U dissemination in agricultural soil as the result of fertilizer application has been studied. The reported U concentrations ranged from 1.9 to 315 mg kg^{-1} (average 80.4 mg kg^{-1} (Al-Eshaikh *et al.,* 2016; Alshahri & Alqahtani, 2015; Latif *et al.,* 2014). Different types of P fertilizers showed different U concentrations with triple superphosphate (TSP) and NPK fertilizers showing the highest U concentrations compared to Di-ammonium phosphate (DAP) and mono-ammonium phosphate (MAP). The use of TSP and NPK led to higher annual effective doses for farmers and members of the general public (Alshahri & Alqahtani, 2015). The study even recommended that NPK and TSP fertilizers were unsafe for use as fertilizers in agriculture in Saudi Arabia. Similarly, Latif *et al.* (2014) reported a higher U concentration for imported P fertilizers when compared to P fertilizers produced using local Saudi PR. Overall the studies indicated that the application of P fertilizers with elevated concentrations of U may significantly increase agricultural soil radioactivity and also the radioactivity of the surrounding environment. Khater and AL-Sewaidan (2008) reported U concentrations in P fertilizers that are sold commercially and made the first judgments about radiation exposure from P fertilizer use in Saudi Arabia. The results showed remarkable and wide variations in the radioactivity contents of the different phosphate fertilizer samples and aligned well with other work from Khater (Khater, 2008).

(iv) Africa

In Algeria, the U concentrations in commercial P fertilizers and the radiological impact of fertilized agricultural soils against unfertilized soils were determined using Gamma ray spectrometry. The result showed that U concentrations in P fertilizers ranged from 10.9 to 15.4 mg kg^{-1} while the U concentration in fertilized soil was reportedly 4.3 mg kg^{-1} compared to 3.8 mg kg^{-1} in virgin soil. In Egypt, Ahmed and El-Arabi (2005) investigated U concentration in P fertilizers and agricultural soils in the Qena Governorate and in Upper Egypt using gamma ray spectrometry. The results showed that U concentration in fertilizers ranged from 28. 3 to 31.2 mg kg^{-1} (average 29.6 mg kg^{-1}). The U concentration in arable soil and in Nile Island soil (control) ranged from 0.8 to 1.1 mg kg^{-1} (average 1.0 mg kg^{-1}) and 0.7 to 1.5 mg kg^{-1} (average 1.1 mg kg^{-1}). The result showed that there were no significant differences between fertilized and unfertilized soils. In east Africa, Minjingu P fertilizer is commonly used in Burundi, DRC,

Kenya, Rwanda, Tanzania and Uganda. Minjingu PR shows particularly high U concentration ranging from 200–600 mg kg^{-1} (Banzi *et al.,* 2000; Bianconi, 1987; Makweba & Holm, 1993; Mustonen & Annanmaki, 1988) and the fertilizer produced from this PR also show increased concentrations of U. However, there is scarce information on U levels from phosphate rocks, phosphate fertilizers, its uptake by selected plants and its influence in soil bacteria community in East Africa countries.

(v) Asia

In India, U concentrations in P fertilizers have been reported to range between 0.02 and 42.8 mg kg^{-1} (Chauhan *et al.,* 2013; Chauhan & Kumar, 2015; Gupta *et al.,* 2014; Hameed *et al.,* 2014). Hameed *et al.* (2014) investigated the effect of application of P fertilizers on the U concentration of highly irrigated agricultural soils in Srirangan Taluk, India where paddy, banana and sugar cane are grown. The review reported that, the U concentration in the single superphosphate was 32.1 mg kg^{-1} and in the triple superphosphate was 23.0 mg kg^{-1}. The average activity of U in P fertilized soils was 0.68 mg kg^{-1} which was about 25% higher compared with that of the control soil (0.55 mg kg^{-1}). Therefore, the application of P fertilizers in agricultural soils in India increased U concentration by 25%. According to Chauhan *et al.* (2013) the natural U concentration were below the recommended limits (370 Bq kg^{-1}) in most P fertilizers with only superphosphate fertilizers and potash fertilizers showing lower concentrations. The examined fertilizers did, however, satisfy the UNSCEAR (UNSCEAR, 2000) requirement that sets a limits of 1000 Bq kg^{-1}. India is one of the largest importers of PR and P fertilizers and the use of both has been rising over time (Mew *et al.,* 2023). The number of chemical fertilizers consumed per hectare grew from an average of 86.7 kg in 2001 to 128 kg in 2018.

Punniyakotti *et al.* (2020) investigated the U content in different types of P fertilizers marked in Villupuram District in Tamil Nadu State, India, and its influence on fertilized soils compared to that of unfertilized soils. U concentrations on fertilized and unfertilized soils ranged from 0.8 to 7.3 mg kg^{-1} (average 1.1 mg kg^{-1} and 0.8 to 8.1 mg kg^{-1}), respectively. It was observed that over 70% of the fertilized soil samples had a higher U concentration than the virgin soil samples. In the same country, Kant *et al.* (2006) investigated U concentrations in fertilizers and its potential radiological contamination in agricultural soils. Mixed soil samples from crop fields had a mean U concentration of 1.3 mg kg^{-1}, while the unfertilized soil had a concentration of 0.7 mg kg^{-1}. Ghosh *et al.* (2008) measured U concentrations in P fertilizers as well as non-

P fertilizers and found that the U concentrations ranged from 11.4 to 210.1 mg kg^{-1} (average 68.6 mg kg^{-1}) and 0.2 to 0.4 mg kg^{-1}, respectively. The P fertilizers were used on agricultural soil and the concentration of U in the fertilized soil ranged from 8.8 to 53.6 mg kg^{-1} (average 29.35 mg kg^{-1}).

In Bangladesh, the concentration of U in common P fertilizers used in agricultural soils were studied. U contents in compost, TSP, and DAP ranged from 2 to 8 mg kg^{-1}, 68 to 209 mg kg^{-1}, and 98 to 119 mg kg^{-1}, respectively (Rahman *et al.,* 2017). The study clearly shows that U concentrations are considerably higher in the three most frequently used P fertilizers in the country. However, the U containing fertilizers were not tested on agricultural farms to observe their influence to soil contamination and potential plant uptake as well.

In Pakistan, Faisalabad is a fertilizer intensive agricultural city with a population of over 6 million inhabitants. Nasim-Akhtar *et al.* (2012) examined the difference between the content of U in fertilized and unfertilized soil. The U concentration in fertilized soil ranged from 2.11–3.9 mg kg^{-1} while that of unfertilized soil was 0. 6 mg kg^{-1} (Akhtar *et al.,* 2011; Nasim-Akhtar *et al.,* 2012). The results showed that the fertilized soils showed higher U concentrations than the unfertilized soils.

In Vietnam, Nguyen *et al.* (2018) determined the gross alpha, gross beta, and activity concentration of ^{226}Ra in certain types of fertilizers typically used in the southern region of the country. The work revealed that the U concentrations in the investigated P fertilizers ranged from 0.1 to 47.9 mg kg^{-1}. The P fertilizers were subsequently tested in agricultural soils in field experiments to understand their concentration in soil and their influence on plant uptake. The results showed that the U concentrations ranged from 3.1 to 56.9 mg kg^{-1}, with mean values of 30.5 mg kg^{-1} (Nguyen *et al.,* 2021). Previous results did, however, indicate that the use of fertilizers had no significant effect on soil radioactivity (Servitzoglou *et al.,* 2018).

Yamaguchi *et al.* (2009) investigated the effects of phosphate fertilizers containing U in Japan and also assessed the effects of various agricultural practices on the U content of the soil. The results indicated that the long-term application of P fertilizers had raised the U concentrations in soil. In the surface soil of agricultural areas, U from applied fertilizers has been found to be adsorbed, precipitated, or integrated into the soil organic matter together with minerals carrying iron or aluminum rock (Yamaguchi *et al.,* 2009). In the grazing land and highland field soils, soil organic matter seemed to be a more significant U pool, but in the paddy field soils that experienced periodic shifts in redox conditions, weakly crystalline Fe minerals seemed to be a

more significant U pool (Yamaguchi *et al.,* 2009). These findings implied that the extra U was cemented to the soil particles to prevent crops from readily absorbing it (Tagami & Uchida, 2020).

(vi) Latin America

Intense fertilizer application is frequently required due to the study's findings about the effects of P fertilizer application in Mexico's agricultural sector. In the highlands of central Mexico, where corn is grown, P fertilizers are commonly used. U concentrations in P fertilizers in the region are reported to range from 50 to 200 mg·kg^{-1} depending on the origin of the PR used in their production. The U content in fertilized agricultural soils varied between 19.5 and 50.6 mg kg^{-1}, while in unfertilized soils, it ranged from 0.4 and 1.0 mg kg^{-1}. The results showed that adding P fertilizers to agricultural soils enhanced U concentrations in soils.

A study done in Brazil by de Souza Braz *et al.* (2021) on the long-term usage of P fertilizers in citrus, oil palm, and black pepper crops for 26, 10, and 5 years indicate an increase in U concentrations. Specifically, the study revealed that the concentrations of U in unfertilized soils used to grow oil palm, black pepper and citrus were 1.51 mg kg^{-1}, 1.71 mg kg^{-1} and 2.19 mg kg^{-1} respectively, while in fertilized soils the concentration of U were 2.15 mg kg^{-1}, 3.48 mg kg^{-1} and 2.99 mg kg^{-1} (de Souza Braz *et al.,* 2021). The results indicate that the concentrations of U in these crops generally, were citrus > black pepper > oil palm. This demonstrates that plants have distinct absorption processes for U. The U contents in the fertilized soils on which black pepper and oil palm was grown were slightly higher than global averages. It is noteworthy that the radioactivity concentrations were low in all crops and these crops did not pose a risk to human health. The study did, however, recommend continuous monitoring of radionuclides in agricultural soils if P fertilizers are continuously used.

Yamazaki and Geraldo (2003) investigated U concentrations in the most commonly used P fertilizers applied to Brazilian agricultural land. The results showed that U concentrations ranged from 5.2 to 54.3 mg kg^{-1}. The U concentrations were lower than other values reported in the literature. Investigations of the influence of U containing P fertilizer in agricultural soils is important to ascertain its potential entry into the food chain. For instance, Lauria *et al.* (2009) investigated U concentration in vegetables grown using different fertilizers. The study determined the presence of U and radium in chemical and organic fertilizers, as well as farm soil. U isotope uptake by vegetables grown in Rio de Janeiro under acidic and limed soils was investigated. The results indicate that U from different NPK fertilizers used to grow lettuce,

carrots, and bean crops ranged from 0.3 to 25.9 mg kg^{-1}. There was no statistically noticeable difference in U concentration in vegetables grown using conventional fertilizers compared to those using organic fertilizers.

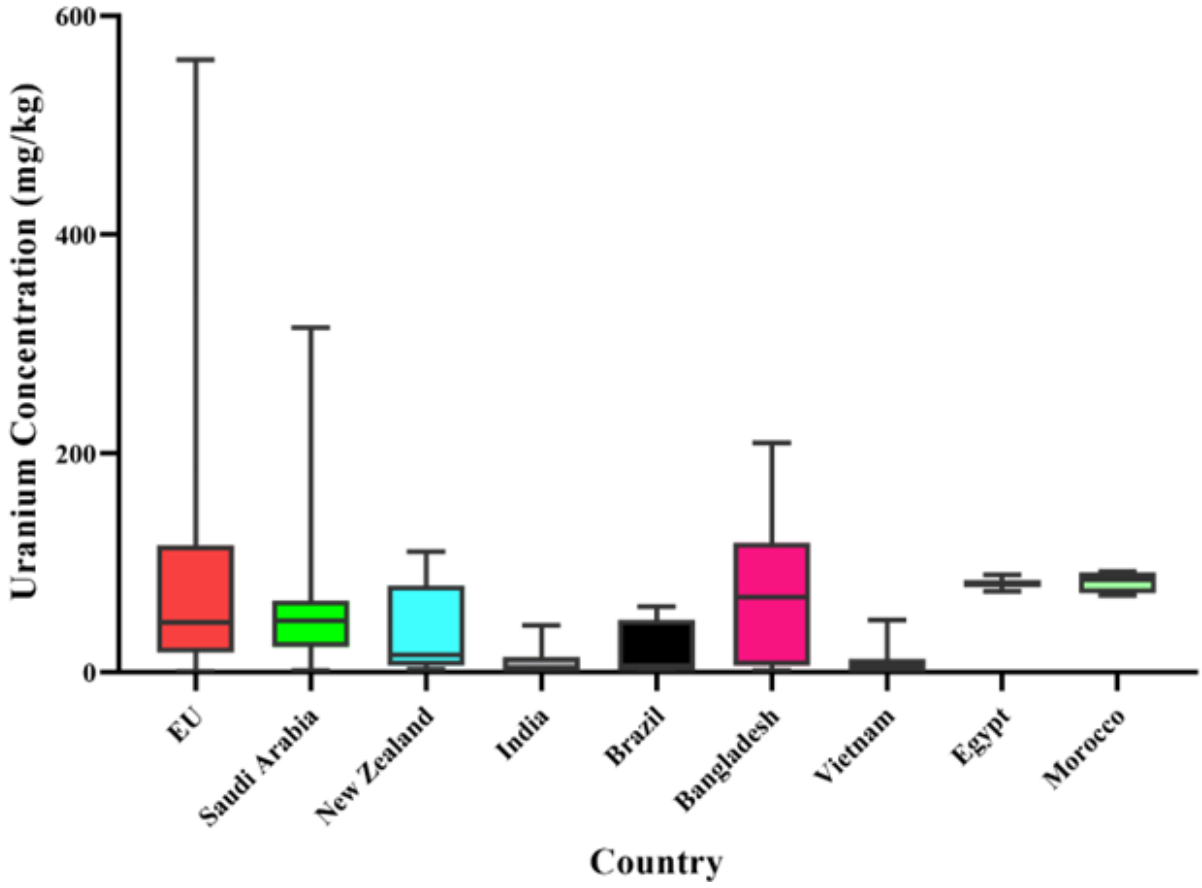

Figure 4: Box & whiskers plots of reported U in P fertilizers from 2003–2022 from peer-reviewed articles in the world, irrespective of the type of P fertilizer and composition

2.5.2 Uranium as a potential agricultural soil contaminant

The application of P fertilizers to agricultural soils results in an elevation of U concentrations in the cultivated soils. It is possible that some of the U applied to topsoil layers leaches into soil bed rocks and subsequently affects surface and ground waters (Birke *et al.,* 2009; Hoyer, 2013; Vodyanitskii, 2011). Schnug and Lottermoser (2013) indicated that U from P fertilizers applied to agricultural soils poses a potential radiological and chemical health risk to humans. However, there is scarce information on U levels from phosphate rocks, phosphate fertilizers, its uptake by selected plants and its influence in soil bacteria community in East Africa countries.

2.6 Reported uranium uptake by plants

Long-term P fertilizer application in agricultural soils increases U build up and potential plant uptake. Even relatively high U concentrations in soils does not mean that the U will be taken up by plants gown on this soil. Different types of plants have for instance different U uptake characteristics. However, the most consumed crops are of high concern, as many people may be affected no matter how small the amount of U that is taken up. For instance, wheat (*Triticum* spp.) and maize (*Zea mays*) are the most frequently produced cereal crops, feeding more than 40% of the global population (Acevedo *et al.,* 2018). Monitoring of these most consumed cereal crops is worldwide is important to ensure that the large population is not exposed to U contamination. For example, in Serbia, maize and wheat crops are the two most important cereal crops where P fertilizer application is intensive (Stanojković *et al.,* 2012). Stojanović *et al.* (2013) reported the assessed U uptake from P fertilized soils into maize and wheat crops. The results showed that relatively high U concentrations in fertilized soils did not enhance U uptake by wheat and maize crops, which was attributed to both high pH (above 6) and organic matter which made U unavailable for plant uptake (Stojanović *et al.,* 2013).

U in soils has no nutritional value to plants but may pose radiological, chemical, and biotoxicity risks to humans and the environment once it gets into the food chain (Gupta *et al.,* 2020). Studies for naturally occurring radionuclides like U from soils to plants have been reported (Gupta *et al.,* 2020). In the environment, the concentrations of U are enhanced by anthropogenic activities like mining activities and the application of phosphate ore-derived fertilizers in agricultural soils (Charro & Moyano, 2017).

In Thailand, U uptake by common foods and commercial crops such as paddy, maize, sugar cane, and cassava was studied (Porntepkasemsan *et al.,* 2018). The application of P fertilizers with high U concentrations raised the concentration in agricultural soils and affected plant absorption. The results showed increased concentration of U in cassava > paddy field > maize > sugar cane > natural forest (control site sample). Several studies have reported a linear relationship between added U in soils during fertilizer application and the potential uptake by plants. It should, however, be noted that, the difference between U concentrations in arable land and grassland has been an important indicator of whether accumulation is possible or not. There are essentially two major possible scenarios when P fertilizers containing U are applied in agricultural soils, either they accumulate in the soils or they leach to the ground and surface waters. However, few studies have been conducted to evaluate U leaching and speciation in

soil solutions, as well as transport in arable soils. Bigalke *et al.* (2018) in Switzerland examined the speciation of U in arable soil solutions and probable U leaching along preferred flow pathways in Switzerland. The result showed that, when compared to manure application, P fertilizer application resulted in the largest U input on agricultural soils and effects on leaching and accumulation of U to greater carbonate ions.

The U radionuclide's interaction between soils and plants depends on soil properties such as nutrient concentrations, pH, mineral composition, and organic matter content (Stojanović *et al.,* 2013). The mechanism of U radionuclide transfers from soils to crops depends on various factors, such as the physicochemical properties of the U radionuclides of interest, the type of crops, the soil management practices, the wastes, and the time after the fallout and soil properties (Tagami & Uchida, 2020). The uptake of U radionuclides from soils to plants follows the natural process of essential nutrient uptake (Semioshkina & Voigt, 2021). The analogous chemical properties between essential and nonessential elements can enter the plant's body through similar mechanisms. The factors influencing soil to plant nutrient uptake are agroclimatic conditions, pH, soil properties, organic matter content, and soil microbial activity (Gupta & Walther, 2019). However, there is little information reporting the transfer of radionuclide from the agricultural soils to plants. Therefore, there is a need to investigate the transfer of radionuclides from agricultural soils to plants.

Moreover, U is not an essential element for crops, but it has a detrimental effect on ecosystems, including the health of human beings due to its carcinogenic effect (Sheppard *et al.,* 2005). The U possesses chemical and radioactivity toxicity to organisms in soils. Several studies have revealed the U uptake by plants from the soils, which is eventually transferred into the food chains (Guillén & Gómez-Polo, 2020; Ratnikov *et al.,* 2020). For example, Mohammed and Nkuba (2014) indicated that, there is higher natural radioactivity and radiation transfer from soils into maize and bean crops grown near Minjingu phosphate mines (Leonid & Najat, 2014). Therefore, this, indicates that crops have the ability to take up U from the contaminated soils. Also, other studies reported that, sorghum, rice, cassava, sunflower, and vegetables have the ability to increase U uptake from the soils grown in U mineralized sites which has health effects on human consuming foodstuffs with elevated U concentrations (Amaral *et al.,* 2005).

U in agricultural soils can enter plants through roots and get distributed in different parts of the plants. Different plant species have different U uptake behavior. The behavior of U in plants follows the behavior of calcium (Asaduzzaman *et al.,* 2015). It is reported that the uptake and

distribution of U radionuclides in plants depends on the type of crop, soil pH, exchangeable Ca and K and organic matter contents, physicochemical properties of the radionuclide, fertilizer application, irrigation, plowing, liming, and climate conditions (Asaduzzaman *et al.,* 2015; Laurette *et al.,* 2012).

Little information is reported about the U uptake from contaminated soils into *Nicotiana tabacum*, which sparked interest in investigating the uptake of the U from the soils into *Nicotiana tabacum*. Also, little information exists on the U uptake from the soils into the crops in the East Africa region, and it has become important to investigate the concentrations of radionuclides uptake by *Nicotiana tabacum*. To our understanding, research studies on screening potential plants and organic materials for removing U toxicity levels from soils have not been studied in the East Africa Region. Therefore, there is a need to conduct screening for potential crops and organic materials to be aware of the concentrations of the radionuclides transferred from agricultural soils to plants.

2.7 The fate of Uranium added to agricultural soils

The assessment of the fate of U in soil is challenging because of the complex nature of its interaction in soil (Chen *et al.,* 2021; Takeda *et al.,* 2006; Yamaguchi *et al.,* 2009). In the soil, fertilizer derived U has been reported to leach out because U is mobile in surface soils as a uranyl complex depending on prevailing oxidation-reduction (redox potential) and acid–base reactions (pH) (Bigalke *et al.,* 2018; Liesch *et al.,* 2015; Schnug & Lottermoser, 2013). Therefore, the fate of U in the surface and sub-surface soil environments depends entirely on U valency states. The U in soils is fairly soluble as a U(VI) cation or (UO_2^{2+}) in an oxidizing environment (Gavrilescu *et al.,* 2009; Guillén & Gómez-Polo, 2020; Gupta *et al.,* 2020). In soil with a reducing environment and a pH between 4.0 and 7.5, U is immobile because UO_2 mostly binds to oxidized organic matter (OM) and precipitates on the surface of iron/aluminum (Fe/Al) minerals that are weakly or non-crystalline (Tagami & Uchida, 2020). Iron/Aluminum are regarded as significant U sinks (Takeda *et al.,* 2006; Yamaguchi *et al.,* 2009). The U is mostly found as UO_2^{2+} in oxidative environments, and as the pH goes up, more of it sticks to negatively charged soil particles (Campos *et al.,* 2021). However, the creation of soluble and negatively charged compounds with substances like carbonate improves the mobility of U (Echevarria *et al.,* 2001).

2.8 Potential regulatory limits of U in P fertilizers

In the environment, U is regarded as a chemical and radioactive element. Along with Arsenic and Chromium, it is one of the three most dangerous naturally occurring pollutants in groundwater (Bigalke *et al.,* 2017). There is no regulatory limit on the concentration of U in P fertilizers worldwide. However, some countries are considering setting limits. The European Union has set a regulatory limit for Cd concentration, while U is not yet considered. In Germany, the German Commission for the Protection of Soils advocated setting a limit for the amount of U in fertilizers at 50 mg per kg P_2O_5 (Kratz *et al.,* 2016), or 167 mg kg^{-1} for fertilizers with a 30% P_2O_5 concentration. The potential existence of a regulatory limit for U in P fertilizer has not encouraged P fertilizer producers to recover U from WPA in the past (Haneklaus *et al.,* 2017) but increasing U prizes could make *U recovery from phosphates great again*, as speculated by Steiner *et al.* (2020). According to United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) the average amount of U in soil is reported to vary from 0.05 to 10 mg kg^{-1}. However, depending on the geology of the place, the concentrations can be as high as 200 mg kg^{-1} (UNSCEAR, 2000).

This literature review has evaluated 232 articles from throughout the world that reported on the significant quantity of U in P fertilizers, their reported accumulation in agricultural soils, and some that explained the possible uptake of U by plants worldwide. The review also examined the fate of U in soil. The results showed that P fertilizers used in different countries around the world contained varying concentrations of U. However, P fertilizer from Minjingu P fertilizers had a significantly higher U concentration comparable to that of commercial low-grade U mines as they are for instance operated in Namibia. Studies have clearly shown that long-term application of mineral P fertilizers increases U concentrations in agricultural soils. While there are substantial restrictions on the content of Cd in P fertilizer, there are presently none for U.

CHAPTER THREE

MATERIAL AND METHODS

3.1 Uranium in phosphate rocks and mineral fertilizers applied to agricultural soils

3.1.1 The study area

Uranium concentrations from four major phosphate rocks and four commonly used phosphate fertilizers in East Africa were determined. The study involved phosphate rocks collected from the following deposits: Matongo (Burundi), Minjingu (Tanzania), Mrima Hill (Kenya), and Sukulu Hill (Uganda). In addition, commonly used phosphate fertilizers were collected from traders in Arusha and Dar es Salaam (Tanzania), Bujumbura (Burundi), Kampala (Uganda), Kigali (Rwanda), and Nairobi (Kenya). The locations of the sample sides are depicted in Fig. 5.

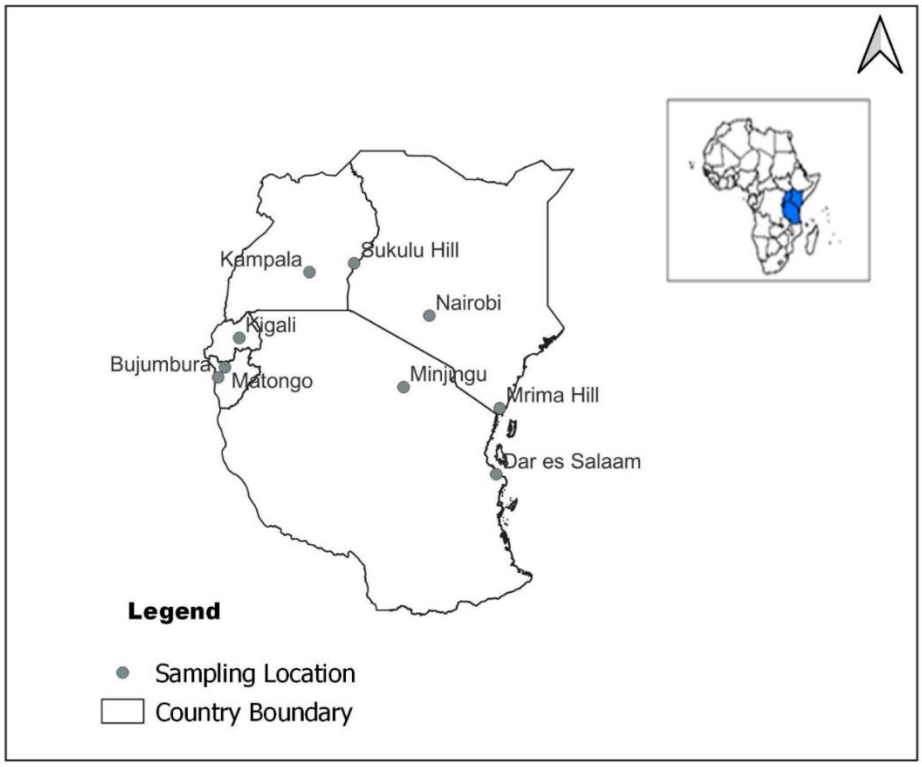

Figure 5: Phosphate rock and mineral fertilizer sampling sites of this study

3.1.2 Sample collection and preparation

The phosphate rocks were collected from five randomly selected sampling points at about 2 m depth measured from the surface from each phosphate deposit with the aim of getting representative samples and minimizing potential influences from weathering or vegetations.

The five samples were placed in a clean polythene sheet, and a composite sample of about 1 kg mass was drawn and carried to the Tanzania atomic Energy Commission (TAEC) laboratory for further processing. In the laboratory, the samples were crushed, ground, homogenized, and sieved using a 60 μm diameter sieve. Three replicates were drawn, packed, and labeled in clean airtight zip-lock polythene bags for further laboratory processing. The phosphate fertilizer samples were collected from fertilizer dealers in each country. In addition, 5 kg of phosphate fertilizers were collected from the capital city of each country making a total of 25 samples.

