ASSESSMENT OF RADIOLOGICAL HAZARDS AROUND RIRIWAI TIN MINES, KANO STATE, NORTH WESTERN NIGERIA

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A thesis submitted to the School of Post Graduate Studies, Ahmadu Bello University, Zaria in partial fulfillment of the requirements for Award of Doctor of philosophy in Nuclear Physics, Department of Physics, Faculty of Physical Sciences Ahmadu Bello University, Zaria Nigeria.

JUNE, 2017

DECLARATION

I declare that the work in this thesis entitled "Assessment of Radiological Hazard AroundRiriwai Tin Mines, Kano State, North Western Nigeria", has been carried out by me in the Department of Physics. The information derived from literature has been acknowledged in the text and a list of references provided. No part of this thesis was previously presented for another degree or diploma at this or any other institution.

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CERTIFICATION

This thesis entitled "Assessment of Radiological Hazard Around Ririwai Tin Mines, Kano State North Western, Nigeria" by Muhammad Attahiru Abdullahi meet the regulations governing the award of Doctor of Philosophy in Nuclear Physics of the Ahmadu Bello University, and is approved for it contributions to knowledge and literacy presentation.

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Abstract

Mining industry in Nigeria provides economic benefits of wealth creation and employment opportunities. Presently there are numbers of artisanal and large scale mining activities going on across Nigeria and most of these artisanal miners currently under take only surface mining and the process produced large volumes of tailings and waste that may containnaturally occurring radioactive materials (NORMs). Some of the NORMs are soluble in water and have the tendency to leach into water bodies and farm lands. This study assessed the radiation exposure to the public from NORMs around Ririwai Tin mine in Kano state Nigeria. A total of one hundred and four (104) environmental samples comprising of 28 soil, 15 cereals, 11 vegetables, 10 dust and 40 water samples were collected. The samples were analysed using Direct Gamma Spectroscopy (NaI (Tl)), Instrumental Neutron Activation Analysis (INAA) and Liquid Scintillation Analysis (LSA). The exposure pathways considered were external irradiation due to activity concentration of 40 K, 226 Ra and 232 Th in soil and dust, ingestion of food (cereals and vegetables) containing 40 K, 226 Ra and 232 Th and ingestion of 222 Rn in domestic water. The results show that the mean activity concentrations of ⁴⁰K, ²²⁶Ra and ²³²Th in soil samples were 296.87±8.25Bq/kg 49.66±6.56Bq/kg and 257.24±6.53Bq/kg respectively. For the cereals samples the mean activity concentrations of ⁴⁰K, ²²⁶Ra and ²³²Th were 59.99 ± 2.76 , 25.95 ± 2.55 and 46.81 ± 1.99 Bq/kg respectively. The mean activity concentration in vegetable samples were 261.84±4.93, 28.65±4.92 and 56.3±1.66Bq/kg respectively for 40 K, 226 Ra and 232 Th, in dust the activity concentration were 385.90±5.70, 54.31±2.51 and 146.64±0.91Bq/kg for 40 K, 226 Ra and 232 Th respectively. The results in this study are higher when compared withthe worldwide average concentrations of 420Bq/kg, 33Bq/kg and 45Bq/kg for ⁴⁰K, ²²⁶Ra and ²³²Th respectively. The high values obtained in cereals and vegetables could be attributed to the fact that they are also used as phytoremediators of uranium contaminated soil due to their high bioaccumulation of ²²²Ra and ²³²Th. The mean absorbed dose for the soils cereals, vegetables and dust sample were 170.04±4.61, 32.562±1.450, 4632±1.43 and 119.85±9.700nGyh⁻¹ respectively. The absorbed dose rate obtained in this study are also higher than the worldwide average values of 60nGy/h (UNSCEAR). The total annual effective dose calculated in this study was 4.23mSv/year with the highest contribution of 60.20% (2.540mSv/year) coming from cereals, 31.20% (1.320mSv/year) from vegetable, soil contributed 4.93% (0.209mSv/year), dust 3.47% (0.147mSv/year) and the least is 0.04% (0.0163mSv/year) ²²²Rn in water. The total annual effective dose is higher than 1mSv/year dose limit recommended by ICRP. The mean ²²²Rn concentration in soil samples was 78.799±10.197kBq/kg while the mean ²²²Rn Emanation fraction was 1.22±0.013. The average value of the emanation fraction obtained in soil in this study is more than the typical range of 0.05 to 0.7. Similarly, ²²²Rn concentration in three (3) water sources in the study area were determined but high consideration was given to the domestic water sources which has a mean ²²²Rn concentration of 2.23±0.11Bq/L. The result obtained in this study is below the ²²²Rn concentration of 10Bq/L recommended by WHO, UNSCEAR and the maximum permissible value of 11.1Bq/L by USEPA and adopted by Standard Organization of Nigeria (S.O.N). The result of hazard indices shows that the mean Ra_{eq} values in these samples were 436.92±16.950 and 266.98Bq/kg respectively for soil and dusts, only soils samples has Ra_{eq} value above 370Bq/kg recommended maximum value while the dust samples are below the maximum recommended value. Similarly the values of external and internal hazard indices for dust are below unity (1) which is the recommended maximum value, soil samples that has mean values of 1.192±0.051 and 1.326±0.062 for external and internal hazard indices respectively are higher than the recommended values. The total fatality cancer risk from all the exposure pathways considered was 2.33 x 10⁻⁴ ranging from 9.0×10^{-7} to 1.34×10^{-4} the total lifetime fatality cancer risk was 1.63×10^{-2} in a

range of $6.30 \times 10^{-5} - 9.38 \pm 10^{-3}$ the total severe hereditary effect was 8.44×10^{-6} ranging from $3.20 \times 10^{-8} - 5.10 \times 10^{-6}$ while the total lifetime severe hereditary effect was 5.92×10^{-4} in a range of $2.24 \times 10^{-6} - 3.57 \times 10^{-4}$. The negligible cancer fatality risk value recommended by USEPA is in the range of 1×10^{-6} to 1×10^{-4} . The total risks estimated in this study were above the acceptable limit by the UNSCEAR, however, public in the study area may not necessarily be exposed to all the exposure pathways considered in this study at the same time. On the basis of the results from this study, water for consumption do not pose any significant radiation hazards to the population. However consumption of food (cereals and vegetables) grown around the study area could pose radiological hazard due to the bioaccumulation of 226 Ra and 232 Th in them.

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CHAPTER ONE

INTRODUCTION

1.1.0 General Introduction

Mining is a global industry undertaken for its economic benefits of wealth creation and employment. In Africa, 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 (Hayumbu and 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 diseases can be attributed to the heavy metal contamination and naturally occurring radioactive materials NORMs (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 ofheavy-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 organisms when leached into water bodies. Metals contamination in aquatic environmental has received huge concern due to their toxicity, abundance and persistence in the environment 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 that killed more than 400 childrenin Zamfara State(Galadima, 2012).

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).

Human 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) 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 (²³⁵U, ²³⁸U and ²³²Th) 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 ²³⁵U, and ²³²Th decay series and from ⁴⁰K. 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). ²³⁸U and its daughter product are responsible for the major fraction of the internal dose received by human from naturally occurring radionuclides.

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).

Uranium like most heavy metals is chemically toxic and accumulates in kidneys (soluble) and also on bones. The dominant uranium valence states that are stable in geologic environments are uranous (U^{4+}) and uranyl (U^{6+}) states with uranyl being

more soluble than the uranous (NRC, 1999). The transport of uranium occurs generally in oxidising water and ground water as uranyl ion (UO_2^{2+}) or as uranyl fluoride, phosphate, or carbonate complexes. In oxidising and acidic waters, UO_2^{2+} and uranyl fluoride complexes dominate whereas the carbonate and phosphate complexes dominate in near-neutral to alkaline oxidising conditions. Maximum absorption of uranyl ions on natural materials occurs at pH 5.0-8.5. For uranium to be fixed, and thereby accumulate, it requires reduction to U^{4+} by the substrate or by a mobile phase such as H_2S (NRC, 1999).

The relative mobility of the ions of the primordial nuclides in water is of the order of $U^{6+}>U^{4+}>>Th^{4+}$ (Malcolm, 2005). The +6 oxidation state forms soluble uranyl complex ions which play the most important role in uranium transport during weathering. Uranium occurs in numerous minerals such as pitchblende (UO₃.UO₂.PbO) and carnotite (K₂O.2U₂O₃.V₂O₅.3H₂O).

Uranium-238 (238 U) isotope decays by α -emission to 234 Th which also undergoes β -decay to form protactinium-234 (234 Pa) as shown in this expression.

$$\frac{238}{92}U \to \frac{234}{90}Th + \frac{4}{2}He \to \frac{234}{91}Pa + \frac{0}{1}\beta \to \cdots \to \frac{226}{88}Ra + \frac{4}{2}He \to \frac{214}{83}Bi + \frac{0}{1}\beta \dots \to \frac{206}{82}Pb + \frac{4}{2}He \dots \dots (1.1)$$

This reaction involves 14 nuclear decay steps resulting in the emission of eight (8) α -particles and six (6) β -particles. In addition gamma photons are also emitted at energies of 1001 keV (234 Pa), 186 keV (226 Ra), 352 keV (214 Pb) and 609 keV (214 Bi) and finally producing stable 206 Pb to end the decay process. In natural undisturbed soil, 226 Ra is generally in equilibrium with uranium but in disturbed soils they might not necessarily be in equilibrium. The health implications of any metal (uranium, thorium and potassium) depend on the intake and the chemical form (speciation). The

main pathway of uptake of uranium is via the food chain. For a better assessment of uranium transfer from geo-to bio-system and accumulation and distribution in the bio-system, knowledge about the chemical behaviour of uranium is important (Bernhard, 2005). The exact knowledge of the quantity of uranium is a prerequisite for calculation and spectroscopic determination of chemical speciation. Uranium is present in the earth's crust in concentration of about 2.7 ppm (Adekanmi *et al*, 2007).

In the near-surface environment, U and Th may both be mobilised but in different ways. Even though in a naturally undisturbed environment, uranium is generally more soluble than thorium. At low pH, such as in acid-leach uranium mill, thorium becomes more soluble. For instance acid-leach milling might dissolve 30-90 % of the thorium in the ore (NRC, 1999). Thorium has extremely low solubility in natural waters and is entirely transported in particulate matter. Thorium is adsorbed onto the surface of clay minerals. It is a naturally occurring radionuclide and is slightly metallic. When the metal is pure, it is silvery-white and air stable, but tarnishes in air becoming gray and finally black when contaminated with the oxide. Chemically, it is slowly attacked by water and also does not dissolve readily in most common acids, except hydrochloric acid. It also dissolves in concentrated nitric acid containing a small amount of catalytic fluoride ion. Thorium compounds are more stable in +4 oxidation state in aqueous systems. Thorium in the +4 state (Th⁴⁺) undergoes hydrolysis in aqueous solutions above pH 2-3 and is subject to extensive absorption by clay minerals and humic acid at near neutral pH. Thorium-232 (²³²Th) isotope decays very slowly, its half-life is comparable to the age of the Universe. Other thorium isotopes occur in the thorium and uranium decay chains. Most of these are short-lived and hence much more radioactive than ²³²Th though on a mass basis they are negligible. The primary source of thorium is Monazite, a rare-earth and thorium

phosphate mineral. It is also found in small amounts in most rocks and soils, where it is about four times more abundant than uranium. Thorium is adsorbed on the surface of clay minerals. It occurs in several minerals including thorite (ThSiO4), thorianite (ThO₂⁺UO₂) and monazite a phosphate mineral ({Ce, La, Nd, Th} PO₄) and the most common being monazite and may contain up to about 12 % thorium oxide. Thorium like uranium and plutonium can be used as a fuel in a nuclear reactor. Thorium-232 (²³²Th) absorbs slow neutrons to produce ²³³U which is fissile (this technique is employed in the determination of ²³²Th by neutron activation analysis). It undergoes radioactive decay emitting predominantly alpha radiation, beta radiation and some gamma radiation. The alpha radiation emitted through the decay of ²³²Th cannot penetrate human skin, however, if the exposure is internal through ingestion or inhalation there is an increased risk of cancers of the lung, pancreas, blood and liver diseases. In the decay series of ²³²Th gamma photons are also emitted at energies of 239 keV (²¹²Pb), 583 keV (²⁰⁸Tl) and 911 keV (²²⁸Ac) which are used to determine the activity concentrations of ²³²Th by gamma spectrometry. Amplified decay reaction of ²³²Th is shown as expression

$$\frac{232}{90}Th \to \frac{228}{88}Ra + \frac{4}{2}He \to \frac{228}{89}Ac + \frac{0}{1}\beta \to \cdots \to \frac{208}{81}Tl + \frac{4}{2}He \to \frac{208}{82}Pb + \frac{0}{-1}\beta \quad \dots \dots (1.2).$$

Potassium has 24 known isotopes three of which occur naturally: ³⁹K (93.3%), ⁴⁰K which is the radioactive isotope of terrestrial importance (0.0117%) and ⁴¹K (6.7%). Naturally occurring ⁴⁰K decays to stable ⁴⁰Ar (11.2%) by electron capture and by positron emission, and decays to stable ⁴⁰Ca (88.8%) by beta emission. During the decay process out of 100 disintegrations, 89 results in the release of beta particles with maximum energy of 1.33 MeV and 11 results in the release of gamma photons with maximum energy of 1.46 MeV. Potassium-40 (⁴⁰K) decays by beta (β-) emission to ⁴⁰Ca and by electron capture (E. C.) to ⁴⁰Ar as shown in figure 1-1.

$${}^{40}_{19}K \rightarrow {}^{40}_{20}Ca + {}^{0}_{-1}e$$

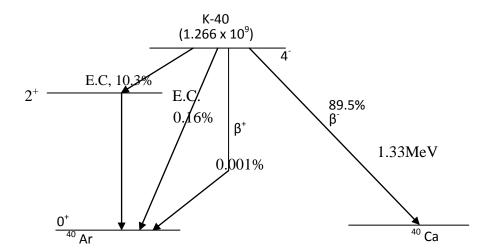


Figure 1-1: Decay scheme of ⁴⁰K

It has a half-life of 1.250×10⁹ years. The decay of ⁴⁰K to ⁴⁰Ar enables a commonly used method for dating rocks. Besides the dating, potassium isotopes have been used extensively as tracers in studies of weathering. They have also been used for nutrient cycling studies because potassium is a macronutrient required for life. The potassium content in the body is under homeostatic control and is little influenced by environmental variations and as a result the dose from ⁴⁰K in the body is reasonably constant (NRC, 1999). Potassium (and thus in some commercial salt substitutes) in sufficient quantity that large amounts of those substitutes can be used as a radioactive source for classroom demonstrations. In healthy animals and people, ⁴⁰K represents the largest source of radioactivity, greater even than ¹⁴C. In a human body of 70 kg mass, about 4,400 nuclei of ⁴⁰K decay per second. The activity of natural potassium is 31 Bq/g (Knoll, 1989).

1.2.0 Statement of the problem

Mining activities have been identified as one of the major source of exposure to NORMs, in recent times, there is an increased awareness of the potential problems of NORMs and this has resulted in most countries taking steps to implement regulations dedicated to radiation protection and safety. Hence the establishment of Nigeria Nuclear Regulatory Authority (NNRA).

In the last decade, a number of international meetings have been dedicated to the radiological consequences of NORMs and these have contributed to world-wide cognition of the issues involved (Van der Steen and Van Weers, 1996). Despite these studies and meetings on NORMs, there is still lack of enough information on the awareness on the radiological hazards and levels of exposures of NORMs in many countries by legislators, regulators and operators.

The Nuclear Safety and radiation protection act 1995 no. 19 and the federal Government of Nigeria Official Gazette No 123 vol. 90 of 2003 empowered NNRA to make regulation on the exposure limit of ionizing radiation dose to different category of persons and organs. Sequel to the current Nigeria's economic dwindling due to global fall in the oil prices, the Federal Government is now diversifying to other sector such as the exploration and exploitation of solid minerals, therefore, it is imperative to carry out radiological assessment of all the area where artisanal mining are taking place so as to have an insight of the radiation levels.

Data/information on radiation level due to NORMs around mining areas across Nigeria are very scanty. It is only around Plateau old tin mine that comprehensive and relevant works on radiation due to NORMs were carried out.(Nasiru 1993, Ibeanu, 1995, Mangset and Sheyin 2009).

The study area of this work is Ririwai, the headquarters of Doguwa local government area, Kano state, Nigeria. Ririwai houses the old international commercial underground tin mine which was closed in 1985. Beside tin other industrial and energy minerals such as uranium, thorium, lead, e.t.c were reported to be in commercial quantities in the area (Kinnaird and Bowden 1985). Similarly ECOPHOENIX an exploration company shows that the area contained high grade of niobium and uranium. This therefore makes it necessary to assess the level of ionizing radiation as a results of the current surface Artisanal tin mining taking place in the area prior to the resumption of large scale commercial tin mine and other industrial and energy minerals present in the area.

1.3.0 Aim and Objectives

1.3.1 Aim

The aim of this study is to assess the radiological hazard due to naturally occurring radioactive materials around Ririwai tin mines, Kano state with the following objectives:

- i. To determine the activity concentrations of 40 K, 226 Ra and 232 Th in soil, cereals vegetables and dust.
- ii. To determine 222Rn concentrations and emanation fraction in soil
- iii. To determine ²²²Rn concentrations in water sources
- iv. To determine absorbed dose rate and annual effective dose due to (i) (iii) above.
- v. To determine the hazard and risk indices due to (i) (iv) above.
- vi. To provide necessary suggestions and recommendations.

1.4.0 Justification

The result from this work would give a base line information on the level of exposure to ionizing radiation from natural sources to the artisanal miner and the general public in the study area and other areas with similar geochemical settings. Geological studies have shown that the area has high deposit of niobium, uranium and thorium this further calls for detailed study of radioactivity levels in the area.

Information from this type of study would help the NNRA to effectively monitor potential NORM producing activities such as mining. Moreover, availability of data from this study is very vital to all stakeholders involved since it will add to the body of knowledge required for the enforcement of regulations meant for safety and radiation protection in Nigeria and elsewhere.

CHAPTER TWO

LITERATURE REVIEW

2.1.0 Background

In the developed countries such as members of the European Union (EU), each member country is obliged to identify work activities that cannot be ignored from the radiological protection point of view. This action has increased the awareness of the potential problems enormously, and most of the EU member states have now implemented regulations dedicated to natural sources of exposure (EC, 1996). Recently there are a lot of reports on NORM with respect to occupational and public exposure situations that have been published recently (Van der Steen and Van Weers, 1996; IAEA, 2003 and ICRP, 2007) that have all contributed significantly to the recognition of the radiological consequences and risk associated with NORM. On the average, the annual global effective dose due to exposure to 36 NORM has been estimated to be 2.4 mSv with a typical range between 1-10 mSv (UNSCEAR, 2000). The main sources giving rise to this dose has been identified to be; cosmic rays, terrestrial gamma rays (referred to as external exposure), inhalation mainly of radon gas and ingestion of materials with NORM (referred to as internal exposures) [UNSCEAR, 2000]. Also 50 % of this global annual effective dose has been estimated to arise from radon exposure with a value of about 1.2 mSv (UNSCEAR, 2000).

Studies have also established that, radiation exposure above certain threshold limits can damage living cells, causing death in some of them and modifying others [UNSCEAR, 2000]. If the repair of the damage or modified cells is not perfect, the resulting modification will be transmitted to other cells and this may eventually lead

to cancer. The biological damage due to radiation exposure could lead to somatic stochastic effect or hereditary stochastic effects.

Stochastic effect is radiation effects, generally occurring without a threshold level of dose, whose probability is proportional to the dose and whose severity is independent of the dose (IAEA, 1996). Radiation exposure has also been associated with most forms of leukaemia and other types of cancers affecting various organs such as lungs, breast and thyroid glands. It is also worth noting that radiation-induced cancer may manifest itself decades after exposure (UNSCEAR, 2000). Radiation exposure also has the potential to cause hereditary effects in the offspring of persons exposed to radiation.

Human activities such as mining and use of ores containing natural radioactive substances and the production of energy by burning coal that contain such substances are known to have enhanced the exposure from natural sources of radiation (UNSCEAR, 2000). Such human activities generally give rise to radiation exposures that are only a small fraction of the global average level of natural exposure. However, specific individuals residing near installations releasing radioactive materials into the environment may be subjected to higher exposures. It should be noted that, should some of the sites with high levels of radioactive residues be inhabited or re-inhabited, the settlers would incur radiation exposures that would be higher than the global average level of natural exposures (UNSCEAR, 2000).

There are several pathways by which the radioactive material can reach humans. The pathway largely depends on the processes involved and can be broadly categorised into; on-site, off-site, airborne, waterborne, food products, etc (O'Brien and Cooper, 1998). The dominant exposure pathways in most situations are external gamma radiation,

inhalation of radon gas and its decay products, ingestion of contaminated food and/or water (O'Brien and Cooper, 1998).

2.2.0 Sources of exposure to NORM

All living organisms are continually exposed to ionizing radiation from natural sources.

The levels of exposure vary depending on location and altitude. The main sources of exposure are:

- i. Cosmic rays that come from outer space and from the surface of the sun;
- ii. Terrestrial radionuclides that occur in the earth crust;
 - In building materials and in air,
 - Water and foods and,
 - In the human body.

Table 2-1 shows the world wide average annual effective doses for the various sources.

Table 2-1: Average radiation exposure from natural sources

Source	Worldwide average annual effective dose, mSv	Typical range
External		
Cosmic rays	0.4	0.3 - 1.0
Terrestrial rays	0.5	0.3 - 0.6
Internal		
Inhalation (radon)	1.2	0.2 - 10
Ingestion	0.3	0.2 - 0.8
TOTAL	2.4	1-10

Source: (UNSCEAR, 2000).

Cosmic radiation has been identified to be intense at higher altitude. The terrestrial Radionuclides which have been in existence since the creation of the earth are known as primordial radionuclides. They include: 40 K with a half life of 1.28 x 10^9 years, 232 Th with a half life of 1.41 x 10^{10} years, and 238 U with a half life of 4.47 x 10^9 years. Other primordial radionuclides of secondary importance include: 235 U with a half life of 7.04 x 10^8 years and 87 Rb with a half life of 4.70 x 10^{10} years. Of these radionuclides, thorium

and uranium lead a series of several radionuclides, many of which contribute to human radiation exposure.

2.2.1 Exposure from U, Th and K

The uranium atom, consists of three different isotopes: about 99.3% of naturally occurring ²³⁸U, about 0.7% ²³⁵U and trace quantities of (about 0.005%) ²³⁴U. The ²³⁸U and ²³⁴U belong to one family called the uranium series (4n⁺²), while the ²³⁵U isotope belongs to another series called the actinium series (4n⁺³). The most abundant (about 100%) of the naturally occurring radioisotopes ²³²Th, is the first member of another long series called the thorium series (4n). The identification numbers are based on the divisibility of the mass numbers of each of the series by 4 (Cember, 1996).

The natural radionuclides exist in secular equilibrium in natural undisturbed environments (Cember, 1996). Due to physicochemical processes in the earth crust, such as leaching and emanation, the radiological secular equilibrium in each series may be disturbed (UNSCEAR, 1993, 2000). Under normal undisturbed secular equilibrium conditions, it has been established that, the mass ratio of ²³⁵U to ²³⁸U is about 0.0073 and activity ratio of 0.046 (UNSCEAR, 1993). In the case of ⁴⁰K they undergo beta decay to stable species (⁴⁰Ca). These radionuclides are present in varying degrees in water, air, soil and in living organisms. As a result, human beings are exposed to external and internal irradiations by gamma rays, beta particles and alpha particles with varying ranges of energies (UNSCEAR, 1993).

The activity concentrations of ⁴⁰K in soil have been found to be higher than ²³⁸U and ²³²Th. According to the UNSCEAR report (UNSCEAR, 1982), activity concentrations of 370 Bq/kg, 25 Bq/kg, 25 Bq/kg have been reported for ⁴⁰K, ²³⁸U and ²³²Th respectively. Other sources have reported activity concentrations of 35 Bq/kg, 30 Bq/kg, and 400 Bq/kg for ²³⁸U, ²³²Th and ⁴⁰K respectively (Bozkurt et al., 2007; UNSCEAR, 2000).

