GAMMA-RAY SPECTROMETRY OF NATURALLY OCCURRING RADIOACTIVE MATERIALS (NORM) FROM BIRNIN GWARI ARTISANAL GOLDMINE OF KADUNA STATE, NIGERIA

BY

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MSc/SCIE/07169/2008 – 2009

MAY, 2013.

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A thesis submitted to the School of Post Graduate Studies, Ahmadu Bello University, Zaria. In Partial fulfillment of the requirement for Award of Masters Degree in Physics, Department of Physics, Faculty of Science Ahmadu Bello University, Zaria Nigeria

May, 2013.

DECLARATION

I hereby declare that the work in this thesis titled Gamma-Ray Spectrometry of Naturally Occurring Radioactive Materials (NORM) from Birnin Gwari Artisanal Goldmine of Kaduna State, Nigeria was performed by me in the Department of Physics under the supervision of Dr. Rabiu Nasiru and Dr. Zakari I. Yusuf.

The information derived from the literature has been acknowledged in the text and a list of references provided. No part of this work has been presented for another degree or diploma at any institution.

CERTIFICATION

This thesis titled Gamma-Ray Spectrometry of Naturally Occurring Radioactive Materials (NORM) from Birnin Gwari Artisanal Goldmine of Kaduna State, Nigeria meets the regulations governing the award of the degree of Masters of Physics of the Ahmadu Bello University, and is approved for its contributions to acknowledge and literary presentation.

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ABSTRACT

Mining has been identified as one of the potential sources of exposure to naturally occurring radioactive materials (NORM). Whilst the developed countries have identified NORM as potential problems and measures are being taken to address the issue very little is being done in the developing countries. However artisanal miners and mining companies are not being regulated for NORM in Nigeria. Birnin Gwari is one of the largest goldmines in Nigeria and has been in operating for the past 80 years with no data on radioactivity levels. This study was carried out to determine the activity concentrations of ^{238}U, ^{232}Th and ^{40}K from the gold ores mine at Birnin Gwari gold mine using instrumental neutron activation analysis (INAA) technique. The mean activity concentrations for ^{238}U, ^{232}Th and ^{40}K in soil/rock samples were found to be 37.36 ± 5.45, 62.69 ± 6.33 and 997.57 ± 199.77Bq/kg respectively. The results from this study are slightly higher when compared with typical world average of 33, 45 and 420 Bq/kg for ^{238}U, ^{232}Th and ^{40}K respectively. The mean absorbed dose rate due to these radionuclides is 96.52 ± 9.62nGy/h which is also higher than 60nGy/h world average. The mean annual effective dose rate to the public in the area was estimated to be 0.118 ± 0.012mSv. The mean annual effective dose rate obtained in this work is lower than 1mSv/year dose limit recommended by ICRP for public radiation exposure control.

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ABBREVIATIONS

WHO	-	World Health Organization
ICRP	-	International Commission For Radiation protection
BSS	-	Basic Safety Standard
IAEA	-	International Atomic Energy Agency
UNSCEAR	-	United Nation Scientific Committee on the effect of Atomic Radiation
NORM	-	Naturally Occurring Radioactive Material
NNRA	-	Nigeria Nuclear Regulatory Authority
INAA	-	Instrumental Neutron Activation Analyses
MMSD	-	Ministry for Mine and Steel Development
BIF	-	Basic Instruction Formation
RIR	-	Reduce Instrusion Related
TAG	-	Thermal Aureole Gold
OIR	-	Oxidized Instrusion Related
VMS	-	Volcano Magnetic Supper enrichment
UV	-	Ultra Violet
BG	-	Birnin Gwari
GPS	-	Global Positioning System
NRR	-	Nigeria Research Reactor
PPM	-	Part Per Million

CHAPTER ONE

1.0 INTRODUCTION

1.1 General Introduction

Mining is a global industry undertaken for its economic benefits of wealthy creation and employment. In African commercial scale mining provides important benefits in terms of exports/foreign exchange earnings and tax receipt to nineteen African countries (Hayumbu, and Mulenga, 2004).

Beside the socio-economic benefits of the mining industry in the developing countries such as Nigeria, the industry may be faced with three potential negative effects. The first one is the socio-economic dislocation all ill-prepared mining communities go through at mine closure, which arise from exploitation of a non-regenerative resources (Hayumbuand Mulenga, 2004). The second and third undesirable aspects arise when non-optimal management of mining operations results in environmental degradation and /or negative health impacts on miners and mining communities. Principal health problems among miners and mining communities from various countries that have been cited by the literature include respiratory disease, neoplasm/cancer, chronic hypertension, mental health and genetic impact (WHO, 1999) The major cause of these disease can be attributed to the heavy metal contamination and naturally occurring radioactive materials (ICRP, 1994).

Mining and industrial processing are among the main sources of heavy metal contamination in the environment. Mining activities, through milling operations

coupled with grinding, concentrating ores and disposal of tailings, along with mill wastewater provide obvious sources of heavy metal contamination of the environment. It is, therefore, not surprising that the degree and extent of heavy-metal pollution as a result of human activities has been one of the main topics studied in environmental geochemistry. Heavy metals can cause health problems at higher exposures and destroy aquatic organism when leached into water bodies. Metals contamination in aquatic environmental has received huge concern due to their toxicity, abundance and persistence in the environmental and subsequent accumulation in the aquatic habitats. (Boamposem *et al.,* 2010).

Heavy metal residues in contaminated habitats may accumulate in microorganisms, aquatic flora and fauna, which in turn may enter the human food chain and result in health problems like the lead poisoning problems in Zamfara State that kills more than 400 children. (Weekly Trust, 2011).

Human beings are continually being exposed to ionizing radiation from natural sources. There are two main contributors to natural radiation exposures: high-energy cosmic ray particle incident on the earth's atmosphere and radioactive nuclides that originated from the earth crust and are present everywhere in the environment, including the human body (UNSCEAR, 2000).

Humans are exposed to radionuclides through ingestion and inhalation (internal exposure) and/or irradiation from external gamma rays emitted from the radionuclide (external exposure).

The International Basic Safety Standards (BSS) for protection against ionizing radiation and the safety of radiation sources (IAEA, 1996) specify the basic

requirement for the protection of health and the environment from ionizing radiation. These are based on the latest recommendations of the International Commission on Radiological Protection (ICRP, 2007) on the regulation of Practices and interventions. The BSS is applied to both natural and artificial sources of radiation in the environment and the consequences on living and non-living species.

Naturally occurring radioactive materials are present in several types of materials. Materials which may contain any of the primordial radionuclides or radioactive elements as they occur in nature, such as radium, uranium, thorium, potassium, and their radioactive decay products, that are disturbed as a result of human activities. However the concentration of NORM in most natural substances is so low that the risk is generally regarded as negligible. Higher concentrations may arise as a result of human activities. In most NORM, several or all of the radioactive isotopes of the three primordial decay series (^{235}U, ^{238}U and ^{232}Th) are present in small concentrations in the natural matrix.

Irradiation of the human body from external sources is mainly by gamma radiation from radionuclides of the ^{235}U, and ^{232}Th decay series and from ^{40}K. these radionuclides may be present in the body and irradiate various organs with alpha and beta particles as well as gamma rays [Cember, 1996; UNSCEAR, 2000; IAEA; 2005].

The radionuclides in the decay series are more or less in radiological equilibrium however, this equilibrium becomes disturbed through human activities such as mining and mineral processing, resulting in either an enrichment or depletion of

some of the radionuclides concentrations compared to the original matrix. This disequilibrium is as a result of differences in the properties of the radionuclides in the series, due to geochemical migration processes and differences in their half-lives [Cember, 1996; UNSCEAR, 2000; Sato and Endo, 2001].

Mining has been identified as one of the potential sources of exposure to NORM [UNSCEAR, 2000]. However, mining companies are not being regulated for NORM in most countries including Nigeria since there are no guidelines for their regulation by the Nigerian Nuclear Regulatory Authority (NNRA). With the recent increase in awareness of the potential exposure situations of NORM, many countries are amending their legislations and putting in place measures to address the problems of NORM.

Whilst the developed countries have identified NORM as potential problems and measures are being taken to address the issues, very little is being done in the developing countries. It is also worth noting that most of the NORM industries such as mining and mineral processing, oil and gas exploration and extraction etc are located in developing countries such as Nigeria. Some studies in some countries have also reported elevated activity concentrations levels during mining and mineral processing (IAEA, 2005).

1.2 Present Work

In this work soil samples were collected from artisanal gold mine areas in Brinin Gwari local government area of Kaduna State. Gamma –ray spectrometry analyses for Naturally Occurring Radioactive Materials (NORMS) was carried out using Instrumental Neutron Activation Analysis (INAA).

1.3 Statement of the Problem

NORM producing activities such as artisanal mining have been in operation for a long period without any knowledge of the radiological aspects of the mining activity. In recent times, there is an increased awareness of the potential problems of NORM and this has resulted in most countries taking steps to implement regulations dedicated to natural sources of radiation in their national legislations, Hence the establishment of Nigeria Nuclear Regulatory Authority [NNRA]

Radionuclides, such as ^{226}Ra and ^{228}Ra are known to have high mobility in the environment due to their high comparative solubility in water. Most of these radionuclides are predominantly alpha emitters and alpha particles tend to cause more internal hazard.

Brinin Gwari Goldmine is one of the largest gold mining sites in Nigeria and has been in operation since 1930s (MMSD 2010). The mine currently undertakes only surface mining and the process produces large volumes of tailings and waste that may contain NORM. Some of the NORM are soluble in water and have the tendency for leaching into water bodies and farm lands. The mine

operates within the Tsoho Brinin Gwari and other smaller communities whose inhabitants depend on surface water and boreholes as their source of water. Farming is also an important activity within the mine's operational area. The soil, water bodies, dust and crops could be potential sinks for these radionuclides. The ultimate substrate of these radionuclides is the human body, which is the main concern of this study.

The above reasons are the basis for the choice of the Brinin Gwari Goldmine for this study since NORM is identified as the major source of human exposure to ionizing radiations. It is therefore essential to conduct studies on NORM in all mining industries in order to establish reference data which will be useful for all stakeholders of the industries in Nigeria.

1.4 Aim of the study

The aim of this study is to determine the activity concentrations and dose level of Naturally Occurring Radioactive Materials (NORM) at all gold mining locations in Brinin Gwari Local Government Area of Kaduna State, Nigeria.

1.5 Objectives of the Study

The study has the following specific objectives.

- To determine the activity concentrations of ^{238}U, ^{232}Th and ^{40}K
- To determine the radiation doses from the activity concentrations.
- To assess the hazard, risk or otherwise to the public within thestudy area.
- To make recommendation for further work in the study area.