The collected phosphate fertilizers from each sampling site were again combined to get four representative samples. Two fertilizers are locally produced in northern Tanzania and used in East Africa region for fertilizing the agricultural soils. These are Minjingu organic hyper phosphate (MOHP) with the following sale specifications: P_2O_5:28%, MgO: 2.5%, CaO: 36%, and Minjingu Nafaka Plus (NPS) with the following sale specifications: N: 9%, P_2O_5: 16%, K_2O: 6%, CaO: 25%, S: 5%, MgO: 2%, Zn: 0.5%, and B: 0.1%. Diammonium phosphate (DAP) (18:46:00) and nitrogen phosphorus and potassium (NPK) (23:23:00) are also locally used but are mostly imported from Morocco and Saudi Arabia. Samples of these imported fertilizers were also analyzed for determing of the levels of U. The phosphate rocks and phosphate fertilizer samples were oven-dried at 100 °C to remove moisture to a constant weight. The phosphate rocks were then crushed, milled, and sieved using a 60 μm sieve size, while the phosphate fertilizers were milled using a RETSCH Cross Beater mill machine and sieved using a 63 μm sieve.

An aliquot of 4 g was subsampled for energy-dispersive X-ray fluorescence (EDXRF) measurements at TAEC laboratory. The subsample was processed by mixing with a 0.9 g clean wax binder (FluXANA CEREOXR, Germany). The mixture was poured into an 80 mL polytetrafluoroethylene grinding bowl with 3 agate grinding balls of 20 mm diameter each, inserted into a Pulverisette 6 planetary mono mill R (Fritsch GmbH, Germany), and set to 150 rpm for 120 s to achieve a fine milling powder that can then be used in the subsequent experiments (Mwalongo & Mohammed, 2013). The homogenized mixture was poured into a cylindrical pressing die with an inside diameter of 32 mm. The mixture was pressed using a manual hydraulic press, and tablet-like pellets were formed for further analysis.

Plate 1: A bench top Spectro Xepos ED-XRF at Tanzania Atomic Energy Commission

3.1.3 Performance Check of Energy Dispersive X-ray Fluorescent system

The EDXRF measurement system was calibrated using multi-elemental standard reference material from the National Institute of Science and Technology (NIST 2711a—Trace Metals in Soil) and re-confirmed by using International Atomic Energy Agency (IAEA) certified reference materials (IAEA 312 and 314) of a similar matrix.

3.1.4 Statistical analyses

The statistical data analysis was performed using STATISTICA 8th Edition software (StatSoft, Inc., Tulsa, OK, USA). U concentrations were analyzed based on the interactions among the phosphate rocks and phosphate fertilizers and each factor individually. The two-way ANOVA statistical analyses were performed with treatments being phosphate rock sources as the main factor and phosphate fertilizer type as a subfactor. For the isolation of interaction and individual effects of sites (East African countries), phosphate rock, and phosphate fertilizers, a post hoc Least significant difference multiple comparison test was used due to a higher degree of freedom (five countries × four fertilizer types = 20 for the measured variables). The significance threshold was set at $p = 0.05$ and $p = 0.001$ for high significance.

3.2 Influence of phosphate fertilizers on the radioactivity of agricultural soils and tobacco plants in east Africa

3.2.1 Experimental design and location of study areas

Three experiments were monitored, which included one-year experiments, ten years of guided experimental farms monitored by the Tobacco Research Institute of Tanzania (TORITA) and smallholder farmer's practices' farms. The one-year experiment was a completely randomized design with four treatments. One district in each East African country (Kenya, Tanzania and Uganda) was selected for smallholder farmer's practice.

The tobacco plant was chosen to assess soil-to-plant transfer of U as one of radionuclides based on its hyper-accumulative property. Furthermore, tobacco was grown using NPK fertilizers rich in the macro-nutrients: nitrogen (N), phosphorous (P), and potassium (K) (NPK 10:18:24). The *YaraMila* Compound (YC) $N_{10}P_{18}K_{24}$ fertilizer containing three component nutrients: N, P, and K, while the YaraMila Blended (YB) $N_{10}P_{18}K_{24}$ fertilizer was blended mechanically to supply balanced nutrients of N, P and K individually (Lisuma *et al.,* 2022). In addition, the Golden Leaf Tobacco (GLT) $N_{10}P_{18}K_{24}$ fertilizer was blended from Minjingu organic hyper-phosphate (MoHP) rock. Therefore, these mineral fertilizers can contain elevated concentrations of naturally occurring radionuclides associated with the phosphate ore processed for its P content.

Three studies were carried out. First, a one year-control experiment with tobacco grown in plots was performed at the TORITA in western Tanzania in the 2021/2022 cropping season. Second, a long-term experiment was carried out in different tobacco fields in Tanzania under TORITA management for the previous ten years (from 2012/2013 to 2021/2022) and with annual tobacco cropping. Lastly, an assessment of radioactivity levels in small hold farmers tobacco farms in the East Africa region which was carried out in Urambo farm (Tanzania), Migori (Kenya), and Kanungu (Uganda).

(i) The One year-controlled experiment

The experimental plots for the one-year controlled experiment were set up at the TORITA experimental farm in Tumbi (Tabora region) during the 2021/2022 cropping season. The tobacco plant *Nicotiana tabacum L.* (variety K326) was planted, and different brands of NPK fertilizers (NPK 10:18:24) were applied. The fertilizers used in this experiment were YC, YB, and GLT. These fertilizers were selected for all the experiments because they are commonly used by tobacco growers. The first two fertilizers were imported from Sweden, while the last

one is locally manufactured at the Minjingu Mines and Fertilizers Limited company in Tanzania, calcium ammonium nitrate (CAN - 27%) was applied to the experimental plots after two weeks as it is usually done in tobacco plantations. This fertilizer is based on synthetic ammonia and is practically radioactivity free. The three phosphate fertilizers (YC, YB, GLT) were analyzed for gamma-emitting radionuclides at the gamma-ray spectrometry laboratory at the Department of technical support and radiation protection service of the Tanzania Atomic Energy Commission (TAEC) Northern zone.

The experimental plots were established in agricultural soils that had not been cultivated and grown any crop for about ten years (a forest before) and received no influence from phosphate fertilization. The site was prepared by clearing the forest, and the soil was prepared using hand hoes. Tobacco seeds (variety K326) were sown in four seedbeds with four treatments (without fertilizer, fertilized with YC, YB and GLT), each with 6 m x 6 m size after application of 1 kg of respective NPK fertilizers per treatment as is the standard procedure (Lisuma *et al.,* 2022).

Seedling seedbeds and experimental plots were prepared for one cropping season. Each experimental plot was replicated three times, thus obtaining a total of 12 plots (three plots per treatment). A control plot (no fertilizer) was also included in the experiment. The ridge and fallow field plots were 6 m x 6 m in size, with a 2 m spacing between blocks and a 1 m spacing within plots. A 3-meter-wide pathway was established around the entire experimental area to reduce potential outside influences. After 10 - 14 days, germinated tobacco seedlings were transplanted to new seedbeds and then re-transplanted into the experimental plots 60 days after sowing with an intra-row spacing of 0.5 m x 0.5 m.

Seven days after transplanting, the seedling's growth was boosted with application of the three fertilizer brands YC, YB, and GLT in the respective plots, each with three replications, as shown in Plate 2. Twenty-one days after seedling transplanting, the CAN fertilizer (27% N) was applied at the rate of 30 grams per plant and agronomic practices were managed throughout the experiment. The tobacco leaves were sampled 12-15 weeks after topping.

The lower, middle, and upper-end leaves were sampled and mixed to get a composite sample for each sampling point for radionuclide measurement. In addition, a composite soil sample, combining soil from the ridge and fallow, was taken from each plot after reaping the leaves. The leaves and soil samples were packed in labeled plastic bags and transported to the laboratory for further preparation and analysis.

Plate 2: A tobacco experimental plot showing replication of the controlled experiment

(ii) Ten-years semi-controlled experiment

The experimental sites used in this investigation were tobacco farms managed under the supervision of TORITA for the previous ten years (ten cropping seasons). One of the core functions of TORITA is to perform long term monitoring of the practices and performances of tobacco plantations in Tanzania. The tobacco farms selected had applied over the previous decade NPK (10:18:24) fertilizers, including NPK derived from YB and GLT, during the year 2021/2022. Therefore, farming practices for ten years duration were slightly different to the one year-control experiments. The tobacco farms investigated were located in 7 districts in Tanzania: Urambo (Tabora), Kahama (Shinyanga), Bukombe (Geita), Biharamulo (Kagera), Kakonko (Kigoma), Manyoni (Singida), and Namtumbo (Ruvuma) as shown in Fig. 6.

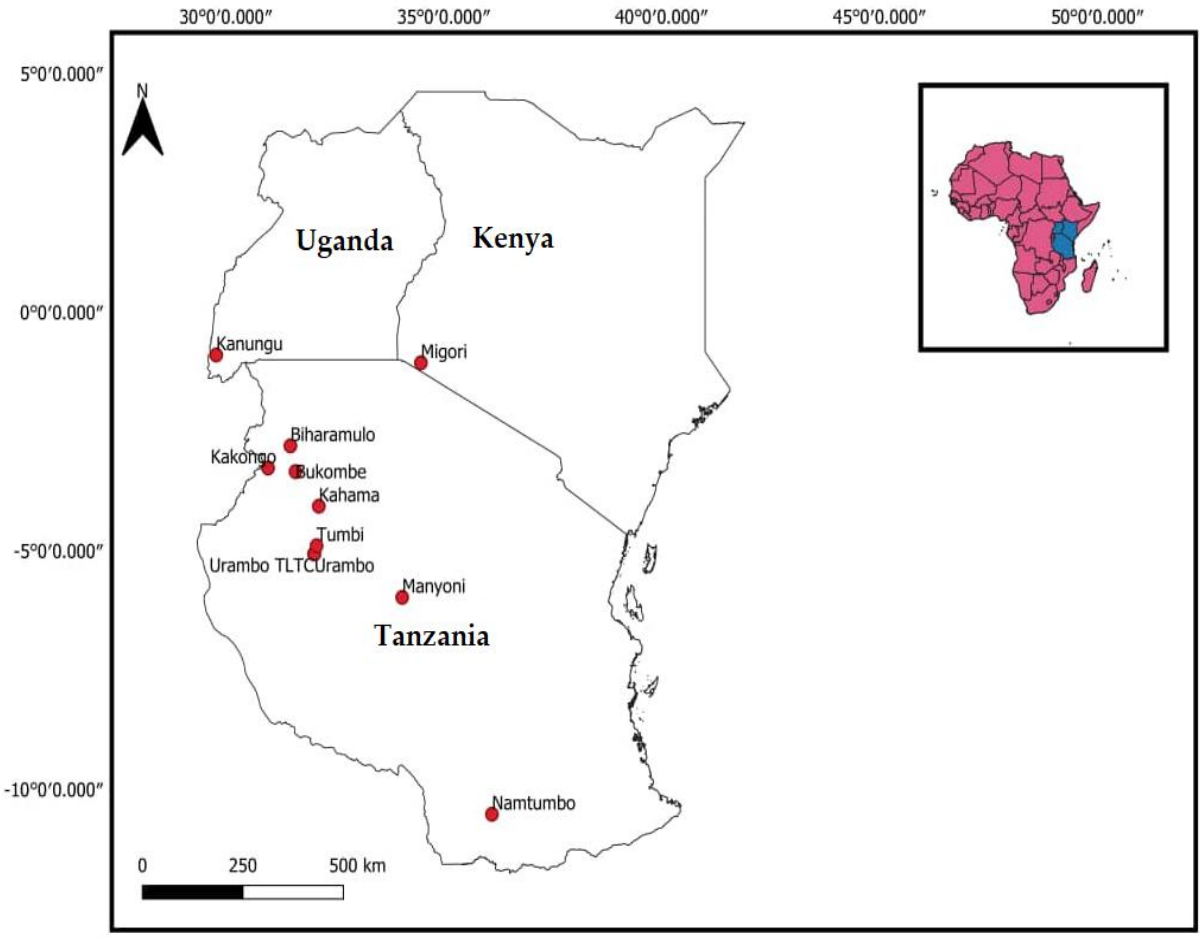

Figure 6: Map of Kenya, Tanzania and Uganda showing the soil and tobacco leaf sampling sites

(iii) Radioactivity levels in selected tobacco farms in Kenya, Tanzania, and Uganda

This experiment was conducted in the three countries of East African regions and it was carried out to assess the radioactivity levels in the selected long-standing tobacco farms tobacco growing districts/counties in Kenya (Migori), Tanzania (Urambo farm), and Uganda (Kanungu). Tobacco cultivation practices and NPK fertilizers application in the smallholder farmers field could not be controlled and might be slightly different from those conducted in the one-year and ten-years experimental farms. Those experimental sites in the three East African countries tobacco growing tobacco were aimed at giving a picture of the levels of the radioactivity and radiation exposure in the commercial tobacco farms. The districts/counties in those East Africa Countries were chosen to represent leading tobacco producers. For instance, Urambo district in Tanzania accounts for over 50% of Tanzania's tobacco production Lisuma *et al.* (2022). Migori in Kenya accounts for over 80% of tobacco production in Kenya (James, 2019) and Kanungu district in Uganda, is responsible for about 20% of the Uganda tobacco production (Chune *et al.,* 2022; Wanyonyi *et al.,* 2020). Soil and tobacco leaf samples were

collected from these farms and analyzed for the levels of radionuclides by using gamma spectrometry at the TAEC laboratory in Arusha.

3.2.2 Sampling and sample preparation

Soil samples from the three sites in the three East African countries at a depth of 0 - 30 cm were collected using an open-end stainless-steel hand-held Auger corer (70 mm diameter) manufactured by Thermo-Fisher Scientific, Germany. Ten soil core samples were collected between the tobacco plants in the plots at 0 – 30 cm depth from each sampling site to make a layer of composite samples of about 500 g each. Large plant debris and stones were removed, and soil samples were subsequently homogenized. Also, ten (10) tobacco leaves from the lower, middle, and upper portions of the tobacco plants were collected during the harvesting of the 2021/2022 cropping season from each plot, as described above. In the laboratory, the tobacco leaves were thoroughly washed with deionized water. Soil and tobacco samples were oven-dried at 80°C until a constant weight was reached. The dried soil and tobacco leaves were ground and sieved using a 200 µm mesh screen, transferred to metal canisters, and sealed airtight. The average weight of the sample materials in the canisters for gamma spectrometry was approximately 250 g for soil samples and 186 g for tobacco leave samples. The samples were stored for 30 days in the laboratory to acquire secular equilibrium before measurements.

3.2.3 Sample analysis

(i) Analysis of radionuclides

Before analysis procedures, analytical control check was carried out. The calibration of the spectrometry equipment was evaluated for accuracy and reliability. It was assessed using IAEA certified reference material (CRM) IAEA soil 375. The performance was assessed based using Equation 1 (IAEA, 2009).

$$\text{Relative biase (\%)} = \frac{\text{Laboratory Measured Value} - \text{IAEA reference Value}}{\text{IAEA reference Value}} \times 100 \quad \ldots\ldots\ldots\ldots\ldots\ldots (1)$$

After 30 days of storage to allow for the formation of secular radioactive equilibrium of ^{238}U and ^{232}Th with their respective short-lived progenies, while ^{40}K was measured from the 1640 keV distinct peak, the samples were counted for 36 000 s on a large volume high purity germanium (HPGe) gamma spectrometer (manufactured by ORTEC), inside lead shielding and connected to a multichannel analyzer (Plate 3). The Gamma Vision® software was used in spectrum analysis (ORTEC, 2020). The energy and efficiency calibration of the TAEC gamma

spectrometry system was performed for lower and higher energies using multi-nuclide calibration sources type CBSS 2, Certificate No. 1035-Se-40202-17, Serial No. 270217-1621040, traceable to the Czech Metrology Institute. The average concentrations of radionuclides ^{212}Pb (238.6 keV) and ^{228}Ac (911.1 keV) were used to calculate the concentration of ^{232}Th in the sample, while the peak gamma energies of radionuclides ^{214}Bi (764.8 keV) were used to calculate the concentration of ^{238}U. The gamma lines 609.3 keV and 1120 keV for the ^{238}U series, and 583.2 keV, 727.3 keV, and 795 keV for the ^{232}Th series were removed from the estimation of radionuclide activity concentration because they are impacted by coincidence summation (Newman *et al.,* 2008; Polouckova, 2021). The activity concentration for each radionuclide of interest was calculated using Equation 2 (Kovacs *et al.,* 2017).

$$A = \frac{N}{P_\gamma \times \varepsilon \times W} \quad \ldots (2)$$

Where: A is the activity concentration of radionuclide per unit mass of dry weight of the sample in Bq kg^{-1}, N is the net counts per second (cps), P_γ is a gamma-line emission probability of a particular radionuclide, ε is the gamma line emission intensity (%), and W (kg) is the weight of the sample.

Plate 3: The HPGe Ortec Gamma ray Spectrometry System

(ii) Analysis of Physical-chemical characteristics

The physical-chemical characteristics of the soil samples collected, namely cation exchange capacity (CEC), organic matter content (OM), soil pH, and soil textures (sand, silt, and clay), were determined in using standard scientific methods described by Moberg (2001).

(iii) Calculation of radiological parameters

The radioanalytical results of the three primordial radionuclides (^{232}Th, ^{238}U, ^{40}K) were used to quantify several radiological hazard indexes that are briefly introduced in the following subchapters.

Radium equivalent activity in soil

The radium equivalent (Ra_{eq}) for soil samples was calculated using Equation 3 (Tufail, 2012):

$$Ra_{eq} = A_U + 1.43A_{Th} + 0.077A_K \quad \text{..... (1)}$$

Where A_u, A_{Th}, and A_k are the activity concentrations (Bq kg^{-1}) of radionuclides in soil samples.

External and internal hazard indexes

The H_{ex} is a single index that measures the exposure to gamma radiation from ^{232}Th, ^{238}U, and ^{40}K radionuclides that are found naturally in soil. Using Equation 4, the H_{ex} was computed (Akpanowo *et al.,* 2020). It is noteworthy that the soil is commonly used as a construction material to build earthen houses in the study area.

$$H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad \text{..... (2)}$$

Where A_u, A_{Th}, and A_k are the activity concentrations (Bq kg^{-1}) of radionuclides in soil samples.

The H_{in} was calculated with Equation 5:

$$H_{in} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad \text{.....(3)}$$

Radioactivity level index for farmers

The radioactivity level index, noted by Iγ, was estimated using Equation 6 (Purnama & Damayanti, 2020).

$$I_{\gamma} = \frac{A_U}{150} + \frac{A_{Th}}{100} + \frac{A_U}{1500} \quad \text{..... (4)}$$

A_U, A_{Th}, A_K are the activity concentrations (Bq kg^{-1}) of ^{238}U, ^{232}Th and ^{40}K, in soils respectively. Radiation safety is considered acceptable if $I\gamma \leq 1$.

Absorbed dose rate

The absorbed dose rate by human beings (D) from exposure to external gamma radiation as the result of radioactive decay of ^{238}U, ^{232}Th and ^{40}K contained in soils was estimated using the activity-to-dose conversion factors (from Bq kg^{-1} in soils to nGy h^{-1} absorbed dose) and the activity concentrations A_U, A_{Th}, A_K in the soil samples was determined by gamma spectrometry. The activity-to-dose conversion factors used were 0.462 (^{238}U), 0.621 (^{232}Th), and 0.0417 (^{40}K). The absorbed dose rate was calculated using Equation 7 (UNSCEAR, 2020):

$$D\ (nGyh^{-1}) = 0.462\ A_U + 0.462A_{Th} + 0.0417A_K \text{..... (5)}$$

Where D is the absorbed dose rate (nGy h^{-1}) and A_U, A_{Th}, A_K are the activity concentrations (Bq kg^{-1}) of ^{238}U, ^{232}Th and ^{40}K in soil samples.

Annual effective dose for tobacco farmers from tobacco leaves

The annual effective dose to tobacco farm workers outdoors (AED_o) and indoors (AED_i) were calculated using Equation 8 and 9 (Akpanowo *et al.,* 2020; Taskin *et al.,* 2009):

Outdoor:

$$AEDo((\mu Sv\ y^{-1}) = D(nGy\ h^{-1}) \times 8760\ h\ y^{-1} \times 0.8\ \times 0.7(Sv\ Gy^{-1}) \times 10^{-3} \text{..... (6)}$$

Indoor:

$$AEDi((\mu Sv\ y^{-1}) = D(nGy\ h^{-1}) \times 8760\ h\ y^{-1} v \times 0.2\ \times 0.7(Sv\ Gy^{-1})\ \times 10^{-3}\ \text{.....(7)}$$

The total annual effective dose AED is the sum of AED_o and AED_i. It should not exceed an annual dose limit of 1 mSv y^{-1} to be considered acceptable for the general public.

Annual effective dose from tobacco inhalation

Tobacco can enter the human lungs through either snuffing or smoking. A distinction was made as follows: the calculation of radiation dose from tobacco snuffing assumes that the gamma-emitting radionuclides present in tobacco reach the lungs. In contrast, in the case of smoking, only a fraction of the present radionuclides reaches the lungs, and other fractions escape through the smoke main stream into the air or are retained by the cigarette filter.

The annual effective dose of tobacco leaf consumption by snuffing was calculated using Equation 10.

$$E_{snuffing} = A(Bq\ kg^{-1}) \times M(kg) \times D_{cf}(Sv\ Bq^{-1}) \quad \text{.....(8)}$$

Where A is the activity concentration of individual radionuclides in tobacco leaves, M is the total mass of the snuffed tobacco in one year, and D_{cf} is the activity-to-dose conversion coefficient (Sv Bq^{-1}).

The annual effective dose inhaled from tobacco/cigarette smoke was calculated using Equation 11.

$$E_{smoking} = 0.75 \times 0.5 \times A(Bqkg^{-1}) \times M(kg\ y^{-1}) \times D_{cf}(Sv\ Bq^{-1}) \quad \text{.....(9)}$$

Excess lifetime cancer risk (ELCR) for tobacco consumers

Adoption of the linear non-threshold (LNT) dose-response model proposed by the International Commission for Radiological Protection (ICRP) implies that low radiation doses from naturally occurring radioactive materials can potentially induce cancer in exposed individuals. Therefore, the risk of developing cancer from low doses over the person's lifetime, or the excess lifetime cancer risk (ELCR) for smoking and snuffing were calculated using Equation 12 and Equation 13 (Kadhim & Ridha, 2019).

$$ELCR_{\ smoking} = A_{ls} \times E_{smoking} \times 0.05Sv^{-1} \quad \text{..... (10)}$$

$$ELCR_{snuffing} = A_{ls} \times E_{snuffing} \times 0.05Sv^{-1} \quad \text{..... (11)}$$

where ELCR is the excess lifetime cancer risk, A_{ls} is the average life expectancy (75 years) for snuffers and smokers, 0.05 Sv^{-1} is the mortality risk coefficient for inhalation, and $E_{smoking}$ and $E_{snuffing}$ are the annual effective doses from smoking and snuffing, respectively.

Therefore, the ELCR from snuffed and smoked tobacco and tobacco products was calculated using an A_{ls} value of 65 years, the average life expectancy across the three countries.

(iv) Statistical analyses

Following descriptive statistical evaluation using the Shapiro-Wilk normality test, the distribution of all data was determined to be normally distributed ($p < 0.05$). Then, further analysis was done using Fisher Least Significant difference (LSD) to show the differences between the means. Finally, the degree of the relationships between the radionuclide's concentration (^{238}U, ^{232}Th and ^{40}K) and soil physical-chemical properties were determined using Pearson's correlation analysis ($p < 0.05$). The statistical analyses were performed using the STATISTICA (8th Edition) software.

3.3 Effects of fertilizers derived U on soil bacteria diversity

3.3.1 Location of the study area and collection of soil samples

The experiment was carried out in Tumbi, Tabora region, Mid-Western Tanzania (GPS coordinates 5º30'44.4" S, 32º40'7.40" E; 1,151 m a.s.l.) in sandy loam soil with very low organic carbon (0.16%) and low calcium (0.10 C mol (+) kg^{-1}) during the 2021-22 cropping season. This soil had not been used for agriculture for more than 10 years. The mean air temperature and rainfall recorded during the ten years cropping season were 27ºC and 952 mm, respectively. Each treatment corresponded to a plot size of 6 m x 6 m, with a spacing of 0.75 m between ridges and 0.30 m between plants. The DKC 8053 variety of maize plants was sown on November 15, 2021. Four treatments were applied on maize (M), each one replicated three times, Table 1 shows the treatments and composition of fertilizers used in the experiments.

Table 1: Composition of fertilizers used in the experiment along with measured U mass concentration in IAEA-Soil 7 CRM and certified reference value

Treatment code	Fertilizer/Treatment component	Fertilizer formulation	Uranium Concentration (mg kg^{-1})
T1	Not fertilized (NF + maize)	No fertilizer used	-
T2	YaraMila Cereal (YC+ maize)	23N:10P:5K+3S+2MgO+0.3Zn	38.84 ± 1.24
T3	Nafaka Plus (NP+ maize)	N: 9%, P_2O_5: 16%, K_2O: 6%, CaO: 25%, S; 5%, MgO; 2 %, Zn:0.5%, B: 0.1%)	147.65 ± 8.61
T4	Minjingu powder (MP + maize)	P_2O_5: 28%	159.67 ± 10.48

Measured U mass concentration

Reference Material	U Concentration (mg kg^{-1})		Bias (%)
	Measured	**Reference**	
IAEA Soil 7	2.78 ± 0.43	2.6 ± 0.6	5.8%
NIST 2711a	3.10	2.96	5.1%

The (NF + M) treatment plot, designated as the control plot, did not receive any fertilizer. Maize plants in the other three treatments received 5 g fertilizer per plant in three equal applications at 2, 4, and 8 weeks after sowing. Sixteen soil samples were collected between the plants using an Auger sampler inserted in the soil to the maize root zone depth of 30 cm below the surface. Rhizosphere soil (soil firmly adhering to the maize roots) was removed using forceps, sum dried and sieved through a 2 mm sieve. Sieved rhizosphere soil samples were used for extraction of bacteria genomic DNA for evaluation of the bacterial diversity in soil, determination of soil pH, and U.

3.3.2 Bacteria DNA extraction and gene sequencing

About 0.25 g of each rhizosphere soil sample was used for DNA extraction using DNeasy® PowerSoil® Kit (Qiagen, Hilden, Germany) according to the manufacturer's instructions (QIAGEN, 2021). The extracted DNA was quantified using Qubit™ 3.0 Fluorometer using dsDNA High Sensitivity assay. The DNA was visualized through 1.0% agarose gel

electrophoresis. Extracted DNA was stored at -20ºC until use (Lear *et al.,* 2018). The purified DNA was transported on dry ice to Inqaba Biotec™, in Pretoria, South Africa, for bacteriota analysis. There, a sequencing run number 220803 was performed. The V3–V4 hypervariable regions of the 16S rRNA gene were amplified during the PCR step using the universal primer pair of 341F forward primer (5'-CCTACGGGNGGCWGCAG-3') and 785R reverse primer (5'-GACTACHVGGGTATCTAATCC-3') for each sample. The amplicons were gel-purified, end-repaired and Illumina TruSeq adapters were ligated to each amplicon. Then, samples were individually indexed, and another bead-based purification was performed. Following quantification and equimolar pooling, amplicons were sequenced on Illumina's MiSeq using MiSeq reagent kit v3 (600 cycles) (Guenay-Greunke *et al.,* 2021).