The reported worldwide annual average absorbed dose rate in air from terrestrial gamma radiation is estimated to be 59 nGy/h in a typical range of 10 to 200 nGyh-1 (UNSCEAR, 2000). The direct measurements of the indoor and outdoor absorbed dose rates in air in some countries have reported average values of 59 and 57 nGy/h respectively (UNSCEAR, 1993). High dose rates have been measured in different parts of the world such as the Nile Delta where dose rates in air have been estimated to be in a range of 20-400nGy/h and also in the Ganges Delta in a range of 260-400nGy/h. Dose rates of the order of 12,000nGy/h have also been measured in thorium-bearing carbonatite in an area near Mombasa on the coast of Kenya. In Brazil, where there is mixed thorium and uranium mineralization, dose rates measured are roughly in a range of 100-3500 nGy/h (UNSCEAR, 2000).

According to literature, thorium bearing and uranium bearing materials have resulted in higher absorbed dose rates around the world (UNSCEAR, 1993). The estimation of the annual effective doses from these activity concentrations takes into account the conversion coefficient from absorbed dose in air to effective dose of 0.7 Sv/Gy, Indoor and outdoor occupancy factors of 0.8 and 0.2 respectively and these are age and climate (location) dependent (UNSCEAR, 1993, 2000).

Internal exposure to radiation is mainly due to ingestion and inhalation of materials containing ²³⁸U and ²³²Th decay series and ⁴⁰K. The committed effective doses are determined through analysis of the radionuclide contents in foods and water following an intake and in addition to bioassay data and knowledge on the metabolic behaviour of the radionuclides (UNSCEAR, 2000). Concentrations of NORM in foods vary widely because of differences in background levels, climate and the agricultural conditions that prevail. The body content of ⁴⁰K is about 0.18 % for adults and 0.2 % for children. The natural abundance is about 1.17 x 10⁴% and specific activity concentration of 2.6 x 10⁸Bq/kg. The corresponding annual effective doses from ⁴⁰K in the body are 165 and

185 μ Sva⁻¹for adults and children respectively. The total annual effective dose from inhalation and ingestion of terrestrial radionuclides is 310 μ Sv of which 170 μ Sv is from ⁴⁰K and 140 μ Sv from the long-lived radionuclides in the uranium and thorium series (UNSCEAR, 2000). Uranium in the body is retained primarily in the skeleton and the concentrations have been found to be approximately similar in various types of bones. Similarly, thorium is mainly deposited on bone surfaces and retained for a long period following intake by ingestion and inhalation. The annual effective dose from reference values of U/Th series radionuclides has been evaluated to be 130 μ Sv [UNSCEAR, 1988, 1993) and re-evaluated in the year 2000 to be 120 μ Sv (UNSCEAR, 2000).

2.2.2 Exposure from radon

Radon is a gas with three natural isotopes of the radioactive element: Actinon, (219 Rn) from the 235 U decay series; Thoron (220 Rn) from the 232 Th decay series; and Radon (222 Rn) from the 238 U decay series (UNSCEAR, 1993).

Due to the low activity concentration of 235 U and the short half-life of 219 Rn of 3.96s, the radiation exposure from 219 Rn is not significant for human exposure. Radon-220 (220 Rn), with a half-life of 55.6 s is of concern only when the concentration of 232 Th is high.

The isotope of concern in terms of human radiation exposure is 222 Rn which has a relatively longer half-life of 3.82 days. It is a noble gas with a slight ability to form compounds under laboratory conditions. It has a density of 9.73 g/L at 00 C. The solubility of 222 Rn in water at 00 C is 510 cm 3 /L decreasing to 220 cm 3 /L at 25 00 C and 130 cm 3 /L at 50 00 C.

The production of ²²⁰Rn and ²²²Rn in terrestrial materials depends on the activity concentrations of ²²⁸Ra and ²²⁶Ra present, respectively which are predominantly alpha emitters. Radon is the most significant element of human irradiation by natural sources. The most significant mode of exposure is the inhalation of the short-lived products, ²¹⁰Pb and ²¹⁰Po of the parent isotope ²²²Rn (UNSCEAR, 1993; 1996 and 2000).

The concentrations of ²²²Rn in surface air are quite variable with time-average concentrations in the range of 2-30 Bq/m³(UNSCEAR, 1993). In the soil and also in the root zone, radon concentrations may be higher by a factor of about 1000 than in the open air (UNSCEAR, 1996). The average concentration varies widely depending on the composition of the soil and the bedrock. For soil with an average ²²⁶Ra concentration of 40Bq/kg, the average ²²²Rn concentration in the soil, in water would be about 60 Bq/m³. Much higher values of 8000 Bg/m³ and 50, 000 Bg/m³ have been measured in deep ground waters in areas such as Maine in the United States of America and in Finland respectively (UNSCEAR, 1996). The action level of radon recommended by the ICRP for which intervention is necessary is 1000 Bq/m³. This value is based on an assumed occupancy of 2000 hours per year and this is equivalent to an effective dose of 6 mSv per year. This value is also the midpoint of a range of 500-1500 Bq/m³(ICRP, 1993). Radon can present hazards in a wide range of work places including the mining industry and other work places other than the mines. Specific measures need to be put in place to reduce radon concentrations in air and water to prevent concentrations reaching very high levels even in places where the concentrations of uranium and radium in raw materials may be very low (UNSCEAR, 2000). The mechanisms by which radon enters buildings is pressure driven flow of gas from soil through cracks in the floor. In addition, most building materials produce some radon due to the presence of elevated levels of ²²⁶Ra and high porosity of the materials allow for the escape of the gas (Van der Steen and Van Weers, 1996).

2.3.0 Tin and Tin Mineralisation

2.3.1 Tin (Sn)

Tin is a metallic element belonging to group 14 of the periodic table, along with C, Si, Ge and Pb. The element has an atomic number of 50, an atomic mass of 119, three main oxidation states (-2, +2 and +4), of which the +4 state is by far the most

common in nature, and ten naturally occurring isotopes, the largest number of all elements (112 Sn, 114 Sn to 120 Sn, 122 Sn and 124 Sn), of which 120 Sn, 118 Sn and 116 Sn are the most abundant at 32.97%, 24.01% and 14.24% respectively of its mass. Tin shows intermediate characteristics in its chemistry between that of Ge and Pb.

Like Pb, Sn is a relatively rare metal, with an average crustal abundance of 2.1 mg kg¹, but it is well known as a metallic element, because of its use in household products and the relative ease of its extraction from natural sources. It is one of the seven metals known in antiquity. Tin is a siderophile metallic element forming several minerals, including cassiterite SnO₂ and the rarer stannite Cu₂FeSnS₄, but can also be present as an accessory element in biotite, muscovite, amphibole, sphene and rutile.(Mielke,1979)

2.3.2 Tin Mineralisation

Cassiterite, the most common tin bearing mineral is associated with anumber of mineral impurities such as monazites, zircon, thorite andxenotime which contain radioactive U and Th that may constituteradiological hazards.(Rabiu,1993).Accordingly,zircon has a chemical formula ZrSiO₄ and is made up of 67.18 ZrO₂ (Zr 49.58), SiO₂ 32.98. Zircon always has a slight admixture of Fe₂O₃ (up to 0.358) and more), often CaO (0.05 to 48) and sometime Al₂O₃. Generally, zircon occurs as small, rare disseminated crystals in magnatic intrusive rocks, nepheline synenites, granites, diogranitic pegatities.

Thorite has a chemical formula $ThSiO_4$. It contains about 10 to 168^{Th} at times with a higher water content. Also contains rare earths, CaO up to 138 of Fe_2O_3 etc.

It is usually formed in the latest stages of crystallization of certain acidic and alkaline magnetic rocks.

Monazite has a chemical formula (Ce, La, Th and Ca) (PO₄ SiO₄, SO₄). It contains an isomorphous admixture of ThO₂ (5 to 108, sometimes as high as 28%) in some instances ZO₂ (up to 78) with SiO₂ (up to 68). Monazite usually occurs in pegmatites, sometimes in granites and gnemissens in paragenesis with magnetite, ilmenite and other minerals.

Xenotime has a chemical formula YPO₄ (638). Commonly it contains small amounts of rare earth (Eu and Le) sometimes ThO₂, UO₂ (up to 58), ZO₂ (up to 38), SnO₂ (up to 98). Xenotime occurs in granites and pegmatites as small disseminated crystals often in association with zircon.

2.3.3 Tin mining activities in Nigeria

Tin mining in Nigeria started around Jos plateau many decades before the European occupation (Rabiu, 1993). Major exploitation of the tin fields started after the discovery of rich alluvial deposits in Jos while extensive commercial mining and processing started in 1930s (Onuoba, 1992). Another area of large deposit of Tin is Ririwai the head quarter of Doguwa L.G area of Kano state which housed the largest underground tin mine that was close in 1984. Other location where artisanal Tin mine are taking place are Toro in Bauchi state and some parts of Nasarawa State. In these areas extensive quantity of mine tailings have generated by the several mining companies and the artisanal miners operating in the areas, these tailings are dumped around the premises of the companies or in the mining pits (Rabiu, 1993).

2.4.0 Geochemical settings of the study area

The Ririwai Complex is located off Saminaka - Kano road north east of the Kudaru Complex. The complex forms a concentric circle and is made up of pre-caldera agglomerates, ash fall tuffs, crystal-poor ignibrites and minor comenditic ignibrites, basalts, quartz porphyry, granite porphyry, aegirine arfvedsonite granite and porphyry,

biotite granite, biotite microgranite, as well as arfvedsonite albite granite. Rocks here are generally purple to brownish in colour with feldspars, quartz and biotites clearly seen. The texture vary from fine to medium, but coarse in some areas.

The Ririwai complex occupied an area of approximately 180km^2 , it is oval in shape, measures $17 \times 16 \text{km}$ and is situated between latitudes $8.41 - 8.48^0 \text{E}$ and longitudes $10.41 - 10.48^0 \text{N}$. It is enclosed within an outer ring-dyke of Fayalite granite porphyry, rising steeply from the level pan-African plain. Instrusions of peralkaline granite are separated from the outer ring-dyke by a broad, hilly tract of volcanic rocks. It is principally within the biotite granite that the ore deposits of cassiterite, columbite, wolframite and base metal sulphides are found (Kinnaird, 1985). Excepts for the columbite and little dispersed cassiterite the ore minerals occur within a broaded quartz-greisen system termed the Ririwai lode.

The Ririwai lode comprises a series of parallel to sub-parallel or braided quartz veins separated by zones of grey greisens, grading outwards in to a reddened micro-climized wall rock – occasionally through a buff- coloured zone into a pale pink boitite granite.

2.5.0 Mineralization of the study area

The following are the descriptions of minerals sequence of ore formation and the characteristics of each of the ore generations in the Ririwai lode as enumerated by Kinnaird et al., 1984., Szentes 2008, and Oyavoye, 1964.

Monazite occurs in minor amounts as 100×10 to $300 \times 60 \mu m$ yellow lath-shaped crystals, which are associated with cassiterite and molybdenite within the micas of the greisen and wall rock. Often it is haematitically stained or contains traces of enclosed chalcopyrite and pyrite $10\text{-}20 \mu m$.

Ilmenite is rare forming 20×2 to $120 \times 10 \mu m$ laths together with traces of magnetite crystals up to 60 /urn in diameter. Usually it is associated with laths of orange TiO_2 within the wall rock.

Zircon forms euhedral, highly zoned crystals 20-100μm in length within mica, or up to 200μm between quartz crystals, in both the wall rock and greisen. Zircon is common but is especially concentrated at the junction between the greisen and the quartz vein. Collections of between 5 and 30 zircon crystals form aggregates up to 400μm in diameter.

Early generations of cassiterite occur in the wall rock as small $10\text{-}150\mu\text{m}$ euhedral, light coloured, zoned and simply twinned crystals; or as larger darker coloured crystals (50-800 μ m in diameter) within the micas of the greisen. Collections of smaller cassiterite crystals up to $300\mu\text{m}$ in diameter, often surround zircon or monazite, whereas cassiterite itself is often surrounded by, or enclosed within, euhedral Ti0_2 minerals, columbite, wolframite, molybdenite and sphalerite crystals (40- $100\mu\text{m}$), Many cassiterites carry abundant 2- $6\mu\text{m}$ inclusions of columbite and Ti0_2 .

2.6.0 Review of Works Carried Out in the Study Area

The work carried out by Abaa (1975) shows that in Ririwai, two phases of mineralization have been found; a prejoint autometamorphic mineralization in which K-feldspars replaced earlier perthites, while new albite developed. Silicification and recrystallisation changed the original rock texture and introduced dispersed mineralisation with thorite, columbite, xenotime and hafnium-uranium rich zircon as well as enrichment in some trace elements. The second phase of mineralisation was a post-joint replacement greisen-mineralisation which took place in the roof zones of the cooling consolidated biotite granite. It involved a metasomatic introduction of cassitori to and sulphide ores into the crystalline biotite granite along cooling joints, fissures and fractures. The sequence of replacement has been found to be haematisation and kaolinisation - chloritisation and sericitisation greisenisation - silicification.

According to Bowden *et al.*,(1981)Ririwai biotite granite shows that the surface of biotite granite represented a zone in the shallow dipping roof from which the volcanic cover has been removed relatively recently chilled margins remains as isolated areas of microgranite within variably textured biotite granite. Texture range from coarsely porphyritic to medium grained equinular, both on the surface and underground to depth of at least 400m. The work also show a progressive uranium loss relative to Th of surface samples in the roof zone of biotite granite at Ririwai The U-Th Variation for surface samples is interpreted as the result of Uranium mobilization to higher levels of the complex which have subsequently been eroded with Th retailed in accessory minerals. Crystallization of both peralkaline and aluminous granites leads to enriched concentrations of U and Th in residual fluids. There is evidence of considerable mobility of both U and Th during post-magmatic periods of rock-fluid interaction leading to considerable enrichment of Uranium the paper added.

In a related development Kinard *et al.*,(1985) revealed that the interaction between crystal and fluids have been monitored by XRD, XRF, INAA, wet chemical analyses and fluid inclusion studies. Ore mineralogy confirms the paragenetic columbite (pyrochlore)-cassiterite-sphalerite evolution. The metasomatic reactions commence with Na metasomatism followed by K, then H and finally Si. The latter reactions are best displaced in the Ririwai loce where particle track studies have delineated the relative mobility of U and Th, Mineralization in biotite granite can be grouped according to the dominate metasomatics process Columbite, minor cassiterite and sphalerite can be equated with albitite formation. Potash metasomatism generated columibite, wolframite, cassiterite and sphalerite deposition, which continued into greisens formation as H metasomatism. According to isotopic data the source of the

ore metals and the granite magma was probably the Pan-African continental crust with contributions from the mantle.

Abiye (2005) studied the natural radiation levels and distributions of dose rates within the younger granite province of Nigeria, the study indicated that the Ririwai complex contains several distinctive lithologies derived from post-metasomatism of granite by a clear sequence of alterations. Accordingly, high concentrations of radionuclides occur within the different rock types in this complex. The radionuclides include cesium (Cs), hafnium (Hf), thorium (Th), scandium (Sc), rubidium (Rb), zirconium (Zr), strontium (Sr), yttrium (Y), uranium (U), lanthanum (La), neodymium (Nd), samarium (Sm), europium (Eu), gadolinium (Gd), thulium (Tm), ytterbium (Yb), and lutetium (Lu) among others. They mostly belong to the lanthanide and actinide series, and occur in coarse-grained discrete crystals associated with magmatic quartz or as aggregates of small crystals, which are often aligned along the cleavage of micas, or form complex intergrowths outside the micas.

In this complex for instance, zirconium (Zr) concentration of between 680–3,075 ppm have been reported within the riebeckite granite and albite-riebeckite granite as against an average value of only 235 and 175 ppm within the Jos-Bukuru Complex and the Pankshin Complex respectively

The ratio of concentration of the radionuclides found within the Ririwai Complex compared to other complexes such as Jos-Bukuru and Pankshincomplexes as calculated from the result of earlier geochemical analysis by reveal a value of between 2.8 and 26.75. These factors therefore explain why radiation dose rates are higher within the Ririwai Complex compared to other Nigerian Younger Granite complexes. Substantial Uranium mineralization occurs in the Ririwai area of southern

Kano. According to Ogedengbe (1984), uranium occurred in peraluminous and peralkaline granites and the content of uranium in peraluminous granite lies between 16 and 32 ppm .(Karniliyus, *et al.*, 2014).

Table 2.2: Comparative Uranium Content in Nigerian Rocks11

LOCATION		ROCK TYPE	URANIUM CONTENT	U.EST RESERVE
Ririwai, Kano state		Peraluminuous Granite	16 – 32 ppm	
Kaduna, Katand Zamfara		Residual soils	1 – 12 ppm	
Adamawa State	Mika	Pan-African granites	10 ppm	52 tons
	Gumchi	Jurassic rhyolite dykes	114 ppm	
	Mayo	Mylonitized, sheared and brecciated fine to porphyritic granites	2,000 ppm	100 tons
	Lope	Bima Sandstone	1,826 – 2,375 ppm	
	Zona	Bima Sandstone	0.01 - 128 ppm	
Osun, Kwara, Kogi and Ekiti States		Residual soils	> 500 ppm	

Source: Karniliyus, et al., 2014.

EcoPhoenix and CSA Global (2008) considered Ririwai the (Kaffo Valley) to have deposit of potentially huge economic significance. The minerals of interest are pyrochlore (containing niobium and uranium) and cryolite. These minerals are contained as accessory constituents throughout, unusual highly-sodic, albite-arfvedsonite granite. The granite body is roughly circular with a diameter of just over one kilometre. This granite is part of a larger area of volcanic and sub-volcanic rocks forming the Ririwai ring complex that is one of the 'younger granites' of Jurassic age that occur in parts of Kaduna, Plateau, Kano and Bauchi States.

Statistical and geological analysis of British Geological Survey data predicts that 47mt of high grade ore exists at Kaffo Valley. This would support at least a 20 year

mine life at 2mtpa. Given that the predicted high grade zone can be delineated and improvements on recovery using modern metallurgical methods are possible, then CSA Global have demonstrated that a theoretical NPV of US\$776 million is possible for a 20 year mine at 2mtpa.

2.7.0 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 ²¹⁰P_b, ²²⁶R_a and ²²⁸R_ain 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 element presence in the Gold ore samples.

Radioactivity concentration measurements of ²³⁸U, ²³²Th, ⁴⁰K, and ¹³⁷Cs 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 ²³⁸U, ²³²Th and ⁴⁰K 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 ⁴⁰K, ²³²Th and ²³⁶U in appreciable amount. The maximum mean dose equivalent obtained in the study area are lower than the allowable maximum limit.

The activity concentrations of naturally occurring radionuclides (²³⁸U, ²³²Th and ⁴⁰K) were investigated ingold ore mined from Birnin Gwari Artisanal Goldmine in Kaduna state Nigeria using Instrumental NeutronActivation Analysis (INAA) technique. Twelve samples were collected from pits at different depths and thelocations of pits were marked out using Global Positioning System (GPS). The measured activity concentration dueto ²³⁸U range from 6.18±3.7 to 66.69±4.9 Bq/kg with mean value 37.36±5.45, Bq/kg ²³²Th range from 16.65±0.8 to87.29±1.2 Bq/kg with mean value of 62.69±6.33 and ⁴⁰K range from 85.13±4.5 to 1564.69±57.9Bq/kg with a meanvalue of 997.52±11 9.97 Bq/kg. The value compared well with published data from other countries and all valuesfrom this study are higher than the world average values(Zakari *et al.*, 2013, Nasiru *et al.*, 2013).

Girgisu *et al.*,(2014) assessed the radiological levels from Awwal artisanal gold mining exercises in Kebbi State.Results show mean values of activities of 40K>226Ra>232Th numerically as 425.96 ± 5.56 , 23.85 ± 2.01 and $18.80\pm1.21Bq/kg$, respectively. The average outdoor gamma dose was 34.26 nGy/h while the mean annual effective dose rate was $42.15 \,\mu Sv/year$ (= $0.042 \,mSv/year$), which is less than $0.07 \,mSv/year$ benchmark given in UNSCEAR (1993).

In another study, eighteen groundwater samples were collected from wells in villages around Zona, an area reported to host uranium mineralization and these were analyzed for mass/activity concentration of ²²⁶Ra, ²²⁸Ra and ²³²Th. Consumption of groundwater with elevated levels of ²²⁶Ra, ²²⁸Ra and ²³²Th may result to cancer, kidney and/or developmental defects in humans and other animals. The results obtained were compared for compliance with international guidelines for radionuclides in drinking water. Results of the study showed that activity concentration of ²²⁶Ra, ²²⁸Ra and mass concentration of ²³²Th ranged from 0.05 to 6.7pCi/L, 0.2 to 4.8pCi/L and 0.02 to 1.10µg/L, respectively. The higher levels of ²²⁶Ra in the studied samples compared with ²²⁸Ra in same sample might be as a result of ²²⁶Ra being part of ²³⁸U decay series as a results of which ²²⁶Ra is found over wide range of aquifer, it might also be an indication of secondary mineralization of uranium. Based on international guidelines regulating these radionuclides in groundwater, levels recorded at the time of the study fall within permissible limits of 20pCi/L for ²²⁶Ra and ²²⁸Ra while ²³²Th which is insoluble in water, therefore, has no regulated limit. All radionuclides analyzed fall within international guideline despite occurrence of uranium mineralization in the study area.(Arabi et al., 2013).

2.8.0 Review of works carried out in area with similar geochemical settings

The level radiological hazards associated with the activities of tin mining and milling around Jos were analysed by Rabiu 1993. The samples analyzed were soil, minerals plants and water. The results indicated that the level of radiation in all the samples were higher than the maximum permissible dose of 0.5mSv/year (5mSv/year) for the general population.

Measurement of Radionuclides in processed tin mine tailings in Jos were made using NaI(TI) detector by Mangset and Sheyin 2009. The results shows that the mean activity concentration ranged from 364Bq/kg-27930Bq/kg in the tailings. Similarly HPGe detector was used to measure the radioactivity in mine tailings in Jos, the results revealed that ²²⁶Ra have mean activity concentration ranged between 51.36Bq/kg – 512.24Bq/kg while ²³²Th mean concentrations ranged between 378.02Bq/kg – 2635.78Bq/kg (Usikalu *et al.*, 2011).

ODUSOTE *et al.*, (2014) measured the radionuclides in theTin mining soil mounds from the Jos Plateau, Nigeria and evaluated the impact of radiation on the environment where the soils are used as building materials. Gamma spectrometry was employed via a NaI(Tl) detector to determine the activities of the radionuclides ⁴⁰K, ²³⁸U and ²³²Th in ten (10) samples from points within a distance of 20 km along the mining trail. The results of measurements in soil samples show that the concentrations of ²³⁸U, ²³²Th and ⁴⁰K ranged between 1.51 – 4.98, BDL - 8.64 and 10.3 – 35.2 Bq/kg with mean concentrations of 3.20 1.16, 1.31 2.75 and 25.60 8.89 Bq/kg, respectively. The external hazards ranged between 0.008 – 0.044 with mean value of 0.019 while the internal hazards ranged between 0.014 – 0.048 with mean value of 0.028. These hazard values are less than 1. The annual gonadal dose equivalent (AGDE) ranged

between 10.28 – 55.87 Svy⁻¹ with mean value of 24.22 12.16 Svy⁻¹. The radium equivalent activities ranged from 3.07 – 16.23 Bq/kg with a mean value of 7.04 Bq/kg. The external absorbed dose rate ranged from 5.35– 18.76 Gyh⁻¹ with a mean value of 12.95 4.26 Gyh⁻¹.