1.6 Justification of the Study

In many developing countries including Nigeria, mining activities have not been subjected to radiological regulatory control. Data on radionuclide concentrations in raw materials, residues and waste streams and data on public exposure are scanty (Darko *et al.,* 2005; Darko and Faanu, 2007). Consequently, there is general lack of awareness and knowledge of the radiological hazards and exposure levels by legislators, regulators and operators.

There is therefore, the need for more studies to be carried out to cover all the gold and other mines in Nigeria so that a concrete decision can be taken to ascertain the NORM situation in Nigeria as the process of formulating guideline on setting standards for the regulations of NORM in the mining industry has started. The availability of data from such studies is very vital to all stakeholders involved since it will add to the data required for the development of this guideline.

CHAPTER TWO

2.0 LITERATURE REVIEW

2.1 Gold

Gold is a dense, soft yellow mineral; the native metallic element Au has high specific gravity sec tile nature and the slightly different color and luster distinguish it from yellow sulfides pyrites and chalcopyrite. (MacDonald 2003) Gold crystals are octahedral, rarely showing other forms. More typically gold is arbors cent, fills fractures or is found as nuggets, grains or wire scales. The structure of gold is face centered cubic which is the same as those of platinum and copper. Its composition is often close to pure Au, but substantial Ag may be present in solid solution. Small amounts of other element such as Cu and Fe may be present. (Dexter, 2007).

2.2 Gold Deposit Systems

This part reviews the main types of gold deposits and the key elements of their footprints, defined here as the combined characteristics of the deposits themselves and of their local to regional settings. Much has been published on gold deposits in the last decade, leading to (1) significant improvement in the understanding of some models, (2) the definition of new types of sub-types of deposits, and (3) the introduction of new terms. However, significant uncertainly remains regarding the specific distinction between some type of deposits. Consequently, specific giant deposits are ascribed to different deposit types by different authors. In this work, we have adopted the most accepted nomenclature

and models used in important reviews published in the last decade (e.g. Hagemann and Brown, 2000; Sillitoe and Hedenquist, 2003).

Thirteen globally significant types of gold deposits are presently recognized each with its own well- defined characteristics and environments of formation.

Minor types of gold deposits are not discussed in this work. As proposed\ by Robert *et al.,*(1997) and Poulsen *et al.,* . (2000), many of these gold deposit types can be grouped into clans i.e. families of deposits that either are formed by related processes or that are distinct products of large scale hydrothermal systems. These clans effectively correspond to the main classes of gold models, such as the orogenic, reduce intrusion-related, and oxidized intrusion-related ores (Hagemann and Brown. 2000). Deposit types such as Carlin, Au-rich VMS, and low- sulfidation are viewed by different authors either as stand-alone models or as members of the broader oxidized intrusion-related clan. They are treated here as stand – alone deposit types, whereas high and intermediate sulfidation and alkali epithermal deposits are considered as part of the oxidized intrusion-related clan. Witwatersrand deposits are still controversial and viewed either modified paleopalcer or as orogenic deposits.

2.2.1 Main deposit types and clans

The term orogenic has been originally introduced by Groves *et al.,* (1998) in recognition of the fact that quartz-carbonate vein gold deposit in greenstone and slate belts, including those in BIF, have similar characteristics and have formed by similar processes. Originally, the orogenic model applied strictly to

syntectonic vein-type deposits formed at mid-crustal levels in compressional or transpressional settings, i.e. syn-orogenic deposits. However, the term has been progressively broadened to include deposits that are post-orogenic relative to processes at their crustal depth of formation. This has led to significant ambiguity in the definition of the boundary between the orogenic and reduced intrusion-related deposit models, with many type examples being ascribed to one model or the other by various authors (Thompson and Newberry, 2000; Goldfarb *et al.,*, 2001). In this work. The orogenic clan is defined to only include the syn-tectonic quartz-carbonate vein-type deposits and their equivalents, formed at mid-crustal levels. Specific deposit types in this clan include the turbiditehosted and greenstoene –hosted vein deposits, as well as the BIF-hosted vein and slufidic replacement desposit.

As discussed in more detail below, a confusing issue is that greenstone belts also contain gold deposit types that don't fit the orogenic model defined here (Grooves *et al.,*, 2003; Robert *et al.,*., 2005). There is no consensus on the origin of these atypical deposits.

The reduce intrusion-related model (RIR) has been better defined in the last decade (cf. Lang *et al.,*, 2000). Deposits of this clan are distinguished by a Au-Bi- Te metal association and a close spatial and temporal association with moderately-reduced equigranular grantic intrusions. These deposits occur mainly in reduced siliciclasic sedimentary rock sequences and are commonly orogenic deposits. A range of style and depths of formation has been

documented for RIR deposit, including intrusion-hosted deposits of mesozonal to epizonal character, and more distal, sediment-hosted mesozonal equivalents Deposits of the sediment-hosted types correspond to the initial sediment-hosted stockwork-disseminated type of Robert *et al.,*., (1997), as well as to the pluton-related thermal aureole gold (TAG) deposits of Wall (2000) and Wall *et al.,*., (2004). Several deposits of the sediment-hosted IR deposits have also been ascribed to the orogenic clan by Goldfarb *et al.,* (2005).

The oxidized intrusion-related clan (OIR) include the well known porphyry and high-sulfidation epithermal gold deposit types, as well as skarn and manto type deposit, formed in continental and oceanic convergent plate settings. These deposits are best regarded as components of large hydrothermal system centered on high-level, generally oxidized, intermediate to felsic porphyry stocks. In the last decade, the genetic connection between porphyry and high-sulfidation epithermal deposits has been more firmly established (Heinrich *et al.,*., 2004), and it has been suggested that the largest deposits of this clan form in compressional arcs (Silitoe and Hedenquist, 2003). The characteristics and settings of the alkali endmember of porphyry deposit have also been refined, as has their possible connection with low-sulfidation alkali epithermal systems (Jensen and Barton, 2000).

Other types of globally important deposit include low- and intermediate sulfidation epithermal, Carlin, Au-rich VMS, and Witwatersrand types deposit Epithermal deposits are now subdivided into low-, intermediate- and high

sulfidation categories on the basis of mineralization and alternation assemblages (Sillitoe and Hedenquist, 2003). Intermediate sulfidation deposits, like high-sulfidation ones, are interpreted to be a component of large OIR systems, as is the case for the Victor veins in the Far Southern-Lepanto system and at Kelian. These deposit were initially singled out as carbonatebase-metal Au deposit types by Corbett and Leach (1998), and are characterized by a pyrite, low- sphalerite and Mn carbonate ore assemblages accompanied by dominant illite alternation. Mineralization can consists of vein and breccia bodies and commonly display a larger vertical continuity than their low-or high sulfidation.

Carlin-types deposits have been regarded either as being distal parts of large OIR systems or as standalone deposit (Cline *et al.,* 2005). Distinction has also been made between Carlin-type deposit proper and distal disseminated deposits, which occur peripheral to a causative intrusion and have a distinct Ag-rich metal association. However, controversy remains as to whether two groups of deposits are fundamentally different (Muntean, 2003).

Work on the modern seafloor has provided additional insight into the formation of Au-rich VMS deposit, with the identification of a number of favorable settings (Huston, 2000; Hannington, 2004). The recognition that some Au-rich VMS deposits are effectively submarine equivalent of high-sulfidation deposits and has significant explorations. Finally, the controversies remain concerning the origin of Witwatersrand gold deposits, with both modified pale placer and the hydrothermal origins proposed (Frimmel *et al.,* 2005; Law and Philips, 2005).

Although many of the giant deposit conform to one models outlined above, many of them have unique characteristics and are not easily classified in the scheme presented (Sillitoe, 2000). It is therefore likely that the next big discovery could be of different style or mineralization, or perhaps located in an unexpected geologic setting, a fact that obviously has to be taken into account in regional exploration programs. A good example is the discovery of the Las Lagunas Norte deposit in the alto Chicama district of northern Peru, where high-sulfidation epithermal mineralization is hosted in clastic sedimentary rocks rather than in volcanic rocks, as favored by the classical model.

2.3 Gold Deposits in Nigeria

Gold in Nigeria is found in alluvial and eluvia placers and primary veins from several parts of super crustal (schist) belts in the northwest and southwest of Nigeria. The most important occurrences are found in the Maru, Anka, Malele, (Zamfara state) Tsohon Birnin Gwari-Kwaga (Kaduna state), Gurmana (Niger state), Bin Yauri (Kebbi state), Okolom-Dogondaji (Kogi state) and Iperindo (Osun state) areas, all associated with the schist belts of northwest and southwest Nigeria. There are also a number of smaller occurrences beyond these major areas. (Baba *et al.,*2011).

2.3.1 Geological Setting and Mineralization of Gold in Nigeria

Nigeria Archean to Lower Paleozoic basement rocks consist of a migmatite-gneiss-quartzite complex. They bear imprints of Liberian, Eburnean and pan-African tectonic events. The enclosed schist belts lithologies consist of upper Proterozoic fine grained clastics, politic schists, phyllites, banded iron formations, marble and amphibolites with imprints of the Kibaran and Pan-African tectonic events.

Metamorphism within the belt is the of green schist facies grading into amphibolites, especially in the older gneiss migmatite-quartizite complex, with lesser volumes of volcanic rocks.

Primary gold mineralization in the schist belt commonly occurs in quartz veins within several lithologies. Morphologic types include bedding concordant veins or discordant vein system. Mineralized wall rocks are common adjacent to known veins and may be impregnated with fine grained sulphide minerals. (Baba*et al.,*2011).

i. Maru

Two old gold mines are most important, and are within the maru schist belt. Duki mine is hosted by a shear zone traversing a quartzite-schist series, often exploiting the Si schistosity planes. The mineralization apparently made up of gold quartz veins, was exploited by past miners for over 1km of strike length leaving behind series of collapsed ns trending working without any surface exposures of the mineralization.