3.3.3 Bioinformatics analyses for the soil bacteriota composition

As low-quality scores were observed for the reverse end reads, the bacteriota analyses were performed using mainly forward reads. The factor that contributed to the low-quality scores of reverse-end reads could be attributed to the exhausted primers, dNTPs, and other mixture reagents towards the end of sequencing runs. The analysis of demultiplexed forward-end 16S rRNA gene reads was performed based on DADA2 (ver. 1.24.0) (Callahan *et al.,* 2016) in R software (ver. 4.2.1) (R Core Team, 2022). The DADA2 pipeline includes trimming and filtering of quality reads, dereplicating sequences, learning error rates, generating an abundance table of amplicon sequence variants (ASVs), removing chimeric sequences using the "bimera *de novo*" method, taxonomic assignment and classification of the amplicon sequence variants (ASVs) using the SILVA reference (ver. 138) database (Quast *et al.,* 2013). About 378 160 forward reads generated from 4 bacterial DNA samples, one from each treatment, were pre-processed in the DADA2 pipeline by removing low-quality reads using the truncated length set at 240 bp and left trimming at less than 20 bp. Reads were further filtered to remove reads with ambiguous bases as previously reported by Lisuma *et al.* (2020). The DADA2 pipeline detected 6.8% of the relative abundance in all reads as chimeric, which was then removed from the datasets. As a result, the final ASV abundance Table contained 297 865 high-quality non-chimeric reads from the 4 DNA samples (Callahan *et al.,* 2016). The soil pH was determined using standard measuring techniques with a calibrated pH meter (Orion VersASTAR pro) and soil- water suspension (1:2.5) (FAO, 2022).

3.3.4 Determination of U concentration in the soil

The U concentration in rhizosphere soil samples was determined using Energy Dispersive X-ray Fluorescence (EDXRF).The performance of energy dispersive x-ray fluorescence (XRF) spectrometer and accuracy of results were validated through using the commercially available standard reference material offered by the National Institute of Science and Technology (NIST 2711—Montana Soil, Moderately Elevated Trace Element Concentrations) and reconfirmed using certified reference International Atomic Energy Agency (IAEA) certified reference material (IAEA Soil 7 – Trace Elements in Soil) materials (IAEA, 2000; NIST, 2011). Results indicate that the overall bias was within ± 5.8%, which indicates that the accuracy of the measurement method was suitable (<10%) for the purpose of this research.

3.3.5 Statistical analyses

Statistical analyses of the association between soil pH and U concentration were done using one-way analysis of variance (ANOVA), and significant means were compared using Fisher LSD at $p < 0.05$ using STATISTICA ver. 8.0 (Stat-Soft, Inc., Tulsa, OK, USA). Normalization of the sequence '*phyloseq*' was performed on four treatment soil samples by sub-sampling, including only highly abundant samples with means greater than 1000 reads was performed as previously described by Lisuma *et al.* (2020). About 110 Operational taxonomic unit (OTUs) were removed as they were not present in any sample after random sub-sampling. The alpha index groups (Observed, Chao1, and Shannon) were used to estimate species richness and evenness at the phylum level in four treatment plots (unfertilized NF, and fertilized with YC, NP, and MP) were calculated using *phyloseq* package ver. 1.40.0 (McMurdie & Holmes, 2013) and the visualization of unique or common and shared ASVs at phylum, order, and class levels across four treatment plots using *MicEco* package ver. 0.9.18 (Russel & Oksanen, 2021) (ver. 4.2.1) (R Core Team, 2022).

3.4 Effects of *Eucalyptus globulus spp maidenii* organic matter and kaolin in soils

Materials used in this study include the maize seeds type DKC 8053 commercially available in the market, the *Eucalyptus globulus spp maidenii* bark which were left over from timber industry, three fertilizer types namely Nafaka plus, YaraMila cereal and Minjingu powder Kaolin was collected from Pugu hills, Kisarawe district, Pwani region, Tanzania. The Pot experiment and field experiments were organized at Nelson Mandela African Institution of Science and Technology (NM-AIST) and Tabora, respectively. The field experiment was

performed first to assess nominal U uptake by maize after treatment with three phosphate fertilizers containing different concentration of U. The pot experiment was performed using the U contained fertilizers used in field experiments in additional to kaolin and *Eucalyptus globulus ssp maideii.* The field and pot experiments were analysed using inductively coupled mass Spectrometry (ICP-MS). Standard reference materials for geological and biological samples were used to check the quality of the analytical results.

3.4.1 Description of the study area and experimental Setting

The pot and field experiments were conducted during 2022/2023 cropping seasons. The pot experiment was arranged at M-AIST Nambala ward, in Arusha region, located at S 03° 23' 59" and E36° 47' 47", while the field experiment was conducted at Tobacco Research Institute (TORITA) experimental site, located at S 05° 04′ 02.5″ and E 032°06′ 09.7″, Tumbi ward in Tabora region. The avoid influence of residual fertilized induced condition, the field experiment at TORITA was set at the farms which had not been farmed for over ten years. The Pot experiment was watered using distilled water and the field experiment received enough rain from sowing to harvest. The distilled water was produced from wall-mounted PURELAB. The site is characterized by an annual rainfall and air temperature of 950 mm and 26°C, respectively. The water for pot experiment was purified by reverse osmosis utilizing a Purelab Ultra system (type 2004, made in Germany) available at Tanzania Atomic Energy Commission (TAEC).

3.4.2 Performance quality control of ICP-MS

A total of 91 soil and maize compartments (root, stem, leaf and grain) from pot and field experiment were prepared and analyzed for this study. The 0.1 g of the aliquot was digested in an acid mixture (3 mL HNO_3 ultrapure 60% plus 5 ml HF 40%), The solution after ambient cooling was transferred quantitatively in 50 ml flasks and delivered to the desired volume with high purity water. The measurements were conducted using the XSERIES 2 ICP-MS (Thermo Fischer Scientific) at Czech Technical University. The calibration of the ICP- MS was validated using standard reference materials.

3.4.3 Characterization of materials

(i) Analytical quality control

To ensure of accuracy of U and calcium in soil and maize plant compartment (root, stem, leaf and grain), certified reference materials with known concentrations of elements of interest were measured and the results compared with the reference values. The standard reference materials (SRM) measured were from United States National Institute of Standards and Technology (NIST) and US Geological Survey (USGS) for trace elements in spinach leaves (NIST 1570a) and Columbia Rive Basalt (BCR-2); respectively. The data quality was evaluated based on relative bias (%) calculated using eq.2. Blind standard reference materials (SRMs) were sent with samples to laboratory that performed the measurement of samples to check the accuracy and hence reliability of the measured results. The laboratory result showed that the reference value and laboratory reported value were in good agreement (within ± 5% relative bias). The result is shown on Table 2.

Table 2: Measurement of U (mg kg^{-1}) in standard reference materials

Reference material	Reference mass fraction	Measured mass fraction	Relative bias (%)
NIST 1570a	0.155 ± 0.023	0.149 ± 0.023	-4.02%
US GS(BCR-2)	1. 683 ± 0.020	1.76 ± 0.05	4.6%

(i) Soil physical chemical properties

The soil for pot and field experiment were characterized for physical-chemical characteristics of soil as shown on Table 3.

Table 3: Physicochemical characteristic of soil used in this study

Sample Code	Soil Texture	pH	Organic Carbon (g/kg)	CEC (mmol+/kg)
Namb	Clay	7.58 ± 0.78	15.55 ± 0.79	39.71.74 ± 0.78
S00	Sandy Loam	7.31 ± 0.07	13.73 ± 0.25	25.43 ± 0.34
S01	Sand Loamy	7.08 ± 0.02	12.37 ± 0.15	17.87 ± 0.98
S02	Sand Loamy	7.38 ± 0.18	12.30 ± 0.43	24.57 ± 1.56
S03	Sandy Loam	7.45 ± 0.25	12.13 ± 0.12	36.37 2.24

Maize variety

The maize seed variety used in the field and pot experiment was a hybrid seed type DKC 8053 supplied by Monsanto seed Company Limited. The seed was certified by Tanzania Official Seed Certification Institute (TOSCI). The seed variety was chosen because is commercially, commonly used by many farmers and has been reported to have better yield (Lisuma *et al.,* 2020). The seed was also reported to be popularly grown in eastern and southern Africa (Mubanga, 2018).

(ii) Fertilizer characterization

Three fertilizer containing nitrogen (N), phosphorous (P) and potassium (K) compound were used in the pot and field experiment namely YaraMila cereal (YC) (N: 23%, P_2O_5: 10%, K_2O: 5%, S: 3%, MgO: 2%, Zn: 0.3%), Minjingu Nafaka plus (NP) (N: 9%, P_2O_5: 16%, K_2O: 6%, CaO: 25%, S: 5%, MgO: 2%, Zn: 0.5%, and B: 0.1%) and MP (phosphate ore) (P_2O_5:28%). The fertilizers used in the field and pot experiment were characterized for U concentration as shown in Table 4.

Table 4: Type of fertilizers used in the experiment

Fertilizer/Treatment component	Fertilizer formulation	Uranium Concentration (mg kg^{-1})
Not fertilized (NF) (Control)	No fertilizer used	-
YaraMila Cereal (YC)	23N:10P:5K+3S+2MgO+0.3Zn	38.84 ± 1.24
Nafaka Plus (NP)	N: 9%, P_2O_5: 16%, K_2O: 6%, CaO: 25%, S; 5%, MgO; 2 %, Zn:0.5%, B: 0.1%)	147.65 ± 8.61
MP (MP)	P_2O_5: 28%	57± 10.48

(iii) Experimental design

The experiments comprised of the field experiment which was set in 2021/2022 cropping season at TORITA experimental site using complete randomized block design (CRBD), and screen house experiment arranged at NM-AIST in Arusha region.

(iv) Field experiments

In the field experiment, three types of fertilizers were used namely, Nafaka Plus (NP), YaraMila Cereal (YC) and Minjingu powder (MP). In the screen house pot experiments, three fertilizers were used (NP, YC and MP) plus kaolin and organic matter derived from *E. globules ssp maidenii*. Also, the results for pot experiment are also presented. In the field experiment, maize crop was grown in ridges similar to farmers' practices. The size of each experimental plot was 6 m x 6 m and ridge spacing of 75 cm which led to 4 ridges per plot, with 16 ridges and 30 cm plant spacing making a total of 16 plant population (64 plants per plot). Fifteen grams (15 g) of fertilizers per plant was applied during the first split two weeks after sowing, 4 weeks after seedlings emergence and in the third week fertilizers were applied before tasseling (total of 45 g of fertilizer). Also, a control (unfertilized soil) to monitor any potential variation in the treatments was included in the experiment (Table 5).

Table 5: Experimental design on allocation of field treatments

S/No	Treatments
1.	T0 = Control (unfertilized)
2.	T1= Nafaka Plus fertilizer + Maize
3.	T2 = YaraMila Cereal fertilizer + Maize
4.	T3 = Minjingu Powder fertilizer + maize

The maize experimental plots for control (unfertilized), minjingu powder, Yaramila cereal and nafaka plus treatments.

(v) Screen house experimental design

A completely randomized block design (CRBD) was used to set up the experiment, and it was replicated three times. The experimental setup comprised of the control (unfertilized) soil where maize crop was grown without application of fertilizers. The second set of layouts comprised of three pots which were treated with three phosphate fertilizers, namely Minjingu Nafaka Plus (NP), YaraMila Cereal (YC) and Minjingu Powder (MP). The third set comprised of NP, YC and MP fertilizers, each fertilizer was mixed with kaolin. The fourth set of experiment comprised the same three fertilizers (NP, MP and YC) mixed with *Eucalyptus globules ssp maidenii* powder. The fifth set comprised of the same three fertilizers (NP, MP and YC) each mixed with *Eucalyptus globules ssp maidenii* and kaolin. The treatments are schematically shown on Table 6.

Table 6: Treatments applied to the field experiment

S/No	Treatment
0	T0 = Absolute Control (no fertilizer)
1	T1 = Nafaka plus (NP)
2	T2 = YaraMila Cereal (YC)
3	T3 = Minjingu Powder (MP)
4	T1 = Nafaka plus + Kaolin (NP + K)
5	T2 = YaraMila Cereal + Kaolin (YC +K)
6	T3 = Minjingu Powder + Kaolin (MP+K)
7	T1 = Nafaka plus + *Eucalyptus Globulus ssp Maidenii* (NP + E)
8	T2 = YaraMila Cereal + *Eucalyptus Globulus ssp Maidenii* (YC + E)
9	T3 =Minjingu Powder + *Eucalyptus Globulus ssp Maidenii* (MP + E)
10	T1 = Nafaka plus + Kaolin + *Eucalyptus Globulus ssp Maidenii* (NP + K+E)
11	T2 = YaraMila Cereal + Kaolin + *Eucalyptus Globulus ssp Maidenii* (YC + K+E)
12	T3 = Minjingu Powder + Kaolin + *Eucalyptus Globulus ssp Maidenii* (MP + K+E)

The maize (*Zea Mays)* seeds variety DKC 8053 were sown and 15 g of fertilizers mixed with 50 g of both kaolin and blue gum (*Eucalyptus globulus ssp maidenii) were* applied two weeks after seedling mergence. Frequent weeding was done to keep the experimental plots practically weed-free for the bulk of the plant growth period; the same treatment was administered four weeks following planting, and a third application was made two weeks before tussling.

(vi) Sampling and sample preparation

The soil from each pot was sampled from the surface to 30 cm depth using soil auger (76 cm diameter) made by Thermo-Fisher Scientific, Germany. The samples were subsampled for physicochemical and U analysis using standard scientific methods described by Moberg (2001). Five soil samples from the field were also, sampled from each treatment poured in clean well polyethylene plastic and representative sample of 1kg was drawn, placed in labelled plastic bag with sample tag written using a graphite pencil and the bag was labeled using a permanent marker pen. The sample was transferred to Sokoine University of Agriculture (SUA), soil science laboratory for further preparation and analysis.

The plant compartments (roots, steam, leaf and grain) were equally harvested from the same points where the soil was drawn. The bulk plant samples were composited packed in clean well labelled sampling bag with a sample tag inside. The samples were taken to the laboratory for pretreatments and treatments. Maize compartments were soaked into distilled water and washed to eliminate any foreign material, soil and dust. A care was taken for roots, soaked in water for three hours and completely washed to ensure no contamination of the root samples with soil is almost negligible. The samples were oven dried in clean and dust free environment at 105°C until a constant weight was reached. A plant compartment was weighed and placed in a dry crucible and placed in muffle furnace at 350°C temperature for 5 hours to form whitish brown colored ashes. The ashes sample was weighed, with an inside sample label and packed in an airtight well labelled sampling bag to prevent moisture. The ash was performed in order to concentrated U and Ca by reducing the volume of maize compartment with high water content.

(vii) Calculation of Concentration ratios

Equation 14 was used to calculate the Concentration ratios from soil to plant compartments.

$$\text{Concentration ratio(CR)} = \frac{\text{Dry weight concentration in plant(mg/kg or Bq/kg)}}{\text{Dry weight concentration in soil}(^{\text{mg}}/_{\text{kg}} \text{ or Bq/kg})} \quad \ldots\ldots\ldots\ldots\ldots\ldots \textbf{(}12)$$

3.4.4 Data analysis

Following a descriptive statistical review utilizing the Shapiro- Wilk normality test, the distribution of all data was judged to be normally distributed ($p < 0.05$). The Fisher's Least Significant Difference (LSD) test was then used to demonstrate the differences between the means. STATISTICA (8^{th} Edition) software was used for the statistical analyses.

CHAPTER FOUR

RESULTS AND DISCUSSION

4.1 Uranium concentrations in phosphate rocks and phosphate fertilizers in East Africa

4.1.1 Performance check of EDXRF analytical system

The result of the performance of the EDXRF system is shown on Table 7. The validation process aimed at confirming the fit for the purpose of measuring U concentrations in unknown field samples. The ratio of the laboratory-measured value to the certified value ranged from 0.968 to 1.032 and was thus within ± 3%. The detection limit for the EDXRF technique for elemental U was 1.41 ± 0.07 mg kg^{-1}.

Table 7: Measurements of the standard reference materials

Reference Material	U Concentration (mg kg^{-1})		
	Certified Value	Measured Value	Bias (%)
NIST 2711A	3.10	3.20	+ 3%
IAEA-312	16.5	15.97	- 3%
IAEA-314	56.8	55.7	- 2%

4.1.2 Uranium concentrations in major phosphate rocks in East Africa

The concentration of U measured in phosphate rocks from East Africa agricultural soils is shown in Table 8. The results of the U concentration vary significantly among the East African countries and are generally (except for the samples from Mrima Hill, Kenya) high to very high when compared with U concentrations of phosphate rock deposits around the world reported by Haneklaus (2021). The highest U concentration of 631.6 ± 2.5 mg kg^{-1} was recorded at Matongo phosphate rock deposit in Burundi, and the lowest U concentration (10.7 ± 0.2 mg kg^{-1}) was recorded at the Mrima Hill phosphate rock deposit in Kenya. The Minjingu phosphate rock deposit in Tanzania showed a U concentration of 446.1 ± 0.4 mg kg^{-1}, and the Sukulu Hill deposit (Uganda) showed a U concentration of 120.6 ± 0.3 mg kg^{-1}. It is noteworthy that the naturally occurring concentration of U in the earth crust is estimated to be between 1.4 and 2.7 mg kg^{-1} (Earth, 2006; Haynes *et al.,* 2016) and U mines in Namibia on the other side of the continent commercially process ores with low U concentrations (100–400 mg kg^{-1}) (WNA, 2016).

Table 8: Uranium concentration in major phosphate rock deposits in East Africa

Name of the phosphate rock deposit	Deposit Type	Country	Conc. of U (mg kg^{-1})
Matongo	Igneous	Burundi	631.6 ± 2.5
Minjingu	Sedimentary	Tanzania	446.1 ± 0.4
Sukulu Hill	Igneous	Uganda	120.6 ± 0.3
Mrima Hill	Igneous	Kenya	10.7 ± 0.2

Since phosphorus is the primary element for which phosphate rock is mined, the quality of the phosphate rock is usually classified by its P_2O_5 concentration. Phosphate rock with P_2O_5 concentration of 12–16% is considered lower grade and 17–25% medium grade, and in high-grade ores, the P_2O_5 concentration is above 26% (Boujlel *et al.,* 2019). Based on this classification, the analyzed samples from the Sukulu hill and the Minjingu deposit can be considered as high-grade phosphate rocks, while the samples from the Matongo deposit can be considered a medium-grade phosphate rock and samples from the Mrima hill deposit a low-grade phosphate rock (Table 9).

The relatively high U concentration at the Matongo phosphate rock deposit can be attributed to the syenite complex formation that contains thorium-U-potassium anomalies. It was earlier found that the deposit has high impurities that do not support using the raw material for the production of superphosphate fertilizer (Van Straaten, 2002). This can be expressed through the CaO to P_2O_5 ratio which is 0.17. In this study, Matongo phosphate rock was found to contain a P_2O_5 content of 17.65%, which is higher than the 0–15% and 11–13% P_2O_5 content previously reported by Songore (1991) and Van den Berghe (1995). The difference may be attributed to different sampling strategies and variations of P_2O_5 concentration within the deposit. Furthermore, Matongo phosphate rock has the lowest MgO (0.34 ± 0.08%) but a substantial K_2O (1.60 ± 0.03%) content. Matongo phosphate ore is a low-grade phosphate ore whose development could nonetheless be interesting if not only its P_2O_5 content, but the other valuable materials are considered for recovery.

The Minjingu phosphate rock deposit also contained high concentrations of U that are usually attributed to the ores with high organic matter content (Szilas, 2002). The Minjingu phosphate rock deposit is a layered phosphate deposit comprising of remaining organic matter and dead animals sedimented in a paleo-rift valley environment (Schluter Thomas, 1997). Our study also observed that Minjingu phosphate rock has a high P_2O_5 content (> 30%) and a relatively high CaO to P_2O_5 ratio of 1.51. The Minjingu phosphate rock deposit had a MgO concentration of 4.58 ± 0.04% and a K_2O concentration reaching 1.95 ± 0.01%.

The Sukulu Hill deposit had an average U concentration of 120.6 mg kg^{-1} and the second highest P_2O_5 concentration of 30.57%. It is an alkaline igneous carbonatite phosphate rock deposit used to produce phosphate fertilizer in Uganda through the wet phosphoric acid process. The produced fertilizer is mostly used in Uganda (Kisitu, 1991; Nakasango Proscovia, 2021) and not exported. The phosphate rock has a relatively low MgO content and CaO/P_2O_5 ratio making wet phosphoric acid processing possible. In addition, the Sukulu Hill phosphate rock deposit has low reactivity that could be attributed to a relatively high iron oxide content, so that the material is not feasible for direct application (Butegwa *et al.,* 1995). Samples from the Mrima Hill deposit had the lowest U concentrations (10.7 ± 0.2 mg kg^{-1}) and a very low P_2O_5 content of 3.5 ± 0.2 mg kg^{-1} which raises the question if they should be considered a phosphate rock deposit at all. As a result of the low P_2O_5 content, the deposit is presently not mined. It might eventually be developed for its MgO content (9.39 ± 0.20%) rather than the traces of P_2O_5.

The selected macronutrient content (P_2O_5 and K_2O and MgO, CaO) of the common phosphate fertilizers used in East Africa were assessed and are shown in Table 9. Appropriate supply of nutrients is one of the important factors to assess the quality of the fertilizer supplied to farmers for meeting soil requirements and improving yield. Assessing the content of macronutrients such as P and K expressed as P_2O_5 and K_2O, respectively are essential. Depending on soil conditions, crop types, and other agronomical factors, the fertilizers used usually include other secondary macronutrient oxides in the form of CaO and MgO. During manufacturing, fertilizers are produced by either using compound processes where NPK are homogeneously mixed in one granule or bulk blending where the nutrients are in separate granules (Morari *et al.,* 2011).

The average macronutrients for the four common phosphate fertilizers (DAP: 18:46:00; MOHP: P_2O_5: 28%, MgO: 2.5%, CaO: 36%; NPS: 9:16:06, and NPK: 23:23:00) used in East Africa countries agricultural soils was assessed. The measured results were compared with the manufacturer's declared value on the label. In DAP, the average P_2O_5 was 34.61 ± 2.91% compared to 36% provided by the manufacturer. The K_2O was not detected in DAP samples (not present in fertilizer formulation), and the CaO average concentration was 25.89 ± 2.95%.

The average P_2O_5 concentration for MOHP was 26.47 ± 1.19% compared to 28% specified by the manufacturer. This difference is within the tolerable limit of 1.1% set by the Kenya Bureau of Standards (KeBS, 2018). The measured MOHP CaO concentration (25.75%) was compared

with the 36% quoted by the manufacturer. Our assessment suggests that it was overdeclared by about 28.4%, and the concentration of MgO was 1.84 ± 0.03% compared to the manufacturer's quoted value that was 2.5%. This first analysis indicates that the macronutrient may be overstated by as much as 26%, but more systematic studies would be needed to get a clearer picture.

The NPS fertilizer average P_2O_5 measured was 14.83 ± 0.25% compared with 16% stated by the manufacturer. The result is within the tolerable limits specified by East African Authorities. The average concentration of MgO and CaO were 1.73 ± 0.16% and 34.63 ± 1.6%, respectively. Our results are in agreement with data published by Szilas (2002) who observed that the MgO and CaO content varied from 0.17 to 4.05% and 28.91 to 50.72%, respectively. The K_2O was 4.4% compared with the manufacturer's quoted value (6%); Szilas (2002) reported the K_2O to range from 0.1 to 2.59%, which implies that the NPS K_2O was overdeclared by 26.7%. The NPK average P_2O_5 was 21.12 ± 0.32% compared with the manufacturer's quoted value of 23%, which was within the recommended tolerable standards.

The average concentration for macronutrient oxides, P_2O_5, and K_2O in all phosphate fertilizers ranged from 14.83 ± 0.25% to 34.61 ± 2.91%. The macronutrients oxide MgO and CaO ranged from 1.73 ± 0.16% and 1.84 ± 0.03%. These results indicate that fertilizer manufacturer's declared nutrients did not ascertain 100% matching with the fertilizer formulation shown on the manufactured fertilizer labels. Some nutrient formulations were within or not within the tolerance specification given by the East African fertilizer standards regulatory bodies. Overall, the reported major nutrients complied with the East African countries' fertilizer standards. The calculated ratio of CaO/P_2O_5 ranged from 0.74 to 1.6, which is within the acceptable fertilizer value in agriculture (Kawatra and Carlson, 2013). Although the phosphate fertilizers manufactured from Minjingu phosphate rock have higher U concentrations, the quality of the fertilizers is still good and meets the set standard (Table 9).

Table 9: Chemical composition of major phosphate rocks and common phosphate fertilizers in East Africa

Name of deposit	P_2O_5 (%)	K_2O (%)	CaO (%)	MgO (%)	CaO/ P_2O_5
Sukulu Hill	30.57 ± 0.07	0.32 ± 0.01	39.56 ± 1.30	0.63 ± 0.08	1.29
Minjingu	34.23 ± 0.30	1.95 ±0.01	51.81 ± 0.41	4.58 ± 0.35	1.51
Matongo	17.65 ± 0.26	1.60 ± 0.03	13.02 ± 0.30	0.34 ± 0.08	0.74
Mrima hill	3.5 ± 0.01	ND	0.61 ± 0.02	9.39 ± 0.20	0.17
Name of the phosphate fertilizer					
DAP	34.61 ± 2.91	ND	25.89 ± 2.95	1.75 ± 0.25	0.74
MOHP	26.47 ± 1.19	ND	25.75 ± 2.40	1.84 ± 0.03	1.39
NPS	14.83 ± 0.25	4.41 ± 0.33	34.63 ± 2.50	1.73 ± 0.16	2.34
NPK	21.12 ± 0.32	ND	24.32 ± 1.57	1.74 ± 0.32	1.61

ND: Not detected

4.1.3 Uranium concentration in major phosphate fertilizers used in East Africa

The measured U concentrations in the phosphate fertilizers are shown in Table10. Tanzania had 226.48 ± 13.81 mg kg^{-1} the highest U concentration in this study followed by Kenya with 187.07 ± 11.64 mg kg^{-1}, Rwanda with 174.71 ± 16.72 mg kg^{-1}, Uganda with 152.63 ± 11.58 mg kg−1, and Burundi with 136.37 ± 11.67 mg kg^{-1}.