In a related development the activity Concentration of 40 K, 226 Ra and 228 Th were determined in the food crops on the Jos Plateau using gamma-ray spectrometry. The activity concentration of the natural radionuclide in the food crops lied between $(12.36 \pm 0.82 \text{ and } 56.92 \pm 8.84 \text{Bq/Kg})$ for 40 K, $(1.46 \pm 0.05 \text{ to } 10.42 \pm 0.04)$ Bq/Kg for 226 Ra and from $(1.53 \pm 0.08 \text{ to } 6.85 \pm 0.42)$ Bq/Kg for 228 Th. These relativity high values for the activity concentrations maybe attributed to the series of tin mining activities that have taken place in these areas in the past decades. However, the values obtained suggest that the dose taken from intake of these radionuclides in the food crops is low and that harmful effects are not expected (Jwanbot *et al.*,2012)

Similarly soil samples were analyzed for radioactivity levels due to ²²⁶Ra, ²³²Th and ⁴⁰K using gamma-ray spectroscopy while physical-chemical parameters were determined using standard methods. Most of the physical and chemical properties of farm soils indicated low values in heavy mined area (Bitsichi) and relatively high values in low mined areas (Bukuru and Ropp). The farm soils across the locations were essentially acidic. Results also showed no obvious correlation between physical-chemical properties and the radionuclide concentration of ²²⁶Ra, ²³²Th and ⁴⁰K in the farm soils. The outdoor radiation exposure to a farmer during farming operations in these mining areas and the likely associated radiological health risks were considered low and almost insignificant (Jibiri*et al.*, 2011).

Studies of the social and economic impacts of tin mining in Rayfield of Jos Plateau was carried out by Onwuka *et al.*, (2013). The result of the first analysis showed that there is a significant difference between the social components resulting from tin mining, while the second analysis revealed that there is a significant difference between the economic variables resulting from tin mining activities. The work showed that tin mining activities impacted on the environment significantly, leaving behind so much social and economic scars that the government needs to consider and take measures to ameliorate.

2.9.0 Review of Gamma-Ray Spectrometry Analysis

2.9.1 Overview of Gamma-Ray Detector

The types of detectors commonly used can be categorized as:

- a. Gas-filled Detectors
- b. Scintillation Detectors
- c. Semiconductor Detectors

The choice of a particular detector type for an application depends upon the X-ray or gamma energy range of interest and the application's resolution and efficiency requirements. Additional considerations include count rate performance, the suitability of the detector for timing experiments, and of course, price.

Detector Efficiency

The efficiency of a detector is a measure of how many pulses occur for a given number of gamma rays. Various kinds of efficiency definitions are in common use for gamma ray detectors:

a. Absolute Efficiency: The ratio of the number of counts produced by the detector to the number of gamma rays emitted by the source (in all directions).

- b. Intrinsic Efficiency: The ratio of the number of pulses produced by the detector to the number of gamma rays striking the detector.
- c. Relative Efficiency: Efficiency of one detector relative to another; commonly that of a germanium detector relative to a 3 in. diameter by 3 in. long NaI crystal, each at 25 cm from a point source, and specified at 1.33 MeV only.
- d. Full-Energy Peak (or Photopeak) Efficiency: The efficiency for producing full-energy peak pulses only, rather than a pulse of any size for the gamma ray.

Clearly, to be useful, the detector must be capable of absorbing a large fraction of the gamma ray energy. This is accomplished by using a detector of suitable size, or by choosing a detector material of suitable high atomic number(Knoll,1989).

Detector Resolution

Resolution is a measure of the width (full width half max) of a single energy peak at a specific energy, either expressed in absolute keV (as with Germanium Detectors), or as a percentage of the energy at that point (Sodium Iodide Detectors). Better {lower full with half maximum(FWHM) value} resolution enables the system to more clearly separate the peaks within a spectrum.

2.9.2 The NaI(Tl) Detector

The structure of the NaI(Tl) detector is illustrated in Figure 2.1. It consists of a single crystal of thallium activated sodium iodide optically coupled to the photocathode of a photomultiplier tube. When a gamma ray enters the detector, it interacts by causing ionization of the sodium iodide. This creates excited states in the crystal that decay by emitting visible light photons. This emission is called a scintillation, which is why this type of sensor is known as a scintillation detector. The thallium doping of the crystal

is critical for shifting the wavelength of the light photons into the sensitive range of the photocathode.

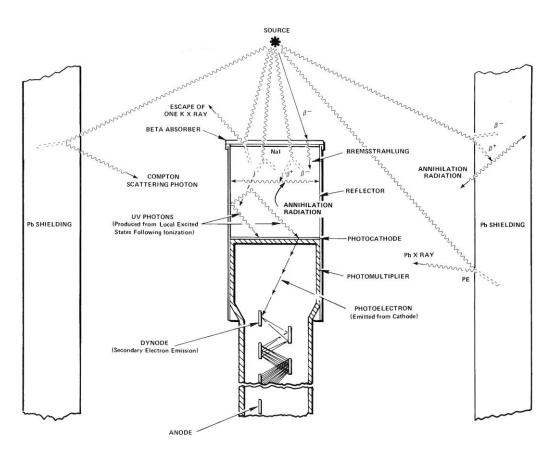


Fig. 2.1: The structure of the NaI(Tl) Detector and Various types of Gamma-Ray Interactions that Occur in the Typical Source-Detector-Shield configuration.(ORTEC, 2003).

Fortunately, the number of visible-light photons is proportional to the energy deposited in the crystal by the gammaray. After the onset of the flash of light, the intensity of the scintillation decays approximately exponentially in time, with a decay time constant of 250ns.

Surrounding the scintillation crystal is a thin aluminum enclosure, with a glass window at the interface with thephotocathode, to provide a hermetic seal that protects the hygroscopic NaI against moisture absorption. The inside of the aluminum is lined

with a coating that reflects light to improve the fraction of the light that reaches the photocathode.

At the photocathode, the scintillation photons release electrons via the photoelectric effect. The number of photoelectrons produced is proportional to the number of scintillation photons, which, in turn, is proportional to the energy deposited in the crystal by the gamma ray.

The remainder of the photomultiplier tube consists of a series of dynodes enclosed in the evacuated glass tube. Each dynode is biased to a higher voltage than the preceding dynode by a high voltage supply and resistive biasing ladder in the photomultiplier tube base. Because the first dynode is biased at a considerably more positive voltage than the photocathode, the photoelectrons are accelerated to the first dynode. As each electron strikes the first dynode the electron has acquired sufficient kinetic energy to knock out 2 to 5 secondary electrons. Thus, the dynode multiplies the number of electrons in the pulse of charge. The secondary electrons from each dynode are attracted to the next dynode by the more positive voltage on the next dynode. This multiplication process is repeated at each dynode, until the output of the last dynode is collected at the anode. By the time the avalanche of charge arrives at the anode, the number of electrons has been multiplied by a factor ranging from 104 to 106, with higher applied voltages yielding larger multiplication factors. For the selected bias voltage, the charge arriving at the anode is proportional to the energy deposited by the gamma ray in the scintillator.

The preamplifier collects the charge from the anode on a capacitor, turning the charge into a voltage pulse.

Subsequently, it transmits the voltage pulse over the long distance to the supporting amplifier. At the output of the preamplifier and at the output of the linear amplifier, the

pulse height is proportional to the energy deposited in the scintillator by the detected gamma ray. The Multichannel Analyzer (MCA) measures the pulse heights delivered by the amplifier, and sorts them into a histogram to record the energy spectrum produced by the NaI(Tl) detector. See Figure 2.2 for the modular electronics used with the NaI(Tl) detector

Fig. 2.2:Electronics Block Diagram of Gamma-Ray Spectroscopy system with NaI(Tl) Detector(ORTEC, 2003).

For an ideal detector and supporting pulse processing electronics, the spectrum of 662-keV gamma rays from a Cs radioactive source would exhibit a peak in the spectrum whose width is determined only by the natural variation in the gamma-ray energy. The NaI(Tl) detector is far from ideal, and the width of the peak it generates is typically 7% to 10% of the 662-keV gamma-ray energy. The major source of this peak broadening is the number of photoelectrons emitted from the photocathode for a 662-keV gamma-ray. For a high-quality detector this is on the order of 1,000 photoelectrons. Applying Poisson statistics (Knoll, 1989 and Ron *et al.*, 1981), 1,000 photoelectrons limit the full width of the peak at half its maximum height (FWHM) to no less than 7.4%. Statistical fluctuations in the secondary electron yield at the first dynode and fluctuations in the light collected from the scintillator also make a small contribution to broadening the width of the peak in the energy spectrum. Because the broadening is dominated by the number of photoelectrons, and that number is proportional to the gamma-ray energy, the FWHM of a peak at energy E is approximately described by

% Resolution (FWHM) =
$$\frac{\delta E}{E} x 100\% \approx \frac{K \times 100\%}{\sqrt{E}}$$
 (2.1) where

E = is the energy of the peak,

 δE = is the FWHM of the peak in energy units, and

K= is a proportionality constant characteristic of the particular detector.

Equation (2.1) indicates that the percentage energy resolution of the NaI(Tl) detector improves as the gamma-ray energy increases.

2.9.3 Neutron Activation Analysis (NAA)

Neutron activation analysis is a two steps analytical procedure in which some components ion 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 (2.2) (Landsberger, 1994).

$$N + {}^{A}Z \rightarrow {}^{A+1}Z \rightarrow {}^{A+1}Z + y \tag{2.2}$$

where;

^AZ is the target nucleus,

 $^{\mathrm{A+1}}\mathrm{Z}$ 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 (2.3) (Landsberger, 1994)

$$R = N/\sigma(E)\phi(E)dE \tag{2.3}$$

Whole Energy Range

where R is the reaction rate, ϕ (E) dE is the neutron flix of neutrons with kinetic energy between E and +dE in cm⁻²s⁻¹, σ (E) is the neutron capture cross-section in cm² 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 (2.4) (Landsberger, 1994)

$$A = \sigma \phi \left(\frac{m}{M}\right) N_A SDC \theta Py \eta \tag{2.4}$$

where

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

σis the activation cross section of the reaction in cm².

φis the activating neutron flux in n.cm⁻²S⁻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 x 10^{23} atoms/mol

S is the saturation factor which is given by

 $S = (1 - \exp(-\lambda t_1), \lambda \text{ is the constant of the reactive product and } t_1 \text{ is the duration}$ 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

θis the relative natural isotopic abundance of the activated isotope:

 P_{γ} is the probability of emission of photon with energy E; and

ηis the detector efficiency for the measured gamma radiation energy.

Equation (2.4) is simplified to equation (2.5) by the comparator method using the same geometry, equal weights of both samples and standard with the \ same irradiation, decay and counting times.

$$C_{sam} = C_{std} \left(\frac{A_{sum}}{A_{std}} \right) \tag{2.5}$$

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 (2.6) in ppm.

$$C_{sum} = C_{std} \left(\frac{A_{sum}}{A_{std}} \right) \left(\frac{D_{std}}{D_{sum}} \right) \left(\frac{C_{std}}{C_{sum}} \right) \left(\frac{W_{std}}{W_{sum}} \right)$$
(2.6)

where W_{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 it 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 component are then analysed to identify and determine quantitatively, the concentration of each radionuclide applying gamma spectrometry technique.

2.10.0 Liquid Scintillation Analysis

i. Principle

The principle for the determination of radon-222 by liquid scintillation counting is quite specific for this radionuclide. Radon-222 is extracted readily from the water sample by an organic scintillant. The decay products of radon-222 will remain in the water phase whilst radon-222 will be extracted into the organic phase. Before measurement the sample is stored for three hours until equilibrium is reached between radon-222 and its alpha emitting decay products. The alpha activity from radon-222 and its decay products is measured in a liquid scintillation counter (Jorma, 1993).

The literature reviewed in this chapter indicated that the works carried out in the study area were mostly geological, geochemical and mineralogy. Detail and comprehensive radiological and elemental studies were not carried out in the study area like those carried out in areas with similar geological settings (Jos, Plateau). This study therefore intended to quantitatively assess the radioactivity levels in the study area using some environmental samples.

CHAPTER THREE

MATERIALS AND METHODS

3.1.0 Introduction

In this chapter, the location of the study area, the samples, sampling, sample preparation and the methods of analysis are described. Mathematical formalisms used for the calculation of activity concentrations of the natural radionuclides are described in detail.

3.2.0 Study Area

The study area is Ririwai town headquarter of Doguwa Local Government Area in the extreme south of Kano State, Nigeria. It has an area of 1,473 km² and a population of 151,181 at the 2006 census. The study area also housed one of the largest underground tin mine that was closed in 1984. Plate 1 in appendix 8 shows the underground mine.



Fig. 3.1: Map of Nigeria showing Kano State and study area (Ecophoenix, 2008).



Fig. 3.2: Showing map of Kano State and the study area

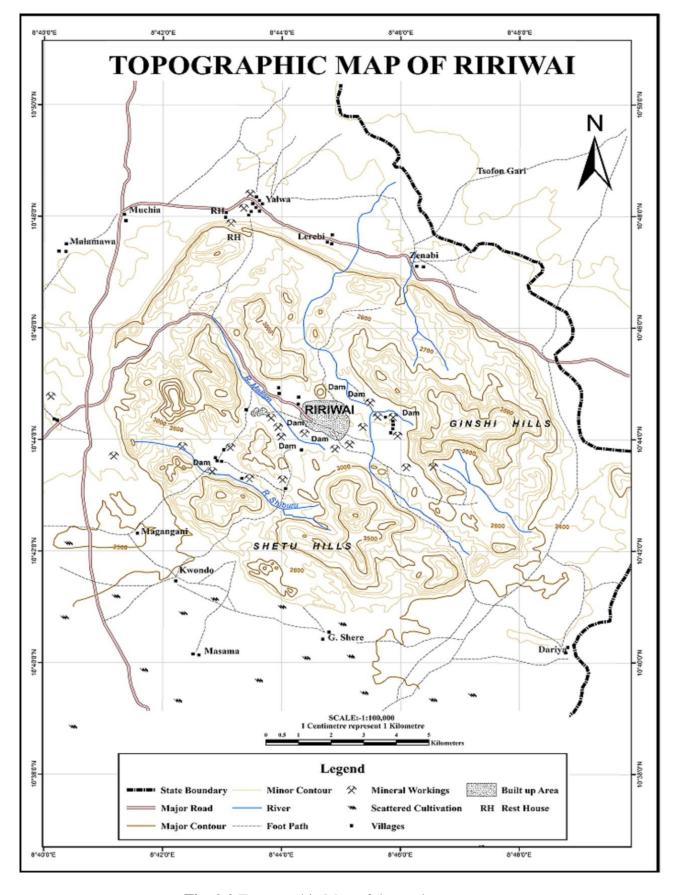


Fig. 3.3 Topographic Map of the study area

3.3.0 Material and methods

3.3.1 The samples used in this work are as follows:

i. Soil sample

28 soil samples were collected and used, 6 from farm land, 5 from irrigation land and 17 across the mining sites.

ii. Cereals

15 cereals sampleswere collected and used which included: 5 maize (*zea maiz*) samples, 5 guinea corn (*sorghum vulgare*) samples and 5 rice (*Orize sativa*) samples.

iii. Vegetables

11 samples of vegetable were collected and used which included 6 samples of tomatoes (*lycopersium esculatum*) and 5 samples of pepper (*Capsicum species*).

iv. Dust

10 samples from active mining pits were collected and used.

v. Water

40 samples were collected and used, 22 dwelling (15 from taps and 7 from hand dug wells), 10 from surface water sources including the one used for irrigation farming and 8 from tailing bearing.

3.3.2 Materials and Equipment

- i. Geographical positioning system (GPS)
- ii. Mechanical hand Auger
- iii. 2.5L plastic bottles
- iv. Nitric Acid
- v. Air sampler
- Power generator

- Filter papers
- Weighing balance
- Oven
- Grinder
- 32μm Sieve
- 231.0cm³ and 115.5cm³ plastic containers
- Candle wax masking tape Vaseline
- 7.0cm³ rabbit capsules
- Instagel scintillation cocktail (Packard instrument company)
- 76 x 76mm NaI(TI) Detector model ORTEC
- Maestro Data acquisition software by Cambera nuclear product
- Nigeria Research Reactor I (NRR1) No. NRRI/DS/JC/09/16

3.4.0 Sample Collection

3.4.1 Soil sample

Soil samples were collected within the mines area, farmlands and irrigation land. The sample location were marked using a geographical positioning system (GPS) and are tabulated in Table 3.1.

 Table 3.1:
 Sampling locations

S/No	North	East	Elevation
1	N10 ⁰ 44′ 35.3″	E008 ⁰ 45′ 16.3″	856m
2	N10 ⁰ 44′ 35.3″	E008 ⁰ 45' 16.4"	856m
3	N10 ⁰ 44′ 35.2″	E008 ⁰ 45' 16.4"	857m
4	N10 ⁰ 44′ 36.7″	E008 ⁰ 45' 15.8"	856m
5	N10 ⁰ 44′ 33.8″	E008 ⁰ 45' 17.8"	856m
6	N10 ⁰ 44′ 32.3″	E008 ⁰ 45' 21.0"	858m
7	N10 ⁰ 44′ 30.3″	E008 ⁰ 45' 27.0"	862m
8	N10 ⁰ 44′ 23.8″	E008 ⁰ 45' 27.3"	864m
9	N10 ⁰ 44′ 25.9″	E008 ⁰ 45' 27.7"	865m
10	N10 ⁰ 44′ 28.2″	E008 ⁰ 45' 26.9"	869m
11	N10 ⁰ 43′ 48.2″	E008 ⁰ 44' 57.1"	896m
12	N10 ⁰ 43′ 49.1″	E008 ⁰ 44' 53.4"	894m
13	N10 ⁰ 43′ 48.5″	E008 ⁰ 44' 53.0"	895m
14	N10 ⁰ 43′ 50.2″	E008 ⁰ 44' 58.7"	892m
15	N10 ⁰ 43′ 49.5″	E008 ⁰ 44' 59.2"	894m
16	N10 ⁰ 43′ 50.4″	E008 ⁰ 44' 41.8"	894m
17	N10 ⁰ 43′ 58.2″	E008 ⁰ 44' 38.7"	893m
18	N10 ⁰ 44′ 21.1″	E008 ⁰ 45' 26.4"	894m

The sampling strategy that was adopted for the soil samples was random (ASTM, 1983, 1986; IAEA, 2004). At each identified location samples were arbitrarily collected within defined boundaries of the area of concern. Each sampling point was selected independent of the location of all other sampling points. By this approach all locations within the area of concern had equal chance of being selected. The soil samples were taken using a mechanical hand auger to a depth of 5-10 cm. At each sampling location, samples of soil were taken from at least five different sections of the area into labeled plastic bags. One kilogram (1 kg) of each sample was collected for analysis.

3.4.2 Water

Water samples were collected from 3 sources. Thus, surface water, domestic water (taps and hand dug wells), and tailing bearing water within the mines. The samples were collected into two and half litres (2.5 L) plastic bottles and then labeled. The bottles were acid washed with Concentrated HNO3 before the bottles were filled with water to ensure radionuclides remain in solution rather than adhering to the walls of the container. The bottles were also filled to the brim without any head space to prevent CO2 frombeing trapped and dissolving in water which might affect the chemistry pH.

Some of the places where water samples were collected are shown in plate 2-4 in appendix 8.

3.4.3 **Dust**

Dust samples were collected mainly within the mines inside an active pits using air sampler. The air sampler was powered using a power generator continuously, dust were collected on a filter papers attached to the suction part of the air sampler. In order to prevent dust being collected on the filter paper emanating from other sources, the air sampler was placed inside the active pits. The sampling was carried out for about 3 hours where an average dust weight

of 0.20mg were collected. Plate 5 and 6 in appendix 8 show the active pits where dust samples were collected.

3.4.4 Cereals

Cereals samples comprising of maize (*zea mais*) guinea corn (*sorghum vulgare*) and rice(*Oriza sativa*) were collected from two farms located about 50.0m and 20.0m from the mine. Each sampling location was divided into 20m x 20m grids and sampled were taken at different point and mixed together to give a sample. Each sampling point was selected independent of other location of other sampling points. By this approach all locations within the area of concern had equal chance of being selected.

3.4.5 Vegetables

Samples of *Lycopersicum esculatum* (tomatoes) and *Capsicum species* (pepper) were collected from a dry season farm within the mine area. Cereals pattern of sampling was adopted with a sample grids area of 10m x 10m. Plate 7 in appendix 8 shows the farm where vegetable samples were collected.

3.5.0 Samples Preparation

3.5.1 Sample preparation for direct gamma spectrometry for soils, cereals and vegetables samples.

The samples were oven dried at temperature of 105^{0} C for soils samples while cereals and vegetables at 55^{0} C (IAEA 1989). The samples were then grinded into fine powder and sieved through $32\mu m$. The samples were then weighted and place into a plastic container which was sealed using candle max versseline and masking tape.

The sealing process included smearing of the inner rim of each container lid with Vaseline jelly, filling the lid assembly gap with candle wax to block the gaps between

lid and container, and tight-sealing lid-container with masking adhesive tape. The plastic containers were selected based on the space allocation of the detector vessel which measure 7.6cm by 7.6cm in dimension. Two types of containers of volume 231.0cm³ and 115.5cm² were used and the mass of each sample was measured and tabulated.

The samples were stored for 30 days to allow for secular equilibrium between the long-lived parent radionuclide and their short-lived daughter radionuclides in the ²³⁸U and ²³²Th decay series before the counting commenced.

3.5.2 Preparation of dust samples for N.A.A

The preparation started from the time of sample collection where the weight of the filter paper was measured before placing it at the suction part of the air sampler. After a period of 3hrs the weight of the filter paper and dust content were measured, the actual weight of the dust content were then recorded and the filter paper containing the dust were then sealed in a polyethylene bag. The filter paper containing the samples were wrapped in a polyethylene and then placed in 7cm³rabbit capsules. The polyethylene and rabbit capsules containing the samples were cleaned by soaking in 1:1 HNO₃ (Nitric acid) and then washed with de-ionised water in order to eliminate every contamination prior to sample irradiation in addition two blank filter papers were also analyzed.

3.5.3 Preparation of water samples for liquid scintillation analysis (L.S.A)

10ml each of the sample were added into a scintillation vial containing 10ml of the instagel scintillation cocktail having been sealed tightly, the vials were then shaken for more than 10 minutes to extract ²²²Rn in water phase into the organic scintilator and the sample so prepared were then taken to the laboratory for measurement.

3.6.0 Samples Analysis

3.6.1 Analysis of samples using direct gamma spectrometry

The analysis was carried out using a 76x76mm NaI(Tl) detect crystal coupled to a photomultiplier tube (PMT). The assembly has a preamplifier incorporate into it and a 1 kilovolt external source. The detector is enclosed in a 6cm lead shield with cadmium and copper sheets. This arrangement is aimed at minimizing the effects of background and scattered radiation.

The data acquisition software is Maestro produced by Canberra Nuclear Products was connected to the arrangement as shown in Plate 3.8. The samples were measured for a period of 29000 seconds each. The peak area of each energy in the spectrum was used to compute the activity concentrations in each sample by the use following equation.

$$C(Bq.kg^{-1}) = \frac{cn}{c_{fk}}$$
 (3.1)

where

C = activity concentration of the radionuclides in the sample given in Bq/kg-1

Cn = count rate (counts per second) count per second (cps) =
$$\frac{Net \ Count}{Live \ Time}$$

 C_{fk} = Calibration factor of the detecting system



Plate 3.1: Experimental Setup for NaI(Tl) Spectrometry Analysis

3.6.1.1 Calibration and efficiency determination

Calibration of the system for energy and efficiency were done by counting two calibration point sources, Cs-137 and Co-60 for 30 minutes each. These were done with the amplifier gain that gives 72% energy resolution for the 66.16keV of Cs-1371.

Standard

The standard materials used for calibrating the gamma-ray spectrometer consist of RGK-1 for K-40, RGU-1 for Ra-226 (Bi-214 peak) and RTG-1 for Th-232 (Ti208). The three formed a set of certified reference materials produced under the auspices of the International Atomic Energy Agency (IAEA) and distribute through its Analytic Control Services (AQCS) programme. The background counts were made for 29000 seconds.

Table 3.2: Spectral energy windows used in analyses

Isotope	Gamma Energy (kev)	Energy window (kev)
R-226	1764.0	1620-1820
Th-232	2614.5	2480-2820
K-40	1460.0	1380-1550

Table 3.3:Energy calibration for quantitative spectral analyses

Isotope	Calibration		Conversion	Detection	Limits
			Factor (Bq/kg ⁻¹		
	10 ⁻³ (cps/ppm) 10 ⁻⁴ (cps/ppm)		PpmBq/k	g
40 K	0.026	6.431	0.032	454.54	14.54
²²⁶ Ra	10.500	8.632	12.200	0.32	3.84
²³² Th	3.612	8.768	4.120	2.27	9.08

Source (Ibeanun, 1995).