Recent explorations drilling by the Nigeria mining corporation have shown the continuity of the gold quartz-sulphide veins below the old workings. The other old gold mine (maraba) is a two sub quartz vein (~300 m long) system. Tourmaline and chlorite in quartz floats and altered wall rocks are the only indicators of mineralization, as there are no in situ exposures due to intensity of past mining.

i. Anka

In this area there are a number of old gold mines such as kwali, jameson, zuzzurfa and kuba, hosted by schist phylities and quartzite of the schist belt. Individual veins or reefs seldom exceed 0.5km of strike length as indicated by the extent of past workings, and are concordant with the host rock foliation. These mineralized wall rock show metal

values in the following range: Cu=647-5410ppm, Pb=7550-22600ppm, Ag=1.5-8.6ppm, and Au=123-6320pp.

ii. Malele

The Malele area is in the extreme southern part of the maru schist belt. Gold mineralization occur in a NNE trending gold quartz vein series cutting both biotite-gneiss and chlorite schist, now represented by a series of sub-parallel surface workings (pits, trenches and water ponds) with the vein exposures seldom observed. The mineralization was discovered in 1934 with the most prominent of the vein being 1.5 m thick, 370 m long and a grade of about 30 ppm Au.

iv. Bin Yauri

Gold-sulphide-carbonate quartz veins occur in a brittle fault zone cutting hornfels of the contact of a pan-African granodiorite batholith intruding phyllites and tourmalines of zuru schist belt in north western Nigeria. Mineralizations have been uncovered over a strike length of 1.5km by past miners (1920s-1940s). Gold occurs associated with pyrite, chalcopyrite, galena, and minor sphalerite, magnetite and bismuth telluride in a gangue of mainly quartz with some carbonates, Seri cites, chlorite and tourmaline. The gold veins are surrounded by a narrow zone of hydrothermal alteration in which a chlorite-tourmaline-pyrite-carbonates association overprints a dominantly sericitic fabric of the hornfels wall rocks. Past mine records, reconnaissance exploration and study have shown gold grades in the range 5-15ppm.

v. Gumana

The Gumana area of gold mineralization is situated in the extreme southern part of the kushaka schist belt. Gold mineralization is in the form of quartz-sulphide veins and stock works hosted by amphibolite's and gneisses. The gold-quartz-sulphide veins seldom extend more than a few tens of meters. Gold occurs with pyrite, chalcopyrite, galena, cove lite and chalcocite.

vi. Okolom-dogondaji

The Okolom-dogondaji area gold mineralization is in the egbe-isanlu schist belt of south western Nigeria, and the primary gold-quartz veins and eluvia/alluvial placers have been mined extensively in the period 1930s-1950s. a series of gold-quartz veins is hosted by N-S and NNE-trending shear zones which cut gneisses, schist and amphibolite's. The most prominent site is the okolom old mine, which a gold-sulphide-quartz reef system with a total strike length of about 3km hosted by gneiss, amphibolite and talc schist. Other site in the (dogondaji)area have relatively smaller veins hosted by amphibolite's, gneisses, mica, schist and phyllites. Gold is associated with pyrite, marcasite, pyrrhotite, chalcopyrite, argentite and galena, with a gangue of quartz, tourmaline, sericite and chlorite.

vii. Iperindo

The Iperindo old gold mine is in the ilesha schist belt of southwestern Nigeria. The Iperindo mineralization comprises a series of auriferous quartz-carbonate veins localized by a subsidiary fault within biotite gneiss and mica schist, presently defined by sub-parallel old working extending overall for about 900m in a NNE direction. Gold occurs with pyrite, pyrrhotite and minor chalcopyrite, galena, sphalerite, magnetite and

limonite. Adjacent to the gold-bearing veins the host granite-gneiss has been hydrothermally altered to a sericite-chlorite-epidote assemblage (also with hematite and pyrite).

viii. Tsohon Birnin Gwari and Kwaga

The Tsohon Birnin Gwari and Kwaga gold sites are within the Kushaka schist belt of northwestern Nigeria. While the Tsohon Birnin Gwari was an active gold mine in the 1930s where over 600 ounces of gold were produced, the Kwaga site is a recent discovery by artisanal miners following the extensions of the Tsohon Birnin Gwari mine.

The Tsohon Birnin Gwari gold reef system is about 7 km long, while the Kwaga reef system extends for about 3 km. Gold is mainly associated with pyrite and minor sphalerite, chalcopyrite, pyrrhotite, galena and magnetite. The gangue is predominantly quartz, but K-feldspar and graphitic matter (from wall rocks) are also constituents. Grades are very variable within the reef system and between oxide and sulphide ore zone, but generally 5-100 g/t Au are found.

Recent core drilling and near-surface mine exposures have provided relatively fresh samples of the gold-quartz reefs and their altered wall rocks from which the geochemical characteristics of the Tsohon Birnin Gwari mineralization were studied. MMSD (2010).

2.4 Elements Associated with Gold

Carlin-type deposits in northern Nevada, which provide~ 9%of the world gold production, are hosted mainly in hydrothermally altered Paleozoic impure carbonate rocks as well as in various types of Jurassic and Eocene intrusive rocks. The main

auriferous event took place in Eocene during the onset of the basin and range extension. Gold occurs in arsenian pyrite and is commonly associated with other element such as Cu, Sb, Ni, Se, Hg, Tl, Ti, Zn, Ag, Co, Pb, and W. Despite the large gold endowment and more than 40 years of research, very little is known about the source of metals and fluids associated with Carlin-type gold deposits. (Olivo*et al.,* 2009).

In the cyanidation method of gold processing pure gold dissolves slowly in cyanide solution than gold containing small quantities of other metals (Jiffrey and Ritchie, 2001).Trace amount of lead, mercury thallium or bismuth reduce the passivation effect on gold surfaces thereby accelerating gold leaching rate (Marsden and House, 2006). These metals can also have a detrimental effect on the adsorption of gold on to activated carbon and its subsequent elution by competing for adsorption sites and by consuming elution reagents.

The long history of gold mining at Obuasi, Ghana, has generated huge environmental legacy issues in the area. The most significant of the environmental challenges is that of the trace elements contamination. Amonoo-Neizer *et al.,* (1995) found significant distribution of As and Hg in the top soil, plantain, water fern, elephant grass, cassava and mud fish at Obuasi and its environs. Other studies have made various findings regarding presence of trace elements in soils water sources and foodstuffs at Obuasi and surrounding areas (Amasa, 1975, Bamford *et al.,* 1990; Golow *et al.,* 1995). So far it appears that as constitutes the major trace element problem in the Obuasi. This has been linked to the considerable level of naturally occurring arsenic bearing gold ores during gold extraction (Amonoo-Neizer *et al.,*l; 1995; Asiam, 1996, Smedley, 1996, Smedley *et al.,*1996, Ahmad and Carboo, 2000 Kumi-Boateng, 2007).

Regarding the divalent metals (Zn, Cu and Pb), it was presumed that their relative abundance in the soils with respect to contamination in the Obuas areas, might be partly dependent on potential intrusions of the elements in to soil and their rate of mobility in the soil. Generally the selectivity of clay minerals and hydrous oxide absorbents surfaces in soil and sediments for divalent metals follow the order Pb>Cu>Zn. In this respect the rate of mobility of these three metals in such soils would increase in the reverse order (Zn > Cu > Pb) which is consistent with the findings in the study area. Overall, the decreasing trend of element concentration in the Obuasi soil i.e. As > Zn > Cu > Pb is contrary to observations by Shumlyanskyy *et al.,* (2005) in which among other metals As Zn, Cu, and Pb were reported to decrease in soils around a gold base metal deposit in Ukrain in the order Pb > Zn > As > Cu at a distance of 400 – 700m from the dump. This disagreement with the study at Obuasi might be attributed to differences in soil properties considering the two different locations involved.

In a related studies at eastern Mangolian, Altay, Aichler *et al.,*(2008) indicated new occurrences and indices of Au, Ag, Cu, Zn, Pb and (Ba) – As – Sb – Hg Mineralization were discovered in some of the prospective gold mine areas. Low gold contents accompany, also the As-Sb-Hg-(Ba) vain disseminated mineralization confined to acid volcanic rocks of the Permian Khar Argalantyn Fm. The Ag-rich base metal quartz veins in silicified marbles of the NeoProterozoic – Palaeozoic Maykhan Tsakhir Fm. In the Gobi Altay terrain,the occurrences and showing of slightly auriferous base metal (C, As, Zn, Pb, Ag) veins were ascertained in siliceous chlorite-sericite schist with layers of queenschists and quartzite's of the Carbon Tugrug Fm.

Similarly, according to Adetoyinbo *et al.,* (2011). The concentration of major element in the soil samples collected from five location in the mining areas of Itagunmodi Osun

state Nigeria and three locations outside the mining areas shows that the concentrations of the elements is higher within the mining site than outside mining site.

The concentration of heavy metals and other elements in antisanal Gold mining soils from Zamfaraand Kebbi State were determined using XRF Technique by Girigisu *et al.*, (2012). Four out of the ten elements (Pb. Si, Rb and Fe) showed relatively high concentration

2.4.1 Radioactive Element Associated With Gold

Naturally occurring radio isotopes occurs in most soil and rocks and by mining and mineral processing some of the radio isotopes are significantly enhanced, this section will look at the occurrences of some of the radio isotopes around Gold mining and processing areas.

Radon emanation from mine dump soil were measured with Electrets Passive Environmental Random Monitor (E-perm) detector by Kotrappa *et al.,* (1988),soils samples from the same site was analyzed using HPGE detector by Venema *et al.,*(2001), the activity concentration of the natural radio nuclides ^{238}U and ^{232}Th were measured indirectly by using the^{214} Pb and ^{214}Bi γ -rays from ^{228}Ac and ^{208}TI (found in the ^{232}Th decay series) for the ^{232}Th determination.

In the related study indoor and outdoor radon concentrations, the build-up factor and indoor and outdoor exposure of midval water and Batshabelo Community Health Centre within Klerksdoup gold areas of the North West province South Africa were determined by Nnenesi *et al.,* (2009).

A report compiled by Coetzee, (2004) indicated that Uranium in gold reefs of South Africa occurs mainly in brown-blackish tetravalent minerals such as uraninite (UO_2) or

pitchblende (an amorphous mixture of UO_2 and U_3O_8), and in oxidized derivates thereof (secondary minerals) such as coffinite ($USiO_4$), brannerite (UTi_2O_6) and leucoxene, the latter being a weathering product of brannerite. The extremely heavy uraninite (density: 10,/cm^3**)** occurs in reefs in the form of small, muffin-shaped weathered particles from uraniferous rock with a typical diameter of 77-100 μm. Uraninite becomes unstable in the presence of oxygen and tends to release uranium into dissolving agents such as fissure water. Other forms of uranium include a wide range of hexavalent fluorescing under UV-light. Such minerals include carnotite, uranophane, beta-uranophane, tyuyamunite, autune and toberite. Uraoorganic compound, as a third form of uranium in ores, are frequently present in sedimentary deposit such as black shale and lignite.

The highest uranium concentration in the area are associated with carbon and were found in kerogen seams and nodules, displaying up to 5800mg/kg uranium (5.8%). Other uraniferous reefs include the Doornfontein Reef (which was never exploited because of low gold grades), Ventersdrop Contact Reef, Elsburg Reef, Composite Reef and quartz-pebble conglomerates of the white Reef, Monarch Reef and Middleville Reef.