The higher U concentration reported in Tanzania can largely be attributed to the common use of Minjingu phosphate rock as a raw material in fertilizer production (Banzi *et al.,* 2000; Makweba & Holm, 1993; Meza *et al.,* 2015) or even in direct application after simple beneficiation (Kalala & Semoka, 2010; Kifuko *et al.,* 2007; Mnkeni *et al.,* 1991; Szilas *et al.,* 2008). The MOHP and Nafaka Plus, a NPS fertilizer produced from Minjingu phosphate rock, both have elevated U concentrations (Table 10 and Fig. 8). These fertilizer products are used on acidic soils and are also exported to neighboring countries. Kenya uses considerable amounts of fertilizer products derived from Minjingu phosphate rock (Kifuko *et al.,* 2007; Ndeleko-Barasa *et al.,* 2021; Ndungu-Magiroi *et al.,* 2015) and thus has the second highest average U concentration among the investigated East African countries in this study as indicated in Table 10.

The imported NPK and DAP fertilizers in East African Countries recorded the lowest U concentrations of 107.9 ± 9.6 mg kg−1 and 108.8 ± 29.0 mg kg^{-1}, respectively. These levels are almost similar to other NPK and DAPs fertilizers from Western and Northern Africa (Yamazaki & Geraldo, 2003). The NPK concentrations for Kenya (Figure 8) are relatively high since the country imports phosphate ores from Minjingu with elevated U content to produce NPK fertilizer.

Table 10: Average U concentrations of the common phosphate fertilizers used in East Africa

Country	U (mg kg^{-1})
Burundi	136.37 ± 11.67
Kenya	187.07 ± 11.64
Rwanda	174.71 ± 16.72
Tanzania	226.48 ± 13.81
Uganda	152.63 ± 11.58
Type of fertilizer	
DAP	107.88 ± 9.60
NPS	203.57 ± 18.40
MOHP	281.57 ± 15.82
NPK	108.79 ± 29.00
2-WAY ANOVA F-Statistics	
Countries (C)	189.72***
Fertilizers (F)	1391.03***
C x F	285.36***

The values on the table are: Mean ± SE (Standard Error), and *** is significant at $P \leq 0.001$

Detailed U concentrations in the different fertilizers by country are provided in Fig. 7. A considerable variance in the U concentration in the same fertilizer types could be observed that may be attributed to different national fertilizer nutrient requirements. The MOHP fertilizer showed the highest U concentrations with 336.6 mg kg^{-1} detected in fertilizer obtained in Tanzania, 359. 2 mg kg^{-1} in Uganda, 267. 67 mg kg^{-1} in Rwanda, 234.0 mg kg^{-1} in Burundi, and 210.4 mg kg^{-1} in Kenya. Similarly, high differences were observed for NPS fertilizer in Rwanda (306.8 mg kg^{-1}), Tanzania (242.6 mg kg^{-1}), Kenya (208.0 mg kg^{-1}), Burundi (148.8 mg kg^{-1}) and Uganda (111.6 mg kg^{-1}).

The NPK and DAP fertilizers used in East African countries are directly imported from China, Egypt, Morocco, and Saudi Arabia (Nations, 2021). Phosphate rocks in these countries show lower average U concentrations than the analyzed phosphate rocks from East Africa. Average U concentrations are approximately 27 mg kg^{-1} for China (though higher concentrations have been measured by Ye *et al.* (2019), 90 mg kg^{-1} for Egypt, 97 mg kg^{-1} for Morocco, and100 mg kg^{-1} for Saudi Arabia (Haneklaus, 2021; Khater, 2012; Tulsidas *et al.,* 2019) so that the resulting fertilizers show lower U concentrations than the fertilizers produced from local phosphate rock with higher U content in East Africa. A recent study from Ramteke *et al.* (2022) on the U content of imported mineral fertilizers marketed in India that are from similar sources than the imported once sold in East Africa is in good agreement with this work.

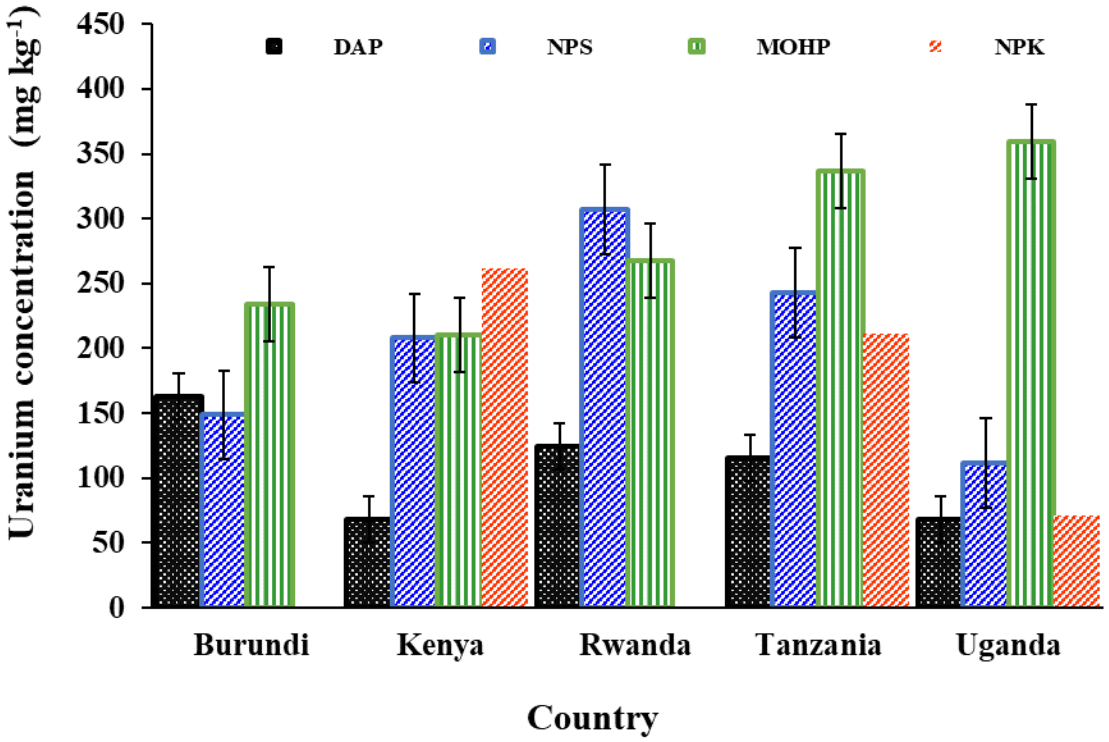

Figure 7: Uranium concentration in common phosphate fertilizers used in East Africa

The frequent use of MOHP and NPS could have resulted in the accumulation of U in East African soils, and further systematic studies as they were for instance reported by Sun *et al.* (2022) and Sun *et al.* (2020) are recommended. First studies by Mlwilo *et al.* (2007) as well as Nkuba and Mohammed (2014) already observed radioactivity above background levels in common crops such as maize and mung beans; respectively. It is strongly recommended to systematically assess the influence of applying phosphate fertilizers on the radioactivity of agricultural soil and plant uptakes.

4.2 Impact of phosphate fertilizers applications on agricultural soil radioactivity

4.2.1 Assessment of accuracy of measurement technique

The measured result of IAEA-375 soil reference materials using gamma spectrometry system are given in Table 11. The relative bias between the IAEA value to the laboratory measured value were within ±10%

Table 11: Measured activity concentrations of reference material and reference values

Radionuclides	Measured value (Bq kg^{-1})	Certified value (Bq kg^{-1})	Relative bias (%)
^{238}U	18.4 ± 0.71	19.8 ± 0.6	-7
^{238}Th	469.2 ± 20.45	434.4 ± 18.9	8
^{40}K	427.6 ± 15.67	406.8 ± 16.7	5

4.2.2 One-year experimental results

The activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in the NPK fertilizers used in the one-year experimental plots are indicated in Table 12. These fertilizer brands are also the most commonly used in tobacco plantations in Kenya, Tanzania, and Uganda. The results show that the GLT fertilizer had the highest ^{232}Th concentration (687.7 ± 1.5 Bqkg^{-1}) and the highest ^{238}U concentration (3216.3 ± 21.7 Bq kg $^{-1}$). The high radioactivity of the GLT fertilizer is associated with the radioactive nature of the local phosphate ores, which serves as the primary raw material for fertilizer production (Mwalongo *et al.,* 2023). Potassium is one of the macronutrients present in the fertilizer. It mainly comprises the stable isotope ^{39}K, but it always contains the radioactive isotope ^{40}K in a fixed proportion, and ^{40}K makes up about 0.012% (120 mg kg^{-1}) of the elemental potassium.

Compared to the other fertilizers, YC showed much lower amounts of ^{232}Th (26.3± 0.3 Bq kg^{-1}) and ^{238}U (198.6 ± 6.1 Bq kg^{-1}), indicating that the source raw material for YC phosphate fertilizer may have low radioactivity levels. Despite the low-level radioactive detection, the product (YC) is sufficiently suitable for use as fertilizers for tobacco. According to the much greater ^{40}K radionuclide concentration (1,566.7 ± 98.5 Bq kg^{-1}) found in YB, the raw materials stable potassium (^{39}K) may include the radioactive ^{40}K (Mike *et al.,* 2017). The crop leaf yield (kg ha^{-1}) was affected by fertilizer application. Applying YB and YC increased leaf yield and K concentration significantly compared to GLT (Table 12). The increase in tobacco leaf K was due to the applied good quality NPK fertilizers.

Table 12: Activity concentrations of radionuclides (Bq kg $^{-1}$ dry weight) in NPK fertilizers used in this study and their leaf yield

Fertilizers trade name	Activity Concentration (Bq kg $^{-1}$)			Dry leaf yield (kg ha^{-1})
	^{238}U	^{232}Th	^{40}K	
Yaramila Blended (YB)	514.6 ± 37.3 ± 0.9b		2179.7± 102.7a	2144.18 ± 77.15a
Yaramila Compound (YC)	198.6 ± 26.3 ±0.3c		1566.7 ± 98.5b	1995.25 ± 65.27b
Golden Leave Tobacco (GLT)	3,216.3 ± 21.7a	687.7 ± 1.5a	760.7 ± 34.8c	1515.43 ± 66.77c
Unfertilized				478.70 ± 58.63d

In Table 12, the letters indicate the Fisher significant difference among the average concentrations of ^{232}Th, ^{238}U, and ^{40}K across the column. The Fisher Least Significant

Difference (FLSD) statistical test, the column with different letter(s) are significantly different at $p \leq 0.0\ 5$ confidence level.

Results from the one-year experimental plots are shown in Fig. 8. The concentrations of the ^{238}U, ^{232}Th, and ^{40}K activity in NPK-treated soils were significantly ($p < 0.05$) higher than the soil from the control plot where no fertilizers were applied (Fig. 9). The results also showed that the soil treated with YB displayed significantly ($p < 0.05$) higher ^{40}K activity concentration than soil from the other plots. High ^{238}U levels in the soil are assumed to be caused by the high ^{238}U content of the phosphate rock used in fertilizer production. The soil treated with GLT fertilizer contained significantly ($p < 0.05$) higher ^{238}U activity concentration (30.57 ± 1.3 Bq kg^{-1}) compared with the other plots.

There were no significant differences in activity concentrations of ^{238}U and ^{232}Th in soils treated with YB and YC fertilizers (Fig. 9). Low ^{238}U and ^{232}Th in YB and YC fertilizer is attributed to low ^{238}U and ^{232}Th in the source rock or the radionuclides were washed out by rainfall. Furthermore, the Fisher Least Significant Difference (FLSD) post hoc test revealed that the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K after fertilizer application did not vary significantly with soil depth, i.e., the tillage of the soil distributed the fertilizers in the 30 cm topsoil layer.

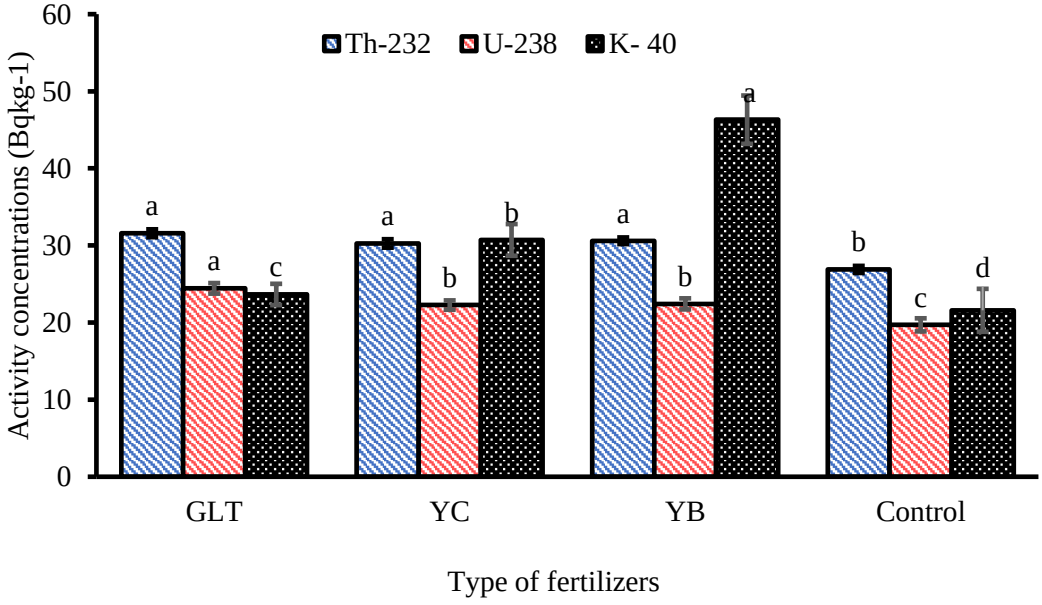

Figure 8: Activity concentrations of ^{232}Th, ^{238}U, and ^{40}K in soils after application of different NPK fertilizers (GLT, YC, and YB) in the one-year experimental plot

In Fig. 9, the letters indicate the Fisher Least Significant Difference (FLSD) among the average concentrations of ^{232}Th, ^{238}U, and ^{40}K across the column. The Fisher's multiple pairwise honestly significant (THSD) statistical test, a different letter (s) on each similar bar chart are significantly different at $p \leq 0.0\ 5$ confidence level.

The activity concentrations of ^{232}Th, ^{238}U, and ^{40}K in tobacco leaves from the one-year experimental plots are shown in Fig. 10. The activity concentrations of ^{232}Th, ^{238}U, and ^{40}K in tobacco leaves from plots treated with NPK fertilizers were in the ranges of 5.97 ± 0.36 Bq kg^{-1} (GLT) to 74.45 ± 2.8 Bq kg $^{-1}$ (YC) for ^{232}Th, 24.56 ± 0.78 Bq kg $^{-1}$ (YB) to 28.58 ± 1.60 Bq kg $^{-1}$ (GLT) for ^{238}U, and 1438.79 ± 12.1 Bq kg $^{-1}$ (GLT) to 1539.38 ± 52.6 Bq kg $^{-1}$ (YB) for ^{40}K. The results showed that the leaves of tobacco plants grown with NPK fertilizers displayed significantly higher ^{238}U concentrations than those of the control plants. The tobacco leaves showed a similar concentration of ^{232}Th for YB and GLT but a statistically higher ^{232}Th concentration for the YC fertilizer.

The ^{238}U uptake was significantly highest for tobacco leaves applied with GLT, followed by YC and YB. Conversely, the unfertilized tobacco leaves (control) had the significantly lowest U content (Fig. 10). It is also noteworthy that the fertilized tobacco leaves showed considerably higher radionuclide concentrations than tobacco leaves analyzed elsewhere (Landsberger *et al.,* 2015; Söğüt *et al.,* 2014).

The activity concentrations of ^{40}K in tobacco leaves were significantly higher than those of ^{238}U and ^{232}Th (Fig. 10) in plots treated with YB and YC fertilizers. The higher concentration of ^{40}K is likely due to the hyperabsorption of potassium by tobacco plants in comparison with other macronutrients (Çalişkan & Çalişkan, 2018; Eke & Ishfaq, 2021). The activity concentrations of ^{232}Th were significantly higher in tobacco leaves from plots treated with YC fertilizer compared with GLT, YB and the control group. Except for the plot treated with YB fertilizer, the activity concentrations of ^{238}U and ^{232}Th in soils were still lower than the worldwide average concentrations of ^{238}U (35 Bq kg^{-1}) and ^{232}Th (30 Bq kg^{-1}) in soils reported by the United Nations Scientific Committee on Atomic Radiations estimates (United Nations Scientific Committee on the Effects of Atomic Radiation [UNSCEAR], 2020). These experimental results show that when NPK fertilizers with high ^{238}U, ^{232}Th, and ^{40}K concentrations are applied, a transfer of these radionuclides from the soil to the tobacco plant is taking place.

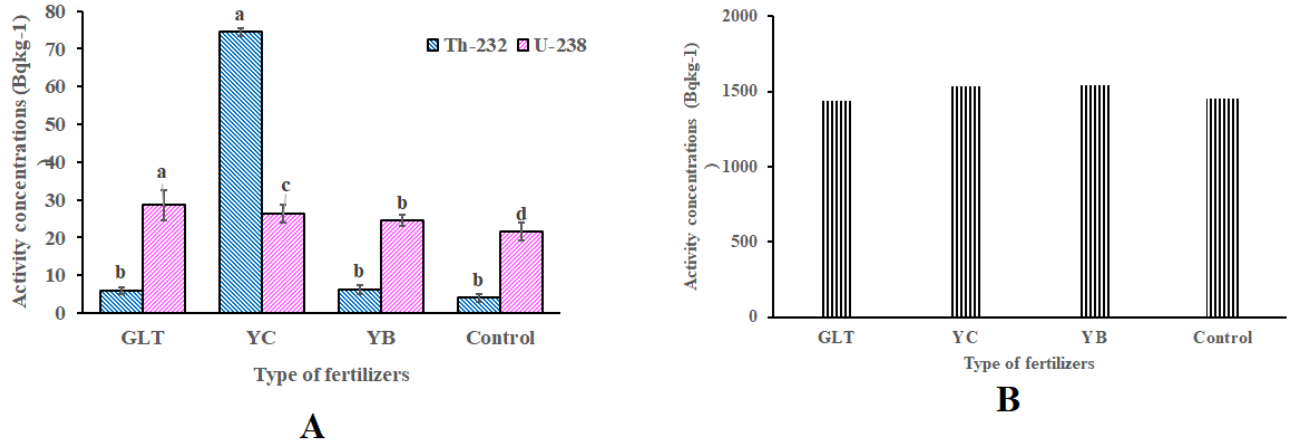

Figure 9: The activity concentration for ^{238}U, ^{232}Th (A), and ^{40}K (B) in tobacco leaves after NPK fertilizer application in the one-year experiment

The physical and chemical properties of soil from the one-year experimental plots were determined, and the relationship between ^{232}Th, ^{238}U, and ^{40}K activity concentrations and soil physical-chemical properties was investigated. The results of a simple correlation matrix between the radionuclide concentrations and the physical-chemical properties are indicated in Table 13. The correlation matrix revealed a moderately positive relationship between ^{232}Th and the soil sand fraction, and a moderately negative relationship with electrical conductivity, silt and clay fractions in soil, and an extremely weak relationship with soil pH. Thus, a strong positive correlation of ^{238}U with sandy soils could also reflect the ability of tobacco plant preference in sandy soils (Lisuma *et al.,* 2020; Lisuma *et al.,* 2021).

The ^{238}U displayed a strong positive correlation with sandy soil and a strong negative correlation with EC, silt and clay in the soil, and a moderate positive correlation with soil pH and organic carbon content. Furthermore, the ^{238}U concentration displayed a strong positive correlation with ^{232}Th and is moderately correlated with the ^{40}K concentration. Potassium (^{40}K) displayed a strong negative correlation with the soil pH, a moderately negative relationship with electrical conductivity, silt and clay fractions in soil and an extremely weak relationship with organic carbon.

It has been reported that the behavior of U and other radionuclides in tropical soils depends upon radioelement solubility, soil pH, redox potential, cation exchange capacity (CEC), organic matter content, and soil texture (sandy, silted, and loamy) (Ribeiro *et al.,* 2018; De Souza Braz *et al.,* 2021). Similarly, according to Hu *et al.* (2020), soil physicochemical characteristics such as pH for sunflowers and peas showed the highest U uptake at pH 3-5.

Another factor that affects U uptake by plants is the presence of carbonates and phosphates in the soil, that have the propensity to interact with U to create complexes, which affects the absorption of U by plants (Sokolik *et al.,* 2020; Tagami & Uchida, 2020).

Table 13: Pearson's correlation coefficients between ^{232}Th, ^{238}U and ^{40}K concentrations with some soil physical-chemical parameters for the one-year experiment

Parameter	^{232}Th	^{238}U	^{40}K	pH	EC	OC	sand	Silt	Clay
^{232}Th	1								
^{238}U	0.66 ns	1							
^{40}K	-0.99*	-0.55ns	1						
pH	0.91*	0.44 ns	-0.93*	1					
EC	0.98*	0.54 ns	-0,94*	0.94*	1				
OM	0.97*	0.51 ns	-0.99*	0.97***	0.99*	1			
Sand	-0.84*	-0.56 ns	0.82*	-0.7*	-0.81*	-0.78*	1		
Silt	0.94*	0.52ns	-0.96*	0.92*	0.97*	0.97*	-0.71*	1	
Clay	0.76*	0.38 ns	-0.77*	0.78*	0.74*	0.76*	-0.83*	0.59 ns	1

Significant at $P \leq 0.05$ (*), $P \leq 0.01$ (**), $P \leq 0.001$ (***); ns = no significant differences

4.2.3 Ten-years NPK fertilizer application experiment

The results showed that the concentration of ^{232}Th, ^{238}U, and ^{40}K in agricultural soils used in ten years experiments were significantly higher *($p < 0.05$)* compared with annual application of phosphate fertilizers and the control samples (no application of fertilizer) (Fig.10 and Fig. 11).

After the annual application of NPK fertilizers for ten years, the activity concentrations of ^{232}Th varied from 25.76 ± 0.9 Bq kg^{-1} (Bukombe district) to 111.58 ± 6.8 Bq kg^{-1} (Manyoni District). The activity concentrations of ^{40}K varied significantly from 100.41 ± 7.5 (Bukombe district) to 1074.79 ± 86.0 Bq kg^{-1} (Bukombe District). The activity concentrations of ^{238}U ranged from 16.1 ± 0.6 Bq kg^{-1} (Bukombe district) to 69.99 ± 2.8 Bq kg^{-1} (Manyoni District).

The higher ^{238}U concentration from Manyoni was related to the prolonged phosphate fertilizer application, but the naturally occurring U in soils in this area is also slightly above average (Kasoga *et al.,* 2015). Indeed, the Manyoni region hosts some surficial U occurrences, which

may have contributed to the higher radioactivity in local agriculture soils than other farms (Anon, 1970). The activity concentrations of ^{238}U, ^{232}Th, and ^{40}K in the agricultural soils at different depths are shown in Fig.10 and Fig. 11.

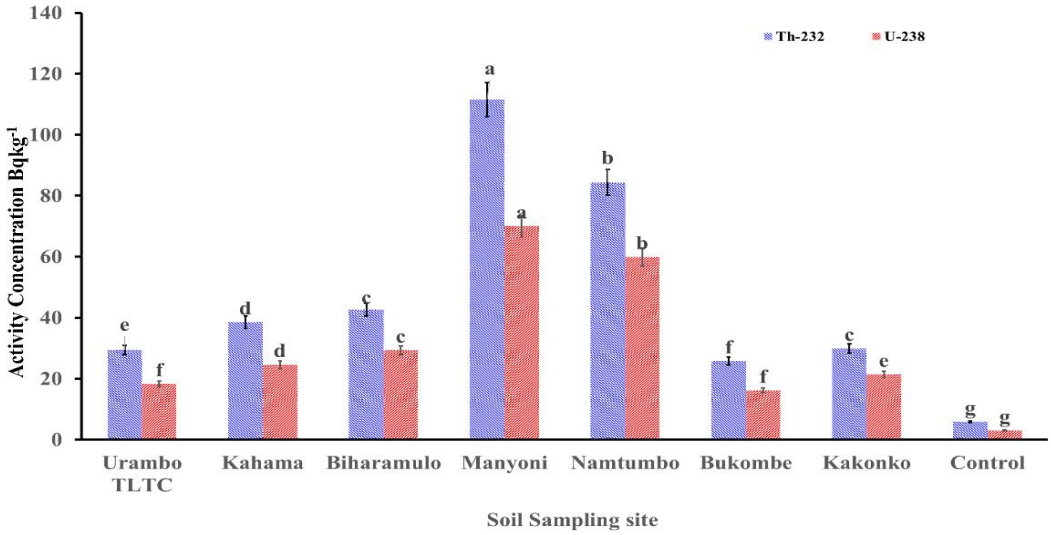

Figure 10: Concentration of ^{232}Th and ^{238}U in agricultural soils after ten-year application of NPK fertilizers application

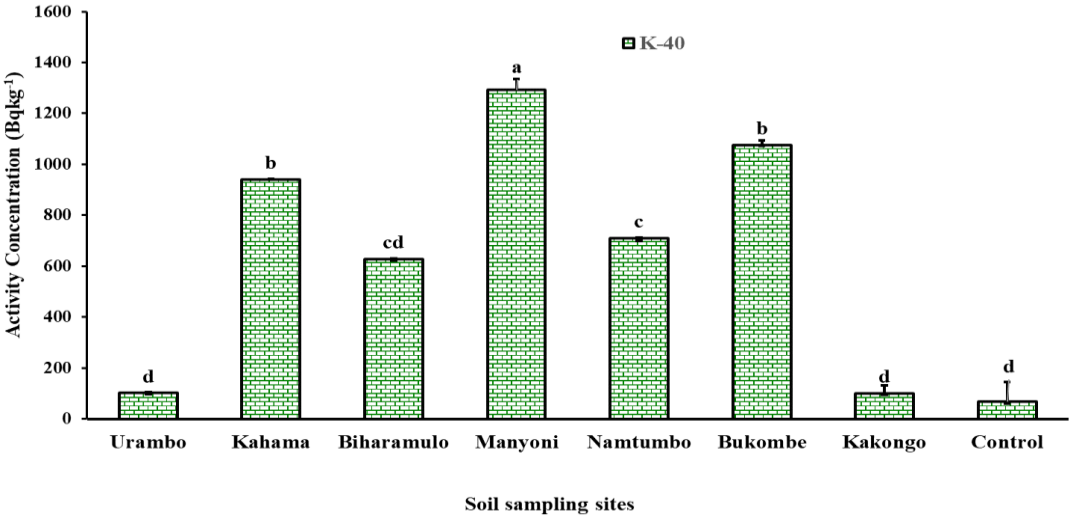

Figure 11: Concentration of ^{40}K in agricultural soils after ten-year application of NPK fertilizers

The results indicate that ^{232}Th was not detected in tobacco leaves from the Biharamulo and Bukombe farms meaning that the value was below the method's detection limit of 1.56 ± 0.12 Bq kg^{-1}. Tobacco leaves from Kahama (33.73 ± 1.60 Bq kg^{-1}) showed the highest ^{232}Th concentration, followed by those from the Urambo farm (26.02 ± 1.50 Bq kg^{-1}), Kakonko (15.73 ± 1.42 Bq kg^{-1}), Manyoni (12.83 ± 1.03Bq kg^{-1}), and Namtumbo (12.43 ± 0.76 Bq kg^{-1}). The range of ^{232}Th concentrations in tobacco leaves reported in this study agrees well with those reported by Mkhaiber *et al.* (2020), who assessed the presence of ^{232}Th in tobacco leaves

from the UAE, France, Iraq, Armenia, Türkiye and South Korea, and reported ^{232}Th concentrations ranging from 4.8 Bq kg^{-1} to 36. 0Bq kg^{-1}.