3.6.2 Neutron activation analysis (NAA) for dust sample

The concentrations 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.

3.6.2.1 Long Irradiation

Samples were irradiated for 6 hours (the element of interest have long half-life) 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/cm². 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 days for the first lap counting and ten days for the second lap of counting.(Jonah, et al, 1993).

3.6.2.2 Measurements of Gamma Rays

Two laps of measurements were conducted.

The 1st lap of counting was carried out after a waiting period of 3 days for 30mins using the H1 sample holder for radioelements that have short decay chain. 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 for radioelement that have longer decay chain.

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 using eqn. (2.4).

3.6.3 Liquid scintillation analysis

Instal-gel scintillation cocktail (SC) a commercial product of Packard instrument Company was used to measured the ²²²Rn concentration in the prepared water samples. The ²²²Rn concentration in the prepared samples were determined using

$$A = \frac{(Cs - CB)x \, 100}{Cf \, x \, D(v)} \tag{3.2}$$

where A = radon concentration in Bq/l

 $C_S = Count \ per \sec of \ sample$

 $C_B = background CPS$

CF = conversion factor, $Cf = \frac{CPS(^{226}Ra)}{dps(^{226}Ra)}$, D = Decay constant

3.6.3.1 Calibration procedure of LSA for measurement of ²²²Rn in water

A secondary calibration material was prepared by dissolving 1ml of the Ra-226 in 10ml of distilled water. The solutions were stored for one month for equilibration to be attained between the long lived parent radionuclide and their short lived daughter

radionuclide in the ²²⁶Ra decay series. Later, 10ml of the solution was mixed with 10ml of Instal-gel scintillation cocktail and counted with a window setting of 25-900 in region C of the LSA for 60 minutes.

The calibration factor was determined by using equation 3.3 below.

Calibration Factor =
$$\frac{SC - BC}{C - V}$$
 (3.3)

where:

SC = Standard ²²²Rn concentration (count/min)

BC = Background count (count/min)

C = Standard ²²⁶Ra concentration (pCi/L) and,

V = Volume of standard ²²⁶Ra used (L).

3.6.3.2 Detection Limit of LSA for Measurement of ²²²Rn in water

The value of LD (Bq/L) of ²²²Rn water with the LSA was found to be 0.02Bq/L using equation 3.4, and assuming that day time (t) is zero.

$$LD = \frac{1}{(CF)x(D)xV} \left(\frac{2.71}{T} + 4.65\sqrt{\frac{CB}{T}}\right) \text{ (ASTM, 1999)}$$
(3.4)

3.6.3.3 Decay correction Factor

The decay correlation term is calculated by using equation 3.5. below

$$D - \exp\left(-\frac{0.693T}{t_{1/2}}\right). \tag{3.5}$$

where:

D = Decay correction

T = Time in days from samples collection time to midpoint of sample counting and $T_{1/2}$ = Radon radiological half-life (3.82 days).

3.7.0 Calculations

3.7.1 Determination of activity concentration

The absorbed dose rate for the soil, cereals and vegetables samples were calculated from the activity concentrations of the relevant radionuclides using equation (3.6).

$$D(nGyh^{-1}) = 0.0417A_K + 0.0462A_U + 0.604A_{Th}$$
(3.6)

where;

AK, AU and ATh are the activity concentrations of ⁴⁰K, ²³⁸U and ²³²Th respectively, and Table 3.3 shows the dose conversion factors of ⁴⁰K, ²³⁸U and ²³²Th.

Table 3.4: Activity to dose rate conversion factors

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

Source (UNSCEAR, 2000)

3.7.2 Calculation of annual effective dose

The annual effective dose was calculated from the absorbed dose rate by applying the dose conversion factor of 0.7 Sv/Gy and an outdoor occupancy factor of 0.2 for the soil samples using equation 3.7. In the case of cereals and vegetables the committed effective dose (E_{ing}) were estimated from the activity concentrations of each individual radionuclide and applying a yearly consumption rate for adult using equation 3.8. For the food samples, annual effective dose was calculated by applying the consumption rate of cereals of 140 kg/year for adult and for vegetables applying the consumption rate of 60 kg/year (UNSCEAR, 2000).

$$H_E = DTF (3.7)$$

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

T =the occupancy factor (0.2).

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

$$E_{ing} = A_{sp}. I_n. \sum_{j=1}^{3} DCF_{ing}(K, Ra, Th)$$
 (3.8)

where, A_{sp} is the activity concentration of the radionuclides, I_n annual consumption rate and DCF_{ing} is the ingestion dose coefficient in Sv/Bq.

3.7.3 Calculation of annual effective dose in water samples

The annual effective dose in water samples was calculated using equation 3.9

$$E_{Rn} = DF_{Rn} \ x I_{in} x A_{Rn} \tag{3.9}$$

where

 E_{Rn} = annual effective dose in μ Sv/year

 DF_{Rn} = effective dose per unit intake for adult = 10^{-8} Sv/Bq

 I_{in} = water consumption rate = 2 litre per day

 A_{Rn} = radon concentrations

3.7.4 Activity concentration of dust for ²³⁸U, ²³²Th, ⁴⁰K from NAA

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 3.6 below was used to convert from ppm to specific activity in Bq/kg.

Table 3:5: Conversion of radio element concentration to specific activity

1%k in rock/soil = 313 Bq/kg 40 K

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

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

Source (IAEA 2003).

3.7.5 Determination of ²²²Rn concentration and ²²²Rn emanation fraction in soil samples

Radon concentrations in the soil (kBqm-3) were calculated using a proposal (Eqn 3.10) in UNSCEAR report from the activity concentrations of ²²⁶Ra (UNSCEAR, 2000).

$$C_{Rn} = C_{Ra} f \rho_s \varepsilon^{-1} (1 - \varepsilon) (m[K_T - 1] + 1)^{-1}$$
(3.10)

Where;

CRa is the activity concentration of ²²⁶Ra in soil (Bq/kg),

f is the radon emanation factor, (0.2),

ρs is the density of the soil grains, (2700 kgm-3),

 ε is the total porosity, (0.25),

m is the fraction of the porosity that is water filled, (0.95), m is zero if the soil is dry, and k_T is the partition coefficient of radon between the water and air phases, (0.23). Since the soil samples were dried before the activity concentrations were measured then, m is zero and the last term of equation (3.10) is omitted (UNSCEAR, 2000).

²²²Rn emanation fraction was calculated using eq 3.11

$$E = \frac{VC}{MR} \tag{3.11}$$

where

E = emanation fraction

V = effective volume sampling container

 $C = {}^{222}Rn$ concentration

M = mass of the sample

 $R = {}^{226}Ra$ activity concentrations.

3.7.6 Calculation of total annual effective dose

For the purpose of verifying compliance with dose limits, the total annual effective dose was determined. The total annual effective dose (ET) to members of the public was calculated using ICRP dose calculation method (ICRP, 1991). The analytical expression for the total annual effective dose is determined by summing all the individual equivalent doses for the exposure pathways considered in this study. These include:

- External gamma irradiation from the gamma emitting radionuclides in the soil and dust samples ($E\gamma(Ra, Th, K)$;
- Committed dose from ingestion of cereals and vegetables containing 226 Ra, 232 Th and 40 K radionuclides E_{ing} (W);
- Inhalation of radon gas from water, E_{ing} (Rn) and
- External irradiation from dust containing the ²²⁶Ra, ²³²Th and ⁴⁰K radionuclides.

Thus:

 $E_T = Esoil + Evegetables + Ecereals + Edust + Ewater$ (3.12)

3.8.0 Determination of Hazard indices and risks

The radiological risk of NORM in soils in the study area which may be used as building materials was assessed by calculating the radium equivalent activity (Ra_{eq}), the external hazard and internal hazard indices. The Ra_{eq} is a widely used hazard index and it was determined using equation 3.13(Xinwei et al., 2006):

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.0077C_k (3.13)$$

Where; C_K , C_{Ra} and C_{Th} are the activity concentrations of ^{40}K , ^{226}Ra and ^{232}Th respectively. In the definition of Ra_{eq} , it is assumed that 370 Bq/kg of ^{226}Ra , 259 Bq/kg of ^{232}Th and 4810 Bq/kg of ^{40}K produce the same gamma ray dose rate. The above criterion only considers the external hazard due to gamma rays in building materials. The maximum recommended value of Ra_{eq} in raw building materials and products must be less than 370 Bq/kg for safe use. This means that the external gamma dose must be less than 1.5 mSv/year.

Another criterion used to estimate the level of gamma ray radiation associated with natural radionuclides in specific construction materials is defined by the term external hazard index (Hex) as shown equation (3.14) (OECD/NEA, 1979; Alam *et al.*, 1999; Higgy *et al.*, 2000).

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_k}{4810} \tag{3.14}$$

where C_{Ra} , C_{Th} and C_{K} are the activity concentrations of 40 K, 226 Ra and 232 Th respectively. The value of the external hazard index must be less than unity for the external gamma radiation hazard to be considered negligible. The radiation exposure due to the radioactivity from construction materials is limited to 1.5 mSv/y (OECD/NEA, 1979; Beretka and Mathew, 1985). Another hazard index known as internal hazard index due to radon and its daughters was calculated from equation.

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_k}{4810} \tag{3.15}$$

This is based on the fact that, radon and its short-lived products are also hazardous to the respiratory organs.

where C_K , C_{Ra} and C_{Th} are the activity concentrations of ^{40}K , ^{226}Ra and ^{232}Th respectively. For construction materials to be considered safe for construction of dwellings, the internal hazard index should be less than unity.

3.9.0 Risks

The cancer and hereditary risks due to low doses without threshold dose known as stochastic effect were estimated using the ICRP cancer risk assessment methodology (ICRP, 1991; 2007) were estimated. In its 1990 recommendations, risks from radiation induced cancers were derived from observations of people exposed to high doses using a dose and dose rate effectiveness factor (DDREF). Risk estimates based on the observations of people exposed to low doses has associated large uncertainties and therefore will contribute to quantitative risks estimates (ICRP, 1991). The lifetime risks of fatal cancer recommended in the 1990 recommendations by the ICRP are 5 x 10^{-2} Sv⁻¹ for the members of the public and 4 x 10^{-2} Sv⁻¹ for occupationally exposed workers (ICRP, 1991).

In its latest recommendations of 2007, the Commission has retained its fundamental hypothesis for the induction of stochastic effects of linearity of dose and effect without threshold and a dose and dose-rate effectiveness factor (DDREF) of 2 to derive the nominal risk coefficients for low doses and low dose rates. In its latest recommendations, the system of regulations for radiological protection based on the 1990 recommendations has not changed (ICRP, 2007).

However, a new set of nominal risk coefficient has been derived to be used for the estimation of fatal cancer as well as hereditary effects. The recommended nominal risk coefficients in its 2007 recommendations are given in table 3.7. The new nominal risk coefficients were derived based upon data on cancer incidence weighted for lethality and life impairment whereas the 1990 values were based upon fatal cancer risk weighted for non-fatal cancer, relative life years lost for fatal cancers and life impairment for non-fatal cancer. However the combined detriment from stochastic effects in the new values has remained unchanged at around 5 % Sv⁻¹(ICRP, 2007).

Table 3.6: Detriment-adjusted nominal risk coefficients for stochastic effects after exposure to radiation at low dose rate (10⁻²)

Exposed	Ca	Cancer		Hereditary		detriment
Population	2007	1990	effects		2007	1990
			2007	1990		
Whole	5.5	6.0	0.2	1.3	5.7	7.3
Adult	4.1	4.8	0.1	0.8	4.2	5.6

Source (ICRP, 2007).

The risk of exposure to low doses and dose rates of radiation to members of the public in the Ririwai and surrounding from the mining and mineral processing activities of Ririwai Tin mine were estimated using the 2007 recommended risk coefficients (ICRP, 2007) and an assumed 70 years lifetime of continuous exposure of the population to low level radiation as in equation 3.16 and 3.17.

Fatality cancer risk = total annual effective dose (Sv) x cancer nominal risk factor. (3.16)Hereditary effect = total annual effective dose (Sv) x hereditary nominal effect factor (3.17)

3.10.0 Statistical analysis of samples

Paired Sample t-test statistical technique of Statistical Package for Social Scientists (SPSS) statistical software was used to compare the Means of the radionuclides concentrations in the samples. If the probability value P is greater than the

significance level at 5 % (P>0.05), then it implies that the paired sample Means are insignificant or the Mean of the two paired samples are equal. On the other hand if the P-value is less than the significance level at 5 % (P<0.05) then there is a significant difference between the means of the two sets of data. The paired sample t-test computes the difference between two variables for each case, and tests to find out if the average difference is significantly different from zero at 95 % Confidence level.

The paired sample t-test is calculated from the expression below:

$$t = \frac{\bar{d}}{\sqrt{s^2/n}} \tag{3.18}$$

where \overline{d} is the Mean difference between two samples, S^2 is the sample variance, n is the sample size and t is a paired sample t-test with (n-1) being the degrees of freedom. Analysis of variance (ANOVA) was used to compare the means of the elemental concentrations of uranium, Thorium and Potassium in the water samples in order to determine whether the differences in the elemental concentrations of the metals were significant or otherwise.

3.10.1 Mean differencing

Statistical analysis of K, U and Th concentrations within each interpreted unit is useful for extracting subtle information from the data that is not immediately visible. The simplest analysis is to look at deviations from the unit means. The mean of K, U and Th concentrations are calculated for each interpreted unit, which are subtracted to find the deviation from the mean called mean differencing. (Kovarch *et al.*, 1994). Large deviations from the mean could be due to errors in the mapping of the unit boundaries. Alternatively, deviations could indicate alteration/mineralization or other geological processes such as magma differentiation or weathering.

The method is useful for removing gross systematic changes in radioelement concentration within an interpreted unit. Wellman (1998) gives a good example of this type of analysis.

CHAPTER FOUR

RESULTS AND DISCUSSIONS

4.1.0 Introduction

Environmental samples comprising of soils, cereals vegetables dust and water were analyzed using the following analytical methods. Direct Gamma Spectroscope with NaI(TI) detector, instrumental neutron activation analysis with Nigeria Research reactor I and liquid scintillation analysis the results are presented as follows:

4.1.1 Direct gamma spectrometry using NaI (Tl) detector

Twenty eight (28) soil samples, 5 samples each of (*zea maiz*) maize, (*sorghum vulgane*) guinea corn and (*oriza sativa*) Rice cereals and samples of tomatoes (*lycopersicum esculatum*) and 5 samples of pepper (*capsicum species*) were analysed by direct gamma spectrometry using NaI (TI) detector. Table 4.1 shows the weight of the samples while Appendix 1 shows the counting for ⁴⁰K, ²²⁶Ra and ²³²Th for samples RS1 and Appendix 2 shows the counting in CPS, conversion factors and the corresponding activity concentrations for all the samples analysed.

Table 4.1: Weight of prepared samples of soil, cereals and vegetables.

S/N	Samples ID	Weight in	S/N	Samples ID	Weight in
		gram			gram
1	RGC 2	107.21	29	RS 8	237.17
2	RGC 3	96.79	30	RS 3	228.39
3	RGC 6	104.83	31	RS 28	157.66
4	RGC 5	98.88	32	RS 2	120.00
5	RGC 4	106.46	33	RS 5	131.77
6	RGC 7	106.63	34	RS 21	110.34
7	RGC 1	87.92	35	RS 16	114.13
8	RR 1	114.99	36	RS 23	105.51
9	RR 2	99.48	37	RS 24	144.49
10	RR 3	123.67	38	RS 14	133.84
11	RR 4	84.41	39	RS 12	98.09
12	RR 5	137.76	40	RS 11	107.07
13	RS 8	222.28	41	RS 11	131.88
14	RM 3	96.51	42	RS 13	133.81
15	RM 5	97.44	43	RS 10	104.81
16	RM 2	90.40	44	RS 25	134.51
17	RM 1	106.12	45	RS 9	112.37
18	RM 7	53.22	46	RS 4	116.42
19	RM 6	85.75	47	P1	48.35
20	RM 4	96.67	48	P2	64.75
21	RM 17	203.71	49	P3	58.06
22	RS 22	148.57	50	P4	64.48
23	RS 26	232.3	51	T5	77.57
24	RS 15	191.21	52	Т3	61.18
25	RS 1	205.98	53	T2	80.35
26	RS 7	229.07	54	T1	88.49
27	RS 20	206.88	55	T4	90.78
28	RS 2	198.12	56	T6	87.70

Soil samples

4.1.1.1 Activity Concentration of $^{40}\mathrm{K}$ $^{226}\mathrm{Ra}$ and $^{232}\mathrm{Th}$ in soil samples

The activity concentration in Bq/kg for 40 K, 226 Ra and 234 Th radionuclides ion the soil sample are tabulated in Table 4.2.

Table 4.2: Concentration of 40 K, 226 Ra and 232 Th in Bq/kg for soil samples

S/No	Sample ID	⁴⁰ K in Bq/kg	²²⁶ Ra in Bq/kg	²³² Th in Bq/kg
1	RS1	528.82 ± 12.60	47.39 ± 4.64	230.88 ± 5.78
2	RS2	267.56±7.47	107.30 ± 9.96	273.20±11.29
3	RS3	210.89 ± 9.80	98.49 ± 8.46	157.01±9.46
4	RS4	306.99 ± 4.04	29.08±3.47	112.08 ± 2.28
5	RS5	137.79±5.29	55.27±7.42	492.47±9.01
6	RS6	366.72±7.78	19.12±5.68	176.56±4.56
7	RS7	588.18±11.04	52.72±5.21	256.78±5.24
8	RS8	409.82±12.76	30.60±7.91	321.39 ± 6.80
9	RS9	296.42±3.89	28.04 ± 3.36	112.09 ± 2.28
10	RS10	207.15 ± 12.90	48.44±1.74	16.65±6.96
11	RS11	271.3±3.59	28.29±7.91	87.87±3.53
12	RS12	268.89 ± 5.74	14.06±4.16	130.89 ± 3.26
13	RS13	234.60±7.93	68.83 ± 6.60	270.24±5.93
14	RS14	123.48 ± 7.62	57.70±4.98	425.99 ± 8.21
15	RS15	199.84±5.91	23.99±1.26	136.60±12.99
16	RS16	22.82±0.99	14.36 ± 0.88	81.53±7.75
17	RS17	558.34±1.98	28.08 ± 8.45	271.72±6.84
18	RS18	586.15±7.61	55.50±6.55	213.89 ± 4.32
19	RS19	357.39 ± 9.95	26.65 ± 6.83	101.39 ± 3.53
20	RS20	290.98±3.73	28.27±3.34	106.15 ± 2.17
21	RS21	101.57 ± 2.22	67.36±5.91	154.76±3.30
22	RS22	137.17 ± 4.32	64.08±6.49	359.06±9.12
23	RS23	278.22±2.17	26.30±3.12	104.48 ± 2.05
24	RS24	351.63±16.46	103.45±9.91	406.36 ± 8.87
25	RS25	265.72±16.14	62.13±18.10	422.28±7.86
26	RS26	596.26±13.22	53.42±5.21	260.43±6.61
27	RS27	182.92±20.65	85.43±15.94	630.80±12.11
28	RS28	164.79±13.84	66.13±10.42	589.23±10.69
	Mean	296.87±8.27	49.66±6.57	257.24±6.53

The activity concentration in Bq/kg of ⁴⁰K, ²²⁶Ra and ²³²Th for the soil samples in Table 4.2 shows that the mean activity concentration of ⁴⁰K is 296.87±8.27Bq/kg in a range of 22.8 – 596.26Bq/kg. For ²²⁶Ra the mean value of the activity concentration is 49.66±6.56Bq/kg in range of 14.06-107.30Bq/kg and that of ²³²Th is 257.24±6.53Bq/kg in a range of 81.53 – 630.80Bq/kg. The standard deviation of the mean for ⁴⁰K, ²²⁶Ra and ²³²Th are 156.84, 26.64 and 153.23 respectively. The world average value of activity concentrations of ⁴⁰K, ²²⁶Ra and ²³²Th in soil, are 420, 33 and 45 Bq/kg respectively (UNSCEAR 2000). The value of ⁴⁰K in this study is lower than the world average value while the mean value of ²²⁶Ra and ²³²Th in this work were more than the world average value this could be attributed to the geochemical nature and mining activities taking place in the study area. Fig 4.1 shows the percentage distributions of ⁴⁰K, ²²⁶Ra and ²³²Th.

4.1.1.2 Absorbed dose rate and annual effective dose rate

The absorbed dose rate for the soil samples was calculated from the measured activity concentration using Equation 3.6 and the activity to dose rate conversion factor in Table 3.2. For examples sample RS1, has the following activity concentration.

40
K = 528.82, 226 Ra =47.39 and 234Th = 230.88 using equation 3.6

$$D (nGyh^{-1}) = 0.04.7 \times 528.82 + 0.0462 \times 47.39 + 0.604 \times 230.88$$

$$= 22.05 + 2.12 + 139.45$$

$$D = 163.62 \text{ nGyh}^{-1}$$
.

The annual effective dose rate was calculated from the absorbed dose rate using the conversion factor of 0.7Sv/Gy and an out door occupancy factor of 0.2 from equation 3.7. For sample RS1, the annual effective dose rate

$$D = 1163.62 \text{ nGyh}^{-1}$$

$$F = 0.7 \text{ SvGy}^{-1}$$

$$T = 0.2$$

$$H_E = 163.62 \times 10^{-9} \times 24 \times 365 \times 0.7 \times 0.2$$

$$\therefore H_E = 2 \times 10^{+5} \times 10^{-9} \, SV$$

$$= 2 \times 10^{-4} \, Sv$$

$$= 0.2 \, mSv$$

The result for the absorbed dose rate and annual effective dose rate for soil samples is summarized in Table 4.2

Table 4.3: Concentration of ⁴⁰K, ²²⁶Ra and ²³²Th, absorbed dose rate and annual effective dose rate.