Owing to the comparatively high uranium concentration in the gold reefs mined earlier on, uranium production in SA was concentrated on the West Rand and far West Rand goldfields. Uranium in the Far West Rand was exclusively produced form the Carbon Leather Reef, yielding a total of 11,295t U_3O_8between 1952 and 1988 at an average uranium grade of 145mg\kg. The first pilot uranium plant of SA was constructed at Blyvooruitzicht in 1951. Followed by the first uranium producing plant at West Rand Consolidated Estate, Luipaardsvlie (all in the vicinoity of Randfontein), Western Areas (Near Westonaria), West Drrifontein and Western Deep Level (located around

Carletonville). At one stage 9 out of the 22 active goldmine of the region were feeding uraniferous are into a total of seven uranium plants.

2.5 Review of Similar Works

Heavy metals and NORMS were determined using a neutron activation analysis from soil sample collected from oil and gas exploration site in USA by Landsberger *et al.,*(2012). The result successfully established a protocol for the determination of the concentration of $^{210}P_b$, $^{226}R_a$ and $^{228}R_a$in solid and liquid samples from naturally occurring radioactive materials from the oil and gas exploration sites.

In an effort to determine gold concentration in two Egyptian gold ores using instrumental neutron activation analysis El-Taher *et al.,*; (2003) discovered 31 other elements beside gold at different times of irradiation. (Mg, A1, Cu, Ti, V, Na, K, Mn, Ba, Ev, Sr, Ga, Sm, U, Sc, Cr, Fe, Co, Zn, Rb, Zr, Nb, Sn, Cs, Ce, Nd, Yb, Lu, Hf, Ta, Th). U, Th and K were among the elements present in the Gold ore samples.

Radioactivity concentration measurements of ^{238}U, ^{232}Th, ^{40}K, and ^{137}Cs in environmental samples and technologically enhanced product in Tunisia using NAA analysis and gamma ray spectrometry was carried out by Reguigui *et al.,*(2002).

Faanu *et al.,*(2010) assessed the public exposure to naturally occurring radioactive materials from mining and mineral processing activities of Tarkwa Goldmine in Ghana using NAA. The mean activity concentration of ^{238}U, ^{232}Th and ^{40}K in soil rock, water and dust were measured and the total annual effective dose to the public was estimated to be 0.69mSv which compared well with typical world average.

In a related development the concentration of U, Th and K as well as other trace elements, anions and the physical parameters in water and soil samples in a goldmine

and its surrounding in Ghana using NAA, AAS UV-VIS spectrometer and gross alpha/beta was carried out by Faanu *et al.,* (2011). The result shows that the concentration of U, Th, and K and the mean Th/U ratio in all the soil samples compared well with the result of normal continental crust rock and the activity concentration of gross-α and gross-β in the water samples were all below WHO recommended guideline values.

The concentration of naturally occurring radioactive materials in the crystalline bedrocks and soil from abandoned quarries in Abeokuta Nigeria was measured using HPGE by Gbadebo (2011). The result confirmed the presence of ^{40}K, ^{232}Th and ^{236}U in appreciable amount. The maximum mean dose equivalent obtained in the study area are lower than the allowable maximum limit.

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2.6 Neutron Activation Analysis (NAA)

Neutron activation analysis is a two steps analytical procedure in which some components ion in a material are activated (irradiation) with high flux of thermal neutrons. The activation process\ in nuclear reactions between the incident neutrons and target nuclei in to sample being irradiated. When thermal neutrons collide with the nucleus a number of reactions may occur but the most useful in NAA being radioactive capture and the reaction is generally represented by equation (2.1) (Landsberger, 1994).

$$N + {}^{A}Z \rightarrow {}^{A+1}Z \rightarrow {}^{A+1}Z + y _____ 2.1$$

Where;

^{A}Z is the target nucleus,

^{A+1}Z is a compound nucleus in an excited state which de-excites with the emission of gamma ray called prompt gamma,

^{A+1}Z is the product after irradiating the target nucleus which is irradioactive.

The radioactivity produced after the irradiation is governed by the usual decay equation and generally represented by equation (2.2) (Landsberger, 1994)

$$R = N\int_{\text{Whole Energy Range}}\sigma(E)\phi(E)dE _____ 2.2$$

Where R is the reaction rate,ϕ (E) dE is the neutron flux of neutrons with kinetic energy between E and +dE in cm-^{2}s^{-1}, σ (E) is the neutron capture cross-section in cm^{2} defined as the probability of a radioactive capture reaction occurring in a collision between a neutron and a nucleus given in terms of area and dependent on the energy of the incident neutron, N is the number of atoms of element in the sample.

During neutron irradiation, the dominant reaction rates are the thermal and epithermal components and because the neutron cross-section of the fast neutrons (R_{fast}) is negligible the reaction rate of fast is small.

The activity of an element in a sample is given by the following general expression (2.3) (Landsberger, 1994)

$$A = \sigma\phi\,(m/M)\,N_A\,SDC\theta Py\eta _____ 2.3$$

Where

A is the measured activity in Bq from a product of an expected relations;

σ is the activation cross section of the reaction in cm^2.

ϕ is the activating neutron flux in $n.cm^{-2}S^{-1}$;

m is the mass of element in g ;

M is the atomic weight of the element to be determine in g/mol,

N_A is the Avogadro's constant, which is 6.022×10^{23} atoms/mol

S is the saturation factor which is given by

$S = (1 - \exp(-\lambda t_1)$, λ *is the constant of the reactive product and* t_1 *is the duration* of irradiation.

D is the decay factor and it is given by

$D = \exp(-\lambda t_d)$ *and* t_d*) and* t_4 *is the duration of decay.*

C is the correction factor for nuclide decay during the counting time given by

$C = (1 - \exp(-\lambda t_c)$ *and* t_c *is the duration of counting*

θ is the relative natural isotopic abundance of the activated isotope:

P_γ is the probability of emission of photon with energy E; and

η is the detector efficiency for the measured gamma radiation energy.

Equation (2.3) is simplified to equation (2.4) by the comparator method using the same geometry, equal weights of both samples and standard with the \ same irradiation, decay and counting times.

$$C_{sam} = C_{std}\left(\frac{A_{sum}}{A_{std}}\right) \quad \text{2.4}$$

Where:

C_{sam} is the unknown concentration of the element in the sample

C_{std} is the known concentration of the element in the standard

A_{sam} is the activity of the sample and $A_{sam}$is the activity of the standard.

By using the terms D and C in equation 2.4 and also normalizing the weight between standards and unknowns, the overall equation becomes equation (2.5) in ppm.

$$C_{sum} = C_{std}\left(\frac{A_{sum}}{A_{std}}\right)\left(\frac{D_{std}}{D_{sum}}\right)\left(\frac{C_{std}}{C_{sum}}\right)\left(\frac{W_{std}}{W_{sum}}\right) \quad \text{2.5}$$

WhereW_{sum} and W_{std} are the weights of the samples and the standard respectively.

The product is required to be radioactive and capable of emitting at least one gamma-ray photon. The gamma ray photon emitted is detected on a gamma ray detector using HPGE. If the activation product is stable, it cannot be detected. The duration of the irradiation depends on the neutron flux density, mass of the sample and the efficiency of the gamma detector. The sample to be irradiated are specially sealed in capsules and transferred to the reactor core and irradiated with high flux neutrons. The activated components are then analysed to identify and determine quantitatively, the concentration of each radionuclide applying gamma spectrometry technique

CHAPTER THREE

3.0. METHODOLOGY

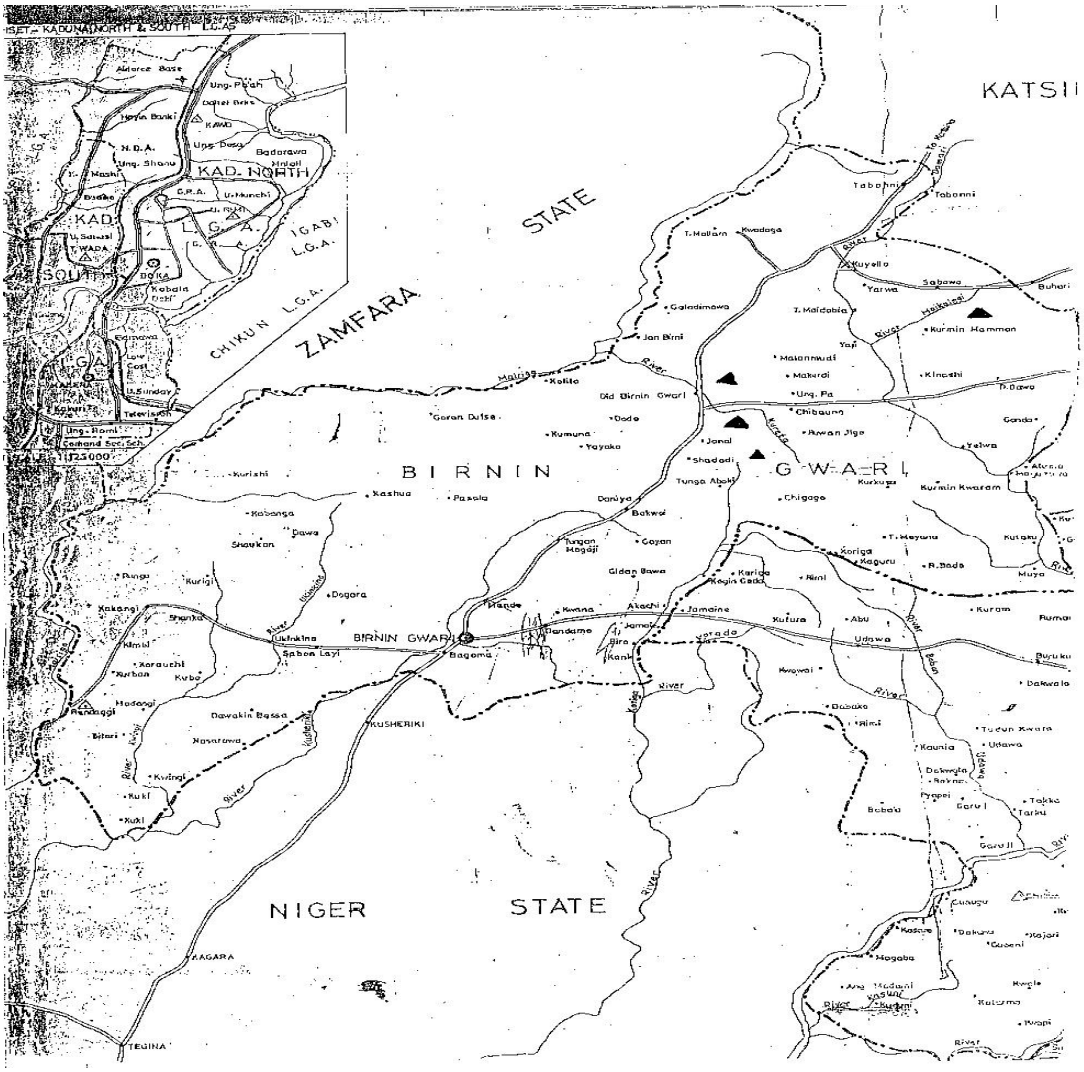
In this section the location of the study area, the climate, sampling, sample preparation and analysis methods are described, mathematical formalism used for the calculation of specific activity dose rate and annual effective dose rate of the naturally occurring radionuclides are described in details.