The concentrations of ^{238}U in the same tobacco leaves (Fig. 12) ranged from 7.83 ± 0.1 Bq kg^{-1} to 14.52 ± 0.62 Bq kg^{-1}. Tobacco leaves from Manyoni showed the highest concentrations of ^{238}U compared to other studied areas, followed by Namtumbo (9.98 ± 0.51). The concentration did not differ significantly for Kakonko (8.80 ± 0.45), Bukombe (8.26 ± 0.49), Urambo farm (8.21 ± 0.65), Biharamulo (8.14 ± 0.37) and Kahama (7.83 ± 0.86). The presence of surficial and sedimentary U mineralization in Manyoni and Namtumbo (Banzi *et al.,* 2015; Ngulimi & Ishiga, 2016), respectively, may have influenced the high ^{238}U concentration in the soil as this can increase uptake by tobacco leaves. The concentrations of ^{238}U in tobacco leaves from the Bukombe and Biharamulo sampling sites were not detected (below the detection limit).

The results show that the concentration of ^{232}Th decreased from Kahama down to Urambo, Kakonko, Manyoni, and Namtumbo (located in the southern part of Tanzania). The ^{232}Th concentration with decreasing trend may be attributed to the variation of the physical-chemical properties of the soils. Thus, an increase in acidity from Kahama to Namtumbo influenced the increase of ^{238}Th. A similar observation was reported by Gupta *et al.* (2020), whose results showed an increase of Th in more acidic soils and U decreasing in acidic soils.

The concentration of ^{40}K (Fig. 13) was significantly higher in tobacco leaves from Biharamulo (1545.30 ± 58.87 Bq kg^{-1}) and Kahama (1538.14± 32.3 Bq kg^{-1}) followed by Bukombe (1497.83 ± 78.89 Bq kg^{-1}), Kakonko (1434.3 ± 78.7 Bq kg^{-1}), Urambo (1255.1 ± 60.9 Bq kg^{-1}), Manyoni (1081.8 ± 46.6 Bq kg^{-1}) and Namtumbo (823.9 ± 75.9 Bq kg^{-1}). However, the ^{40}K concentration of Namtumbo was statistically similar to the control sample.

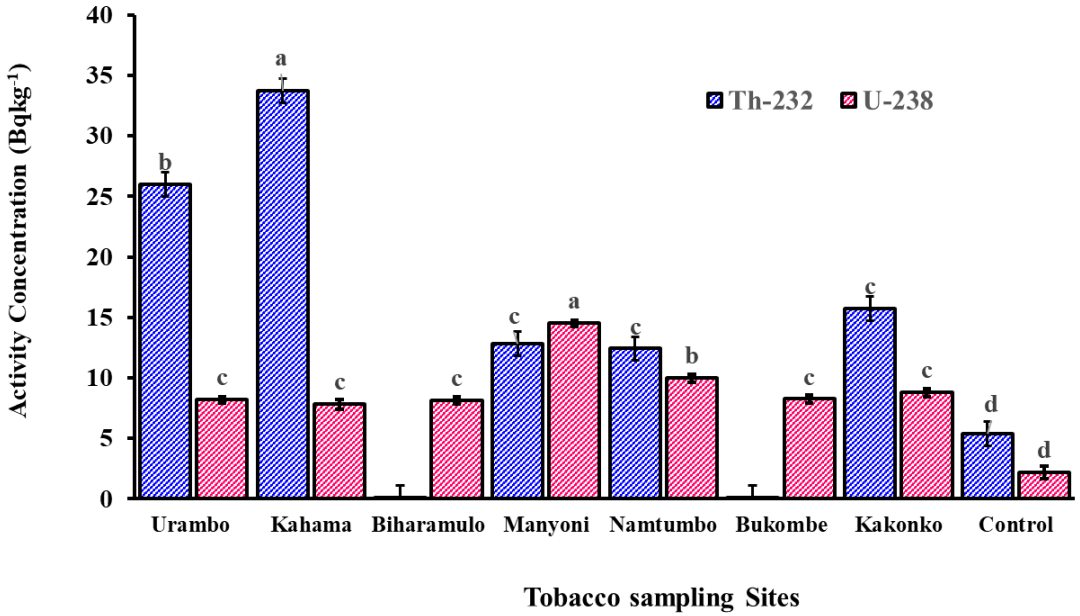

Figure 12: Activity concentrations of 232Th and 238U in tobacco leaves grown in experimental farms with soils fertilized in the previous ten years

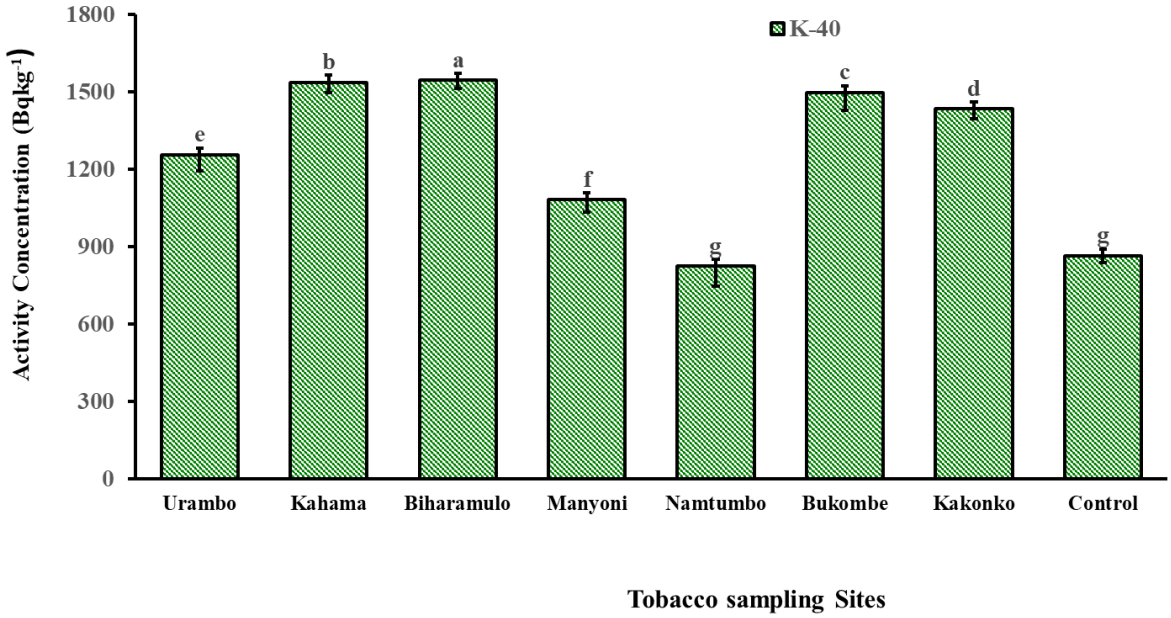

Figure 13: Activity concentrations of 40K in tobacco leaves grown in experimental farms with soils fertilized in the previous ten years

The physical-chemical soil factors influence the concentration of ^{238}U, ^{232}Th and ^{40}K in the soils. Studies have shown that the geology and soil structure influences environmental concentration of radionuclides of natural origin (UNSCEAR, 2020). The concentration of ^{238}U, ^{232}Th and ^{40}K can also be influenced by soil weathering, sediment accretion, sorption, leaching and circulation of groundwater and surface water (Gupta & Walther, 2019; Ratnikov *et al.,* 2020). All these factors certainly played a role in the varying radionuclide concentrations among regions, as found in this investigation. In the ten (10) year's experiment, it was, however, impossible to determine the exact role of each factor. Nevertheless, it was found that

there is a strong and positive correlation between ^{238}U and ^{232}Th, and both displayed a strong negative correlation with organic matter in the soil. Furthermore, the sandy soil was positively correlated with ^{238}U and ^{232}Th, and negatively correlated with organic matter and silt, as indicated in Table 14.

Table 14: Pearson's correlation coefficients between ^{232}Th, ^{238}U and ^{40}K concentrations with some soil physical-chemical parameters for the ten years experiment

Parameter	^{232}Th	^{238}U	^{40}K	pH	EC	OC	sand	Silt	Clay
^{232}Th	1								
^{238}U	-0.24 ns	1							
^{40}K	-0.16 ns	-0.13 ns	1						
pH	0.61*	-0.61*	-0.30 ns	1					
EC	-0.43 ns	0.49 ns	0.28 ns	-0.94*	1				
OM	-0.76*	0.66*	0.27 ns	-0.83*	0.59*	1			
sand	-0.42 ns	-0.38 ns	0.19 ns	-0.27 ns	0.30 ns	0.14 ns	1		
Silt	-0.38 ns	-0.36 ns	0.09 ns	-0.24 ns	0.28 ns	0.11 ns	0.99*	1	
Clay	-0.08 ns	-0.36 ns	0.40 ns	-0.06 ns	0.13ns	-0.08 ns	0.47*	0.43 ns	1

Significant at $P \leq 0.05$ (*), $P \leq 0.01$ (**), $P \leq 0.001$ (***); ns = no significant difference

4.2.4 Radioactivity in selected tobacco farms in Kenya, Tanzania, and Uganda

The activity concentration of ^{232}Th, ^{238}U, and ^{40}K was determined in the agricultural soils of farms grown tobacco in Kenya, Tanzania, and Uganda for the soil layer with 0–30 cm depth.

The concentrations of ^{232}Th, ^{238}U, and ^{40}K in soils were significantly higher ($p < 0.05$) than the control sample from the non-fertilized farm in Tabora, Tanzania (Fig. 15 and Fig. 16). Migori (Kenya) had a significantly higher ^{238}U concentration, followed by Kanungu (Uganda), which showed the lowest value that was still significantly larger than the control group. A similar result trend was observed for ^{232}Th (Fig. 14). However, for ^{40}K, Kanungu (Uganda) had a significant concentration, followed by Migori (Kenya), and Urambo (Tanzania) had the lowest ^{40}K concentration (Fig. 15), which was similar to the control group. The variations In natural radioactivity in soil depend on the unique natural radionuclide present on the sampling sites, the parent rock that formed the soil, and other forming variables (UNSCEAR, 2020).

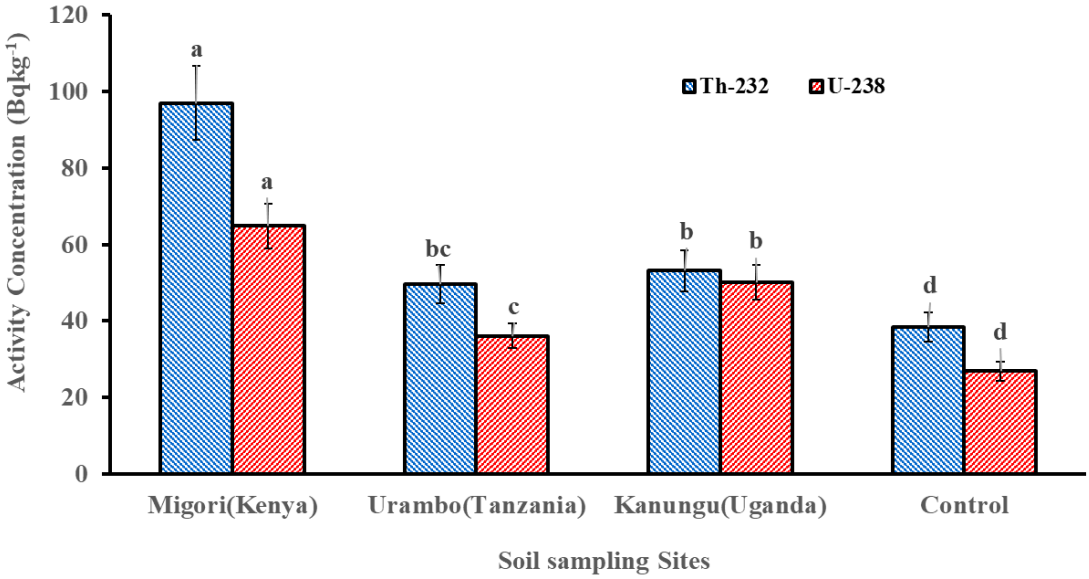

Figure 14: Activity concentrations of 232Th, 238U in soils from tobacco fields from Kenya, Tanzania, and Uganda

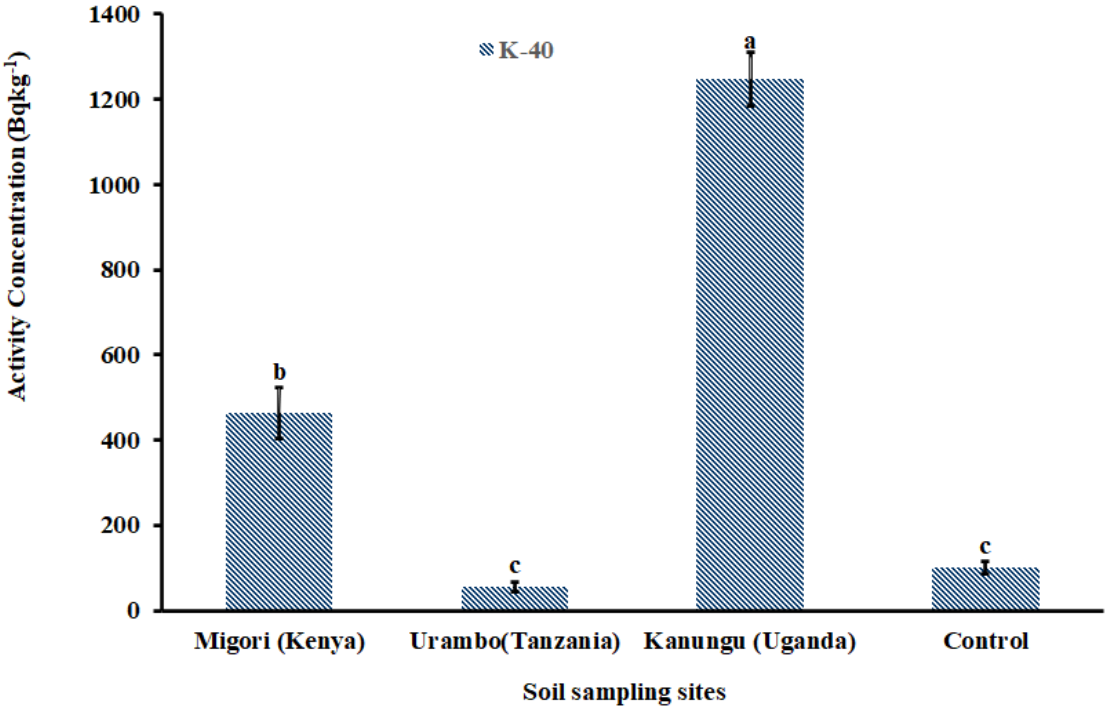

Figure 15: Activity concentrations of 40K in soils from tobacco fields from Kenya, Tanzania, and Uganda

The concentration of radionuclides in tobacco leaves from Kenya, Tanzania, and Uganda ranged from 49.6 ± 3.9 Bq kg^{-1} to 96.9 ± 8.4 Bq kg^{-1} for ^{232}Th, 36.6 ± 3.2 Bq kg^{-1} to 64.8 ± 5.8 Bq kg^{-1} for ^{238}U, and 55.0 ± 4.5 Bq kg^{-1} to 1247.5 ± 87.8 Bq kg^{-1} for ^{40}K as depicted in Fig. 16 and Fig.18. Tobacco leaves from Migori (96.9 ± 4.8Bq kg^{-1}) displayed the highest ^{232}Th concentrations, followed by those from Kanungu (53.1 ± 5.0 Bq kg^{-1}), and with the lowest concentrations measured in leaves from Urambo (49.6 ±3.1 Bq kg^{-1}). Samples from Migori

also displayed the highest ^{238}U concentration (64.8 ± 2.8 Bq kg^{-1}) followed by those from Kanungu (50.1 ± 4.1 Bq kg^{-1}), and lowest concentrations were again measured in leaves from Urambo (36. 1 ± 3.4 Bq kg^{-1}).

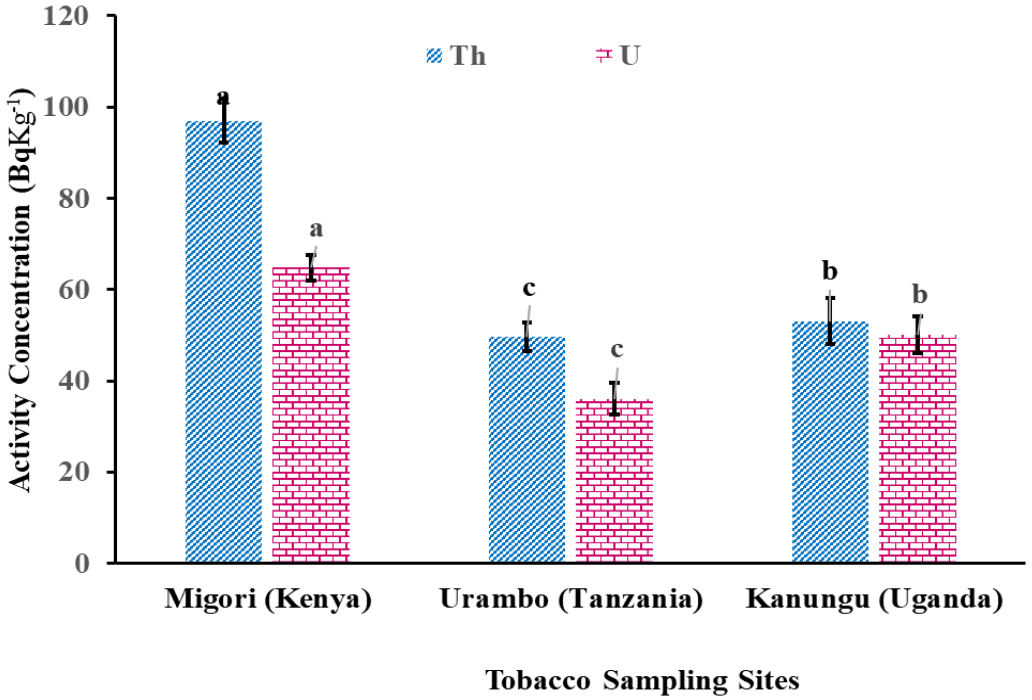

Figure 16: Activity concentrations of 232Th and 238U in t**obacco leaves from Kenya, Tanzania, and Uganda**

Tobacco leaves from Kanungu showed the highest ^{40}K concentration (1247.5 ± 24.0 Bq kg^{-1}), followed by those from Migori (464.5 ± 15.8 Bq kg^{-1}) and Urambo (55.1 ± 4.3 Bq kg^{-1}) as shown in Fig. 17. In these three countries, the radionuclide concentrations in tobacco leaves were globally higher than in the 1-year and 10-year experiments for ^{238}U and ^{232}Th.

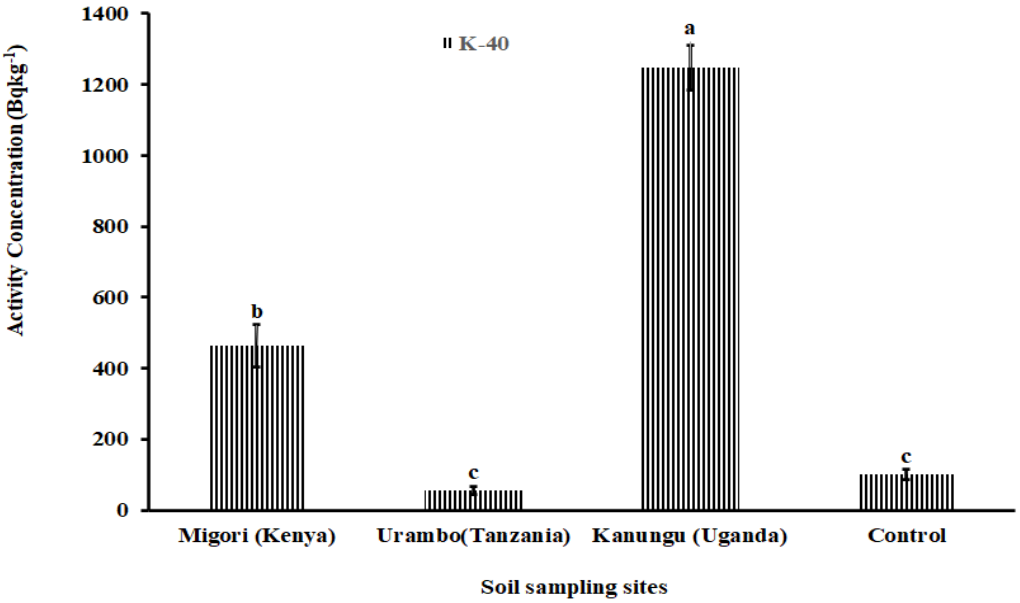

Figure 17: Concentration of 40K in tobacco leaf from selected tobacco farms in Tanzania, Kenya, and Uganda

The Pearson's correction matrix on the association between ^{232}Th, ^{238}U and ^{40}K concentrations and the physical-chemical parameters for agricultural soils of Kenya, Tanzania, and Uganda are presented in Table 15. The results showed a negative association of sand and silt with that of OM, indicating that the OM content was higher in clay soils. The negative association may be because sand and silt lose OM more rapidly than clayey soils through natural and anthropogenic processes (Arunrat *et al.,* 2020). Another possible reason is that indigenous tobacco farmers uproot stalks from the field after harvest and burn them, thus reducing organic carbon from the decay of plant materials in soils (Lisuma *et al.,* 2022). This result is similar to that Lisuma *et al*. (2022a) reported earlier, who found that Tabora and Urambo soils are mainly composed of sand and silt and contain the lowest OM content.

The results further indicate a strong positive association between ^{238}U and OM. The association between ^{238}U and OM results from soil OM reducing the water-soluble and mobile hexavalent U U(VI) to the insoluble tetravalent U(IV), and thus, over time, the U concentrations in soils increases (Sokolik *et al.,* 2020). There was a positive correlation between the ^{40}K concentration and the soil pH and a negative correlation between ^{40}K and the clay soil fraction.

Table 15: Pearson's correlation coefficients between ^{232}Th, ^{238}U and ^{40}K dry weight concentrations with soil physical-chemical parameters at selected tobacco farms in Kenya, Tanzania, and Uganda

Parameter	pH	EC	OM	Sand	Silt	Clay	^{232}Th	^{238}U	^{40}K
pH	1								
EC	0.42								
OM	-0.12	0.69*	1						
Sand	0.38	-0.49	-0.96*	1					
Silt	-0.05	0.63	0.97*	-0.92*	1				
Clay	-0.51	0.39	0.91*	-0.99*	0.87	1			
^{232}Th	-0.41	-0.74*	-0.83*	0.68	-0.86	-0.57	1		
^{238}U	-0.07	-0.7	0.97*	0.89	0.98*	0.82	0.93	1	
^{40}K	0.94*	0.12	-0.45	0.66	-0.36	-0.76*	-0.1	0.26	1

Significant at $P \leq 0.05$ (*), $P \leq 0.01$ (**), $P \leq 0.001$ (***); ns = no significant difference

4.2.5 Estimation of radiological hazards from agricultural soil and tobacco leaves

It was hypothesized that radionuclides present in soil and tobacco leaves could be a radiological hazard to farm workers and members of the public. Furthermore, tobacco consumers may ingest and inhale the radionuclides contained in the tobacco leaves (IAEA, 2013).

(i) Radium equivalent activity of soils

Radium equivalent (Ra_{eq}) is a radiological health risk parameter resulting from exposures to naturally occurring radioactive materials containing ^{232}Th, ^{238}U, and ^{40}K. It is used by regulatory bodies to set a single regulatory limit rather than setting individual radionuclide limits (Abed *et al.,* 2022). The Ra_{eq} of agricultural soils from all study areas ranged from 68.17 ± 3.5 Bq kg^{-1} (Urambo) to 329.2 ± 12.8 Bq kg^{-1} (Manyoni) and is shown in Table 16. These values are below the Ra_{eq} value of 370 Bq kg^{-1}, corresponding to delivering an annual dose of 1 mSv y^{-1} to public members (UNSCEAR, 2020). The low Ra_{eq} implies that these soils can be used as a construction material, as is common in the investigated regions.

(ii) Absorbed dose rate due to radioactivity in soils

The obtained results show that the absorbed dose rate from agricultural soils ranged from 32.38 nGy h^{-1} (Urambo) to 282.93 ± 14.67 nGy h^{-1} (Bukombe) (Table 16). The absorbed dose rate for the Urambo (32. 38 nGy h^{-1}), the Yaramila compound experimental site (33.14 nGy h^{-1}), Kakonko (40.5 nGy h^{-1}), Kanungu (41.44 nGy h^{-1}) and Migori (54.37 nGy h^{-1}) were lower than the global average value of 60 nGy h^{-1}(UNSCEAR, 2020). Therefore, the farmers and public members using these lands are exposed to low doses of gamma radiation from the agricultural soils. However, the absorbed dose rate at the Tanzania Leaf Tobacco Company Limited (TLTC) farm (Urambo, Tanzania 128.09 nGy h^{-1}) was about twice the recommended value. On the other hand, agricultural fields such as Namtumbo (154.16 nGy h^{-1}), the YB field experiment (167.86 nGy h^{-1}), Kahama (175.26 nGy h^{-1}) and Biharamulo (178.94 nGy h^{-1}) were about three times higher than the worldwide average dose rate value. In comparison, Manyoni (231.89 nGy h^{-1}) was four times higher than the recommended value, and Bukombe (282.93 nGy h^{-1}) was even more about five times the worldwide average global value.

(iii) The annual effective dose rate from radioactivity in soils

The calculated outdoor and indoor annual effective dose rate from soils in the tobacco plantations, as summarized in Table 16 ranged from 37.0 $nSvh^{-1}$ (GLT experimental site) to

190.0 $nSvh^{-1}$ (Manyoni) and 150 $nSvh^{-1}$ (GLT, YC, Urambo, Kakonko) to 750 $nSvh^{-1}$ (Manyoni), respectively. The annual effective dose rates were below the worldwide mean annual effective dose, which is about 500 μSv y^{-1}. Moreover, the total annual effective dose has been found to vary depending on the geological parameters of the soil matrix where tobacco plants are grown and are further influenced by anthropogenic activities such as applying mineral phosphate fertilizers (Prno & Scott, 2012; UNSCEAR, 2020).