S/No	Sample ID	⁴⁰ K in Bq/kg	²²⁶ Ra in Bq/kg	²³² Th in Bq/kg	Absorbed dose rate in nGyh ⁻¹	Annual effective dose in mSv/Year
1	RS1	528.82±12.60	47.39±4.64	230.88±5.78	163.62±4.18	0.20±0.005
2	RS2	267.56±7.47	107.30±9.96	273.2±11.29	181.13±7.59	0.22 ± 0.009
3	RS3	210.89 ± 9.80	98.49±8.46	157.01±9.46	108.17±6.51	0.13 ± 0.008
4	RS4	306.99±4.04	29.08±3.47	112.08±2.28	81.61±1.70	0.09 ± 0.002
5	RS5	137.79±5.29	55.27±7.42	492.47±9.01	305.75 ± 6.00	0.37 ± 0.007
6	RS6	366.72±7.78	19.12±5.68	176.56±4.56	122.81±3.33	0.15 ± 0.004
7	RS7	588.18±11.04	52.72±5.21	256.78±5.24	182.07±3.87	0.22 ± 0.005
8	RS8	409.82±12.76	30.60±7.91	321.39±6.80	212.62±5.01	0.26 ± 0.006
9	RS9	296.42±3.89	28.04±3.36	112.09±2.28	81.13±1.69	0.10 ± 0.002
10	RS10	207.15±12.90	48.44±1.74	316.65±6.96	202.14 ± 4.82	0.25 ± 0.006
11	RS11	271.3±3.59	28.29±7.91	87.87±3.53	65.69 ± 2.65	0.08 ± 0.003
12	RS12	268.89 ± 5.74	14.06±4.16	130.89±3.26	90.92 ± 2.40	0.11 ± 0.003
13	RS13	234.6±7.93	68.83±6.60	270.24±5.93	176.18±4.21	0.22 ± 0.005
14	RS14	123.48±7.62	57.70±4.98	425.99±8.21	265.12±5.51	0.33 ± 0.008
15	RS15	199.84±5.91	23.99±1.26	136.6±12.99	91.95±8.16	0.11 ± 0.001
16	RS16	22.82 ± 0.99	14.36±0.88	81.53±7.75	50.85 ± 4.76	0.06 ± 0.006
17	RS17	558.34±1.98	28.08±8.45	271.72 ± 6.84	188.70 ± 5.02	0.23 ± 0.006
18	RS18	586.15±7.61	55.50±6.55	213.89 ± 4.32	156.19±3.23	0.19 ± 00.003
19	RS19	357.39 ± 9.95	26.65±6.83	101.39 ± 3.53	77.37 ± 2.86	0.10 ± 0.003
20	RS20	290.98±3.73	28.27±3.34	106.15 ± 2.17	77.34 ± 1.62	0.10 ± 0.004
21	RS21	101.57 ± 2.22	67.36±5.91	154.76±3.30	100.83 ± 2.77	0.12 ± 0.003
22	RS22	137.17±4.32	64.08±6.49	359.06 ± 9.12	255.55±5.99	0.31 ± 0.007
23	RS23	278.22 ± 2.17	26.30±3.12	104.48 ± 2.05	65.49 ± 1.47	0.08 ± 0.002
24	RS24	351.63±16.46	103.45 ± 9.91	406.36±8.87	264.88 ± 6.51	0.33 ± 0.008
25	RS25	265.72±16.14	62.13±18.10	422.28 ± 7.86	269.02 ± 6.26	0.33 ± 0.007
26	RS26	596.26±13.22	53.42±5.21	260.43±6.61	184.63 ± 4.78	0.23 ± 0.006
27	RS27	182.92±20.65	85.43±15.94	630.80±12.11	392.78 ± 8.91	0.48 ± 0.011
28	RS28	164.79±13.84	66.13±10.42	589.23±10.69	365.82±7.53	0.45±0.009
	Mean	296.87±8.27	49.66±6.57	257.24±6.53	170.04±4.61	0.21±0.006

The mean absorbed dose rate in the soil sample is 170.72±4.62nGyh⁻¹in a range of 50.85 – 392.78nGyh⁻¹ with a standard deviation of 93.80±2.099. The absorbed dose rate due to the activity concentration of ⁴⁰K, ²²⁶Ra and ²³²Th in the soil sample in this study is about 3 times more than the world wide average value of 60nGyh⁻¹ (UNSCEAR 1993, 2000) Fig. 4.2 shows the percentage contribution of ⁴⁰K, ²²⁶Ra and ²³²Th in the absorbed dose rate in soil. ²³²Th contributed about 91.01% while ⁴⁰K and ²²⁶Ra contributed 7.25% and 1.34% respectively. The corresponding mean annual effective dose calculated from the soil activity concentration in this study is 0.209±0.005mSv/year in a range of 0.060 – 0.480mSv/year with a standard deviation of 0.115±0.003. The annual effective dose obtained in this study for soil is less than 1mSv/year recommended value by UNSCEAR. The summary of the Descriptive statistics of all the parameters determined in the soil samples are presented in Appendix 5A.

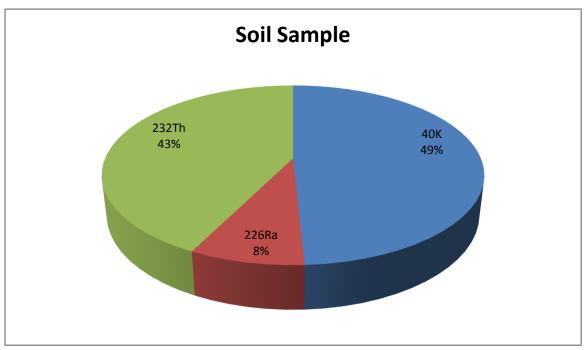


Figure 4.1:Percentage Distribution of 40K, 226Ra & 232Th Activity conc. in Soil

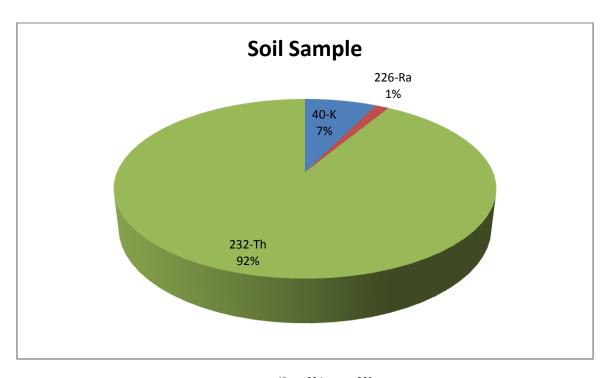


Figure 4.2: Percentage Contribution of ⁴⁰K, ²²⁶Ra & ²³²Th in absorbed Dose Rate for Soil

4.1.1.3²²²Rn Concentration and ²²²Rn Emanation fraction in soil samples

²²²Rnconcentration in K Bq/m⁻³ in soil sample were calculated using Equation 3.10. Since the soil samples were dried before the activity concentrations were measured i.e. m is zero. Equation 3.10 now reduced to

$$C_{Rn} = C_{Rn} f P_s \Sigma^{-1} (1 - \Sigma)$$

For sample RS1,

$$C_{Rn} = 47.39 \times 0.2 \times 2700 (0.25)^{-1} (1 - 0.25)$$

$$= \frac{47.39 \times 0.2 \times 27.00 \times 0.75}{0.25}$$

$$= 47.39 \times 0.2 \times 2700 \times 3$$

$$= 76.77kBq m^{-3}$$

The ²³²Rn concentrations in other samples are tabulated in Table 4.3. The ²³²Rn emanation coefficient were calculated using equation 3.12 and using that weight of each sample and the volume of the sample container for sample RS1.

$$E = \frac{V \times C_{Rn}}{M \times R_a}$$

where E = emanation fraction

 C_{Rn} = Radon concentration

V = volume of sample container

Ra = activity concentration²²⁶ Ra

Table 4.4: Concentration of ²²⁶Ra, ²²²Rn and Radon emanation fraction

S/No	Sample ID	²²⁶ RaBq/kg	²²² Rn kBq/m ³	Emanation fraction
1	RS1	47.39±4.64	76.77±7.52	1.54±0.015
2	RS2	107.3±9.96	173.83±16.14	1.11±0.011
3	RS3	98.49±8.46	159.55±13.71	1.18±0.018
4	RS4	29.08±3.47	47.11±5.62	1.11±0.011
5	RS5	55.27±7.42	89.54±12.02	0.97 ± 0.010
6	RS6	19.12±5.68	30.97±9.20	0.97 ± 0.009
7	RS7	52.72±5.21	85.41±8.44	1.33 ± 0.012
8	RS8	30.60±7.91	49.57±12.81	1.14 ± 0.010
9	RS9	28.04±3.36	45.42±5.44	1.11±0.011
10	RS10	48.44±1.74	78.47±2.82	1.20 ± 0.012
11	RS11	28.29±7.91	45.83±12.81	1.44 ± 0.012
12	RS12	14.06±4.16	22.78 ± 6.74	1.29 ± 0.013
13	RS13	68.83±6.6	111.50±10.69	0.97 ± 0.009
14	RS14	57.70±4.98	93.47±8.07	0.97 ± 0.010
15	RS15	23.99±1.26	38.86 ± 2.04	1.62 ± 0.016
16	RS16	14.36±0.88	23.26±1.43	1.11 ± 0.011
17	RS17	28.08±8.45	45.49±1.30	1.54 ± 0.017
18	RS18	55.50±6.55	89.91±10.61	1.40 ± 0.014
19	RS19	26.65±6.83	4317±11.06	1.54 ± 0.015
20	RS20	28.27±3.34	45.80 ± 5.41	1.50 ± 0.015
21	RS21	67.36±5.91	109.12±9.57	1.20 ± 0.012
22	RS22	64.08±6.49	87.61±10.51	0.87 ± 0.010
23	RS23	26.30±3.12	42.61±5.05	1.20 ± 0.012
24	RS24	103.45±9.91	167.59±16.05	0.91 ± 0.009
25	RS25	62.13±18.10	100.65 ± 29.32	0.98 ± 0.010
26	RS26	53.42±5.21	56.54 ± 8.44	0.87 ± 0.013
27	RS27	85.43±15.94	138.40 ± 25.82	1.54 ± 0.015
28	RS28	66.13±10.42	107.13±16.88	1.71 ± 0.017
	Mean	49.66±6.57	78.799±10.197	1.222±0.013

4.1.1.4 ²²²Rn activity concentration and ²²²Rn emanation fraction in soil

The ²²²Rn activity concentration in soil sample was calculated from the ²²⁶Ra activity concentration Table 4.3 shows that the mean ²²²Rn activity concentration in soil samples is 78.799±10.197KBq/m³ in a range of 22.780-173.830KBq/m³ with standard deviation of 43.151±6.543. The mean ²²²Rn Emanation fraction is 1.226±0.0120 in a range of 0.870-1.710 with standard deviation 0.200±0.003. Table 4.25 the value of emanation fraction obtained in this study is higher than 0.05- 0.7 recommended by UNSCEAR

4.1.1.5 Hazard indices in soil sample

The radium equivalent activity Ra_{eq} internal (H_{in}) and External (H_{ex}) hazard indices due to the activity concentration of ^{40}K , ^{226}Ra and ^{232}Th in soil samples were calculated using Equation 3.13, 3.14 and 3.15 respectively as shows below using sample RS1.

a.
$$Ra_{eq} = 47.39 + 1.43 C_{th} + 0.0077 C_{K}$$

 $= 47.39 + 1.43 \times 230.88 + 0.0077 \times 528.82$
 $= 47.39 + 330.16 + 4.07$
 $= 381.62Bq/kg^{-1}$
b. $H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_{K}}{4810}$
 $= \frac{47.39}{370} + \frac{230.88}{259} + \frac{528.82}{4810}$
 $= 0.128 + 0.900 + 0.110$
 $= 1.133$
c. $H_{in} = \frac{C_{Ra}}{187} + \frac{C_{Th}}{259} + \frac{C_{K}}{4810}$
 $= 0.253 + 0.891 + 0.110$
 $= 1.256$

The hazard indices for the samples were calculated and are indicated in Table 4.5.

 $\label{eq:table 4.5} \textbf{Table 4.5} : \mbox{Hazard Indices, radium equivalent activity, external hazard index (H_{ex})} \\ and internal hazard index (H_{in})}$

S/No	Sample	Ra _{eq} in Bq/kg	H _{ex}	H _{in}
	ID			
1	RS1	381.62±13.76	1.133 ± 0.04	1.256 ± 0.05
2	RS2	418.78±36.68	1.408 ± 0.07	1.688 ± 0.09
3	RS3	339.25 ± 22.73	0.920 ± 0.06	1.176 ± 0.08
4	RS4	212.99±7.04	0.577 ± 0.02	0.652 ± 0.03
5	RS5	770.11±20.71	2.097 ± 0.06	2.261 ± 0.08
6	RS6	299.84±12.74	0.814 ± 0.04	0.863 ± 0.05
7	RS7	465.21±13.55	1.133 ± 0.04	1.398 ± 0.05
8	RS8	521.68±18.61	1.418 ± 0.05	1.497 ± 0.07
9	RS9	211.15±6.92	0.572 ± 0.02	0.645 ± 0.03
10	RS10	517.20±12.68	1.406 ± 0.04	1.533 ± 0.04
11	RS11	174.83±13.24	0.472 ± 0.04	0.545 ± 0.06
12	RS12	221.93±9.26	0.602 ± 0.03	0.639 ± 0.04
13	RS13	473.33±15.69	1.287 ± 0.04	1.466 ± 0.06
14	RS14	676.38±17.31	1.842 ± 0.05	1.992 ± 0.06
15	RS15	234.72±20.30	0.637 ± 0.06	0.699 ± 0.06
16	RS16	132.71±12.04	0.362 ± 0.03	0.399 ± 0.04
17	RS17	459.63±19.15	1.248 ± 0.05	1.321 ± 0.07
18	RS18	406.49±13.32	1.101 ± 0.04	1.245 ± 0.05
19	RS19	199.16±12.65	0.538 ± 0.03	0.607 ± 0.05
20	RS20	202.47±6.73	0.548 ± 0.18	0.622 ± 0.03
21	RS21	296.49±11.57	0.806 ± 0.03	0.981 ± 0.05
22	RS22	588.10±19.86	1.600 ± 0.06	1.767 ± 0.07
23	RS23	197.13±6.23	0.534 ± 0.02	0.603 ± 0.03
24	RS24	711.62±23.86	1.934 ± 0.07	2.203±0.09
25	RS25	686.45±30.58	1.868 ± 0.08	2.029 ± 0.13
26	RS26	471.75±15.68	1.279 ± 0.04	1.418 ± 0.06
27	RS27	1001.55±34.86	2.728 ± 0.094	2.950 ± 0.135
28	RS28	924.42±26.78	2.510 ± 0.073	2.681 ± 0.100
	Mean	436.92±16.95	1.192±0.051	1.326±0.062

Table 4.5 gives the summary of the hazard indices in soil samples. The results shows that the mean (Ra_{eq}) radium equivalent activity is 436.92±16.95Bq/kg with minimum and maximum values of 132.710-1001.550Bq/kg and standard deviation of 233.42±8.055. Table 4.26 the compares the radium equivalent activity in soil from this study with other works. Soil samples have the highest value of Ra_{eq} as shown in Fig. 4.14. The mean external and internal hazard indices (H_{ex} and H_{in}) in the soil are 1.192±0.015 in a range of 0.362–2.728 with standard deviation of 0.637±0.032 for H_{ex} and 1.326±0.062 in a range of 0.399-2.590 for H_{in} . The result of Ra_{eq} obtained in this study is slightly higher than 370Bq/kg considered to be the safe value similarly the mean H_{ex} and H_{in} obtained in this study are higher than unity (1). The external and internal hazard indices must be less than unity for the radiation hazard to be considered as insignificant (UNSCEAR 2000).

4.1.2 Cereals Samples

Five (5) samples each of Maize (*Zea maiz*), Guinea corn (*Sorghum vulgare*) and Rice (*Orize sativa*) were analyzed.

4.1.2.1 Activity concentration

The activity concentration for ⁴⁰K ²²⁶Ra and ²³²Th in Bq/kg for Maize, Rice and Guinea Corn extracted from Appendix 2 are tabulated in Table 4.6.

Table 4.6: Activity concentration for ⁴⁰K, ²²⁶Ra and ²³²Th, for cereals in Bq/kg.

S/N	Samples	⁴⁰ K in Bq/kg	²²⁶ Ra in Bq/kg	²³² Th in Bq/kg
	ID			
1	RR1	39.81±2.17	55.85±4.75	78.90±2.16
2	RR2	32.19±3.42	13.22±0.46	55.87 ± 2.28
3	RR3	32.92 ± 2.35	11.82±0.43	23.43±2.59
4	RR4	29.23±1.56	40.99±3.48	54.96±1.65
5	RR5	44.67±1.60	18.26±0.71	7.79 ± 0.31
6	RM1	69.46±4.58	9.74 ± 0.78	27.59 ± 3.46
7	RM2	14.59±1.13	12.47±4.99	41.09±1.97
8	RM3	76.52±1.55	36.84±2.71	29.02±1.65
9	RM4	50.79 ± 1.82	20.78±0.16	51.42±1.48
10	RM5	76.31±0.16	29.05±1.48	51.43±3.19
11	RGC1	108.39 ± 2.79	30.59±3.13	51.99±1.82
12	RGC2	32.50±1.61	18.54±3.94	53.51±1.62
13	RGC3	30.02±1.39	17.15±3.59	49.37±1.48
14	RGC4	131.39±2.57	37.04±3.88	63.07±2.16
15	RGC5	131.26±2.64	36.96±3.82	62.94 ± 2.08
Mean		59.99±2.76	25.95±2.55	46.81±1.99

The activity concentration of ⁴⁰K, ²²⁶Ra and ²³²Th in cereals samples are presented in Table 4.05. The results shows that the mean activity concentration for ⁴⁰K in cereals is 59.99±2.76Bq/kg in range of 44.590 – 131.39Bq/kg with a standard and deviation of 37.84±2.2 for ²²⁶Ra the mean activity concentration is 25.95±2.55Bq/kg in a range of 9.74 – 55.85Bq/kg with standard deviation of 13.46±1.70 while ²³²Th has a mean value of 46.81±1.99Bq/kg in a range of 7.79 – 79.90Bq/kg and standard deviation 18.15±0.749. Table 4.27 is a comparison of ²²⁶Ra and ²³²Th concentration in cereal in this study with publish data (UNSCEAR 2000). The values obtained in this study are very high when compared with the reference values of 80mBq/kg and 3.0mBq/kg for ²²⁶Ra and ²³²Th respectively this could be attributed to geochemical nature of the soil in the study area where the cereals were grown. Also the cereals have high bioaccumulation tendency for thorium and uranium; they are often use as phytoremediators in a uranium contaminated soils (Evan 2011). Fig 4.3shows the percentage distributions of ⁴⁰K, ²²⁶Ra and ²³²Th.

4.1.2.2 Absorbed dose rate

The absorbed dose rate for the cereals were calculated as in 4.1.1.2 the results for Rice, Maize and Guinea Corn are tabulated in Table 4.7.

4.1.2.3 Committed effective dose

The committed effect dose for the cereals were calculated for ⁴⁰K, ²²⁶Ra and ²³²Th using equation 3.8. The annual grain intake of 140kg/year (UNSCEAR 2000). The effective dose coefficient (e) for ingestion of radionuclides for members of the public for 70 years for ⁴⁰K, ²²⁶Ra and ²³²Th (ICRP 2003 and UNSEAR 2000). Appendix 6 were used. For sample Rm1 the committee effective dose were calculated as follows.

$$H_{ing}$$
 ⁴⁰ $K = Asp \ x \ I_{in} \ DCF_{ing}$
 $= 69.46 \ x \ 140 \ x \ 6.2 \ x \ 10^{-9}$
 $= .060 \ mSv/year$
 $H_{ing} \ Ra = Asp \ x \ I_{in} \ DCF_{ing}(^{226}R)$
 $= 9.74 \ x \ 140 \ x \ 2.8 \ x \ 10^{-7}$
 $= 380 \mu Sv = 0.380 mSv/year$
 $H_{ing} \ Th = 27.59 \ x \ 140 \ x \ 2.3 \ x \ 10^{-7}$
 $= 0.888 mSv/year$

The results of all the cereals are tabulated in Table 4.8.

Table 4.7: Activity concentration for ⁴⁰K, ²²⁶Ra and ²³²Th, for cereals in Bq/kg and absorbed dose rate in nGyh⁻¹

S/N	Samples	⁴⁰ K	²²⁶ Ra	²³² Th	Absorbed dose rate
	ID				nGyh ⁻¹
1	RR1	39.81±2.17	55.85±4.75	78.90±2.16	59.90±1.61
2	RR2	32.19±3.42	13.22±0.46	55.87 ± 2.28	35.70±1.54
3	RR3	32.92±2.35	11.82±0.43	23.43 ± 2.59	16.42±1.68
4	RR4	29.23±1.56	40.99±3.48	54.96±1.65	36.31±1.22
5	RR5	44.67±1.60	18.26±0.71	7.79 ± 0.31	7.41 ± 0.89
6	RM1	69.46±4.58	9.74±0.78	27.59 ± 3.46	20.01±2.32
7	RM2	14.59±1.13	12.47±4.99	41.09 ± 1.97	26.50±1.48
8	RM3	76.52±1.55	36.84±2.71	29.02 ± 1.65	22.42±1.18
9	RM4	50.79±1.82	20.78±0.16	51.42±1.48	34.14±0.98
10	RM5	76.31±0.16	29.05±1.48	51.43±3.19	35.58±2.01
11	RGC1	108.39 ± 2.79	30.59±3.13	51.99±1.82	37.33±1.35
12	RGC2	32.50±1.61	18.54±3.94	53.51±1.62	34.45±1.22
13	RGC3	30.02±1.39	17.15±3.59	49.37±1.48	31.86±1.12
14	RGC4	131.39±2.57	37.04±3.88	63.07±2.16	45.28±1.60
15	RGC5	131.26±2.64	36.96±3.82	62.94±2.08	45.20±1.55
	Mean	59.99±2.76	25.95±2.55	46.81±1.99	32.56±1.45

Table 4.8: Committed annual effective dose from ingestion of ⁴⁰K, ²²⁶Ra and ²³²Th series radionuclide in cereals in mSv/Year

S/N	Samples ID	$^{40}{ m K~H_{ing}}$	²²⁶ Ra H _{ing}	²³² Th H _{ing}	Total H _{ing}
		mSv/year	mSv/year	mSv/year	mSv/year
1	RR1	0.034 ± 0.002	2.190±0.190	2.541±0.069	4.765±0.0261
2	RR2	0.028 ± 0.003	0.520 ± 0.018	1.799 ± 0.073	2.347 ± 0.094
3	RR3	0.029 ± 0.002	0.463 ± 0.017	0.754 ± 0.083	1.246 ± 0.102
4	RR4	0.025 ± 0.001	1.046±0.136	1.770 ± 0.053	2.841 ± 0.190
5	RR5	0.038 ± 0.001	0.715 ± 0.028	0.250 ± 0.009	1.003 ± 0.038
6	RM1	0.060 ± 0.004	0.380 ± 0.030	0.888 ± 0.111	1.328 ± 0.145
7	RM2	0.013 ± 0.001	0.488 ± 0.195	1.323±0.063	1.824 ± 0.259
8	RM3	0.066 ± 0.001	1.444 ± 0.106	0.934 ± 0.053	2.444 ± 0.160
9	RM4	0.044 ± 0.002	0.814 ± 0.006	1.656±0.047	2.517±0.055
10	RM5	0.066 ± 0.001	1.138±0.058	1.656±0.103	2.860 ± 0.162
11	RGC1	0.094 ± 0.002	1.199±0.123	1.674 ± 0.059	2.967 ± 0.184
12	RGC2	0.028 ± 0.001	0.727 ± 0.154	1.723 ± 0.052	2.478 ± 0.207
13	RGC3	0.026 ± 0.001	0.672±0.141	1.589 ± 0.047	2.287 ± 0.189
14	RGC4	0.114 ± 0.002	1.452±0.152	2.031±0.069	3.597±0.223
15	RGC5	0.113 ± 0.002	1.449±0.149	2.027±0.067	3.589±0.218
	Mean	0.052±0.002	0.980±0.100	1.508±0.064	2.540±1.500

The mean absorbed dose rate in the cereals is 32.56±1.44nGyh⁻¹ in range between 7.41±0.89 and 59.90±2.32nGy/h with a standard deviation of 12.88±0.38 as shown in Table 4.06. ²³²Th contributed 88.42% to the total absorbed dose rate in cereals while 40 K and 226 Ra contributed 7.82% and 3.44% respectively as shown in Fig. 4.4. Cereals has the lowest absorbed dose rate when compared with soil dust and vegetable as shown in Fig. 4.12. The committed annual effective dose for cereals was calculated using effective dose coefficient for ⁴⁰K, ²²⁶Ra and ²³²Th Appendix 6. Table 4.8 shows that the mean committed annual effective dose due ⁴⁰K is 0.052±0.002 mSv/year in a range of 0.013 - 0.114mSv/year with standard deviation of 0.033±0.001. ²²⁶Ra mean committed effective dose is 0.980±0.100mSv/year in a range of 0.380 – 2.190mSv/year with standard deviation of 0.503±0.067. The mean committed effective dose for ²³²Th in cereals is 1.508±0.064mSv/year in a range of 0.254 - 2.541mSv/year with standard deviation of 0.585±0.024. The men total committed effective dose in the cereals is 2.540±0.150mSv/year in a range of 1.003 – 4.765mSv/year with standard deviation of 0.988±0.072. The percentage contribution to the total committed effective dose for ⁴⁰K, ²²⁶Ra and ²³²Th are 2.047%, 38.582% and 59.370% respectively as shown in Fig. 4.5. Cereals sample contributed 60.23% to the total annual effective dose in the study are as indicated in Table 4.21 while Appendix 5B gives the summary of the descriptive statistic of all the parameters analysed in cereals samples.