3.1 Study Area

The study area is Tsohon Birinin Gwari and its surrounding communities where artisanal gold mining activities are taking place in Brinin Gwari local government area Kaduna State. The local government was created in September, 1978. The local government is situated in thewestern part of Kaduna State between latitude $12^{0}20^{1}$N and longitude $90^{0}0^{1}$E and has a population of 252,363 (NPC 2006). Brinin Gwari has fourteen (14) districts, namely; Brinin Gawri central, Gayin, Bugai, Kutemeshi, Tabanni, Dogon Dawa, Kakangi, Randagi, Kazage, kungi, Maganda, Saulawa, Saminaka, and Kuyello. The main ethnic groups are Hausa, Fulani, Gwari and Kamuku and a handful of settled Yoruba and Igbo.

3.1.1 Climate

The study area lies within the tropical savannah region, which is characterized by well distinct wet and dry seasons. The wet season lasts from October to March. The mean annual rainfall is about 109.22mm. The highest peak of the dry season is experienced in March/April with temperature as high as (35- 39^{0}c). The map of the study area is as shown in figure 3.1.



LEGEND

═══	Road
— · · —	Local governemnt boundary
———	*River*
▲	Study area

FIG 3.1: Map of Birnin Gwari Local Government Area Showing the study Area

3.2 Sample Collection

Twelve samples were collected from 12 gold pits at different depth between (2.5m – 43.0m) in the study area which comprised of the following artisanal gold mining sites: Tsohowar Gwari Farinruwa and Kakini. Global position system (GPS) was used to determine the location of each pit and a type rule was used to determine the depth of each pit. Table 3:1 shows the location, depth and elevation of each pit where samples were collected, plate 3:1 – 3:12 shows the sample locations plotted on the geological map of the study area while plate 3.13 shows the examples of pits where samples were collected.

Table 3.1: Sample locations

PLACE	SAMPLE CODE	DEPTH IN m	LOCATION N	E	ELEVATION IN m
KAKINI	BG1	2.70	11^0.10" 25'	06^0 58" 0'	663
	BG2	4.30	11^0 10" 23'	06^0 59" 15'	685
F/RUWA	BG3	7.00	11^0 04" 14'	06^0 47" 34'	595
	BG4	7.50	11^0 04" 16'	06^0 47" 33	594
TSBG	BG5	19.00	10^0 59" 45'	06^0 48" 27'	560
KANO	BG6	43.00	10^0 59" 37'	06^0 48" 25'	562
TSBG	BG7	28.00	11^0 00" 43'	06^0 48" 28'	547
JINEER	BG8	26.00	11^0 00" 37	06^0 48" 23'	542
TS.BG	BG9	24.00	11^0 01" 14'	06^0 48" 20'	550
KATSINA	BG10	28.00	11^0 01" 10'	06^0 48" 21'	546
TS.BG	BG11	10.20	10^0 59" 19'	06^0 48" 31'	558
ABUJA	BG12	8.50	10^0 59" 17'	06^0 48" 32'	559