(iv) The external and internal hazard indexes

The results in Table 16 show that the H_{ex} and H_{in} of tobacco agricultural soils ranged from 0.18 (YC, Urambo) to 0.89 (Manyoni) and 0.23 (Urambo) to 1.08 (Manyoni), respectively. For the radiation hazard to be tolerable, it should be lower than 1, which is (mostly) the case here. An index result greater than 1 corresponds to an annual dose of more than 1 mSv y^{-1} (Imani *et al.,* 2021). The results imply that if the soil is used as a building material this practice can exceed the public annual effective dose constraint of 1 $mSvy^{-1}$ (UNSCEAR, 2020). Therefore, the external and internal hazard indices are acceptable (i.e., not exceeding the dose limit of 1 mSv y^{-1}) when the calculated index is ≤ 1 (Purnama & Damayanti, 2020).

Table 16: Summary of calculated radiological hazard indices and excess lifetime cancer risk from agricultural soils in Tanzania, Kenya and Uganda

Site	Ra_{eq} (Bq/kg)	D (nGyh^{-1})	AED (nSvy^{-1}) outdoor	AED (nSvy^{-1}) indoor	H_{ex}	H_{in}	ELCR $_{Outdoor}$ (10^{-6})	ELCR indoor (10^{-6})
One-year experiment								
GLT	68.69	30.14	37	150	0.19	0.25	0.13	0.52
YC	68.20	30.01	40	150	0.18	0.25	0.13	0.52
YB	**71.51**	33.59	40	160	0.19	0.25	0.14	0.58
Ten-years Experiment								
Urambo	68.17	30.45	40	150	0.18	0.23	0.13	0.52
Kahama	152.11	73.87	90	360	0.41	0.48	0.32	1.27
Biharamulo	138.56	65.46	80	320	0.37	0.45	0.28	1.12
Manyoni	329.18	153.73	190	750	0.89	1.08	0.66	2.64
Namtumbo	235.17	108.24	130	530	0.64	0.80	0.46	1.86
Bukombe	135.70	67.83	80	330	0.37	0.41	0.29	1.16
Kakonko	70.38	31.45	40	150	0.19	0.24	0.14	0.54

Ra_{eq}: radium equivalent activity; D: radiation absorbed dose; AED: Annual effective dose; nSvy^{-1}: nano Sievert per year; H_{ex}: External radiation exposure health index**;** H_{in} : internal radiation exposure health index; $ELCR_{Smoking}$: Excess life time cancer risk for smoking; and $ELCR_{Snuffing}$: Excess life time cancer risk for snuffing

(v) Tobacco leaf radium equivalent activity

Tobacco plants were assumed to constitute an above ground reservoir of ^{232}Th, ^{238}U and ^{40}K and thus a source of radiation exposure to human beings, and surface soils. Accordingly, the values of Ra_{eq} with the lower value of 90.20 ± 4.6 Bq kg^{-1} in Namtumbo and the highest value of 174.50 Bq kg^{-1} at the Kahama sampling location were measured (Table 17). Compared to the worldwide average of 60 Bq kg^{-1} of Ra_{eq} all investigated tobacco plant leaves exceeded the worldwide average Ra_{eq} value for soils. Nevertheless, the equivalent radium activity ranged from 90.20 Bq kg^{-1} to 174.5 Bq kg^{-1}, which is significantly lower than the UNSCEAR recommended combined activity concentration of 370 Bq kg^{-1} (Mostafa *et al.,* 2020; UNSCEAR, 2020) that again corresponds to an annual dose of 1 mSv y^{-1} for the general public. Therefore, the Ra_{eq} activity indicates that the tobacco crops do not pose a significant radiological risk to farm workers and relatives living nearby.

(vi) Annual effective dose due to tobacco inhalation

The results shown in Table 17 indicate that the annual effective dose due to tobacco snuffing ranged from 0.01 to 6.53 mSv y^{-1}, varying with the producing region. The tobacco leaves from the one-year experimental plots may lead to effective doses higher than the recommended safety limit value of 1 mSv y^{-1} if snuffed and may pose a severe radiological health hazard.

Through the root uptake of radionuclides from soils, tobacco plants can become an additional reservoir of radionuclides above ground. Therefore, It was hypothesized that tobacco plantations (i.e., the plant biomass) might be a source of external radiation to farmers and workers of the plantations. Applying the ICRP activity-to-dose conversion factors (Sievert per gray (Sv/ Gy)), occupancy factors for indoor and outdoor (0.8 and 0.2, respectively), the time of a year (hours per year (h/y), and the absorbed dose rate D (nGy h^{-1}), the yearly effective dose was calculated (UNSCEAR, 2020).

It was estimated and assumed that about 75% of the radionuclides present in tobacco and tobacco products (cigars, cigarettes, etc.) pass into smoke. However, only 50% of that smoke reaches the lungs, while the other 50% is released into the air (side smoke) (UNSCEAR, 2020). Therefore, the smokers and snuffers' inhalation dose conversion coefficients for individual radionuclides used were 2.9 10^{-6} Sv Bq^{-1} for ^{238}U, 4.5 10^{-5} Sv Bq^{-1} Th^{232}, and 2.1 10^{-9} Sv Bq^{-1} for ^{40}K.

The annual effective dose from tobacco smoking and snuffing was calculated using local statistics available for cigarette consumption in Tanzania (URT, 2018). The calculations were based on the consumption of 8.5 cigarettes per day per smoker/snuffer (URT, 2018). The average weight of tobacco rolled by the smoker/snuffer in a piece of paper, banana leaf, maize leaf (traditionally known as "roll-your-own tobacco"), or proper cigarette paper is approximately 1.3 grams. Therefore, the weight of tobacco consumed annually in Tanzania was calculated considering the weight of tobacco in a cigarette multiplied by the number of cigarettes smoked per day (1.3 g x 8.5 = 11.05 g per day), which yields the annual mass of tobacco consumed of approximately 4 kg (11.05 g per day x 365 days per year = 4.033 kg y^{-1}). Unfortunately, the information on partitioning the amount of tobacco consumed as smoked or snuffed per year was not available, and it was assumed that the amount of tobacco snuffed is equal to the amount smoked.

The annual effective dose equivalent for tobacco smoking ranged from 0.1 mSv y^{-1} to 2.45 mSv y^{-1}, varying again with the tobacco producing regions. Tobacco smokers may receive an annual dose over 50-194% of the recommended limit of 1.26 mSv y^{-1}, the average annual global exposure to natural radiation sources from inhalation (UNSCEAR, 2020; Zlobina *et al.,* 2022). Therefore, snuffing and smoking tobacco can produce relatively high annual effective doses.

(vii) Excess lifetime cancer risk

According to United Nations Commodity Trade Statistics Database (2022), tobacco and tobacco products are consumed in the producing nations (Kenya, Tanzania, and Uganda), as well as other African nations (Burundi, Malawi, Rwanda, and South Africa), with average life spans of 65 years.

Table 17: The estimated radiometric parameters for the consumption of tobacco leaves from the 10 years experiment and traditional tobacco farms in Kenya, Tanzania, and Uganda

Site	Ra_{eq}	AED (Smoking) mSv y^{-1}	AED (Snuffing) mSv y^{-1}	$ELCR_{Smoking}$ (10^{-3})	$ELCR_{snuffing}$ (10^{-3})
10-years experiment:					
Urambo	142.06	1.89	5.04	7.08	18.89
Kahama	174.5	2.45	6.53	9.18	24.48
Biharamulo	127.27	0.01	0.03	0.05	0.12
Manyoni	115.16	0.93	2.49	3.50	9.33
Namtumbo	90.2	0.90	2.41	3.39	9.03
Bukombe	123.74	0.01	0.03	0.05	0.12
Kakonko	145.73	1.14	3.05	4.29	11.44
Traditional farms:					
Migori	106.7	0.87	2.31	3.25	8.67
Urambo TLTC	130.16	0.00	0.01	0.02	0.05
Kanungu	109.13	1.18	3.16	4.44	11.85
Worldwide average 0.29					0.29

Ra_{eq}: radium equivalent activity; D: radiation absorbed dose; AED: Annual effective dose; $nSvy^{-1}$: nano Sievert per year; $H_{ex:}$ External radiation exposure health index**;** H_{in}: internal radiation exposure health index; $ELCR_{Smoking}$: Excess life time cancer risk for smoking; and $ELCR_{Snuffing}$: Excess life time cancer risk for snuffing

The excess lifetime cancer risk (ELCR) values for tobacco snuffing ranged from 5.0 x 10^{-5} to 24.48 x 10^{-3} for snuffing and from 0.02 x 10^{-3} to 9.18 x 10^{-3} for smoking, respectively.

Therefore, as shown in Table 17, tobacco snuffing and smoking increases the likelihood of cancer significantly if compared to the worldwide average exposure of non-smokers/snuffers.

The study indicated that application of phosphate fertilizers increased radioactivity in agricultural soil and enhanced radionuclides transfer from soil to plants. The study recommends further research on radionuclide transfers in cereal crops which are phosphate fertilizer use-intensive and advocates for the use of fertilizers with lower radioactivity levels to conserve soil quality and minimize crop uptakes.

4.3 Soil bacterial diversity in sandy loam soils under varying Uranium concentrations

The results for U concentration and bacterial community diversity from sandy loam soil experimental sites are presented in this section.

4.3.1 Quality control of uranium in soil

The accuracy and reliability of the analytical methods in determining U concentration in the samples was checked by measurement of NIST Standard Reference Material (SRM)® 2711a (Montana II Soil) and verified by IAEA soil 7 certified reference material (trace elements in soil). The result of the measurement system average relative bias was within ± 5% as indicated in Table.18

Table 18: Measured U mass fraction in reference materials

Reference Material	U Concentration (mg kg^{-1})		Bias (%)
	Measured	Reference	
IAEA Soil 7	2.72 ± 0.43	2.6 ± 0.6	4.6%
NIST 2711a	3.10 ± 0.01	2.96	5.1%

4.3.2 Effect of soil pH and uranium content in different fertilizer application plots

The goal of measuring standard reference material was to verify the suitability of the methodology for characterizing U in PFs. Since soil pH influences the development of bacteria, measurements of soil pH were made to evaluate changes both before and after fertilizer application. Results of U concentrations detected in fertilizers used in the research study are shown in Table 19, and the accuracy of the measurements proved to show good precision. Minjingu powder fertilizer had the highest U content (159.67 ± 48.48 mg kg^{-1}), followed by Nafaka Plus fertilizer (147.65 ± 8.61 mg kg^{-1}), and YaraMila cereal fertilizer had the lowest U content recorded at 38.84 ± 1.24 mg kg^{-1}. The results of pH and U concentrations in soil samples from the experimental plots, before and after application of fertilizers. Before maize

planting, the soil pH and soil U content did not differ significantly among plots and averaged 5.22 ± 0.51 mg kg^{-1} for pH and 2.02 ± 0.07 mg kg^{-1} for U. After maize planting and application of fertilizers several changes were observed among the plots. The soil pH in the control treatment (Nafaka Plus) and in the treatment YC+M did not change (Table 19). The soil pH increased significantly ($p < 0.001$) from pH 5.21 to pH = 5.26 in the maize plot fertilized with Nafaka Plus, and slightly increased but not changed significantly in the maize plot fertilized with Minjingu Powder (pH = 5.24). The U concentration in soils in the maize plot fertilized with NP increased significantly from 2.04 mg kg^{-1}(unfertilized) - 3.93 mg kg^{-1}, followed by the maize plot fertilized with MP fertilizer (to 3.06 mg kg^{-1}). The maize plot fertilized with YaraMila Cereal and the not-fertilized maize plot (control plot) did not differ significantly in soil pH and U content.

Table 19: Soil pH and U content before and after fertilization

Treatments (T)	Soil pH before fertilization	Soil U (mg kg^{-1}) before fertilization	Soil pH after fertilization	Soil U (mg kg^{-1}) after fertilization
T1 = NF + M	5.21 ± 0.01a	2.04 ± 0.01a	5.21 ± 0.01c	2.04 ± 0.02c
T2 = YC + M	5.22 ± 0.00a	2.02 ± 0.02a	5.22 ± 0.01bc	2.04 ± 0.01c
T3 = NP + M	5.22 ± 0.00a	2.01 ± 0.02a	5.26 ± 0.01a	3.93 ± 0.28a
T4 = MP + M	5.22 ± 0.00a	2.02 ± 0.01a	5.24 ± 0.01ab	3.06 ± 0.15b
F-statistics	0.27ns	0.48^{ns}	0.02^{*}	0.00^{***}

Means in the same category of evaluated interface sharing similar letter(s) do not differ significantly (ns=not significant) based on their respective Standard error (SE) at 5% error rate. Values presented are means ± SE $\bar{x}$ (Standard error of means); *, ** means significant at $P < 0.05$, $P < 0.001$.

The not-fertilized plot had the highest total operational taxonomic units (OTUs), reaching 822, followed by the plot Minjingu Powder and YaraMila Cereal plots, which had 795 and 648 amplicon sequence variants (ASVs), respectively (Fig. 23a). The higher the U concentration in soils the lowest the number of ASVs. The NP fertilizer application in maize plots significant ($p < 0.001$) increased the U content in soil to 3.93 mg kg^{-1}, indicating that the fertilizer had a substantial content and influence of U derived from the Minjingu phosphate rocks. On the other hand, maize plots fertilized with Minjingu Powder (the natural rock in powder used as fertilizer) had the lowest (3.06 mg kg^{-1}) soil U concentration, which was lower by 0.87 units compared to the NP fertilizer (Table 19). This indicates that the Minjingu powder, despite its higher U concentration content, had the lowest U concentration in soils compared with NP because most of its U concentration could have been easily lost through leaching as it was applied in powder form.

The organic carbon and calcium content of the soils under the study site was very low, which could have resulted in the low retention of U released by Nafaka plus and Minjingu powder in

soils. A study by Ratnikov *et al.* (2020) also observed that organic carbon and calcium contents in soils contribute the mobility of U concentrations significantly. Following Nafaka plus and Minjingu fertilizers applications, soil pH was increased in two treatments, T3 and T4, respectively. The pH increase was attributed to the CaO content of fertilizers, which was 36% and 25% from MP and NP fertilizers; respectively. The U concentration in agricultural soils also increased in the treatments with these fertilizers and it was associated with the soil pH increase in Nafaka plus and Minjingu treatments (Table 19). Soil pH is the most determinant factor for the U distribution coefficient as it increases to pH 6 (Cui *et al.,* 2023; Manoj *et al.,* 2020; Vandenhove *et al.,* 2007; Xiao *et al.,* 2019). The no change of soil pH and U content observed in not-fertilized (NF) plots and YC fertilized plots signifies that YC fertilizer has minor U impurities.

Several studies have reported the affinity between U and organic matter in soils for contaminated sites (Ahmed *et al.,* 2014; Blanco *et al.,* 2004), also U is reported to binds to soil containing high organic matter contents (Gupta & Walther, 2019; Vandenhove *et al.,* 2007). The studies further established those soils organic matter served as a significant pool of importantly in agricultural (paddy) soils, U derived from application of PFs were found to adsorbed on weakly crystalline Fe/Al minerals(Tagami & Uchida, 2020).

4.3.3 Abundance of soil bacteria at Phylum level in maize plots

The total number of classifiable sequence's was 297 865 and correlated with ten relative abundance bacterial phyla from each treatment, i.e., MP + M, NP + M, NF+M, and YC + M (Fig. 18). In all fertilizer treatments, the dominant three phyla were *Actinobacteriota* (41.64%), *Proteobacteria* (22.00%), and *Acidobacteriota* (8.44%), together accounting for 72.08% of all the phyla. Next to these phyla were *Chloroflexi* (7.00%), *Gemmatimonadota* (6.76%), *Planctomycetota* (4.02%), *Myxococcota* (3.96%), *Firmicutes* (3.66%), followed by *Bacteroidota* (1.49%) and *Verrucomicrobiota* (1.01%). Bacterial phyla represented by <1% were omitted and hence not considered in the relative abundance calculation.

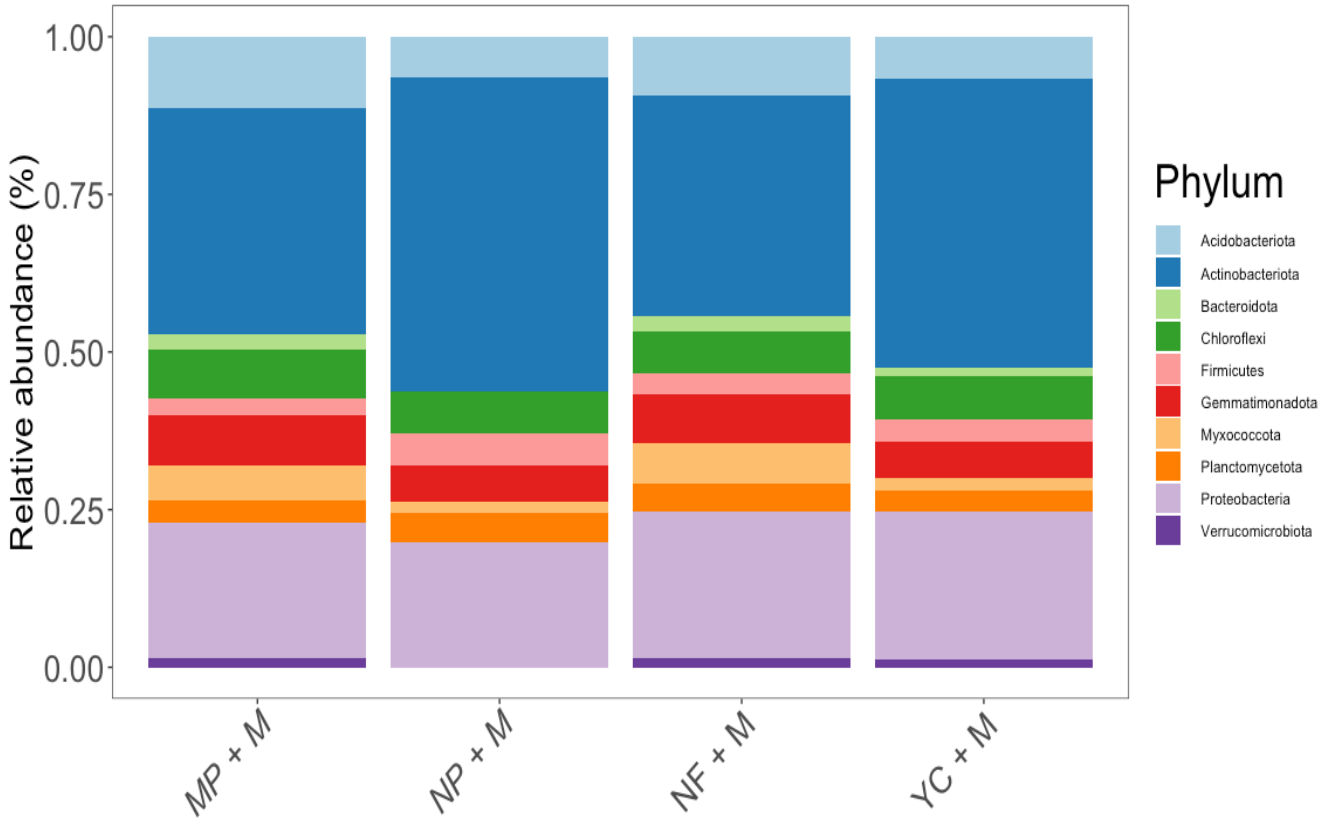

Figure 18: Relative abundances (%) of soil bacteria Phyla in each treatment applied to maize plantation

The distribution of bacteria at the phylum level and their relative abundance varied with the fertilizer treatments. Bacteria from *Actinobacteriota* phylum were abundant in NP + M (12.42%), YC + M (11.50%), MP + M (8.98%), and NF+M (8.74%). *Proteobacteria* were abundant in YC + M (5.89%), followed by NF+M (5.82%), MP + M (5.35% and NP + M (4.92%). *Acidobacteriota* were remarkably found in MP + M (2.85%), NF+M (2.32%), followed by YC + M (1.67%), and NP + M (1.59%). The plot fertilized with Minjingu Powder (MP + M) had 1.97% *Chloroflexi* phylum followed by 1.73% in YC + M, 1.66%, and 1.65% in NF+M and NP + M, respectively. Minjingu Powder fertilizer (MP + M) also displayed the highest abundance for *Gemmatimonodota* (2.02%), followed by the not-fertilized plot (NF+M) with 1.92%, 1.44% in the treatment (YC + M) and 1.38% in the treatment NP + M. The fertilized plot (NP + M) had 1.17% of *Planctomycetota* phylum, followed by NF+M (1.08%), MP + M (0.91%), and YC + M (0.86%). The not-fertilized plot (NF+M) had a higher abundance of *Myxococcota* (1.64%), followed by MP + M (1.36%), YC + M (0.98%), and NP + M (0.46%). *Firmicutes* were abundant in NP + M, reaching 1.29%, followed by YC + M at 0.91%, NF+M at 0.83%, and MP + M at 0.63%. *Bacteroidota* and *Verrucomicrobiota* phyla were very few; in an average of 1%, NF+M had 0.60 and 0.36% of *Bacteroidota* and *Verrucimicrobiota*

phyla, respectively. YC + M had 0.31, and 0.29%, and MP + M had 0.58 and 0.36% of *Bacteroidota* and *Verrucimicrobiota* phyla, respectively.

4.3.4 Abundance of soil bacteria at the Order level in maize plots

In relative abundance, the top five Orders of soil bacteria in the maize treatments were *Frankiales* (15.91%), *Rhizobiales* (13.47%), *Burkholderiales* (12.96%), *Gemmatimonadales* (12.20%), and *Gaiellales* (10.02). Following these five orders in relative abundance, the *Solirubrobacterales* (9.6%), *Micromonosporales* (8.56%), and *Pseudonocardiales* (8.49%) were represented (Fig. 19). Moreover, in lower abundances orders were detected; *Propionibacteriales* (2.63%), *Acidobacteriales* (2.55%), *Ktedonobacterales* (1.80%), and 1.74% of *Bacillales*.

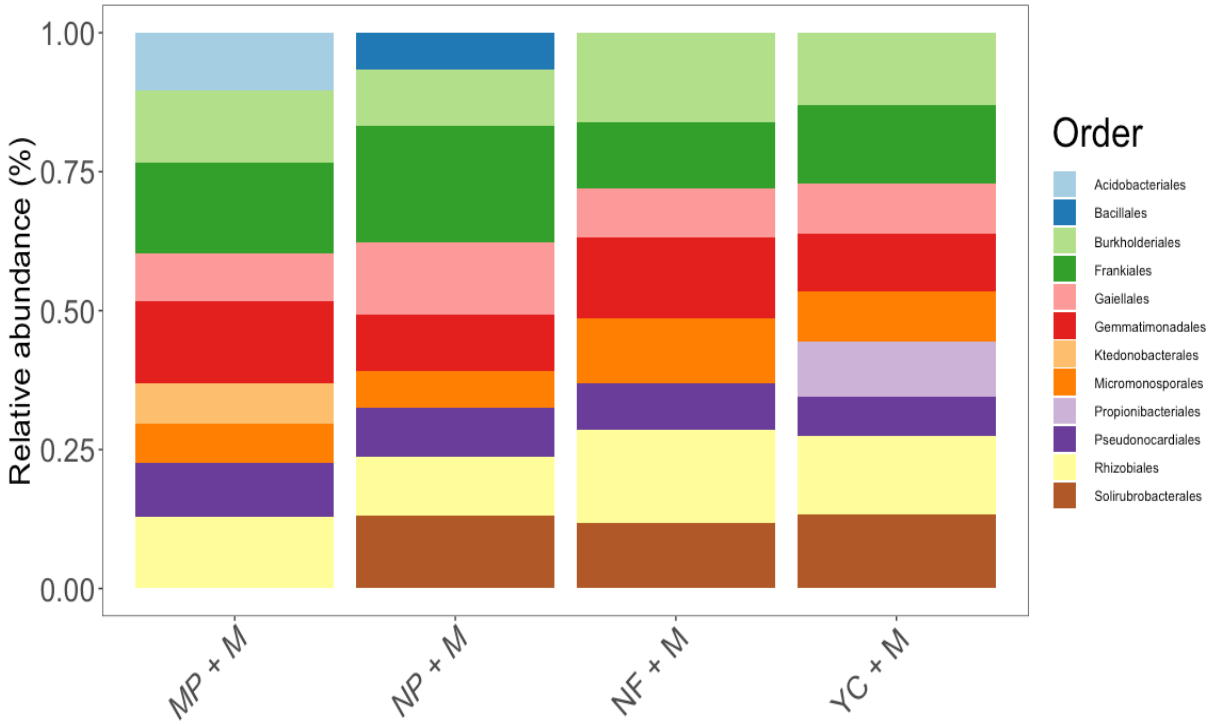

Figure 19: Relative abundances (%) of soil bacteria at Order level for each treatment on maize plantation

Similarly, to the relative abundance of bacteria at Phylum level, the abundance of bacteria at the Order level varied also with the fertilizer treatments. The *Frankiales* were abundant in NP + M by 5.57%, followed by MP + M at 3.99%, YC + M at 3.75%, and NF+M at 2.60%. The *Rhizobiales* next to *Frankiales* were abundant in YC + M (3.80%), followed by NF + M (3.69%), MP + M (3.15%), and NP + M (2.82%). *Burkholderiales* were rich in NF+M by 3.58%, YC + M by 3.50% and MP + M by 3.18%. *Gemmatimonadales* were abundant in MP + M by 3.64%, followed by NF+M by 3.19%, YC + M by 2.73%, and NP + M by 2.65%.

Gaiellales were found at a higher level in NP + M, reaching 3.45%, YC + M by 2.47%, MP + M by 2.12%, and NF + M by 1.98%. Other Orders observed were, *Solirubrobacterales* in YC + M (3.56%), NP + M (3.46%), and 2.63% of NF+M. *Micromonosporales* Order in decreasing abundance was 2.60% (NF + M), 2.46% (YC + M), 1.77% (NP + M), and 1.72% (MP + M), while *Pseudonocardiales* were abundant in MP + M by 2.40% followed by NP + M by 2.32% and YC + M by 1.90%. The Order of *Propionibacteriales* was represented only in YC + M by 2.63%, while *Acidobacteriales* and *Ktedonobacterales* were found only in MP + M treatment by 2.55 and 1.80%, respectively. Finally, the order of *Bacillales* was found to be NP + M only by 1.74%.

4.3.5 Abundance of soil bacteria at the Class level in maize plots

Figure 20 shows the relative abundance of the twelve Classes of soil bacteria that were sequenced. The seven more represented Classes were *Actinobacteriae* (33.36%), *Alphaproteobacteria* (15.81%), *Thermoleophilia* (12.95%), *Gammaproteobacteria* (10.98%), *Gemmatimonadetes* (7.43%), *Acidobacteriae* (5.88%) and *Bacilli* (3.58%). Other Classes in lesser abundance were *Polyangia* (3.02%), *Acidimicrobiia* (2.92%), *Chloroflexia* (1.81%), *Ktedonobacteria* (1.22%), and *Planctomycetes* (1.02%). Six Classes were present in all treatments: *Actinobacteria*, *Alphaproteobacteria*, *Thermoleophilia*, *Gammaproteobacteria*, *Gemmatimonadetes,* and *Acidobacteriae*. The treatment with Nafaka Plus (NP + M) displayed the highest abundance of *Actinobacteriae* Class (10.11%) followed by (YC + M) treatment with 9.11%, (MP + M) with 7.55%, and (NF+M) with 6.59%. The (YC + M) treatment had 4.37% of *Alphaproteobacteria*, followed by (NF+M) at 4.08%, (MP + M) at 3.92%, and NP + M at 3.43%. *Thermoleophilia* class was higher in NP + M treatment, reaching 4.24%, followed by YC + M at 3.67%, NF+M at 2.82%, and MP + M at 2.21%.