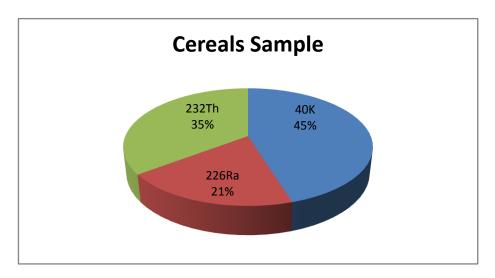


Figure 4.3: Percentage Distribution of ⁴⁰K, ²²⁶Ra & ²³²Th Activity conc. in Cereal

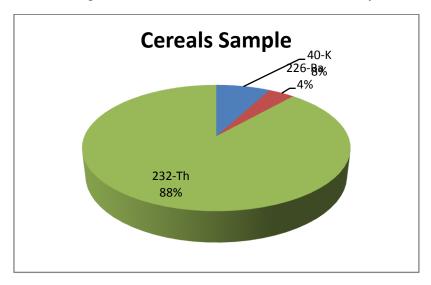


Figure 4.4:Percentage Contribution of ⁴⁰K, ²²⁶Ra & ²³²Th in absorbed Dose Rate for Cereal

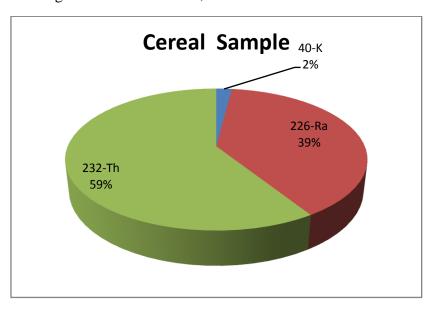


Figure 4.5: Percentage Contribution of ⁴⁰K, ²²⁶Ra & ²³²Th in Total Committed Dose Rate for Cereal Sample

4.1.3 Vegetables

Six (6) samples of tomatoes (*lycopersium esculatum*) and five (5) samples of pepper (*capsicum species*) were analysed.

4.1.3.1 Activity concentration

The activity concentration of ⁴⁰K, ²²⁶Ra and ²³²Th in Bq/kg for the vegetable sample analyzed extracted from Appendix 2 are summarized in Table 4.9 for vegetables samples.

Table 4.9: Activity concentration of ⁴⁰K, ²²⁶Ra and ²²²Th for vegetables in Bq/kg

S/N	Samples	⁴⁰ K	²²⁶ Ra	²³² Th
	ID			
1	T1	314.42±3.49	16.4±1.19	35.66±2.32
2	T2	191.13±0.96	7.75 ± 0.78	25.40±0.39
3	T3	228.72±2.30	36.56±9.19	37.98±1.14
4	T4	216.12±1.34	23.06±7.67	42.74±1.14
5	T5	127.26±4.02	18.78±5.75	35.15±3.22
6	T6	323.05±1.56	28.93±8.83	38.38±1.41
7	P1	459.16±15.44	19.35±8.23	81.21±11.32
8	P2	193.49±0.48	31.84±3.52	39.24±1.14
9	P3	137.39±4.07	32.13±4.99	56.10±2.67
10	P4	208.02±4.34	25.65±1.12	55.99±1.14
11	P5	453.03±13.51	68.12±3.43	152.28±2.50
Mean		261.84±4.93	28.65±4.92	56.43±1.66

The activity concentration of ⁴⁰K, ²²⁶Ra and ²³²Th in vegetable samples are presented in Table 4.09. The mean activity concentration of ⁴⁰K is 259.254±4.683Bq/kg in a range of 127.26–459.160Bq/kg with a standard deviation of 114.578±5.041. The mean activity concentration for ²²⁶Ra obtained was 28.052±4.973 in a range of 7.750 - 68.120Bq/kg with standard deviation of 15.652±3.200 while the mean activity concentration of ²³²Th is 54.557±2.588Bq/kg in a range of 25.400-152.280Bq/kg with standard deviation of 35.720±3.022. Fig. 4.6shows the percentage distributions of ⁴⁰K, ²²⁶Ra and ²³²Th

4.1.3.2 Absorbed dose rate

The absorbed dose rate calculated for vegetable samples are presented in Table 4.10.

4.1.3.3 Committed effective dose (E_{ing})

The committed effective dose (E_{ing}) using equation 3.8 due to ^{40}K , ^{262}Ra and ^{232}Th for the vegetable samples were calculated based on the annual consumption rate of 60kg/year for vegetable (UNSEAR, 2000) for adults and the effective dose coefficient in Appendix 6 for ^{40}K , ^{226}Ra and ^{232}Th are tabulated in Table 4.11.

Table 4.10: Activity concentration of ⁴⁰K, ²²⁶Ra and ²²²Th for vegetables in Bq/kg and absorbed dose rate in nGyh⁻¹

S/	Samples	⁴⁰ K in Bq/kg	²²⁶ Ra in Bq/kg	²³² Thin Bq/kg	Absorbed dose
N	ID				rate nGyh ⁻¹
1	T1	314.42±3.49	16.4±1.19	35.66±2.32	35.41±1.60
2	T2	191.13±0.96	7.75 ± 0.78	25.40±0.39	23.67±0.32
3	Т3	228.72±2.30	36.56±9.19	37.98±1.14	34.17±1.20
4	T4	216.12±1.34	23.06±7.67	42.74±1.14	35.89±1.10
5	T5	127.26±4.02	18.78±5.75	35.15±3.22	27.40±2.39
6	T6	323.05±1.56	28.93±8.83	38.38±1.41	37.99±1.33
7	P1	459.16±15.44	19.35±8.23	81.21±11.32	69.08±7.86
8	P2	193.49±0.48	31.84±3.52	39.24±1.14	32.78 ± 0.87
9	Р3	137.39±4.07	32.13±4.99	56.10±2.67	41.09±2.01
10	P4	208.02±4.34	25.65±1.12	55.99±1.14	43.67±0.97
11	P5	453.03±13.51	68.12±3.43	152.28 ± 2.50	114.32±2.23
	Mean	261.84±4.93	28.05±4.92	54.55±1.66	45.04±1.98

Table 4.11: Committed annual effective dose from ingestion of ⁴⁰K, ²²⁶Ra and ²³²Th series radionuclide in vegetables in mSv/Year

S/N	Samples ID	$^{40}\mathrm{K}~\mathrm{H}_{\mathrm{ing}}$	²²⁶ Ra H _{ing}	²³² Th H _{ing}	Total H _{ing}
		mSv/year	mSv/year	mSv/year	mSv/year
1	T1	0.117±0.0012	0.275±0.0200	0.492±0.0320	0.884±0.0532
2	T2	0.017±0.0003	0.130 ± 0.0131	0.351±0.0054	0.552 ± 0.0188
3	Т3	0.085 ± 0.0008	0.614 ± 0.1540	0.524 ± 0.0157	1.223±0.1705
4	T4	0.080 ± 0.0005	0.387 ± 0.1280	0.590 ± 0.0157	1.057±0.1442
5	T5	0.047 ± 0.0015	0.315 ± 0.0966	0.485 ± 0.0444	0.847 ± 0.1425
6	Т6	0.120 ± 0.0006	0.486 ± 0.1483	0529±0.0195	1.135±0.1684
7	P1	0.171 ± 0.0057	0.325 ± 0.1382	1.120±0.1562	1.616±0.3451
8	P2	0.072 ± 0.0002	0.535 ± 0.0591	0.542 ± 0.0157	1.149 ± 0.0750
9	Р3	0.050 ± 0.0015	0.539 ± 0.0838	0.774 ± 0.0368	1.364±0.1221
10	P4	0.077±0.0016	0.430 ± 0.0188	0.772±0.0157	1.279±0.0361
11	P5	0.169 ± 0.0050	1.144±0.0576	2.101±0.0345	3.414±0.0971
	Mean	0.091±0.002	0.471±0.083	0.779±0.023	1.320±0.107

The absorbed dose rate for vegetable samples as indicated in Table 4.10 shows that the mean absorbed dose rate is 45.043±1.989nGyh⁻¹ in a range of 23.670-114.320nGyh⁻¹ with a standard deviation of 25.803±2.044. Fig 4.7 shows that ⁴⁰K, $^{226}\mathrm{Ra}$ and $^{232}\mathrm{Th}$ contributed 24%, 3% and 73% respectively to the absorbed dose rate. Fig. 4.12 shows a comparison of absorbed dose rate in all the samples analyzed in this study. Committed annual effective dose due to ⁴⁰K, ²²⁶Ra and ²³²Th in vegetable are presented in Table 4.11. The results shows that the mean committed effective dose for ⁴⁰K is 0.091±0.002mSv/year in a range of 0.017– 0.171mSv/year with a standard deviation of 0.049±0.002. ²²⁶Ra has a mean committed effective dose of 0.471±0.083mSv/year ranges between 0.130-1.144mSv/year with a standard deviation of 0.263±0.054. ²³²Th has a mean committed effective dose of 0.753±0.036mSv/year in a range between 0.351-2.101mSv/year with standard deviation of 0.493±0.042. The total committed effective dose in vegetable has a mean value of 1.320±0.125mSv/year in a range of 0.552 - 3.414mSv/year with standard deviation of 0.750±0.090. Fig. 4.8 shows the % contribution of ⁴⁰K, ²²⁶Ra and ²³²Th the total annual committed effective in vegetable ⁴⁰K has 6.90% ²²⁶Ra has 35.90 and ²³²Th has 57.40 %. Fig. 4.13. Presented the plot of annual effective dose of all the sample analyzed in this study while Table 4.21 gives the percentage contribution of vegetables sample to the total annual effective dose as 31.2%. The high value obtained in cereals and vegetables samples can be attributed to the fact that some of the cereals (corn) and vegetables grown in the study area are also used as phytoremediators in a uranium contaminated soil because of their high bioaccumulation of uranium (Ivan, 2011). Appendix 5C give the summary of the descriptive statistics of all the parameters analyzed in vegetable samples.

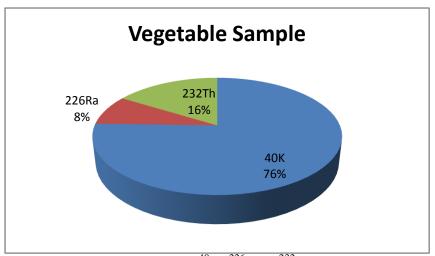


Figure 4.6:Percentage Distribution of ⁴⁰K, ²²⁶Ra & ²³²Th Activity conc. in Vegetable

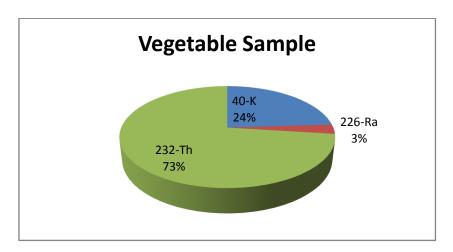


Figure 4.7:Percentage Contribution of ⁴⁰K, ²²⁶Ra & ²³²Th in absorbed Dose Rate for Vegetable

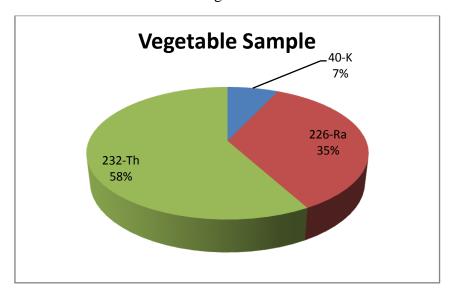


Figure 4.8:Percentage Contribution of ⁴⁰K, ²²⁶Ra & ²³²Th in Total Committed Dose Rate for Vegetable Sample

4.2.0 Instrumental Neutron Activation Analyses.

Dust samples were analysed using instrumental neutron activation Analysis with Nigerian Research Reactor 1 (NRRI) at the centre for energy research and training Ahmadu Bello University Zaria.

The result of the analysis is indicated in Appendix 3.

4.2.1 Activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K.

The N.A.A analysis gives the elemental concentrations in part per million (ppm) from where the radioelement concentration of U, Th and K were extracted and the concentration of K in ppm was converted to percentage Table 4.13 give the radioelement concentration in ppm U, Th and K%.

Using the conversion factors for radioelement concentration to specific activity in Table 3.3 the activity concentrations in Bq/kg for 226 Ra, 232 Th and 40 K in Dust were calculated and tabulated in Table 4.12

 $\textbf{Table 4.12:} \ \ \text{Neutron activation analysis for Dust sample U and Th in ppm and K in \%}$

S/N	Sample Code	K in %	U in ppm	Th in ppm
1	RP ₁	0.98 ± 0.150	2.53 ± 0.280	40.20 ± 0.200
2	RP_2	0.54 ± 0.007	1.19 ± 0.140	52.20 ± 0.200
3	RP ₃	0.65 ± 0.017	1.10 ± 0.180	18.20 ± 0.200
4	RP ₄	0.45 ± 0.005	1.15 ± 0.120	41.60 ± 0.200
5	RP ₅	1.27 ± 0.190	1.88 ± 0.260	37.60 ± 0.230
6	RP ₆	0.91 ± 0.010	BDL	49.30 ± 0.300
7	RP ₇	0.63 ± 0.008	1.49 ± 0.150	31.90 ± 0.200
8	RP ₈	1.78 ± 0.027	2.79 ± 0.360	48.50 ± 0.300
9	RP ₉	1.79 ± 0.032	8.35 ± 0.440	11.10 ± 0.100
10	RP_{10}	3.32 ± 0.047	2.14 ± 0.100	30.60 ± 0.300
	Mean	1.23±0.0493	2.62±0.191	36.12±0.223

Table 4.13: Activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K in Bq/Kg in dust samples

S/N	Sample Code	⁴⁰ K(Bq/kg)	²²⁶ Ra(Bq/kg)	²³² Th(Bq/kg)
1	RP ₁	306.74± 4.70	31.25 ± 3.46	163.21 ± 0.81
2	RP_2	169.02 ± 2.19	14.70 ± 1.73	211.9 ± 0.81
3	RP ₃	203.45 ± 5.32	13.5 ± 2.22	73.89 ± 0.81
4	RP_4	140.85 ± 1.57	14.20 ± 1.48	168.890 ± 0.81
5	RP ₅	397.51 ± 5.95	23.22 ± 3.21	152.66 ± 0.93
6	RP ₆	287.83 ± 3.13	BDL	200.16 ± 1.22
7	RP ₇	197.19 ± 2.50	18.40 ± 1.85	129.51 ± 0.81
8	RP ₈	557.14 ± 8.45	34.46 ± 4.45	196.91 ± 1.22
9	RP ₉	560.14 ± 8.45	103.12 ± 5.43	45.07 ± 0.41
10	RP_{10}	1039.16 ± 14.71	290.23 ± 0.86	124.24 ± 1.22
	Mean	385.30±5.70	54.31±2.51	146.64±0.91

Dust samples were analyzed for activity concentration of ⁴⁰K, ²²⁶Ra and ²³²Th using instrumental Neutron Activation Analysis (INAA) wit Nigeria research reactor the result of NAA analysis for dust (Appendix 3) from where the concentration in ppm of ⁴⁰K, ²²⁶Ra and ²³²Th were extracted Table 4.12 shows that U, Th and K have mean concentration of 2.262±0.191ppm, 36.120±0.223ppm and 1.232±0.0493% respectively.

The radioelement concentration in ppm for U and Th and K% were converted to activity concentration in Bq/kg using the conversion factor in table 3.3. Table 4.13 gives the activity concentration of ⁴⁰K, ²²⁶Ra and ²³²Th. The mean activity concentration of ⁴⁰K is 385.903±5.697Bq/kg in a range of 140.850-1039.160Bq/kg with standard deviation 274.557±3.996. ²²⁶Ra has a mean activity concentration of 54.318±4.512Bq/kg in a range of 0.00-290.25Bq/kg with standard deviation of 48.23±1.626 while ²³²Th has a mean activity concentration of 146.644±0.905Bq/kg with standard deviation 54.630±0.256 in range between 45.07- 211.90Bq/Kg. Fig. 4.9shows the percentage distributions of ⁴⁰K, ²²⁶Ra and ²³²Th in dust.

4.2.2 Absorbed Dose rate and annual effective dose

The absorbed dose rate in nGy/hr was calculated from the activity concentration of ²²⁶Ra, ²³²Th and ⁴⁰K using equation 3.7 and the activity to dose rate conversion factor in Table 3.2. Similarly the annual effective dose due to ²²⁶Ra, ²³²Th and ⁴⁰K were calculated from the calculated absorbed dose rate using equation 3.9. The occupancy factors of 0.2 out door and 0.7 Sv/Gy conversion factors were applied.

Table 4.14 gives the values for absorbed dose rate and annual effective dose due to ²³⁸U, ²³²Th and ⁴⁰K concentrations in dust samples.

Table 4.14: Absorbed dose rate and annual effective dose due to ⁴⁰K, ²²⁶Ra and ²³²Th concentrations in dust samples

S/N	Sample Code	Con	ı/kg	Absorbed Dose rate nGy/h	Annual of effective dose	
		⁴⁰ K ²²⁶ Ra, ²³² Th			mSv/year	
1	RP1	306.74 ± 4.70	31.25 ± 3.46	163.21 ± 0.81	112.81 ± 0.84	0.138
2	RP2	169.02 ± 2.19	14.70 ± 1.73	211.9 ± 0.81	135.74 ± 0.66	0.166
3	RP3	203.45 ± 5.32	13.5 ± 2.22	73.89 ± 0.81	53.74 ± 0.80	0.066
4	RP4	140.85 ± 1.57	14.20 ± 1.48	168.890 ± 0.81	108.53 ± 0.61	0.133
5	RP 5	397.51 ± 5.95	23.22 ± 3.21	152.66 ± 0.93	109.86 ± 0.95	0.135
6	RP 6	287.83 ± 3.13	BDL	200.16 ± 1.22	132.90 ± 0.87	0.163
7	RP 7	197.19 ± 2.50	18.40 ± 1.85	129.51 ± 0.81	87.29 ± 1.00	0.107
8	RP 8	557.14 ± 8.45	34.46 ± 4.45	196.91 ± 1.22	147.75 ± 1.78	0.181
9	RP 9	560.14 ± 8.45	103.12 ± 5.43	45.07 ± 0.41	55.34 ± 0.92	0.068
10	RP 10	1039.16 ± 14.71 290.23 ± 8.60		124.24 ± 1.22	254.46± 3.71	0.312
	Mean	$\textbf{385.90} \pm \textbf{5.70}$	54.31± 4.51	146.64 ± 0.91	$\textbf{119.85} \pm \textbf{9.70}$	0.147

The mean absorbed dose rate in the dust samples from Table 4.14 is 119.85 ± 9.700 nGyh⁻¹ in a range between 53.74–254.46nGy/h with standard deviation 32.059 ± 0.343 . ⁴⁰K, ²²⁶Ra and ²³²Th contributed 15%, 1% and 84% respectively to the absorbed dose rate in dust as shown in Fig. 4.10 while Fig. 4.12 shows the plot of the absorbed dose rate against all the samples. Table 4.14 also gives the values for annual effective dose in the dust sample with mean value of 0.147 ± 0.0119 mSv/year in a range of 0.066-0.312mSv/year Table 4.21 shows that dust samples contributed 3.47% to the total annual effective dose in the study area.

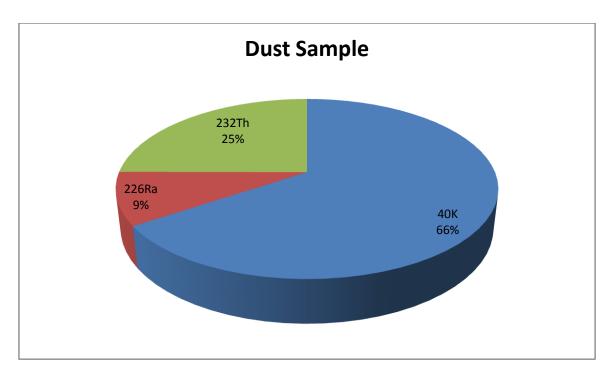


Figure 4.9:Percentage Distribution of ⁴⁰K, ²²⁶Ra & ²³²Th Activity conc. in Dust

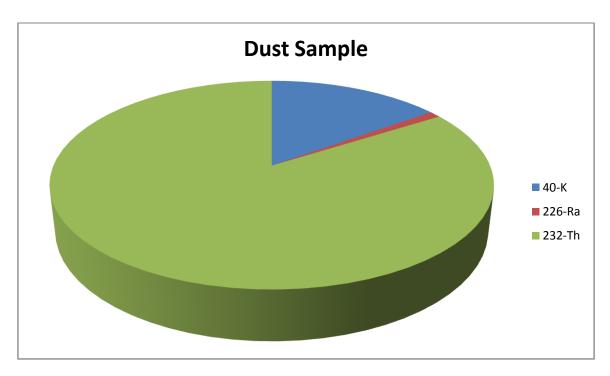


Figure 4.10: Percentage Contribution of ⁴⁰K, ²²⁶Ra & ²³²Th in absorbed Dose Rate for Dust

4.2.3 Hazard indices

The radiological hazard indices calculated from the concentrations of natural radionuclides in dust samples were radium equivalent activity Ra_{eq} external (H_{ex}) and internal hazards indices (H_{in}) . These hazards indices were calculated using Equations 3.13 Radium equivalent activity (Ra_{eq}) , 3.14 external hazard (H_{ex}) and equation 3.15 internal hazard (H_{in}) and the results is summonsed in Table 4.15

Table 4.15:Radium equivalent activity external and internal hazard indices for dust samples

S/N	Sample Code	Ra _{eq} Bq/kg	External Hazard	Internal Hazard
			$(\mathbf{H}_{\mathbf{ex}})$	(H_{in})
1	RP ₁	267.000	0.777	0.860
2	RP_2	319.060	0.892	0.932
3	RP_3	120.820	0.364	0.400
4	RP_4	256.790	0.719	0.757
5	RP ₅	244.580	0.735	0.796
6	RP_6	288.450	0.833	0.833
7	RP ₇	205.120	0.591	0.639
8	RP_8	320.330	0.969	1.060
9	RP ₉	171.780	0.569	0.841
10	RP_{10}	475.890	1.486	2.157
	Mean	266.980	0.793	0.927

Table 4.15 gives the hazard indices for the dust samples. The results shows that the mean value for Ra_{eq} in the dust samples is 266.982Bq/kg in a range between 120.820 – 475.890Bq/kg with standard deviation of 96.908. The mean value of H_{ex} is 0.794 in range between 0.364-1.486 with standard deviation of 0.299 while H_{in} has mean value of 0.928 in a range of 0.400 – 2.157 with standard deviation of 0.466. The mean values in dust are lower than those in soil. However these values are slightly lower than the recommended value of unity and are within the safety levels.

4.3.0 Water analysis with liquids scintillation counter

Three (3) different water sources were analyzed for ²²²Rn these include tailing water, surface water and domestic water sources.

4.3.1 ²²²Rn concentration

The activity concentration in Bq/L for the 3 sources of water was calculated from the result of L.S.A. Appendix 4. Equations 3.2 - 3.5 were used as follows.

$$A = \frac{(Cs - CB) \times 100}{Cf \times D (v)}$$

where A = radon concentration in Bq/l

 $C_S = Count \ per \sec of \ sample$

 $C_B = background CPS$

CF = conversion factor

$$Cf = \frac{CPS(^{226}Ra)}{dps(^{226}Ra)},$$

 $CPS = for standard = {}^{226}Raw$

$$Dps = N(t) = N_0 e^{-\lambda t}$$

$$\lambda = \frac{0.693}{T_{\frac{1}{2}}} T_{\frac{1}{2}} = \text{half life of radium}$$

$$D = exp\left(\frac{0.693t}{T_{\frac{1}{2}}}\right)$$

Where

D = decay constant

 $T_{\frac{1}{2}}$ half life of radon = 3.82day

t = time from the date of sample collection to the mid-time of conuting

$$= 3 days$$

$$D = e^{-\frac{0.693}{3.82}} \times 3 = 0.580$$

$$Cf = \frac{CPSof^{226}Ra}{dpsof^{226}Ra}$$

Cps of standard ²²⁶Ra

Where CPS count per sec

$$=\frac{Cps}{60}$$

but C.P.M = 58.15

$$CPS = \frac{58.15}{60}$$

= 0.963

$$= Dps = N(t) = N^{0}(t)e^{-(\lambda t)}$$

Where
$$\lambda = \frac{0.693}{\frac{T_1}{2}}$$

$$T_{\frac{1}{2}}$$
 half life of ²²⁶Ra = 1600 years

t= time elapse between N(t) and N(0) since date of production of the Ra source to the date of counting.