PLATE 3.1: Sampling Location



PLATE 3.2: Sampling Location



PLATE 3.3: Sampling Location



PLATE 3.4: Sampling Location



PLATE 3.5: Sampling Location

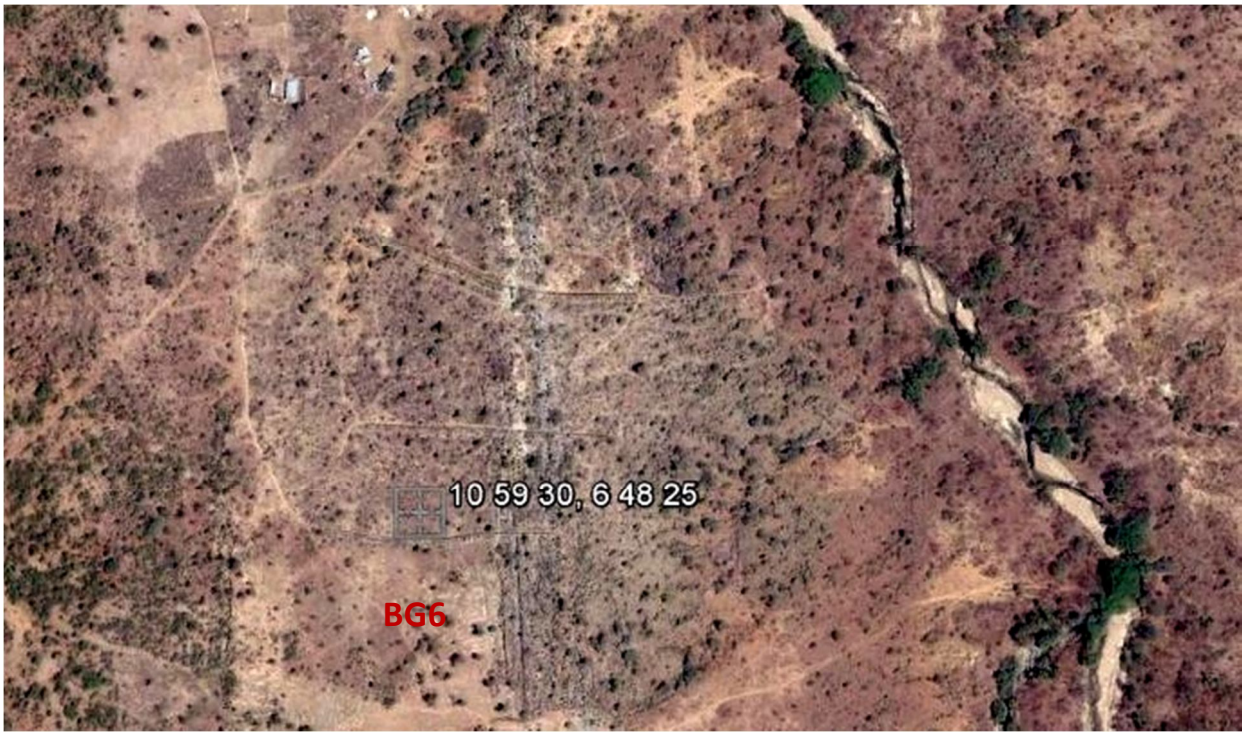


PLATE 3.6: Sampling Location



PLATE 3.7: Sampling Location



PLATE 3.8: Sampling Location

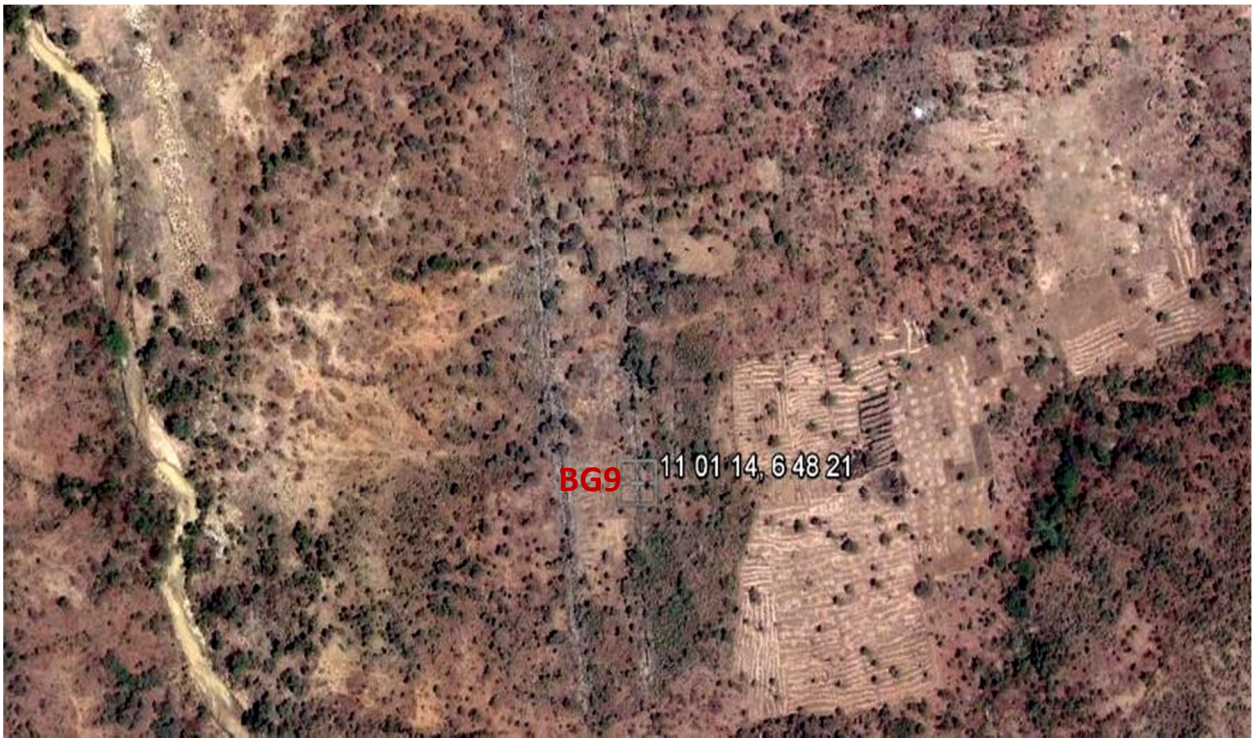


PLATE 3.9: Sampling Location



PLATE 3.10: Sampling Location



PLATE 3.11: Sampling Location



PLATE 3.12: Sampling Location

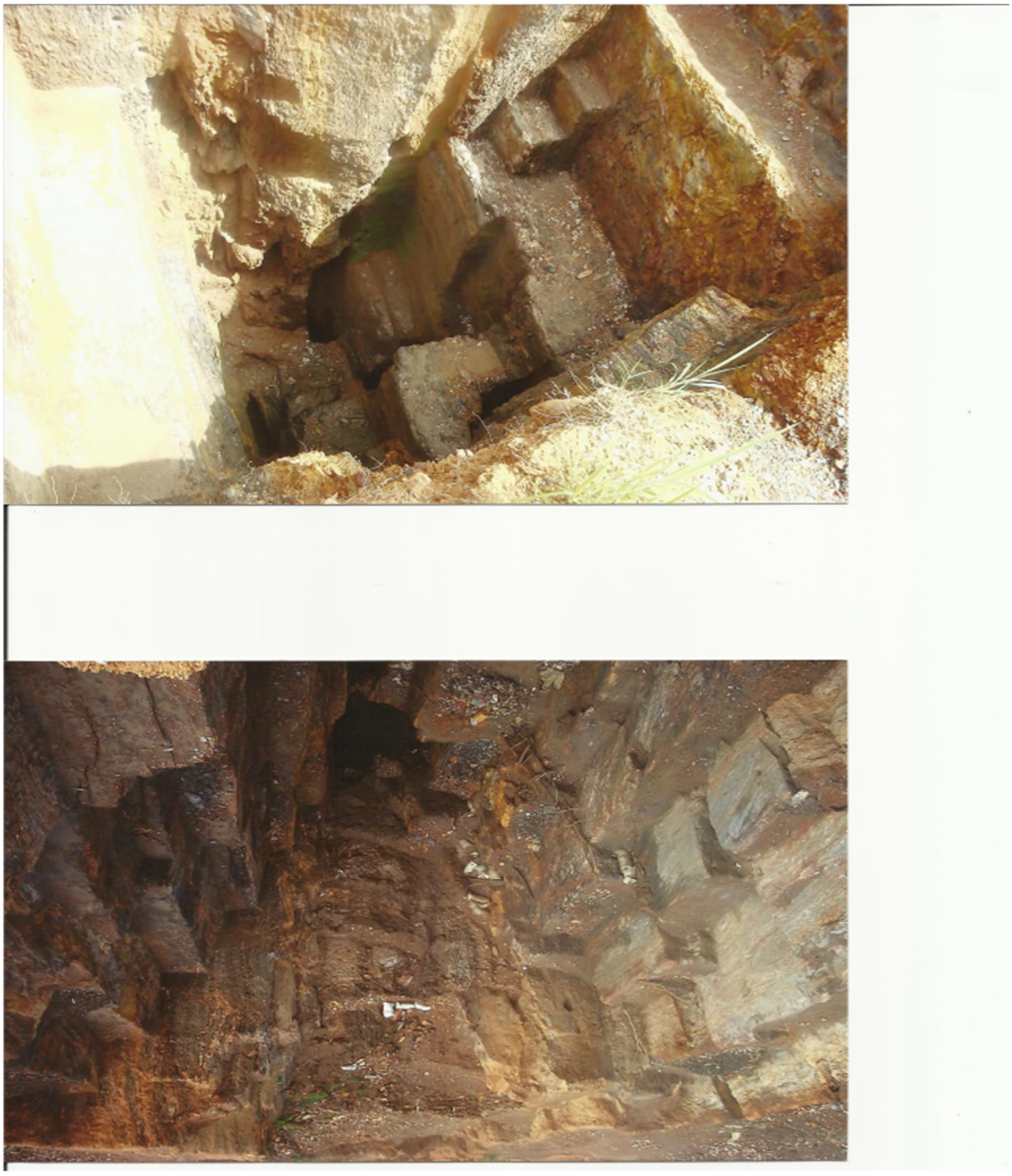


PLATE 3.13: Examples of pits where samples were collected.

3.3.0 Sample Preparation

The samples collected were taken to the Laboratory of Mineral Resources Engineering Department of Kaduna Polytechnic where they were crushed and sieved separately to a tiny bits of 38µm (Kogo *et al.,*2009). The crushed samples were then oven dried at about 100^0C to a constant weight and then taken to Centre for Energy Research and Training Ahmadu Bello University Zaria for Neutron Activation Analysis where 0.150g – 0.180g of the powdered samples were wrapped in a polyethylene and then placed in $7cm^3$rabbit capsules. The polyethylene and rabbit capsules containing the samples were cleaned by soaking in 1:1 HNO_3 (Nitric acid) and then washed with de-ionised water in order to eliminate every contamination prior to sample irradiation (El-Taher *et al.,*2003).

3.4.0 Sample Analysis

The concentration of element of interest from the collected and prepared samples were investigated using neutron activation analysis technique (NAA) with the Nigeria Research Reactor 1 (NRR1) No NRR1/DS/JC/09/16. At the Centre for Energy Research and Training, Ahmadu Bello University Zaria.

3.4.1 Short Irradiation

Sample were sent for irradiation one after the other through the outer irradiation channels B4 where the neutron flux is $5 \times 10^{11} n/cm^2$ and the irradiation time was 60s (1min). The outer irradiation channels was chosen in order to eradicate corrections that may arise from nuclear interferences caused by threshold

reactions notable; Mg in the presence of Al, Al in the presence of Si and Na in the presence of P, which are all as result of closeness of the inner channels of the Miniature Neutron Source Reactor (MNSR) to the core leading to the relatively higher ratio of fast to thermal neutrons (Jonah*et al.,* 2006).

3.4.2 Long Irradiation

Samples were irradiated for 6 hours in the smaller inner irradiation channels B_2 and B_3 that enable exposure to the maxima value of thermal neutrons flux of $5x10^{11}$ n/cm^2. The flux was kept constant by monitoring the neutron flux reading from a fission chamber connected to the microcomputer controlled room. After irradiation the samples were retrieved via the same pneumatic transfer to the control chamber where they were collected and kept in a glass chamber for three and ten days.

3.4.3 Measurements of Gamma Rays

Two laps of measurements were conducted for both short and long irradiation of the samples.

3.4.3.1 Short Irradiation

(a) After the short irradiation a waiting period of 10mins was given. The sample was placed on a flexi – glass sample holders designated as H2 which refers to the source detector geometry of 5cm the counting was carried out for 10mins.

(b) The second counting was carried out after a waiting period of three hours. The sample was placed on a flexi – glass sample holder denoted as H1 referred to as source detector geometry of 1cm, the counting was also carried out for 10mins.

3.4.3.2 Long Irradiation

The 1st lap of counting was carried out after a waiting period of 4 days for 30mins using the H1 sample holder. While the second lap of counting was carried out after a waiting period of 10 days for duration of 1hr also using H1 sample holder.

3.4.3.3 Gamma Ray Concentration

With the help of gamma ray spectrum software known as WINSPAN 2004, the gamma ray of product radio nuclides were identified by their energy spectrum and also quantitative analyses of their concentration were obtained.

3.5 CALCULATIONS

3.5.1 Specific Activity

The results obtained from the neutron activation analysis were the concentrations of elements present in each sample and was given in part per million (ppm). Table 3.4 below was used to convert from ppm to specific activity in Bq/kg.

Table3:2: Conversion of radio element concentration to specific activity (IAEA 2003).

1%k in rock/soil	=	313 Bq/kg ^{40}K
1 ppm of U in rock/soil	=	12.35 Bq/kg ^{238}U or ^{226}Ra
1 ppm of Th in rock/soil	=	4.06 Bq/kg ^{232}Th

3.5.2 Calculation of Absorbed Dose Rate and Annual Effective Dose Rate.

(a) Absorbed dose rate

The gamma dose rate from the samples was calculated from the activity concentration of the relevant radio nuclide from equation (3-1). (UNSCEAR, 2000).

$D(nGyh^{-1}) = 0.0417A_k + 0.462\ A_u + 0.604\ A_{Th}$ _____ 3.1

Where A_k, A_u and A_{Th} are the activity concentrations of ^{40}K, ^{238}U, and ^{232}Th respectively and table 3.3 shows the dose conversion factors of ^{40}K, ^{238}U and ^{232}th

Table 3.3: Activity to Dose Rate Conversion Factor (UNSCEAR,2000).

Radionuclide	Dose Coefficient (nGy/h per Bq/Kg)
^{40}K	0.0417
^{238}U	0.462
^{232}Th	0.604

(b) Annual effective dose rate

The annual effective dose rate was calculated from the absorbed dose rate, the dose conversion factor of 0.7Sv/Gy and an outdoor occupancy factor of 0.2 usingequation (3.2) (UNSCEAR ,2000)

$H_E = DTF$_____ 3.2

Where D = the calculated dose rate ($nGyh^{-1}$) x (24 x 365)

T = the occupancy factor (0.2).

F = conversion coefficient (0.7 $Sv\ Gy^{-1}$).

CHAPTER FOUR

4.0 RESULTS AND DISCUSSION

4.1.0 RESULT

4.1.1 Activity Concentration

The concentrations of U and Th in part per million (ppm) and %K extracted from the results of the neutron activation analyses (Appendix I) is shown in Table 4.1 below.

From Table 3.2, the conversion from ppm and % weight Bq/kg is given below.

1%K in rock/soil = 313 Bq/kg ^{40}K

1 ppm of U in rock/soil = 12.35 Bq/kg ^{238}U or ^{226}Ra

1 ppm of Th in rock/soil = 4.06 Bq/kg ^{232}Th

Therefore for sample BG1

U = 1.0 ± 0.3 ppm = 1.0 ± 0.3 x 12.35 = 12.35 ± 3.7 Bq/kg

Th = 7.1 ± 0.2ppm = 7.1 ± 0.2 x 4.06 = 28.83 ± 0.8 Bq/kg

K = 0.272 ± 0.0144% = 0.272 ± 0.0144 x 313 = 85.13 ± 4.5 Bq/kg

The same conversion factors were used and activity concentrations for ^{238}U, ^{232}Th and ^{40}K are presented in table 4.2.

Table 4:1: Concentration of U and Th in PPm and K in %

S/N	Sample Code	U(ppm)	Th (ppm)	K%
1	BG 1	1.0 ± 0.3	7.1 ± 0.2	0.272 ± 0.0144
2	BG 2	0.5 ± 0.3	4.1 ± 0.2	1.056 ± 0898
3	BG 3	4.3 ± 0.5	9.9 ± 0.2	4.999 ± 0.1849
4	BG 4	5.4 ± 0.4	15.8 ± 0.2	4.162 ± 0.2081
5	BG 5	2.7 ± 0.5	16. 9 ± 0.2	4.454 ± 0.1204
6	BG 6	5.1 ± 0.5	16.3 ± 0.2	3.184 ± 0.1204
7.	BG 7	2.4 ± 0.2	18.6 ± 0.3	3.128 ± 0.0313
8	BG 8	2.9 ± 0.3	17.5 ± 0.3	3.092 ± 0.0340
9	BG 9	2.3 ± 0.3	18.9 ± 0.2	3.762 ± 0.0564
10	BG 10	4.5 ± 0.3	18.5 ± 0.2	3.540 ± 0.0566
11	BG 11	2.6 ± 0.3	21.5 ± 0.3	3.209 ± 0.0546
12	BG 12	2.9 ± 0.3	18.5 ± 0.3	3.399 ± 0.0646

Table 4.2: Specific activity of ^{238}U ^{232}Th and ^{40}K in Bq/kg.