The *Gammaproteobacteria* Class was abundant in NF+M by 3.01%, YC + M by 2.78%, MP + M by 2.63%, and NP + M by 2.56%. The MP + M had 2.22% of *Gemmatimonadetes* class, followed by NF+M by 1.94%, YC + M by 1.66%, and NP + M by 1.62%. *Acidobacteriae* class had 2.44% in MP + M treatment, and next to it was the NP + M with 1.32%, followed by NF+M at 1.10% and YC + M at 1.02%. *Bacilli* abundance was observed in NP + M by 1.51%, followed by YC + M by 1.08% and NF+M by 0.99%. *Polyangia* class was only observed in NF+M and MP + M, which had 1.65 and 1.37%, respectively. *Acidimicrobiia* class was observed in MP + M, NF+M, and YC + M with 1.05, 0.99, and 0.87%, respectively. *Chloroflexia* was abundant by 0.94 and 0.86% in YC + M and NF+M treatments, respectively. *Ktedonobacteria* class was

only found in MP + M treatment by 1.22%, while the *Planctomycetes* class was abundant in NP + M treatment by 1.03%.

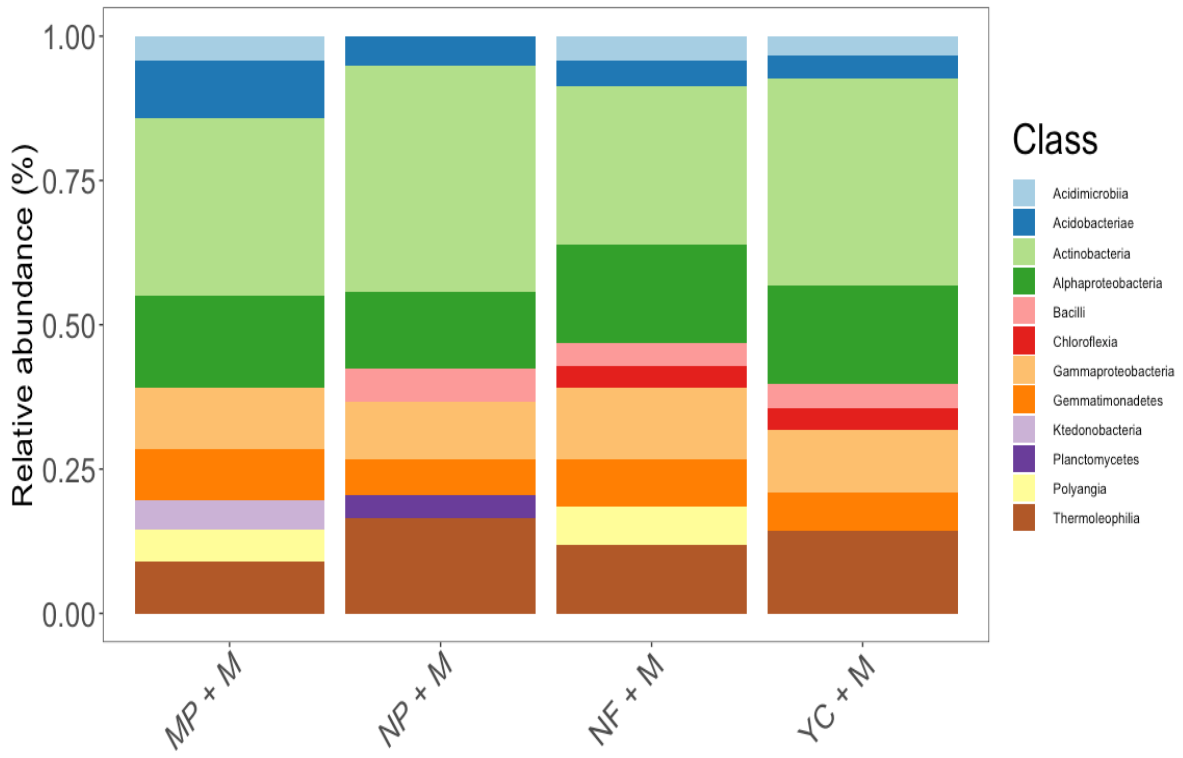

Figure 20: Relative abundance (%) of soil bacteria at Class level for each treatment in maize plantation

4.3.6 Composition of phyla community variation within the treatments

Figure 21 shows the total phyla observed in the maize treatments, including those phyla with <1%, in a heat map graphic representation. The most abundant phyla are indicated in red; the lowest represented phyla are indicated in yellow, and the white color denotes no phyla observed (Fig. 23). The lowest phyla in abundance in all treatments were the *Abditibacteriota*, *Entotheonellaeota*, *Fibrobacterota,* and *Thermoplasmatota*. The *Spirochaetota* were observed in (MP + M), (NF+M), and (YC + M) treatments, and *Elusimicrobiota* Phylum was observed only in (MP + M), (NF+M), and (YC + M) treatments. *Crenarchaeota* was observed only in (MP + M), *Halobacteriota* was observed in (NP + M) only, and *Latescibacterota*, *MBNT-15,* and *NB1-j* were observed only in (NF+M) treatment. Similarly, to Fig. 22, the most abundant phyla were *Actinobacteriota*, *Proteobacteria*, *Acidobacteriota*, *Chloroflexi*, *Gemmatimonadota*, *Planctomycetota*, *Myxococcota*, *Firmicutes*, *Bacteroidota*, *Verrucomicrobiota*, *Armatimonodota,* and *Patescibacteria* (Fig. 23).

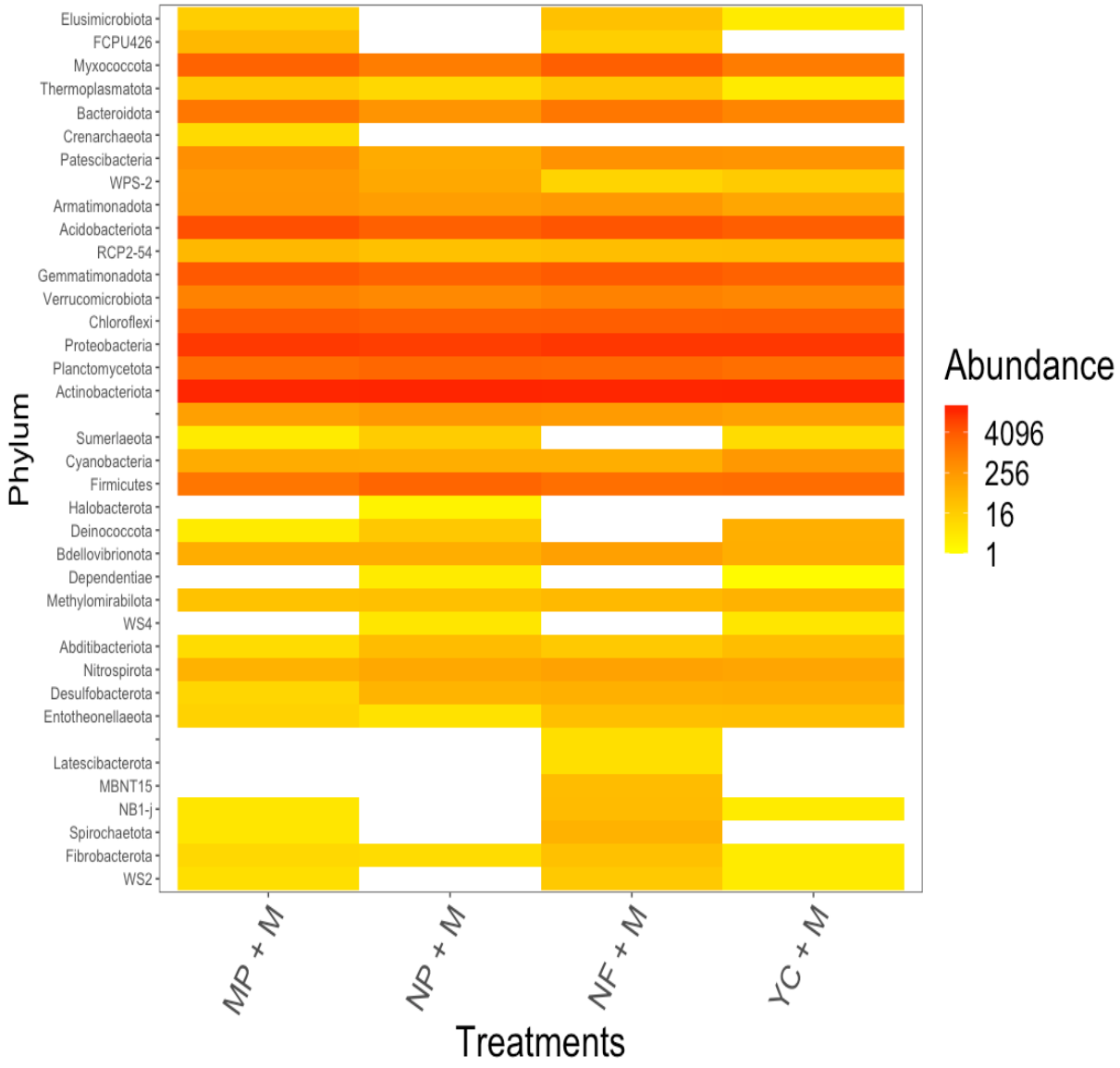

Figure 21: Heatmap indicating the relative abundance of bacteria Phyla in maize treatments

Fertilized with Minjingu Powder (MP + M), fertilized with Nafaka Plus (NP + M), not-fertilized (NF+M), and fertilized with YaraMila Cereal (YC + M).

4.3.7 Venn diagram analysis for unique Phyla, class and order of bacteria

The Venn diagram detected the number of common and unique bacterial species in all treatments (Fig. 22a). The results indicated that 293 species of operational taxonomic units (OTUs) (7.40% of all OTUs) were common to all treatments. The (NF+M) treatment has the highest number of bacterial OTUs reaching 822, and accounting for 20.77% of the total OTUs. The (MP + M) was the second with unique OTUs reaching 795, equivalent to 20.09% of the total OTUs samples. The YaraMila cereal treatment (YC + M) had unique OTUs of 648 (16.38%), while NP + M had 400 OTUs accounting for 10.11%.

Soil bacteria from all treatments belonged to a total of 31 Phyla. The number of shared Phyla in all the treatments is shown in a Venn diagram (Fig. 22b). All maize treatments shared 24 phyla, while three phyla were unique to the (NF+M) treatment accounting for 2.42% of the total phyla. In addition, the Venn diagram showed one phylum unique to the MP + M and another 1 Phylum to (NP + M), accounting for 0.81%, respectively. For the (YC + M) treatment, no unique phylum was observed. A total of 656 bacterial Orders were identified in all the maize treatments (Fig. 22c). Among them, 110 bacterial Orders were shared by all treatments accounting for 16.77% of the total. The most abundant Orders recorded were in the (NF+M) treatment with 23 orders, equivalent to 3.51% of total, followed by the YC + M treatment, with 14 unique orders accounting for 2.13%. The (NP + M) treatment had eight unique orders, equivalent to 1.22% of the total orders. The lowest bacterial orders were recorded in MP + M treatment, with only three orders accounting for 0.46%. The abundance of soil bacteria at Class level in all the treatments is visualized in a Venn diagram (Fig. 22d). The number of bacteria Classes in each intersection of the four treatments had 54 bacterial classes accounting for 17.20% of the total Classes. The (NF+M) treatment had the highest number of bacterial Classes (8), equivalent to 2.55%, followed by YC + M, equivalent to 2.23%. The NP + M had 3 bacterial Classes equivalent to 0.95%, and the MP + M treatment had only 1 unique Class of bacteria that accounted for 0.32%.

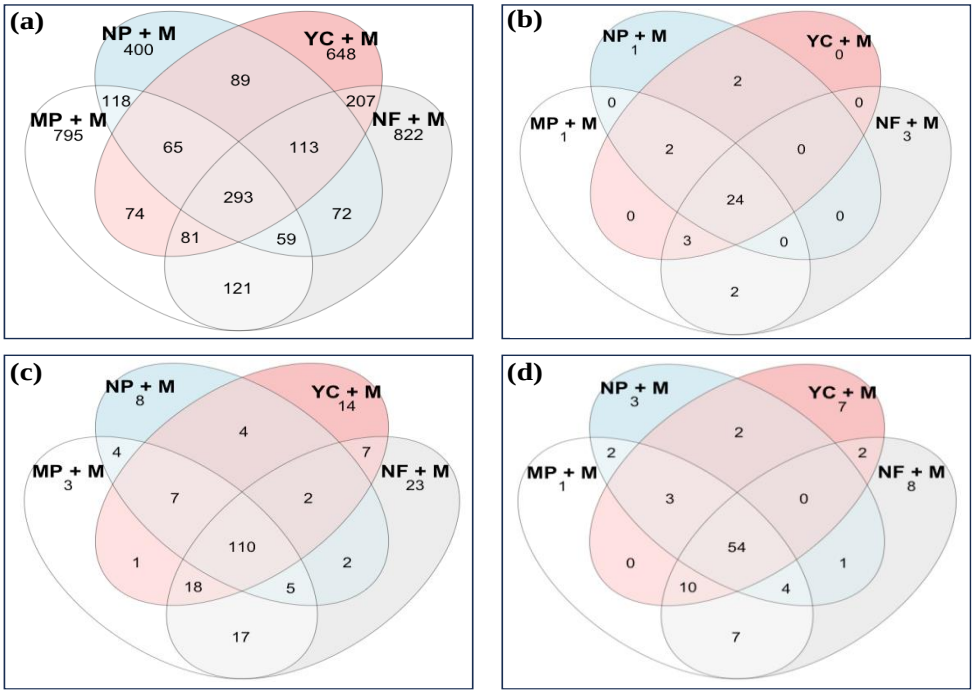

Figure 22: Venn diagram showing operational taxonomic units (a) OTUs, (b) phyla level, (c) bacteria order, and (d) bacteria classes Unfertilized (NF+M), YaraMila cereal (YC + M), Nafaka plus (NP + M), and Minjingu Powder (MP + M).

4.3.8 Observed, Chao1 and Shannon diversity index showing the treatment phylum level diversity

The Observed and Chao1 indices showed no significant change in bacteria phyla and their proportions. However, the Shannon diversity index observed a slight change in bacterial diversity (Fig. 23).

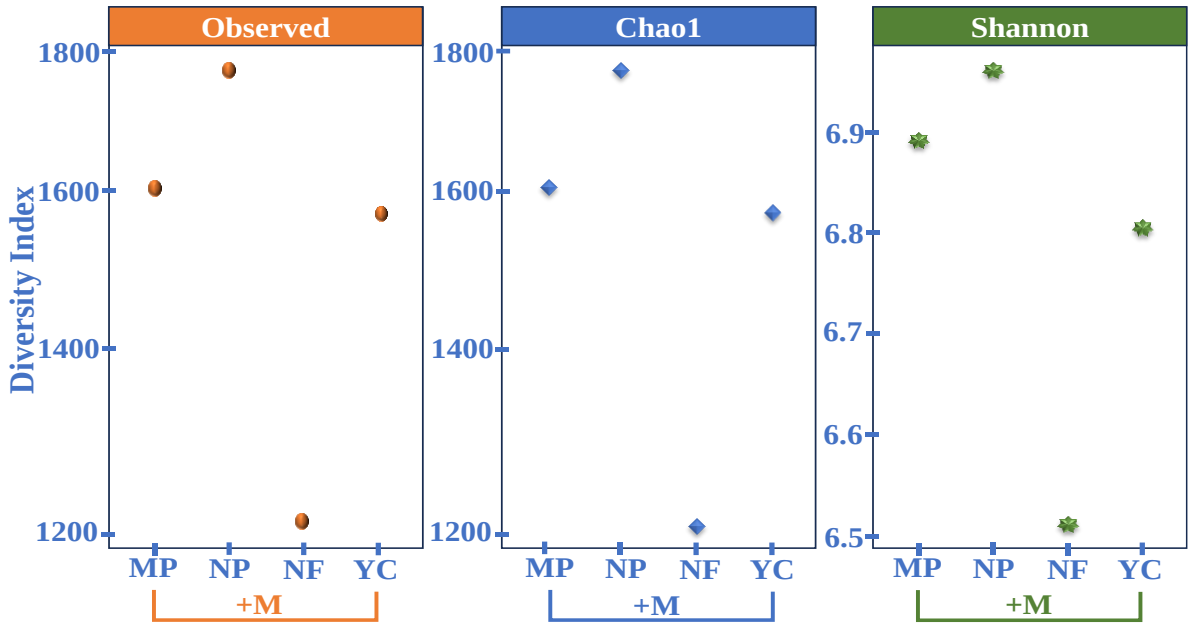

Figure 23: Diversity indices of species richness (Observed, and Chao1), and evenness (Shannon) at phylum level in treatments Not-fertilized (NF+M), YaraMila cereal (YC + M), Nafaka plus (NP + M), and Minjingu Powder (MP + M)

The Shannon index indicated a diversity change while the other indices (Observed and Chao1) show similar patterns. This could be explained due to each index's sensitivity and underlying mathematical properties. Observed index measures species richness by counting different unique taxa present in the samples without considering their abundances. It does not show significant changes if the variations are contributed by species richness and not their relative abundances. The Chao1 index, measure species richness similarly to the Observed index, except it only counts unique, rare taxa and assumes that many taxa are present but not observed. It is less sensitive to changes in the relative abundances of taxa. The Shannon index considers species richness and evenness by assigning higher weights to abundant and rare taxa. It is very sensitive to diversity changes associated with the relative abundances of taxa.

The higher U residual contents in soils due to fertilization with NP and MP showed an abundance of bacteria diversity. The higher U was observed to enhance the abundance of bacteria diversity in (NP + M) treatment (Fig. 22). The *Actinobacteriota* phylum was mainly abundant across the treatments (Fig. 23). However, the NP + M treatment showed more

abundance, followed closely by the YC + M treatment indicating that the more nutrient elements of these fertilizers could have favored the *Actinobacteriota* phylum by 12.42% (Fig. 21). NP + M, besides having N, P, K, S, Mg, and Zn, which were also found in YC + M, had also Ca and B nutrients (Lisuma *et al.,* 2022; Mwalongo *et al.,* 2023). The MP + M treatment was found to release the content of U in soils next to NP + M, composed mainly of P, Ca, and to a lesser amount of Mg (Mwalongo *et al.,* 2023) had 8.98% of *Actinobacteriota* phylum a bit higher than NF+M, which had 8.74%. *Proteobacteria* phylum showed a uniform distribution trend across the fertilizer treatments indicating to be very stable in different types of fertilizer applications.

The NF+M treatment had a higher unique bacterial order and class, followed by YC + M, NP + M, and MP + M. The lowest bacterial orders and classes in NP + M and MP + M could be related to its U residual contents, which negatively affected the bacteria orders compared to the YC + M treatment (Fig. 23c, d). The top three orders of soil bacteria across the treatments were *Frankiales* (15.91%), *Rhizobiales* (13.47%), and *Burkholderiales* (12.96%). The abundance of *Frankiales* bacterial order related to its role of fixing nitrogen to the maize rhizosphere, was higher to the NP + M, MP + M followed by YC + M and NF+M treatments. *Pseudonocardiales* order into the class *Actinobacteria* was more abundant in MP + M and NP + M plots, implying that these fertilizers with U impurities favored mostly the class of *Actinobacteria*. *Acidobacteriales* order a class of *Acidobacteriia*, and *Ktedonobacterales* order a class of *Ktedonobacteria*, were only rich in MP + M treatment, indicating that they could have a role in soil P cycling and regulation of P availability in soil (Bergkemper *et al.,* 2016; Dai *et al.,* 2020; Mason *et al.,* 2021). *Chloroflexia* class was not found in MP + M and NP + M (Fig. 20) as these treatment fertilizers have a substantial amount of P. In addition, NP + M was also rich in N (Mwalongo *et al.,* 2022). *Bacillales* order, a class of Bacilli, was rich in NP + M treatment only, which was also found to have high U soil residual. Tang *et al.* (2021) also revealed a class of *Bacilli* that increases more in higher U concentration.

The bacterial diversity in the fertilizer treatments was observed to be higher than that in the unfertilized treatment, indicating the bacterial species abundance and richness were influenced by fertilization (Fig. 22b- 22d) and increasing their activity in soils. The current results revealed an abundance of *Actinobacteriota*, *Proteobacteria*, *Acidobacteriota*, *Chloroflexi*, *Gemmatimonadota*, *Planctomycetota*, *Myxococcota*, *Firmicutes,* and *Bacteroidota* in soils fertilized with higher U residuals. The higher bacteria richness and diversity for NP + M favored the environment for bacteria growth due to the increase in U concentrations released

to the soils. *Actinobacteriota* phylum was unique and abundant in NP + M. contrarily, the MP + M treatment had a bit lower bacteria richness and diversity as its released U in soil was lower than that of NP + M treatment, and *Acidobacteriota* were unique and abundant MP + M. Moreover, the YC + M fertilizer treatment, which had the lowest insignificant U concentration in soils, had the least bacteria richness and diversity, probably due to its absence of radiological nutrients that did not affect the bacteria environment.

Crenarchaeota was found only in MP + M treatment with higher content of U (Fig. 22a). However, following its application in soils, the large concentration of U could have been leached to the ground and created a favoring environment for *Crenarchaeota*. Weidler *et al.* (2008) revealed *Crenarchaeota* to have a role in the N cycle in subsurface radioactive soil. Furthermore, a recent study by Tang *et al.* (2023) demonstrated that *Crenarchaeota* was among the phylum observed to be better adapted and survived in heavy metal-contaminated areas. *Halobacteriota* was found only in NP + M treatment (Fig. 22a and Fig. 23), indicating tolerating higher U concentration, but it also could be involved in the cycling of S content in soils as NP fertilizer has a higher percentage of sulfur content (Mwalongo *et al.,* 2023).

The unfertilized treatment (NF+M) had the significantly lowest bacteria richness and diversity, indicating that without adding nutrients to soils, it is not affecting bacteria richness and diversity. *Latescibacterota*, *MBNT-15,* and *NB1-j* were unique (Fig. 22a and Fig. 23) and only found in the unfertilized plot (NF + M), signifying to promote the natural rhizosphere microbial interactions (Liu *et al.,* 2022). Recent studies have actually linked *Latescibacterota* to have a significant correlation with microbial interactions (Liu *et al.,* 2022; Yu *et al.,* 2023). Despite all these findings further research studies are needed to explore the agronomic role of *Latescibacterota* and other unidentified bacteria under the unfertilized regime.

4.4 Influence of U derived from phosphate fertilizers on soil to maize uptake

The results from field and pot experiments which assessed the effect of U uptake through the application of fertilizers with different U concentrations. Additionally, the effects of treatment with kaolin and *Eucalyptus globulus ssp*. *maidenii* were evaluated. The findings are presented and discussed in this section.

4.4.1 Uranium Concentration in Soil and maize compartments in the field experiment

The concentrations of U in soil and maize plant compartments (roots, stems, leaves and grains) for the field were assessed. The U concentration in soil and maize compartments (roots, stems,

leaves and grains) were statistically significant (F= 4224.1; $P < 0.001$). The results also showed statistically significant (F =1677.69; $P < 0.001$) variations between different fertilizer treatments. Figure 24 show the post hoc test for U concentration in soils, roots, stems, leaves and grains of the maize plants. The results shows that U concentration trends followed the order of soil > root > Stem > leaf > grain as shown in Fig. 24.

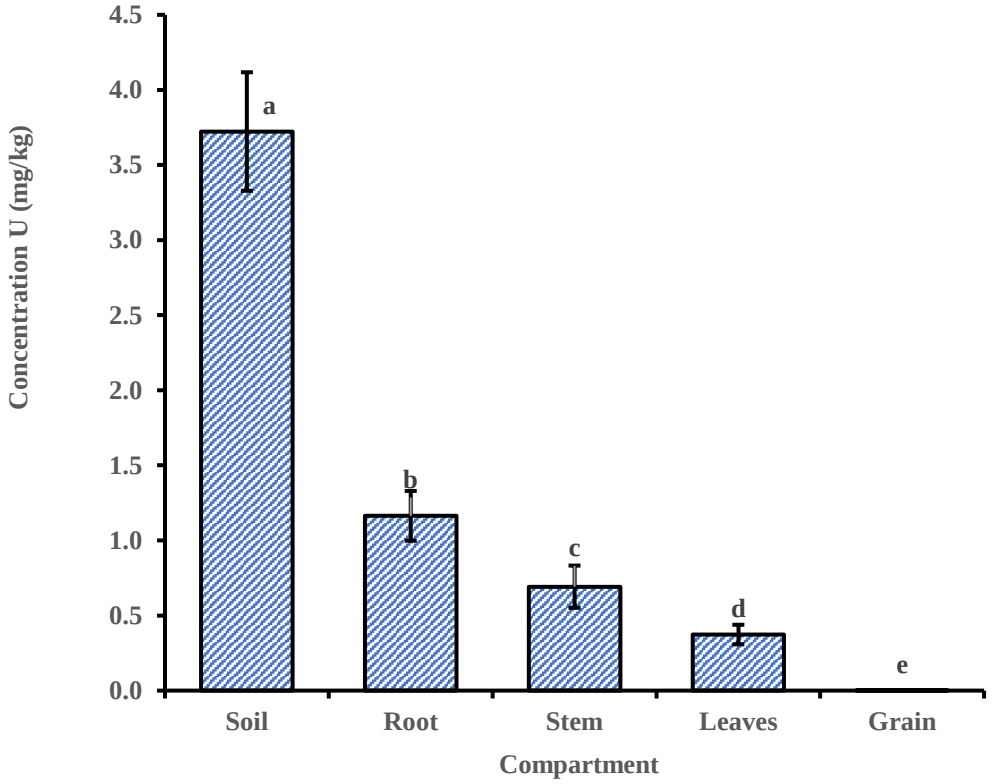

Figure 24: The overall U distribution in the soil and maize compartments

Uranium concentration in soils and maize compartments after treatment with fertilizers in the field experiments are shown in Fig. 25. The results show that U concentrations for fertilized soils were higher than the unfertilized soils (control). It was also demonstrated that there were no appreciable variations in U content between YaraMila Cereal and unfertilized soil. On the other hand, there were significant variations in U concentrations in soils and maize compartment (F=342.1, $P < 0.001$). MP had high U concentration than NP fertilizer, interestingly the result show that nafaka Plus (NP) fertilized soils had higher U concentration than MP fertilized soils. The high U concentrations in NP compared to MP may be attributed to the fact that MP fertilizer was in powder form which might have been easily leached (washed out) compared nafaka Plus which was in the form of granules. The result also shows that there were no significant differences in U content in soils treated with YaraMila Cereal fertilizers compared with the unfertilized soil plots.