Date of manufacturing of the source is 1st of August 1992

Date of counting 23rd February, 2015

Elapse time t = 22years - 7 month

$$t = 7.02 \times 10^8 sec$$

$$T_{1/2}$$
 of $Ra = 5.0 \times 10^{10}$

$$\lambda = \frac{0.693}{5.0 \, x 10^{10}}$$

$$= 1.38 \times 10^{11}$$

$$\lambda = 1.38 \times 10^{11}$$

$$\therefore dps = N(t) = N^{0}(t)e^{-(\lambda t)}$$

$$=\frac{N(t)}{No}=e^{-(\lambda t)}$$

$$t = (22 \times 365 + 7 \times 30) \times 24 \times 60 \times 60$$

$$= (8030 + 330) \times 24 \times 3600)$$

$$= 8360 \times 24 \times 3600$$

$$= 7.02 \times 10^8 sec$$

Elapse time $t = 7.02 \times 10^8 \text{ sec}$

$$Dps = N(t) = N^{0}(t)e^{-(\lambda t)}$$

$$\therefore N_0 = \frac{8.7Bq}{1kg}$$
$$= \frac{8.7Bq}{1000}$$

$$= 0.0087 \text{ x}e^{-(\lambda t)}$$

$$N_0 = 0.0087$$

$$\lambda = 1.38 \times 10^{-11}$$

$$t = 7.23 \times 10^8 sec$$

$$\therefore dps = N(t) = N_0 e^{-(\lambda t)}$$

$$DPS = \frac{N(t)}{No} = e^{-} (1.38 \times 10^{11} \times 7.02 \times 10^{8} =$$

=
$$0.0087 \times e^{-} (1.38 \times 7.02 \times 10^{-3})$$

$$\therefore N(t) = 0.0087 \times 0.990 = 0.0086$$

$$dps = 0.0086$$

 $Cf = conversion \ after$

$$Cf = \frac{Cps}{dps}$$
$$= \frac{0.963}{0.0086}$$
$$= 112.67$$

$$Cf = 112.67$$

For sample RWS1

$$Cpm = 116.95$$

$$Cps = \frac{116.95}{60} = 1.948$$

$$\therefore A = \frac{(1.948 - 0.259)1000}{(112.67 \times 0.58)10} = 65.00$$

$$A = \frac{168.9}{65} = 2.59Bq/l$$

The result for surface water, tailing water and domestic water are tabulated in Table 4.16-4.18.

Table 4.16: ²²²Rn Concentrations in surface water source

S/No	Sample code	Concentration in Bq/L
1	RWS_1	2.59 ± 021
2	RWS_2	2.78 ± 0.12
3	RWS_3	2.55 ± 0.18
4	RWS_4	2.53 ± 0.10
5	RWS_5	2.65 ± 0.09
6	RWS_6	2.75 ± 0.20
7	RWS_7	2.55 ± 0.10
8	RWS_8	2.50 ± 0.08
9	RWS_9	2.66 ± 0.13
10	RWS_{10}	2.53 ± 0.09
	Mean	2.61 ± 0.13

Table 4.17:²²²Rn Concentrations in tailing bearing water

S/No	Sample code	Concentration in Bq/L
1	RWT_1	2.90 ± 0.19
2	RWT_2	3.18 ± 0.24
3	RWT_3	2.89 ± 0.26
4	RWT_4	2.96 ± 0.09
5	RWT_5	3.53 ± 0.33
6	RWT_6	2.88 ± 0.05
7	RWT_7	3.36 ± 0.06
8	RWT_8	2.92 ± 0.20
	Mean	$\boldsymbol{3.08 \pm 0.14}$

 Table 4.18:
 2222Rn concentrations in domestic water

S/No	Sample code	Concentration in Bq/L
1	RWW_1	2.41 ± 0.05
2	RWW_2	2.48 ± 0.13
3	RWW_3	2.61 ± 0.21
4	RWW_4	2.45 ± 0.06
5	RWW_5	2.34 ± 0.09
6	RWW_6	2.38 ± 0.10
7	RWW_7	2.43 ± 0.16
8	RWW_8	2.46 ± 0.11
9	RWP_1	2.33 ± 0.21
10	RWP_2	2.19 ± 0.08
11	RWP_3	2.24 ± 0.10
12	RWP_4	2.23 ± 0.06
13	RWP ₅	2.33 ± 0.14
14	RWP_6	2.34 ± 0.10
15	RWP_7	1.96 ± 0.03
16	RWP_8	2.04 ± 0.06
17	RWP ₉	2.12 ± 0.12
18	RWP_{10}	1.86 ± 0.20
19	RWP ₁₁	1.66 ± 0.16
20	RWP_{12}	2.38 ± 0.07
21	RWP_{13}	2.08 ± 0.09
22	RWP_{14}	1.88 ± 0.01
	Mean	2.23 ± 0.11

The results of the liquid scintillation analysis (L.S.A) as indicated in Tables 4.16 - 4.18 shows that the ²²²Rn concentration in tailing bearing water has a mean concentration of 3.08±0.14Bq/L and ranges between 2.88Bq/L to 3.53Bq/L. The surface water source has a mean ²²²Rn concentration of 2.61±0.13Bq/L with minimum and maximum concentrations of 2.50Bq/L and 2.78Bq/L respectively while the mean concentrations of ²²²Rn domestic water is 2.23±0.11 Bq/L and ranges between 1.66 and 2.61Bq/L.The result shows that the level of ²²²Rn concentrations in (tailing bearing water, domestic water and surface water) differs from each other, hence domestic water has the least mean ²²²Rn concentration, while tailing bearing water has the highest ²²²Rn concentration, as shown in Fig. 4.16.

4.3.2 Annual effective dose

The annual effective dose were collected for domestic and surface water sources only using equation 3.09.

$$E_{Rn} = DF_{Rn} \times I_{in} \times A_{Rn}$$
 3.9

Where

 $E_{Rn} = annual\ effective\ dose\ in\ \mu Sv/year$

 $DF_{Rn} = effective \ dose \ per \ unit \ intake \ for \ adult = 10^{-8} \ Sv/Bq$

 I_{in} = water consumption rate = 2 litres per day

 $A_{Rn} = Radon\ concentration\ in\ Bq/l$

For sample RWS₁

$$A_{Rn} = 2.59Bq/l$$

$$DF_{Rn} = 10^{-8} \, Sv/Bq$$

$$I_{in} = 2 litres \times 365 = 730 \ litres \ per \ year$$

Therefore
$$E_{Rn} = 10^{-8} \text{x} \ 7.30 \ \text{x} \ 10^{2} \text{x} \ 2.59$$

$$E_{Rn} = 18.907 \mu Sv/year$$

The result for surface and domestic water sources are tabulated in Table 4.19 and 4.20.

Table 4.19:²²²Rn concentrations and annual effective dose for surface water source

S/No	Sample code	Concentration in Bq/L	Annual effective dose in
			μSv/year
1	RWS ₁	2.59 ± 021	18.91±1.53
2	RWS_2	2.78 ± 0.12	20.29±0.83
3	RWS_3	2.55 ± 0.18	18.61±1.31
4	RWS_4	2.53 ± 0.10	18.47±0.73
5	RWS_5	2.65 ± 0.09	19.35±0.65
6	RWS_6	2.75 ± 0.20	20.07±1.46
7	RWS_7	2.55 ± 0.10	18.61±0.73
8	RWS_8	2.50 ± 0.08	18.25±0.58
9	RWS_9	2.66 ± 0.13	19.41±0.95
10	RWS_{10}	2.53 ± 0.09	18.47±0.65
	Mean	$\pmb{2.61 \pm 0.13}$	19.04±0.95

Table 4.20:²²²Rn Concentrations and annual effective dose for domestic water

S/No	Sample code	Concentration in Bq/L	Annual effective dose in µSv/year
1	RWW ₁	2.41 ± 0.05	17.59±0.04
2	RWW_2	2.48 ± 0.13	18.10±0.95
3	RWW_3	2.61 ± 0.21	19.05±1.53
4	RWW_4	2.45 ± 0.06	17.89±0.43
5	RWW ₅	2.34 ± 0.09	17.08±0.66
6	RWW_6	2.38 ± 0.10	17.37±0.73
7	RWW_7	2.43 ± 0.16	17.74±1.17
8	RWW_8	2.46 ± 0.11	17.96±0.80
9	RWP_1	2.33 ± 0.21	17.01±1.52
10	RWP_2	2.19 ± 0.08	1.60±0.58
11	RWP ₃	2.24 ± 0.10	16.35±0.73
12	RWP_4	2.23 ± 0.06	16.28±0.43
13	RWP ₅	2.33 ± 0.14	17.01±1.02
14	RWP ₆	2.34 ± 0.10	17.08±0.73
15	RWP ₇	1.96 ± 0.03	14.31±0.21
16	RWP ₈	2.04 ± 0.06	14.89±0.43
17	RWP ₉	2.12 ± 0.12	15.48±0.88
18	RWP_{10}	1.86 ± 0.20	13.58±1.46
19	RWP_{11}	1.66 ± 0.16	12.12±1.17
20	RWP_{12}	2.38 ± 0.07	17.37±0.51
21	RWP ₁₃	2.08 ± 0.09	15.18±0.66
22	RWP ₁₄	1.88 ± 0.01	13.72±0.80
	Mean	2.23 ± 0.11	16.28±0.80

The annual effective dose for surface water and domestic water sources calculated are tabulated in Table 4.19 and 4.20. The result indicated that the surface water source has a mean annual effective dose of $19.04\pm0.95\mu Sv/year$ and ranges between $18.25\mu Sv/year$ and $20.20\mu Sv/year$ while domestic water has a mean annual effective dose of $16.28\mu Sv/year$ with minimum and maximum values of $12.12\mu Sv/year$ and $19.05\mu Sv/year$ respectively.

The result of the independent t-test, indicated that there is significant differences in the mean annual effective dose between surface water and domestic water source, since the p-value (0.00) is less than 0.05 level of significance with surface water having the highest mean annual effective dose (19.04±0.95μSv/year) while domestic water source have the least mean annual effective dose (16.28±0.80μSv/year). The annual effective dose in the domestic water sources contributed 0.04% to the total annual effective dose in the study area as shown in Table 4.20 this mean consumption of water in the study does not pose radiological hazard.

4.4.0 Total Annual effective dose

The total annual effective dose was determined by summing all the individual equivalent doses for the exposure pathways considered in this study using equation 3.12.

$$\begin{split} E_T &= E_t(K,Ra,Th) \ soil + E_{ing}(K,Ra,Th) cereals + E_{ing}(K,Ra,Th) vegetable \\ &+ E_t(K,Ra,Th) Dust + E_{Rn}(water) \end{split}$$

$$E_T = 0.209 + 2.54 + 1.320 + 0.147 + 0.01638$$

 $E_T = 4.232 \, mSv/year$

Table 4.21 gives the summary of annual effective doses and the total annual effective dose from the exposure pathways and % percentage contribution of each pathway.

 Table 4.21:TotalMean annual effective dose and percentage contribution of exposure pathways

S/No	Exposure	Mean Annual Percentage	
	Pathways	Effective Dose in	contribution in %
	- waa wy s	mSv/year	
1	External irradiation K, Ra and Th	0.209	4.93
	in Soil		
2	Ingestion of K, Ra and Th in	2.540	60.20
	Cereals		
3	Ingestion of K, Ra and Th in	1.320	31.20
	vegetables		
4	External irradiation of K, Ra and	0.147	3.47
	Th in dust		
5	Ingestion of Rn in water sources	0.0163	0.04
	Total Annual effective Dose	4.232	100

The total annual effective dose calculated in this study was 4.232mSv/year as indicated in Table 4.21. Soil samples with mean annual effective dose of 0.209mSv/year contributed 4.93% to the total annual effective dose, cereals with mean committed annual effective dose of 2.540mSv/year have the highest contribution of 60.02% to the total annual effective dose in the study area. Vegetables with mean annual committed dose of 1.320mSv/year contributed 31.20% to the total annual effective dose in the study area. Dust with mean annual effective dose of 0.147mSv/year contributed 3.47% to the total annual effective dose in the study area. In domestic water samples the concentrations of ²²²Rn has a mean annual effective dose of 0.0163mSv/year contributed 0.04% to the total annual effective dose in the study area.

4.5.0 Risks Estimate

A summary of the estimated lifetime fatality cancer risk and hereditary disorders for all the exposure pathways studied in this work using ICRP recommended nominal risk coefficient for low dose radiation in Table 3.4 are presented in Table 4.22.

Table 4.22: Estimated Risk components for the various exposure pathways studied

	Exposure Pathways	Mean Annual effective dose in mSv/year	Fatality ca population	ncer risk to per year	Lifetime fatality cancer risk to population	Severe He effect per	•	Lifetime Hereditary effects
1	External irradiation	0.209	Whole	Adult		Whole	Adult	
	K, Ra and Th in		1.15×10 ⁻⁵	.85×10 ⁻⁵	8.05×10 ⁻⁴	4.18×10 ⁻⁷	2.09×10 ⁻⁷	2.93×10 ⁻⁵
	Soil							
2	Ingestion of K, Ra	2.540	1.34×10 ⁻⁴	1.04×10 ⁻⁴	9.38×10 ⁻³	5.10×10 ⁻⁶	2.54×10 ⁻⁶	3.57×10 ⁻⁴
	and Th in Cereals		_	_		_		
3	Ingestion of K, Ra	1.320	7.26×10^{-5}	5.41×10^{-5}	5.29×10 ⁻³	2.60×10^{-6}	1.32×10^{-6}	1.82×10^{-4}
	and Th in							
	vegetables							
4	External irradiation	0.147	8.09×10 ⁻⁶	6.03×10 ⁻⁶	5.16×10 ⁻⁴	2.94×10 ⁻⁷	1.47×10^{-7}	2.06×10^{-5}
	of K, Ra and Th in							
	dust							
5	Ingestion of Rn in	0.0163	0.09×10 ⁻⁵	0.067×10 ⁻⁵	6.30×10 ⁻⁵	3.20×10 ⁻⁸	1.63×10 ⁻⁸	2.24×10 ⁻⁶
	water							
	Total	4.232	2.33×10 ⁻⁴	1.77×10 ⁻⁴	1.63×10 ⁻²	8.44×10 ⁻⁶	2.20×10 ⁻⁷	5.92×10 ⁻⁴

The radiological risks estimated in this study using ICR 2007 recommended normal risk coefficient for law dose radiation were fatality cancer risk to population and adult, life time fatality cancer risk, severe hereditary effect per year to population and adult and life time hereditary effects. Table 4.22 gives the estimated risk components for the various exposure pathways studied. External irradiation from ⁴⁰K, ²²⁶Ra and ²³²Th in soil sample with 0.209mSv/year annual effective dose contributed 4.93% to the total annual effective dose in the study area. The risk component from soil shows that fatality cancer risk to population per year is 1.15x10⁻⁵ meaning that 1 out of 100,000 population is likely to suffer from one from of fatality cancer risk per year and approximately 1 adult out of 100,000 adults is likely to suffer from one form of fatality cancer risk per year. Considering an average life span of 70years the life time fatality cancer risk to population for soil is estimated to be 8.05 x 10⁻⁴ meaning on 8 out of 10000 are likely to have life time fatality cancer risk. The severe hereditary effect par year to whole population was estimated to be 4.18 x 10⁻⁷ while for adult is 2.09×10^{-7} meaning that 4 out of 10,000,000 and 2 adult out of 10,000,000 adult are likely to suffer from one severe hereditary effect per year for the whole population and adult respectively. The life time severe hereditary effects for soil samples was estimated to be 2.93x 10⁻⁵ meaning that approximately 3 out of 100,000 are likely to suffer from one severe hereditary effect in life time.

The risks estimated for the other exposure pathways are summarized in Table 4.23. Ingestion of 40 K, 226 Ra and 232 Th in cereals have the highest estimated risks followed by ingestion of 40 K, 226 Ra and 232 Th in vegetable while ingestion of 222 Rn in domestic water sources has the least estimated risks. The total risks estimated for all the 5 exposure pathways are 2.33 x 10^{-4} fatality cancer risks to population per year 1.74 x 10^{-4} fatality cancer risk to adult per year, 1.63 x 10^{-2} life time cancer risk to

population, 8.44×10^{-6} severe hereditary effects per year to whole population, 2.20×10^{-7} adult severe hereditary risk per year and 5.92×10^{-4} life time severe hereditary effects.

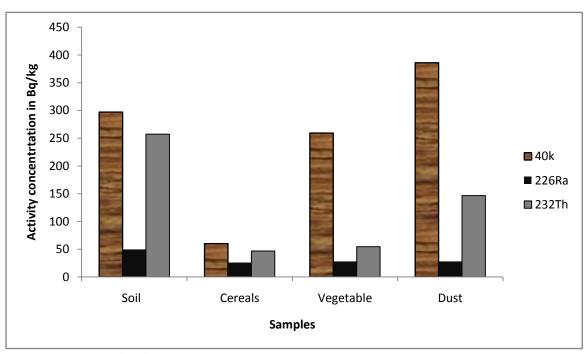


Fig. 4.11: Plot of activity concentration against samples

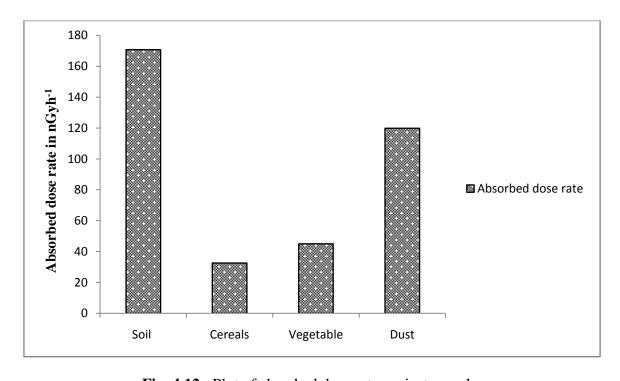


Fig. 4.12: Plot of absorbed dose rate against samples

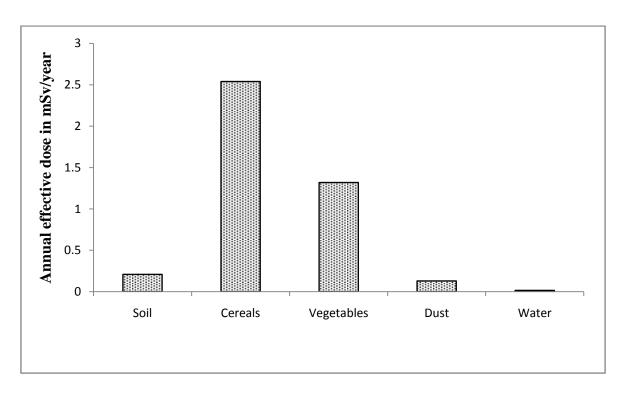


Fig. 4.13: Plot of annual effective dose against samples

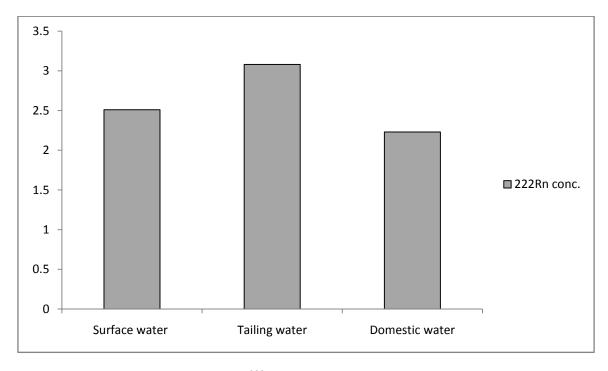


Fig. 4.14: Plot of ²²²Rn concentration against samples

CHAPTER FIVE

SUMMARY, CONCLUSION AND RECOMMENDATION

5.1.0 Summary

In this study 104 environmental samples were analysed for activity concentration of 40 K, 226 Ra and 232 Th and 222 Rn, absorbed dose rate, annual effective dose, hazard indices and risks. Soil sample had the highest activity concentration and absorbed dose rate and hazard indices while cereals and vegetables had the highest values of annual effective dose and risk estimates, water samples had the lowest value of annual effective dose and risks estimates.

²³²Th had the highest % contribution in the total absorbed dose rate and annual effective dose in all the samples analysed.

The activity concentration values of ²²⁶Ra and ²³²Th obtained in soil cereals and vegetable are all above the worldwide average values. The values of committed annual effective dose in cereals and vegetable obtained in this study also exceeded the recommended values. The risks estimated for fatality cancer and severe hereditary effects for cereals and vegetable all exceeded the recommended values by USEPA.

5.2.0 Conclusion

In this study assessment was made on the radiological sources of radiation resulting from artisanal mining mineral processing and other anthropogenic activities around Ririwai Tin mine. The exposure pathways considered for the study were direct gamma ray exposure from natural radioactivity concentrations in soil and dust, internal exposure from ingestion of food (cereals and vegetable) and ingestion of radon gas in water. The study was motivated by the fact that the study area is known to have other industrial and energy minerals. Beside Tin, minerals such as Uranium

(U) Zirconium (Zr) Thorium (Th) etc. were reported to be in the area. High levels of these elements could pose chemical and or radiological hazard.

In this work data on the activity concentrations of ⁴⁰K, ²²⁶Ra and ²³²Th in environmental samples such as soil, cereals, vegetables and dust as well as radiation doses and risk have been established. The activity concentration of ⁴⁰K, ²²⁶Ra and ²³²Th in different media for all the potential exposure pathways through which public could be exposure were quantified using Direct Gamma Spectroscopic Analysis NaI(TI), Instrument Neutron Activation Analysis (NAA) and Liquid Scintillation Analysis (L.S.A).

The result of activity concentrations of ⁴⁰K, ²²⁶Ra and ²³²Th and the corresponding absorbed dose rate in soil samples are higher than world average values of 240 Bq/kg, 33 Bq/kg and 45Bq/kg for ⁴⁰K, ²²⁶Ra and ²³²Th respectively and 60 nGy/h absorbed dose rate. The high values obtained in this study can be attributed to the geology, geochemical settings and mining activities going on in the study area, however, the annual effective dose in soil is less than 1mSv/year recommended value.

The assessment of Emanation Fraction was based on the decay of ²²²Rn from the parent radionuclide ²²⁶Ra content in the soil samples. The 1.22±0.013 mean value of the emanation fraction obtained is very high as expected since the value of ²²⁶Ra in the soil samples is more than the worldwide average value the emanation fraction obtained in this study is more than the typical range of 0.05 to 0.7 (UNSCEAR, 2000).

The mean values of hazard indices in soil are also higher than the recommended values of 370Bq/kg for Ra_{eq} and 1.00 for H_{ex} and H_{in} this can be attributed to the fact that soil has high value of activity concentration of ^{226}Ra and ^{232}Th . The risk

evaluated were based on the ICRP risks assessment methodology (2007) for fatality cancer risk, life time cancer risk, severe hereditary effect and life time hereditary effect, for soil the risk estimated were within the negligible cancer fatality risk of 1 x $10^{-6} - 1 \times 10^{-4}$ value recommended by USEPA. The value for soils obtained in this study falls within the negligible value and does not pose radiological effect.

The values of activity concentration absorbed dose rate and committed annual effective dose obtained in cereals are very high which could be attributed to the fact that the cereals have capacity to bioaccumulation high amount of radioelements present in the soil. The risk estimated for cereals were slightly above the negligible cancer fatality risk recommended by USEPA ($1 \times 10^{-6} - 1 \times 10^{-4}$).

The values of activity concentration, absorbed dose rate and committed annual effective in vegetables are relatively high because of the bioaccumulation of the radionuclides by the vegetable. The risk estimated for fatality cancer, lifetime fatality cancer risk, severe hereditary effect and life time hereditary effect in vegetable were 7.26 x 10⁻⁵, 5.29 x 10⁻³, 2.60 x 10⁻⁶ and 1.84 x 10⁻⁴ respectively these value are also within the negligible values recommended by USEPA.