S/N	Sample Code	^{238}U (Bq/kg)	^{232}Th (Bq/kg)	^{40}K(Bq/kg)
1	BG 1	12.35 ± 3.7	28.83 ± 0.8	85.13 ± 4.5
2	BG 2	6.18 ± 3.7	16.65 ± 0.8	330.53 ± 28.1
3	BG 3	53.11 ± 6.2	40.19 ± 0.8	1564.69 ± 57.9
4	BG 4	66.69 ± 4.9	64.15 ± 0.8	1302.71± 65.1
5	BG 5	33.35 ± 6.2	68.61 ± 0.8	1394.10 ± 37.7
6	BG 6	62.99 ± 2.5	67.52 ± 1.2	996 ± 1379
7.	BG 7	29.64 ± 2.5	75.52 ± 1.2	976.06 ± 9.8
8	BG 8	35. 82 ± 3.7	71.05 ± 0.8	967.80 ± 10.6
9	BG 9	28.41 ± 3.7	76.73 ± 0.8	1177.51 ± 17.7
10	BG 10	55.58 ± 3.7	75.11 ± 0.8	1108.02 ± 17.7
11	BG 11	32.11± 3.7	87.29 ± 1.2	1004.42 ± 17.1
12	BG 12	35.82 ± 3.7	75.11 ± 1.2	1063.89 ± 20.2
	Mean	37.36 ± 5.45	62.69 ± 6.33	997.57 ± 199.97

4.1.2 The activity concentration of ^{238}U, ^{232}Th and ^{40}K

Descriptive statistics and one-way analysis of variance (ANOVA) at the 5% level significance was employed. The results have shown that ^{238}U has a mean concentration of 37.36 Bq/kg with standard error of the mean is 5.45 Bq/kg; which implies that the maximum sampling error in the results cannot exceed 5.45 Bq/kg. The 95% confidence interval for the mean concentration of ^{238}U lies between 6.18 ± 3.7 and 66.69 ± 4.9 Bq/kg. In other words, there is 95% assurance that he mean concentration of ^{238}U in any mining site within the study area will lie between (6.18 ± 3.7, 66.69 ± 4.9) Bq/kg.

Similarly, ^{232}Th has a mean concentration of 62. 69Bq/kg with standard error of the mean is 6.33Bq/Kg; which implies that the maximum sampling error in the results for Thorium cannot exceed 6.33Bq/kg. The 95% confidence interval for the mean concentration of ^{232}Th lies between the limits (16.15 ± 0.8, 87.29 ± 1.2)Bq/kg.

Furthermore, ^{40}K has a mean concentration of 997.57Bq/kg with a standard error of 119.97Bq/kg; meaning that the maximum sampling error in the results for ^{40}K cannot exceed 199.97Bq/kg. Moreover, the 95% confidence interval for the mean concentration of ^{40}K lies between 85.15 ± 4.5 and 1564.69 ± 57.9 Bq/kg. In other words, there is 95% assurance that the mean concentration of ^{40}K in any mining site within the study area will lie between the limits (85.15 ± 4.5,1564.69 ± 57.9)Bq/kg.

The average concentrations of ^{238}U, ^{232}Th and ^{40}K in the mining sites within the study area are not equal. Further results reveal that ^{238}U is the least available radionuclide in the mining sites followed by ^{232}Th which is more prevalent than ^{238}U. While ^{40}K is by far the most abundant radionuclide in the study area as shown infigure 4.1.

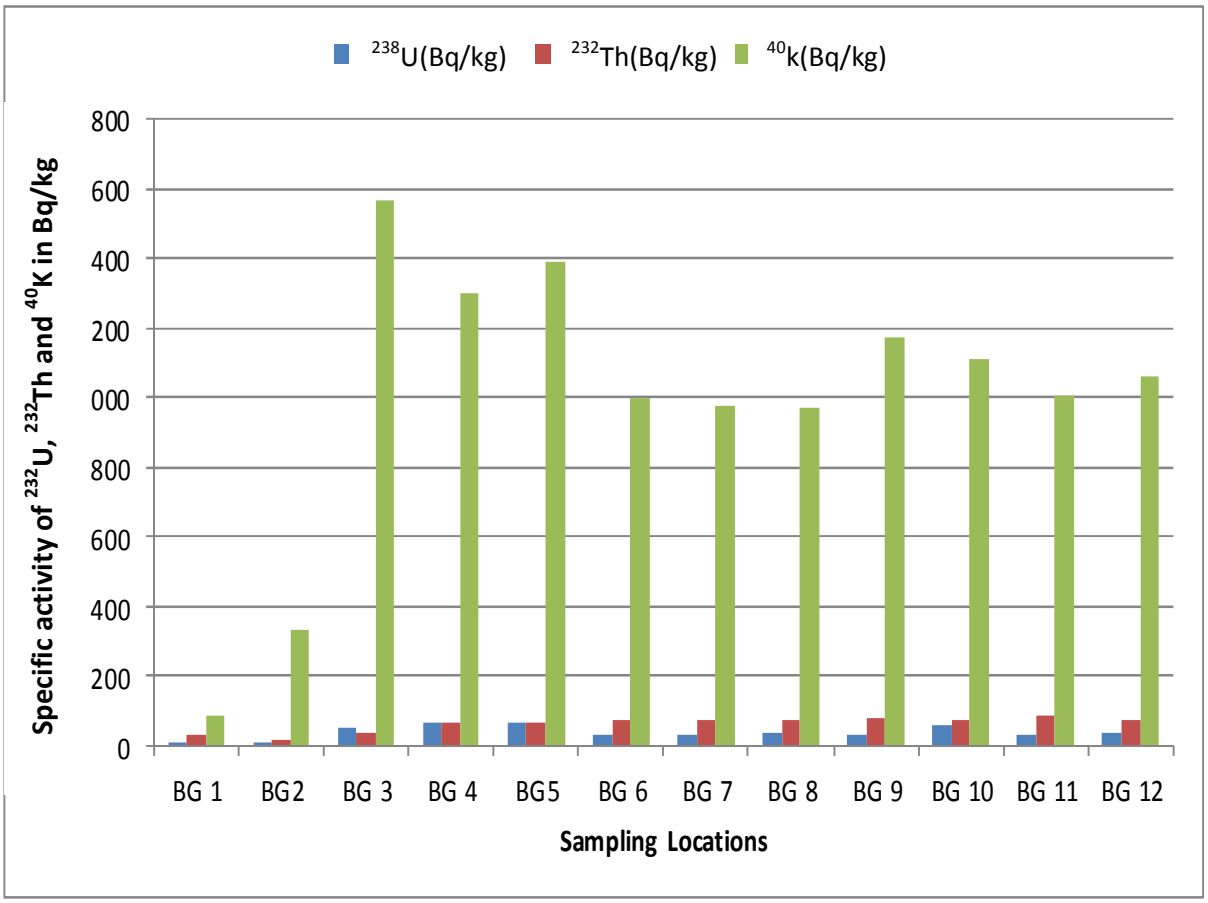


Fig. 4.1: Mean plot for specific activity of ^{238}U, ^{232}Th and ^{40}K per location.

4.1.3 Absorbed Dose Rates

The absorbed dose rate for each of the location were calculated using equation 3.1 below

$D(nGyh^{-1}) = 0.0417A_k + 0.462\ A_u + 0.604\ A_{th}^{-1}$ (3.1)

Where A_k, A_u and A_{Th} are the activity concentrations of ^{40}K, ^{238}U, and ^{232}Th respectively and from table 3.3 the dose conversion factors of ^{40}K, ^{238}U and ^{232}Th are given as 0.0417, 0.462 and 0.604 $nGyh^{-1}$ respectively. Therefore for sample BG 1 A_k, A_u and A_{Th} are 12.35, 28.83 and 85.83.

$= D(nGyh^{-1}) = 0.0417 \times 85.83 + 0.462 \times 12.35 + 0.604 \times 28.83$

$3.578 + 5.71 + 17.41 = 26.67\ nGyh^{-1}$

% contribution of $^{40}K = \frac{3.578}{26.67} x\ 100 = 13.41$

% contribution of $^{238}U = \frac{5.71}{26.67} x\ 100 = 21.39$

% contribution of $^{232}Th = \frac{17.41}{26.67} x\ 100 = 65.29$

The same calculation was done for the remaining samples and the result is summarized in table 4.3.

Table 4.3 : Percentage Contribution to absorbed dose rate due to ^{238}U, ^{232}Th and ^{40}K

S/N	Sample Code	Absorbed Dose in $nGyh^{-1}$	^{238} U%	^{232}Th%	^{40}K%
1	BG 1	26.67	21.39	65.29	13.41
2	BG 2	26.69	10.69	37.68	51.64
3	BG 3	114.03	21.52	21.29	57.22
4	BG 4	123.85	24.88	31.29	43.84
5	BG 5	114.98	13.40	36.04	50.56
6	BG 6	110.63	26.31	36.13	37.56
7	BG 7	100.04	13.69	45.60	40.69
8	BG 8	99.82	16.58	42.99	40.40
9	BG 9	108.57	12.09	42.69	45.23
10	BG 10	117.25	21.90	38.69	39.41
11	BG 11	109.44	13.55	48.18	38.27
12	BG 12	106.24	15.58	42.70	41.76
	Mean	96.52			
	StdErr	9.62			

The result shows that the mean absorbed dose rate in the study area is 96.52 nGy/h with a standarderror of 9.62nGy/h, meaning that the maximum sampling error in the results cannot exceed 9.62nGy/h. The 95% Confidence interval for the mean dose rate in the study area lies between 26.67 and 123.85 nGy/h. In other words, there is 95% assurance that the mean dose rate in the study area lies between the limits (26.67, 123.85)nGy/h.

The percentage contributions to absorbed dose rate due to ^{238}U, ^{232}Th and ^{40}K are shown in figure 4.2 below.