U concentration in roots ranged from 0.53 ± 0.02 to 2.39 ± 0.03 mg kg^{-1}. The roots from fertilizer treated soils contained significantly higher ($P < 0.05$) U concentration than the roots from unfertilized soil. U concentration in roots from fertilizers treated soils were higher than the roots from unfertilized soils. These results agree with other researchers whereby, Stojanović *et al.* (2013) reported that, there is U uptakes by maize plants from different types soils which have been consistently fertilized for over 40 years. The results from this study showed that, roots of maize crop grown in fertilized soils absorbed much more U than those grown in unfertilized soils. Similarly, NP treated fertilizers' soils had higher U concentration followed by MP treated fertilizers soils (Fig. 25). U concentration in the stems of maize crop ranged from 0.35 ± 0.01 to 1.88 ± 0.01 $mgkg^{-1}$. The U concentration in the stems of maize crops in fertilized soils was higher compared with the stems of maize crops in unfertilized soils.

The U concentration in the leaves of the maize crops ranged from 0.24 ± 0.02 – 1.68 ± 0.05 $mgkg^{-1}$. NP treated fertilizers had higher U concentration followed by MP and lowest in YC treated fertilizers. The U concentration in stems of the maize crops in the unfertilized and in stems of the maize crops in YC treated were non significantly different ($p > 0.001$). Moreover, the U concentration in the grain ranged from 0.13 ± 0.03 to 0.96 ± 0.05. The results from this study show that fertilizer treated soils had significantly high U concentration in maize grains than the unfertilized soils Fig. 25). In similar to other compartments, U concentrations in grains was relatively higher in NP treated fertilizers followed by MP treated fertilizers and lowest in YC fertilized soils.

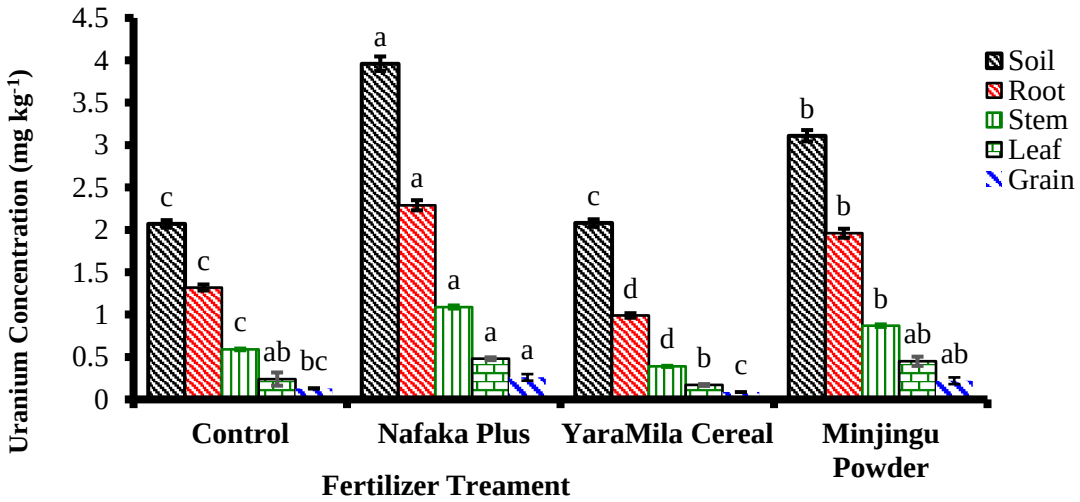

Figure 25: Uranium in Soil and maize crop compartments for the field experiment

The fraction of U transfer from soil to maize compartments (roots, stems, leaves and grains) were calculated using equation 14. The calculated concentration ratios from soils to roots

(CR_{SR}), soil to stem (TS_{SS}), soil to leaves (CR_{SL}) and soil to grain (CR_{SG})) are shown in Table 20. The results show that the CRs from soils to different maize compartment were higher for fertilized soils than unfertilized soils. The overall behavior of the CRs were higher in the order of decreasing for treatment of NP, MP, YC and unfertilized (NP > MP > YC > unfertilized).

The U CR_{SR} ranged from 0.34 ± 0.01 to 0.60 ± 0.02 while that of CR_{SS} ranged from 0.24 ± 0.02 to 0.47 ± 0.02, CR_{SL} ranged from 0.18 ± 0.02 to 0.42 ± 0.02 and CR_{SG} ranged from 0.12 ± 0.01 - 0.24 ± 0.02. The results show that CRs from soil to all maize compartments were highest for NP treated fertilizers and lowest in YC. The U concentration in NP fertilizer was lower (147.65 ± 8.61) compared to MP (159.67 ± 10.48). Since MP was in powder form which might have leached easily, the high concentration could have been caused by the fact that NP remain in the soils for longer time because it was in granules. Studies show that granulated fertilizers release nutrients slowly for long time compared to ungranulated (powder) (Šarauskis *et al.,* 2021). The Concentration ratio calculated using Equation 14 are presented in Table 20.

Table 20: Uranium from soil-to-plant transfer factors dry weight maize crop to dry weight soil

Treatment	Concentration ratios (CR)			
	CR_{SR}	**CR_{SS}**	**CR_{SL}**	**CR_{SG}**
Control (unfertilized)	0.26 ± 0.03	0.17 ± 0.01	0.12 ± 0.01	0.06 ± 0.01
Nafaka Plus (NP)	0.60 ± 0.02	0.47 ± 0.02	0.42 ± 0.02	0.24 ±0.02
YaraMila Cereal (YC)	0.34 ± 0.01	0.24 ± 0.02	0.18 ± 0.02	0.12 ± 0.01
Minjingu Powder (MP)	0.47 ± 0.03	0.31 ± 0.01	0.27 ± 0.01	0.20 ± 0.01
IAEA maize				0.087

4.4.2 Uranium concentrations in soils and maize compartments from screenhouse experiment

The screen house experimental results were obtained and compared based on the three treatments combination and one negative control. The first treatment was where the NP, MP, YC fertilizers were used in the pot experiment. The second treatment results involved NP, MP and YC mixed with Kaolin. The third results involved NP, MP and YC mixed with powder of *Eucalyptus ssp maidenii* bark and the fourth treatment NP, MP and YC fertilizers mixed with kaolin and *Eucalyptus ssp. maidenii* bark. The Combination of the three treatment results is shown on Fig. 26 and Fig. 27.

The results showed that multivariate tests of significance f or U concentration in soils and maize compartments in all treatments in the screen house pot experiment were statistically significant. The post hoc statistical test showed than U concentration in NP and MP alone or with

combination with kaolin and *Eucalyptus globulus ssp. maidenii* bark were higher than unfertilized maize crop. However, the result showed non significance differences between U concentrations in YC and negative control. Overall results showed that MP fertilizer treated soils had higher U concentrations than the NP fertilizer treated soils. Since the MP fertilizers effectively had a higher U content than the NP fertilizers treated soils, this result defies the conclusions from the field experiments. The difference is attributed by the fact that pot experiments were set in closed system whereby watering process did not allow the MP fertilizers to leach.

U concentration in soils with different treatments ranged from 0.63 ± 0.03 to 6.18 ± 0.05 $mgkg^{-1}$. The results indicated that the soils in the screen house pot experiment where maize crop was cultivated treated with individual three fertilizers (YC, MP, and NP) along with kaolin and *Eucalyptus globulus ssp. maidenii* bark were statistically significant ($p < 0.001$). The Post hoc statical analysis showed that U concentrations in the pot experiment was higher in all MP treated fertilizer compared NP and YC.

It was observed that, U concentration in roots ranged from 0.090 ± 0.001 to 3.64 ± 0.02 $mgkg^{-1}$, which were higher compared to other maize compartments. These results are supported by others several studies who reported that different plant species exhibit different levels of U accumulation. According to these studies high concentration of U tended to accumulate more in the roots compared with other part of the maize plant (Charro & Moyano, 2017; Fresquez *et al.,* 1998; Stojanović *et al.,* 2013). The results also, further show that, U concentrations in the roots of MP fertilizer treated with kaolin had highest U uptake compared with other treatments. This may be attributed by adsorption of U by kaolinites which occur due to adsorption interaction with uranyl ion (UO_2^{2+} ions) which has high surface area (Campos *et al.,* 2013; Guerra *et al.,* 2010; Issa *et al.,* 2023).

The root U uptake was lowest in unfertilized soil and in MP and NP fertilizers combined with *Eucalyptus* and kaolin (Fig. 27). It was also, found that, the U concentration was below the detection limit for YC. It is interesting to note that the combination of MP and NP fertilizers with *Eucalyptus globulus spp maidenii* bark powder and kaolin reduced U uptake by almost 4 folds when compared with other fertilizers. This may be attributed to the organic matter derived from *Eucalyptus ssp maidenii* as it has been reported elsewhere to limit the bioavailable fraction of U in soil matrix because it contain carboxylic and phenolic functional groups which has the tendence to bind U at low pH (Cumberland *et al.,* 2018).

U concentration in stem ranged from 0.004 mg kg^{-1} to 2.62 $mgkg^{-1}$. In the stem of maize crop, soils that had been fertilized with MP, had the highest U concentrations, while in soil treated with YC, U concentrations was the lowest (Fig. 28). The trend of U concentrations in the maize crop compartments influenced by fertilizers treatments followed the order of MP < NP < YC < Control.

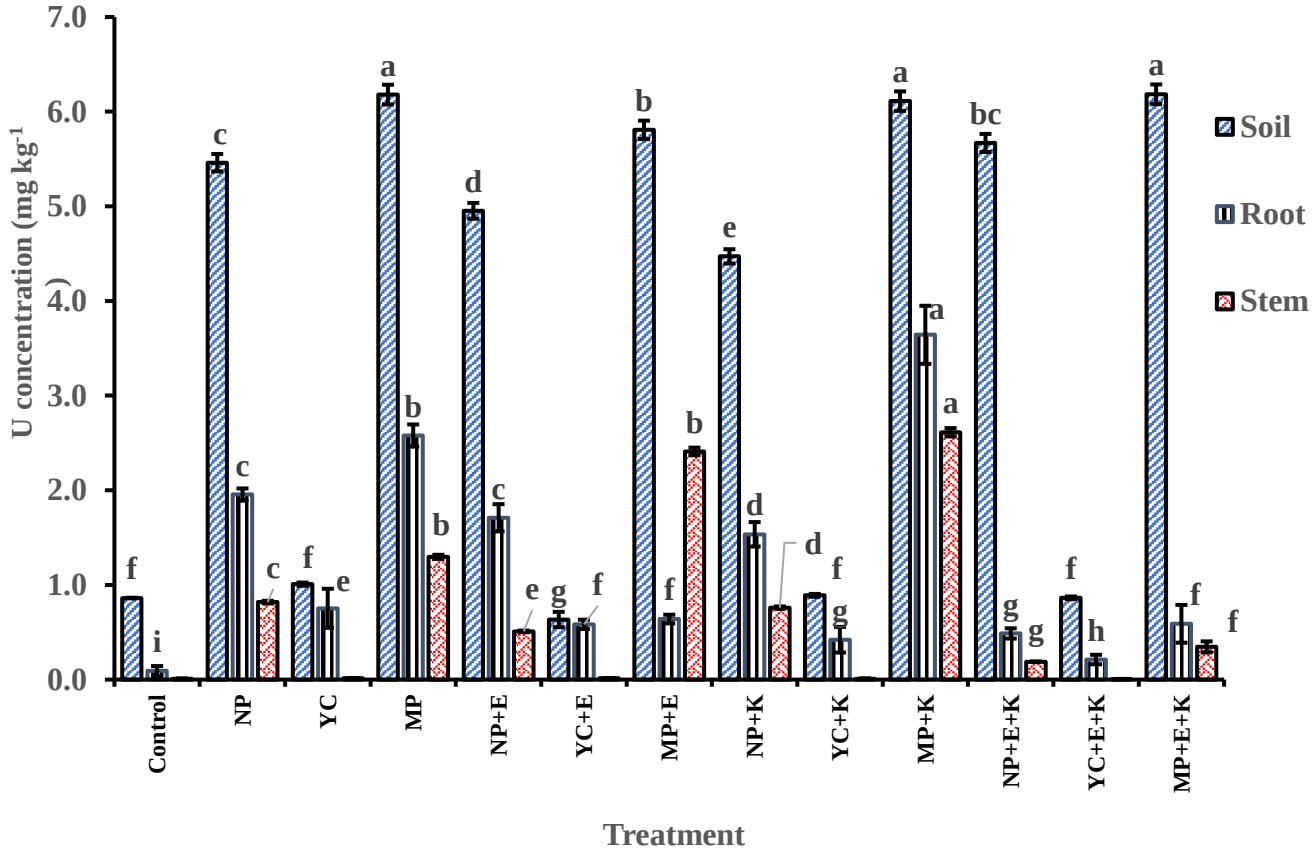

Figure 26: Uranium concentration in soils, roots and stems from the pot experiment

Figure 27 shows U concentration in leaves with different treatments. The U concentrations in leaves of maize crops from screen house experiment ranged from 0. 002 $mgkg^{-1}$ to 1.23 mg kg^{-1}. The U concentrations in the leaves of maize crops grown in the negative control were lower than in the leaves of maize crops grown in the fertilized soils. It was found that U concentration in the leaves of maize crops grown in the soil treated with MP fertilizers were higher than in the leaves of maize crops grown in the NP treated fertilizers.

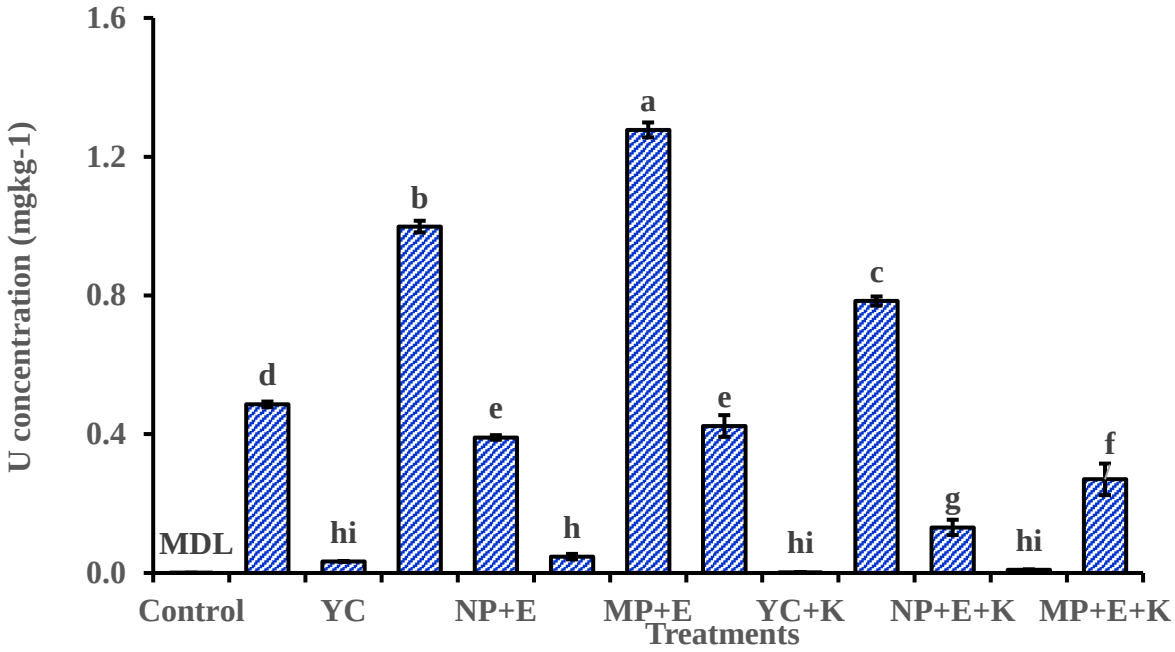

Figure 27: Uranium concentration in Maize leaves from the pot experiment

Moreover, U concentrations in the grains ranged from MDL to 0.006 mgkg^{-1}. The grains from MP and NP fertilized soils where maize crops were grown exhibited slight elevated U concentrations compared with unfertilized soils. Despite high U concentration in grains of maize crops fertilized with MP and NP fertilizers the uptake of U was relatively low. It is worth noting that MP and NP fertilizers combined with *Eucalyptus globulus ssp maidenii* and kaolin showed relative lower U uptake as shown in Fig. 28.

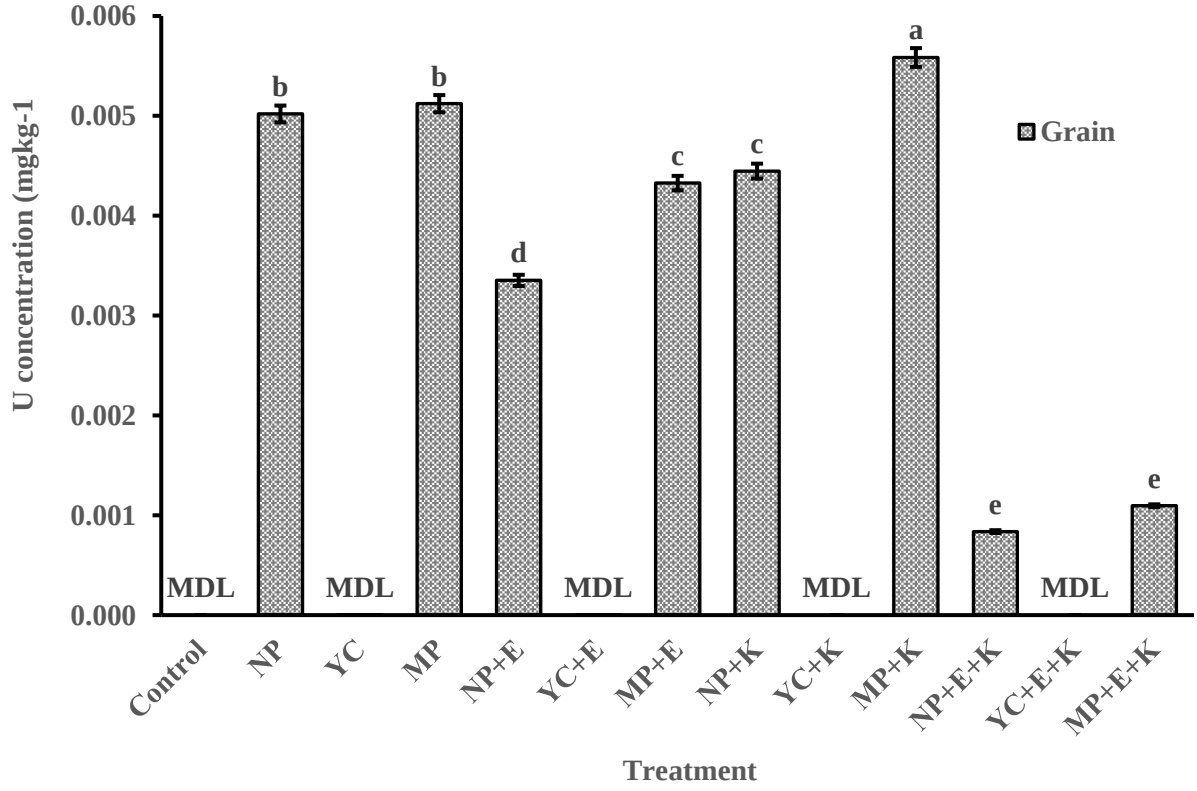

Figure 28: Uranium concentration in maize grain for the pot experiment

Through a variety of human activities, including the mining and mineral processing of all base metals, the burning of coal, and the use of fertilizers based on phosphorus, U heavy and radioactive metals is gradually added to soils. For instance, in Nigeria, Okeji *et al.* (2012) revealed that the main radioactivity of the phosphate ore used for fertilizer production was due to ^{238}U and was responsible to be transferred to the plants after application. Also, it was indicated that the average activity concentration due to ^{238}U in phosphate ore from Nigeria is higher than that from Algeria, Egypt, Sudan and Tunisia.

U concentration in soils endangers both human and animal health once gets into food chain (De Souza Braz *et al.,* 2021). Several governments around the world are reluctant to reduce U contamination in agricultural soils by putting in place regulatory control as it may affect food quality. Extracting of U as mineral commodity by product during phosphate fertilizers is not economically attractive because of low prices in the world market. Extraction of U from P fertilizers as an environmental contaminant is distant preposition.

Reducing U uptake by edible plant parts is an attractive preposition in order to reduce heavy metal and radiation exposure in food chain. The application of *Eucalyptus globulus ssp maidenii* bark powder which is normally discarded as waste during timber production may be a potential candidate for U locking in soils. The result showed lowest U was translocated from soils to maize compartments in pots where *Eucalyptus globules ssp. maidenii* and kaolin were applied together. Therefore, the ratio of U content in soil to U content in maize compartments and plotted as shown in Fig. 27 & 28). The sharp reduction of U to maize compartments may be attributed by *Eucalyptus globules ssp maidenii* because is an organic matter which reduces the mobile of hexa-uranyl ion to immobile tetra-uranyl compound.

Additionally, it has been reported that the uranyl ion UO_2^{2+} has a chelating effects depending on soil pH, Eh, organic and inorganic ligand concentration and type, water movement and mixing rate, and adsorption, desorption, complexation, and precipitation reactions (Chen *et al.,* 2021). These reactions also directly affect U behavior and, as a result, its undergo speciation (Gupta & Walther, 2019). The U reduction ratio from soil to roots was the lowest while from soil to grain was the highest. Implying that most U was absorbed in the roots and less transferred to stem, leaves and maize grains.

The study investigated U uptake in maize plants treated with various fertilizers with varying U concentration in both field and controlled pot conditions. The findings highlight minimal

uptake of U by maize plant and its concentration depends of U content in the fertilizer. Soil amendment using *Eucalyptus globulus ssp maideii* bark can reduce U bioavailability by maize. Implementing such strategies could help manage U entry into food chain safeguarding human and environmental health. The study recommends further studies to understand the mechanisms of kaolin and *Eucalyptus globulus ssp maidenii* in reducing U uptake by plant.

CHAPTER FIVE

CONCLUSION AND RECOMMENDATIONS

5.1 Conclusion

This study assessed U concentrations in major PRs and common PFss used in Burundi, Kenya, Rwanda, Tanzania, and Uganda. The concentration measurements were performed using energy-dispersive X-ray fluorescence spectrometry. The findings showed that the concentrations of uranium in phosphate rock varied from 10.7 mg kg^{-1} (Mrima Hill deposit, Kenya) to 631.6 mg kg^{-1} (Matongo deposit, Burundi). On the other hand, the concentrations in PFs varied from 107.9 mg kg^{-1} for an imported fertilizer to 281.0 mg kg^{-1} for a domestic fertilizer made from Tanzania's Minjingu PRs. In assessing the contribution of PFs application on enhancement of natural radioactivity from primordial radionuclides (^{232}Th, ^{238}U and ^{40}K) in agricultural soils and tobacco plant. The study evaluated the activity concentrations of ^{232}Th, ^{238}U, and ^{40}K after application of NPK fertilizers used for tobacco-production in Kenya, Tanzania, and Uganda.

Additionally, investigation influence of application of NPK fertilizer on the radioactivity in agricultural soils and tobacco plants in Kenya, Tanzania, and Uganda was performed. The study design encompassed three components: a one-year controlled experiment with tobacco growing in plots; a ten-year semi-controlled trial on well-managed tobacco farms; and a farmers' practice field in Migori (Kenya); Urambo (Tanzania); and Kanungu (Uganda). The results showed that the activity concentrations of ^{232}Th, ^{238}U, and ^{40}K in soils and tobacco leaves exposed to NPK fertilizers with elevated radioactivity were significantly greater (at all locations) than in the control samples (which did not use NPK fertilizers). The draw evidence that continuous application of NPK fertilizers raises concentrations of ^{232}Th, ^{238}U, and ^{40}K in agricultural soils and pose radiological hazards to the public. Tobacco snuffing and smoking increase an effective dose from 2.41 to 6.53 and 1.14 to 2.45 times greater than the average yearly dose that the general public receives from inhaling natural radionuclides, respectively. Tobacco consumers may be exposed to serious radiological risks and increase lifetime excess cancer risk for smokers and snuffers.

The influence of U derived from three phosphate fertilizers brands with difference U concentrations on soil bacterial diversity in sandy loam soils in Tabora, Tanzania. The results showed that, the PFs derived from Minjingu powder was noted to have higher U concentration.

The observed rise in U concentration was linked to a notable increase in both the abundance and diversity of bacteria within the soil. Interestingly, certain bacterial groups showed unique responses to these changes. For instance, the *Halobacteriota*, found in the lowest abundance, were distinctive to the Nafaka plus treated soil due to their capability to withstand higher U concentrations. Meanwhile, the *Crenarchaeota,* also present in low abundance, were unique to the Minjingu powder treated soil, which had a slightly lower U concentration compared to Nafaka plus treated soils, indicating their resilience to U concentration. In plots that received no fertilizer (control), specific bacterial groups like *Latescibacterota* were uniquely present, suggesting that these bacteria thrive only in unfertilized conditions. In the treatment that resulted in the highest soil U concentration of 3.93 mg kg^{-1}, the *Bacillales* order within the *Bacilli* class was uniquely persistent, demonstrating its ability to tolerate elevated U levels. Additionally, the *Pseudonocardiales* order in the *Actinobacteria* class were abundant in both Minjingu powder and Nafaka plus treated soil plots, reflecting their tolerance to higher U concentrations. On the other hand, the *Rhizobiales* order within the *Alphaproteobacteria* class was found to be abundant in the YaraMila cereal treated soils, which had an insignificant release of U into the soil.

Finally, the study investigated the effects reducing U concentration from soil to maize plant by treating soil using kaolin and *Eucalyptus globulus* ssp. *maidenii* which contain carboxylic acid functional group. In field and screen house pot experiments. There phosphate fertilizers used were Minjingu powder, YaraMila cereal and nafaka plus which had different U concentrations. In both and field experiments, the results showed that maize grown in fertilized soil exhibited slightly elevated U concentration in soil and maize plant compared to maize grown in unfertilized soil. It was an evident that plots treated by combining phosphate fertilizer with *Eucalyptus globulus* ssp. *maidenii* bark and kaolin showed U concentration reduction from soil to maize plant.

5.2 Recommendations

This study found high uranium concentration in phosphate rocks and phosphate fertilizers used in Burundi, Kenya, Rwanda, Tanzania and Uganda. The influence of application of NPK fertilizers on enhancing radioactivity of agricultural fields and tobacco plant uptakes was also found in selected sites in Kenya, Tanzania and Uganda. The field experiments showed application of fertilizes with difference U concentrations influenced bacteria abundance and diversity. Finally, combination of *eucalyptus globulus ssp maidenii* bark powder and kaolin

was found to reduced U uptake from soil to maize crop. Based on these finding, this study recommends the following:

(i) Investigate both technical and economic aspects of uranium extraction methods to determine their viability.

(ii) Further studies are recommended to investigate the concentration of polonium-210 in tobacco leaves in order to further assess its contribution to radiological risk.

(iii) Further studies should be done to investigate the adsorption and interaction of *Eucalyptus globulus ssp maidenii* and Kaolin in reducing uranium uptake from soil to plants.

(iv) Further detailed studies should be conducted understand influence of fertilizer derived U on bacterial diversity.

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RESEARCH OUTPUTS

(i) Publications

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(ii) Poster presentation