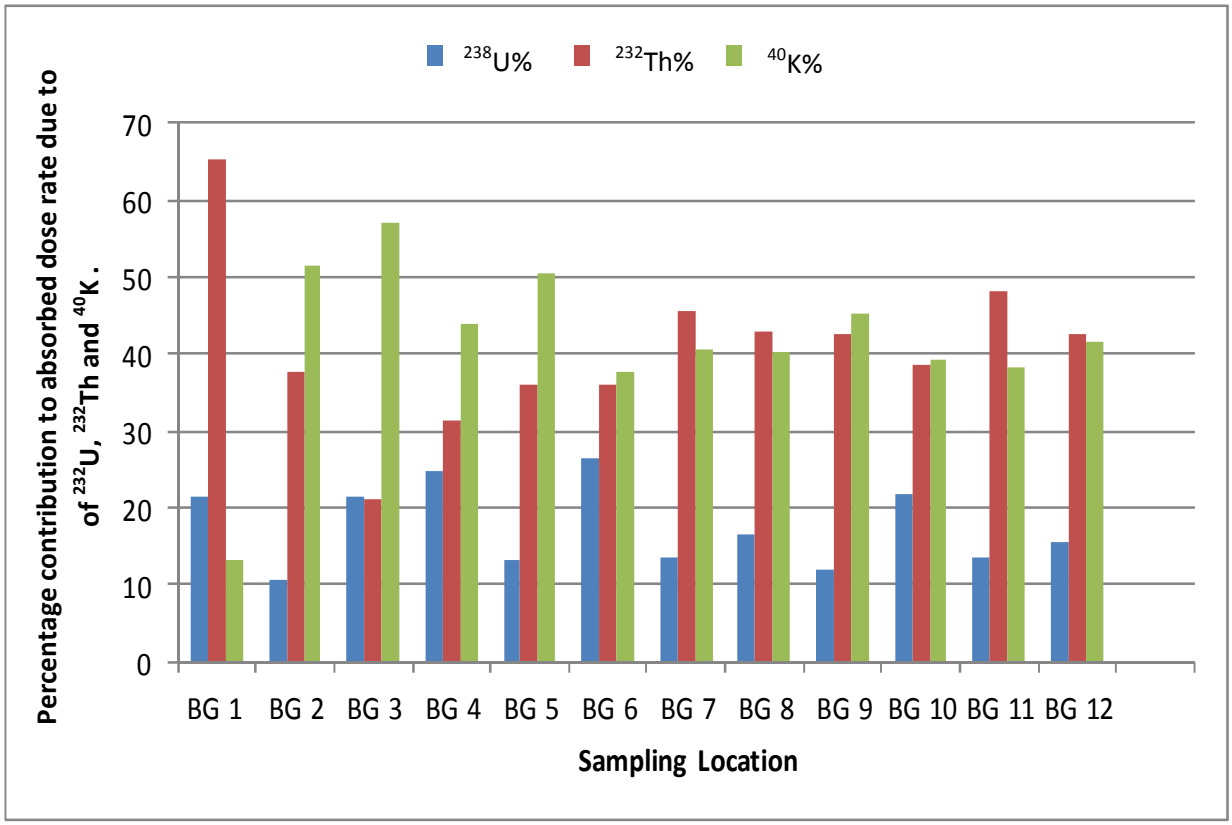


Fig. 4.2: Mean plot for percentage contribution to absorbed dose rate due to ^{238}U, ^{232}Th and ^{40}K per location

4.1.4 Annual Effective Dose Rate

The annual effective dose rate measure in mS_v is calculated from the absorbed dose rate by applying the dose coefficient of 0.7Sv $Gy^{-1}$and outdoor occupancy factor of 0.2 using equation 3.2.

From sample BG1

D = 26.67 $nGyh^{-1}$ =26.67 x 24 x 365 =

T = 0.7Sv and F = 0.2

$H_E = DTF$

$H_E = 26.67\ x\ 24\ x\ 365\ x\ 0.7Sv\ x\ 0.2 = 0.032\ mSv$

The resultfor other samples is summarized in table 4.4.

Table 4.4: Annual effective dose rate in (mS_v)

S/N	Sample Code		Annual Effective Dose Rate In (mS_V)
1	BG 1		0.032
2	BG 2		0.033
3	BG 3		0.140
4	BG 4		0.152
5	BG 5		0.141
6	BG 6		0.136
7	BG 7		0.123
8	BG 8		0.122
9	BG 9		0.133
10	BG 10		0.144
11	BG 11		0.134
12	BG 12		0.130
		Mean	0.118
		StdErr	0.012

The result in table 4.4 shows that the mean effective dose rate is 0.118$mS_v$with standard error of 0.012mS_v meaning that the maximum sampling error in the result from the study area cannot exceed 0.012mS_v. The 95% confidence interval for the mean annual effective dose rate in the study area falls between 0.032 and 0.152mS_v. This shows that there is 95% assurance that the mean annual effective dose in the study area lies between the limits (0.032, 0.152)mS_v/year.

4.2.0 DISCUSSION

Table 4.1- 4.5 show the concentrations of U, Th and % K, the activity concentrations of ^{238}U, $^{232}Th$and ^{40}Kin the soil as well as the calculated absorbed dose rate and the estimated annual effective doses. The percentage contributions of ^{238}U, $^{232}Th$and $^{40}K$to the absorbed dose rates are also provided.

4.2.1 Concentration of U, Th and % K

The mean concentrations of U in the samples is 3.05±0.35ppm the mean values range between 0.5-5.1 ppm, for Th the mean concentration is 15.30±0.23ppm in range between 4.1-21.5ppmwhile the mean % of K is 3.19±0.09% in range between 0.27 to 4.99%. The average crustal abundances of these elements quoted in the literature are in the range 2-3ppm U, 8-12ppm Th and 2-2.5% K (IAEA 2003). This shows that the mean values obtained in this work are slightly higher than the crustal average quoted above.

Table 4.5: Comparison of activity concentration of ^{238}U, ^{232}Th and ^{40}K in soils in the study area and published data (UNSCEAR, 2000, Muhammad *et al.,* 2010, Faanu *et al.,* 2010 and Darko *et al*., 2007).

	Concentration in Soil Bq/Kg					
	^{238}U		^{232}Th		^{40}K	
	Range	Mean	Range	Mean	Range	Mean
This work	6.18-66.69	37.36	16.65-87.29	62.69	85.13- 1564.69	997.57
Nigeria (Zaria)*	4-12	8	15-47	34	317-985	641
Ghana (1)+	8-26	15	9-67	27	60-249	157
Ghana (2)++		29		25		582
Ghana (3)++		35		21		682
Algeria-	2-10	30	2-140	25	66-1150	370
Egypt-	6-120	37	2-96	18	29-650	320
U.S.A-	4-140	35	4-130	35	100-700	370
India-	7-81	29	14-160	64	38-760	400
Malaysia-	49-86	66	63-110	82	170-430	310
Lithaunia-	3-30	20	9-46	25	350-850	600
UK-	2-330		1-180		0-3200	
Hungary-	12-66	29	12-45	28	79-570	370
Spain-			2-210	33	25-1650	470
World- Average		33		45		420

Legend –UNSCEAR 2000 Report, ++ Darko *et al.*, 2007, +Faanu *et al.*, 2010 and * Muhammad *et al.*, 2010.

4.2.2 Activity Concentrations

The mean value of the activity concentrations of ^{238}Uis 37.36±5.45 Bq/Kg in range of 16.18-66.69 Bq/Kg, for ^{232}Th the mean activity concentration is 62.69±6.33 Bq/Kg in range of 16.65-87.29 Bq/Kg and that of ^{40}Kis 997.57±119.97 Bq/Kg in a range of 85.13-156469 Bq/Kg. The highest value (66.69 Bq/Kg) for 238_{U} was measured in soil sample taken from Tsohon Gwari, Kano (BG.4), that of ^{232}Th(87.29 Bg/Kg) was taken from Tsohon Gwari Abuja (BG.11) while that of ^{40}K (1564 Bq/Kg) was taken from Farin ruwa (BG.3).

The mean values of activity concentrations of ^{238}U, ^{232}Thand ^{40}Kare all more than the world average values. (UNSCEAR, 2000). The worldwide average activity concentrations of ^{238}U, ^{232}Thand ^{40}Kin soil samples have values of 33,45 and 420 Bq/Kg respectively (UNSCEAR, 2000).

A comparison of the mean activity concentration of ^{238}U, ^{232}Thand ^{40}Kin soil in the study area with similar studies done in (Zaria)Nigeria and elsewhere are shown table 4.5. The values from this work compared well with published data from other countries and all values were higher than the world average values and other countries with the exception of the activity concentration of ^{232}Th in India and Malaysia that are slightly higher than the mean value of ^{232}Th obtained in this work.

4.2.3 Absorbed Dose Rates and Annual Effective Dose

The mean absorbed dose rate calculated from soil activity concentration was 96.52±9.62 nGy/h in a range of 26.67-123.85 nGy/h which is about $1\frac{1}{2}$ times the world wide average value of 60 nGy/h (UNSCEAR, 2000), about 3 times the value of 29.9 obtained in Ghana (Faanu, 2010) and about twice the value of 46.2 nGy/h obtained in (Zaria) Nigeria (Muhammad *et al*., 2010). The difference could be attributed to the fact that the samples collected in this work were from a mean depth of 17.35m in the range between 2.5-43m below the surface and also the geology and geochemical composition of the sample site.

The mean percentage contribution of ^{238}U, $^{232}Th$and $^{40}K$to the mean absorbed dose rate are 17.63% for ^{238}U, 40.71% for ^{232}Th and 41.66% for ^{40}K. The values show that ^{40}K contribution to the absorbed dose rate is more followed by ^{232}Th while the contribution of ^{238}U is the lowest. Though the study area is K-Feldspar (MMDS 2010) the % contributions of ^{232}Th compared favourable with that of ^{40}K.

The corresponding mean annual effective dose rate in this study is 0.118 ±0.068 mSv and in the range of 0.323-0.867 mSv. In the determination of these values a dose conversion factor of 0.7Sv/Gy and occupancy factor of 0.2 were applied (UNSCEAR,2000). The results from this work shows that the annual effective dose rate is less than the world average value of 0.41mSv/year (UNSCEAR 2000).

CHAPTER FIVE

5.0.0 CONCLUSION AND RECOMMENDATION

5.1.0 CONCLUSION

In this study, data on the activity concentrations of ^{238}U, ^{232}Th and ^{40}K in soil/rocks samples as well as radiation dose in the study area have been established. The activity concentration of ^{238}U, ^{232}Th and ^{40}K in soils/rocks which is one of the potential pathways where members of the public, animal and farm produce could be exposed were quantified using instrumental neutron activation analysis.

In this work, the potential exposure of the population in the study area was assessed by estimating the absorbed dose rate and annual effective dose rate in soil/rocks. The mean absorbed dose rate and the mean effective dose rates were estimated to be 96.52nGy/h and 0.118mS_v/year respectively.The mean annual effectively dose rate is lower than the 1mS_v/year dose rate limit recommended by ICRP for public radiation exposure control. However no amount of radiation is safe because the accumulation of these doses in the study area can have adverse effect over a long period of exposure.

5.2.0 RECOMMENDATIONS

Based on the foregoing the following recommendations are hereby proposed.

- Internal exposure resulting from drinking water ingestion of food and inhalation of radon gas and dust containing U, Th and K should be undertaken in future.
- Determination of gross alpha and beta concentration in drinking water sources in the study area should be done.
- Determination ^{238}U, ^{232}Th and ^{226}Ra, ^{210}Po and ^{210}pb in drinking water from the study area should be carried out.
- Determination of activity concentrations of ^{238}U, ^{232}Th and ^{40}K by gamma spectrometry as well as gross alpha and beta activity concentrations in a variety of food products in the study area (e.g cassava, maize, cabbage etc) is imperative.
- Chemical speciation studies and Assessment of the levels of major and trace metals in a variety of food items in the study area and the mode of translocation from soil to plants should be investigated.

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Appendix I

Result of Neutron Activation Analysis

Appendix II:

Publications from this work