The dust samples analysed have mean activity concentration of ⁴⁰K, ²²⁶Ra and ²³²Th as 385.30±5.70Bqkg, 54.31±2.51Bq/kg and 146.64±0.91Bq/kg respectively while the mean absorbed dose rate and annual effective dose rate were 119.85±9.70nGy/h and 0.147mSv/year respectively. The value of the absorbed dose rate which is higher than 60nGy/h recommended value could be due to high activity concentration of ²²⁶Ra and ²³²Th, while the value of annual effective dose is less than 1mSv/year recommended value. The hazard indices have values of 266.98Bq/kg for Ra_{eq} 0.793 H_{ex} and 0.927

 H_{in} , these values are all below the recommended values. The risk estimated for dust were within the negligible values of $(1 \times 10^{-6} - 1 \times 10^{-4})$ recommended by USEPA. Similarly, 222 Rn concentration in three(3) water sources in the study area were determined but high consideration was given to the domestic water sources which has a mean 222 Rn concentration of 2.23 ± 0.11 Bq/L. The result obtained in this study is below the 222 Rn concentration of 10Bq/L recommended by WHO 1993, UNSCEAR 1993 and the maximum permissible value of 11.1Bq/L by USEPA (1991) and adopted by standard organization of Nigeria (S.O.N). The mean annual effective dose obtained for domestic water sources in the study area was 0.0163 ± 0.0008 mSv/year which falls below the recommended 0.1mSv/yea recommended by UNSCEAR (2000). The risk for water fall below the negligible values of $(1 \times 10^{-6} - 1 \times 10^{-4})$ recommended by USEPA.

The total risks estimated were based on the total annual effective dose of 4.232mSv/year were 2.33 x 10⁻⁴, 1.63 x 10⁻², 8.44 x 10⁻⁶ and 5.92 x 10⁻⁴ for fatality cancer risk, lifetime fatality cancer risk, severe hereditary effect and lifetime hereditary effect respectively.

The total risks estimated in this study were above the acceptable range recommended by USEPA, however, member of public in the study area may not necessarily be exposed to all the exposure pathways considered in this study at the same time. This study considered that the ingestion of food (cereals and vegetable) and water could be the most significant mode of exposure in the study area. On the basis of the results from this study, consumption water do not pose any significant source of radiation hazards to the population. However consumption of food (cereals and vegetable) grown around the mining site in the study area could pose radiological hazard because

they have the capacity to bioaccumulate high amount of ²²⁶Ra and ²³²Th as they are often used as phytoremediators for U contaminated soil (Ivan, 2011).

Finally, the results from this study will serve as reference data for any future studies and also add up to data required for guidelines and regulations of NORM in Nigeria for radiation protection of workers and public.

6.3.0 Recommendation

Based on the foregoing the following recommendations are hereby proposed for the research in future.

- Elemental analysis for heavy metals such as Hg, Pb, Cd, Cu, Zn etc in soil, water, cereals and vegetables grown in the area.
- Determination of Gross alpha and beta activity concentration in domestic water sources in the study area using gross alpha and beta counter.
- Determination of ⁴⁰K, ²²⁶Ra and ²³²Th activity concentration in domestic water.
- Determination of ⁴⁰K, ²²⁶Ra and ²³²Th in other food items grown in the study area especially root vegetable and fruits such as cassava, potatoes, onions, mangoes guava etc.
- The cereals and vegetable grown in the study area as analysed in this study should only be considered as phytoremediators because of their high bioaccumation of ²²⁶Ra and ²³²Th and not for consumptions.
- Chemical speciation studies and assessment of the levels of major and trace metal in variety of food items and domestic water in the study area and the mode of translocation from soil to plant and water should be investigated.

REFERENCES

- Abbaa, S.I. (1975). Geochemistry and petrology of mineralization at Ririwai Gindi Akwati and Dutsen wai in the Nigerian Younger granite province. Unpublished M.sc Thesis, Univ. of St. Andrews.
- Abiye, O.S. (2005). Study of natural radiation levels and distribution of dose rate within the younger granite province of Nigeria. Ph.D Thesis university of Jos Nigeria.
- Adekanmi. A. A, Ogunleye. P.O, Damagum, A.H. and Olaseheinde, O. (2007). Geochemical map of uranium distribution in Nigeria Unpublished Report Nigerian Geological Survey Agency.
- Alam, M. N., Miah, M, M, H., Chowdhury, M. I., Kamal, M., Ghose, S., Islam, M, N., Mustafa, M.N., and Miah, M. S. R. (1999). Radiation dose estimation from radioactivity analysis of lime and cement used in Bangladesh, *J. Environ.Radioact*. Vol. 42 Pp. 3212-3219.
- Arabi, A.S. Funtua, I.I., Dewu, B.B.M., Adeyemo D.J., Abafoni, D.J., Garba, M.L. and Garba, I. (2013). Activity and mass concentration of ²²⁶Ra and ²³²Th in Ground water around the zonal uranium occurrence, peta Gulf syncline, North east, Nigeria. *Int. J. of Adu. Earth Sci. and Engineer* (2).1
- Arabi, S.A., Funtua I.I. Dewu, B.B.M and Muhammad, M.A. (2015).Background radiation and radiological hazard associates with local guiding materials around Zaria, Nigeria. Radiochemistry Vol. No. 2, 207-212.
- ASTM (1999).Standard test method for radon in drinking water ASTM Designation.D5072-98.
- ASTM.(1983). Standard Method for sampling surface soils for radionuclides, American Society for Testing Materials, Report No. C (PA: ASTM).
- ASTM. (1986). Recommended practice for investigation and sampling soil and rock for engineering purposes, In: Annual Book of ASTM Standards; (04/08), American Society for Testing Materials, Report No. D, 420 (PA: ASTM).
- Bamford SA, Osae E, Aboh I, Antwi LA (1990): Environmental impact of the goldmining industry in Ghana. Biol. *Trace Elem. Res.* 26-27: 279-85.
- Bernhard, G. (2005). Speciation of uranium in environmental relevant compartments, Landbautorschung VÖlkenrode, Vol. 55. Pp. 3016-3024.
- Boamponsem, L. K., Adam, J. I., Dampane, S.B, Owusu-Ansah, E. and Addae, G. (2010). Heavy Matals, Lavel in Stream of Tarkwa gold mining areas of Ghana, *J. Chem. Pham res*, 2 (3): 504-527
- Bowden, P. and Turner, D. C. (1974). Peralkaline and associated Ring Complexes in Nigerian-Niger Province, West Africa. In: The Alkaline Rocks. Ed. H. Serensen, pp. 330-351. John Willey.

- Bowden, P., Bennett J.N. Kinnaird, J.A. Whitley J.E. Abba S.I. and Penelope K.H. (1981). Uranium in the Niger-Nigeria Younge granite province, Mineralogy magazine Vol. 44.PP 379-389.
- Cember, H. (1996). Introduction to Health Physics, 3rd Edition, McGraw-Hill, New York. Pp. 46-52.
- Dahlgaard, H. (1996). Polonium-210 in mussels and fish from the Baltic-North sea estuary, *Journal of Environmental Radioactivity*, Vol. 32.Pp.
- Darko E. O., Tettea, G. K., and Akaho, E. H. K., (2005). Occupational radiation exposure to NORMS in gold mine, *Journal of Radiation Protection dosimetry* Vol. 114
- E.C. (1996).Council Directive 96/29/EUROTOM/ of 13 May 1996 Laying Down the Basic Safety Standards for the Protection of the Health of Workers and the General Public against the Dangers Arising from Ionizing Radiation, *Official Journal of EC*, Commission of the European Communities Series L, No. 159.
- ECOPHOENIC LTD. (2008). Investment brochure for industrial and energy minerals in Nigeria.
- El-Taher, A., Kratz K.L., Nossair, A. Azam A.H., (2003): Determination of gold in two Egyptian gold ores using instrumental neutron activation analysis; *Science Direct. Radiation Physics and Chemistry* 68, 751 755.
- Faanu, A., Darko, E. O., and Ephraim, J. H., (2010): Assessment of public Exposure to naturally occurring radioactive materials from mining and processing activities of Tarkwa Gold mine in Ghana *J. Environ. Monit Assess* 180: 15-29
- Faanu, A., Ephraim J. H., Darko, E. O., Kpeglo, D. O Lawluvi; H. and Adukpo, O. (2011); Determination of the concentration of physical chemical parameter in water and soil from a gold mining ore in Ghana *Res J. of Environ. And Earth Sc* 3 (2) 177-186
- Federal Republic of Nigeria (2006). Nuclear safety and Radiations protection act (1995 No. 191 and Nigerian safety and security of radioactive sources regulation 2006.
- Federal Republic of Nigeria official Gazeth (2003). Nigeria Basic Ionizing Radiation Regulation Fourth Schedule.
- Galadama, A. and Garba, Z. N. (2012). Heavy metals pollution in Nigeria; Causal and consequences. Elixer pollution (45) 7917-7922.
- Gbadebo, A. M. (2011): Natural Radionudide distribution in Granitic rocks and soil of abandoned Quarry sites, Abeokuta South Western Nigeria, *Asian J. of App. Sc* 4 (2) 176-185
- Geochemistry and mineralisation of the Ririwai complex, northern Nigeria. *Journal of African Earth Sciences*, 3, 185-222.
- Girgisu, S. Ibeanu I. G.E. Adeyemo D. J. and Okoh; S, (2012)Determination of Heavy metal and other element in Antisanal Mining Soil. *American J. of App. Sc* 9 (7) 1014-1019

- Hayumbu, P. and Mulenga S. (2004): Status of Radon Dosemetry in Zambia underground mine, proceedings of IAEA International Conference (NORM iv) S20Zyrk, Poland.
- Higgy, R. H., El-Tahawy, M. S., Abdel-Fattah, A. T., and Al-Akahawy, U. A. (2000).Radionuclide content of building materials and associated gamma dose rates in Egyptian dwellings, *J. Environ.Radioact*.Vol. 50. Pp. 406-415.
- IAEA. (1989). Measurement of Radionuclides in Food and Environment: A Guidebook, IAEA-Technical Reports Series No. 295, Austria.
- IAEA (2003). Guidelines for radioelement mapping using gamma-ray spectrometry Data, IAEA TECDOC 1363
- IAEA.(1996). International Basic Safety Standards for Protection against Ionising Radiation and for the safety of radiation sources, Safety Series No. 115, IAEA, Vienna.
- IAEA.(2004). Soil sampling for environmental contaminants, IAEA-TECDOC-1415, Austria.
- IAEA. (2005). Naturally Occuring Radioactive Materials (IV), proceedings of an international conference held in Szczyrk, IAEA-TECDOC-1472, Poland.
- Ibeanu, I.G.E (1995). Assessment of Radiological effects of Tin Mining Activities in Jos and its environs. A Ph.D Thesis submitted to the Department of Physics, ABU Zaria.
- ICRP (1994): Protection against Radon-222 at home and at work publication 65, Ann. ICRP23,2Pergamon Press, Oxford and New York.
- ICRP. (1977). Radiation Protection in Uranium and other mines, Vol. 1, No. 1, Pergamon Press, Oxford. Pp. 71-90.
- ICRP. (1991). 1990 recommendations of the International Commission on Radiological Protection, ICRP Publication 60, Pergamon Press, Oxford.
- ICRP.(1993). Protection against Radon-222 at home and work, ICRP Publication 65, Pergamon Press, Oxford.
- ICRP. (2007). 2006 recommendations of the International Commission on Radiological Protection, ICRP Publication 103, Pergamon Press, Oxford.
- Ivan, A.G. (2011). Hard book of phytoremediation. Nova Sci. Pub. Inc. Pp. 93
- Jibiri, N.N., Alausa, S.K.; Owofolaju, A.E. and Adeniran, A.A. (2011). Terrestrial gamma dose rate and physic-chemical properties of form soil from ex-tin mining locations in Jos-Plateau, Nigeria. *African J. of Env.Sci. and Tech* 5(12).
- Jonah S.A. Umar I.M. Oladipo. M.O.A Balogun G.I and Adeyemi D.J. (2006) Standardization of NIRR- 1 irradiation and counting facilities for instrumental Neutron Activation Analysis Centre for Energy Research and Training, Ahmadu Bello University Zaria Pp. 818 822.

- Jorma, S. (1993) Method for determination of Radon ⁻²²² water by liquid scintillation counting. Swidish Radiation Protection Institute (93)-13.
- Karnillyuss J.S., Abafomi, J.D., and Ekedegwa, A. I. G. 2014, History and current status of uranium development Nigeria technical report on uranium for unconventional resources IAEAViena
- Kinnaird J.A., Bowden, P., Ixer, R.A and. Odling N.W.A.(1985). Mineralogy, Geochemistry and mineralization of the Ririwai complex, Northern Nigeria. *J. of Africa Earth Sci.* Vol. (3) 12.
- Knoll, G. F. (1989) Radiation Detection and Measurement, John Wiley & Sons. 2nded., New York
- KOVARCH, A., FLOOD, P.G., TYNE, E., 1994. Geographical information systems for regional scale geological analysis: the Manilla 1:250,000 map area, a case study. Proceedingsof the 7thAustralian Remote Sensing Conference, 1076-1083.
- Kwanbot D.I., Izain, M.M. and Nyan, G.G. (2012) Radionuclide in some food crops from high background radiation area on the Jos plateau, *Nigeria J. of Natural Sci.* (2)6.
- Landsberger, S. (1994). Delayed Instrumental Neutron Activation Analysis, p. 121-139 in: Chemical Analysis by Nuclear Methods, Chapter 6, (Alfassi, Z. B., ed.), John Wiley & Sons. Pp. 461-465.
- Malcolm B. C. (2005). Naturally occurring radioactive materials (NORM) in Australia industrial. Review of current inventories and Future generation. A report for the radiation health and safety advisory council.
- Mangset W.E. and Sheyin A.T. (2009). Measurement of Radionuclides in processed mine tailing in Jos, Plateau. Bayero. *J. of Pure and App. Sc* 2(2) p56-60.
- McDonald P., Baxter M. S. and Scott E. M. (1996). Technological enhancement of natural radionuclides in the marine environment, *Journal of Environmental Radioactivity*, Vol. 32. Pp. 562-569.
- Mielke, J.E., (1979). Composition of the Earth's crust and distribution of the elements. In: Siegel, F.R. (Ed.), Review of Research on Modern Problems in Geochemistry. UNESCO Report, Paris, pp. 13–37.
- Muhammad A.M., I.F., Isa S.P. Mallam and S. A. Arabi (2010). Distribution of Gamma Emitting Radionuclides in soils around the Centre for Energy Research and Training (CERT) Ahmadu Bello University Zaria, Nigeria. *Journal of American Sciences* 2010;10(12).
- Nasiru, R. (1993). Radioactivity levels around the Jos Tin Mines and Mills. An M.Sc Dissertation, ABU Zaria. Pp. 38-45.
- Nasiru, R., Zakari, Y. I., and Abdullahi M. A. (2013). Distribution of Gamma Radionuclides in Gold Ore Mine from Birnin Gwari Artisanal Goldmine Kaduna State Nigeria. Research J. of Applied Science Engineering and Technology 6(17): 3255 3258.

- National Population Commission (2006). National Census Report 2006.
- Nazaroff, W. W., Moed, B. A., and Sextro, R. G. (1988). Soil as source of indoor radon: generation, migration and entry, p. 57-112 in: Radon and its decay products in Indoor Air (Nazaroff, W. W and Nero, A. V. Jr., eds.), John Wiley & Sons, New York.
- NRC. (1999). Evaluation of Guidelines for exposures to Technologically Enhanced Naturally Occurring Radioactive Materials, National Research Council, Washington, DC.
- O'Brien, R. S. and Cooper, M. B. (1998). Technologically Enhanced Naturally Occurring Radioactive Material (NORM): Pathway Analysis and Radiological Impact, Appl. Radiat. Isot, Vol. 49. Pp. 209-220.
- Odusote, O.O., Alausa, S.K. and Gyang, B.N. (2014) Radionuclide concentration and impact assessment of the Jos Tin mining soil residues. The Nucleus 51 No. 1 (1-7).
- OECD/NEA. (1979). Exposure to radiation from natural radioactivity in building materials, Report by NEA Group of Experts, Nuclear Energy Agency (Paris: OECD).
- Ogedengbe.O. (1984).A study of the geochemistry, mineralogy and ore genesis in the mineralized plutonic rocks of the Ririwai comlplex, Kano State, Nigeria. Unpublished M.Sc. Thesis, Univ. of Manchester, United Kingdom
- Ogunleye, P.O., Mayaki, M.C. and Amapu, I.Y. (2002). Radioactivity and heavy metal composition of Nigerian phosphate rocks: possible environment implications. *J. Environ. Radioactivity* **62**, 39 48
- Olivo, G. R., Almeida, C. M., and Chovinard, A, (2009). The sources of gold and associated elements in carlin–type deposits, Northern NevadaU.S.A. *Gold Schmidt conferenceU.S.A*
- Onwoka, S.U., Duluora, J.O., Okoye, C.O. (2013). Socio-economic impacts of Tin mining in Jos plateau state Nigeria. *Int. J. Eng. Sci. Invention* (2)7. Pp. 30-34
- ORTEC (2003) Gamma-Ray Spectroscopy Using NaI(Tl). Experiment 3 Pp. 2-5.
- Oyavoye, M.O. (1964). The geology of the Nigeria basement complex. *J. of Nig. Min. Geol. and Mell. Soc.* Vol. 1.Pp. 7-16
- Reguigui, N Kucera, J., and Benkraiem, (2002), Radioactivity Concentration of 238 U, 232Th 40K and 137Cs in environmental samples and Technologically enhanced products in Tunisia using NAA. *Proceeding of international symposium on environmental pollution control and waste management Tunis* p136-141
- Ron Jenkins, R. W. Gould, and Dale Gedcke, (1981) Quantitative X-ray Spectrometry, Marcel Dekker, Inc., New York, pp
- Sato, J., and Endo, M. (2001). Activity ratios of uranium isotopes in Volcanic Rocks from Izu-Mariana Island-Arc Volcanoes, *Journal of Nuclear and Radiochemical Sciences*, Vol. 2, Japan. Pp. 46-54.

- Szentes G. (2009) Granite formation and granite cavities in Northern Nigeria cadernos Lab. Xeoloxico de laxe corna vol. 34 PP. 13. 26.
- UNSCEAR. (1982). Sources and Biological effects, 1982 report to the General Assembly, with Annexes, United Nations Sales Publication E. 82. IX.8. United nations, New York.
- UNSCEAR. (1988). 1988 Report to the General Assembly with Scientific Annexes, United Nations Sales Publication E.88.IX.7, United Nations, New York.
- UNSCEAR. (1993). Exposures from Natural Sources of Radiation, 1993 Report to General Assembly, Annex A, New York.
- UNSCEAR. (1996). Sources and Effects of Ionising Radiation, 1996 Report to the General Assembly, with Scientific Annex, United Nations, New York.
- UNSCEAR.(2000). Exposures from Natural Sources, 2000 Report to General Assembly, Annex B, New York.
- USEPA (1991) National Primary drinking water regulations for radionuclides.US Government printing office, Washington DC EPA/570/9-9/700.
- USEPA. (1993). Diffuse NORM Waste Characterization and Preliminary Risk Assessment, Prepared by S. Cohen and Associates, Inc., and Rogers & Associates Engineering Corp. for the US Environmental Protection Agency, Office of Radiation and Indoor Air.
- Usikalu M.R., Anoka, O.C. and Balogun, F.A. (2011). Radioactivity measurements of the Jos Tin mine Tailing in Northern Nigeria *Archivesof Physics Research* 2(2) P. 80-86.
- Van der Steen J and Van Weers A.W. (1996), Radiation Protection in NORM industries, NRG, Radiation and Environment, Netherlands.
- WELLMAN, P., 1998. Mapping of a granite batholith using geological and remotely senseddata: the Mount Edgar Batholith, Pilbara Craton. Exploration Geophysics, v. 29, 643-648.
- WHO (1993) Guidelines for drinking water quality vol.1(2nded)
- WHO (1999): Hazard prevention and control in the work environment. Air borne dust WHO/SDE/OEH/199.14, Geneva.
- Yutaka, K. and Micheal (1988). Laboratory manual for liquid scintillation counting Rackard instrument co. inc USA. Pp. 3-15.
- Zakari, Y.I., Nasiru, R., and Abdullahi M.A. (2013). Determination of Absorbed and Annual Effective Doses around Birnin Gwari Artisanal Goldmine, Kaduna State Nigeria. *Research Journal of Environmental and Earth Science* 5(5): 252-255.

NaI(TL) counting for RS1

Appendix 2
Activity concentration values for all the samples analysed with NaI(TL) detector.

Instrumental Neutron Activation Analysis results for dust samples

Liquid scintillation Analysis results for water samples

Appendix 5A

Soil_Descriptive Statistics

	N	Range	Minimum	Maximum	Me	an	Std. Deviation	Variance
	Statistic	Statistic	Statistic	Statistic	Statistic	Std. Error	Statistic	Statistic
40-K Value	28	573.44	22.82	596.26	296.8718	29.58442	156.54604	24506.664
40-K Error	28	19.66	.99	20.65	8.2729	.95899	5.07448	25.750
226-Ra Value	28	93.24	14.06	107.30	49.6600	5.03556	26.64570	709.993
226-Ra Error	28	17.22	.88	18.10	6.5682	.73898	3.91030	15.290
232-Th Value	28	549.27	81.53	630.80	257.2421	28.95919	153.23766	23481.779
232-Th Error	28	10.94	2.05	12.99	6.5286	.59968	3.17322	10.069
AD Value	28	341.93	50.85	392.78	170.7271	17.72706	93.80280	8798.966
AD Error	28	7.44	1.47	8.91	4.6193	.39660	2.09861	4.404
AE Value	28	.42	.06	.48	.2089	.02182	.11548	.013
AE Error	28	.01	.00	.01	.0053	.00048	.00254	.000
Rn Value	28	151.05	22.78	173.83	78.7986	8.15477	43.15100	1862.009
Rn Error	28	28.02	1.30	29.32	10.1971	1.23643	6.54258	42.805
Em Value	28	.84	.87	1.71	1.2257	.04730	.25028	.063
Em Error	28	.01	.01	.02	.0125	.00050	.00265	.000
Raeq Value	28	868.84	132.71	1001.55	436.9157	44.11072	233.41201	54481.167
Raeq Error	28	30.45	6.23	36.68	16.9475	1.52225	8.05501	64.883
Hx Value	28	2.37	.36	2.73	1.1919	.12039	.63706	.406
Hx Error	28	.16	.02	.18	.0508	.00601	.03179	.001
Hin Value	28	2.55	.40	2.95	1.3263	.12931	.68423	.468
Hin Error	28	.11	.03	.14	.0621	.00540	.02858	.001
Valid N (listwise)	28							

Appendix 5B

Cereals_Descriptive Statistics

	N	Range	Minimum	Maximum	Me	an	Std. Deviation
	Statistic	Statistic	Statistic	Statistic	Statistic	Std. Error	Statistic
40-K Value	15	116.800	14.590	131.390	59.99867	9.769041	37.835334
40-K Error	15	9.030	1.130	10.160	2.75600	.577618	2.237105
226-Ra Value	15	46.110	9.740	55.850	25.95333	3.476827	13.465694
226-Ra Error	15	4.830	.160	4.990	2.55400	.439988	1.704066
232-Th Value	15	71.110	7.790	78.900	46.81933	4.686629	18.151236
232-Th Error	15	3.150	.310	3.460	1.99333	.193351	.748844
AD Value	15	52.490	7.410	59.900	32.56740	3.324830	12.877012
AD Error	15	1.430	.890	2.320	1.45000	.098140	.380094
Hin K Value	15	.101	.013	.114	.05187	.008445	.032706
Hin K Error	15	.003	.001	.004	.00173	.000228	.000884
Hin Ra Value	15	1.810	.380	2.190	.97980	.129781	.502638
Hin Ra Error	15	.189	.006	.195	.10020	.017318	.067074
Hin Th Value	15	2.291	.250	2.541	1.50767	.150975	.584724
Hin Th Error	15	.102	.009	.111	.06387	.006266	.024269
Hin T Value	15	3.762	1.003	4.765	2.53953	.255202	.988392
Hin T Error	15	.233	.026	.259	.15013	.018518	.071721
Raeq Value	15	138.910	32.840	171.750	97.53133	9.509145	36.828759
Raeq Error	15	6.800	1.200	8.000	5.55133	.491855	1.904948
Hx Value	15	.377	.088	.465	.26420	.025760	.099768
Hx Error	15	.018	.006	.024	.01773	.001329	.005147
Hin Value	15	.475	.136	.611	.33100	.033909	.131329
Hin Error	15	.034	.005	.039	.02200	.002457	.009517
Valid N (listwise)	15						

Appendix 5C

Vegetables_ Descriptive Statistics

		_					Std.
	N	Range	Minimum	Maximum	Me	an	Deviation
	Statistic	Statistic	Statistic	Statistic	Statistic	Std. Error	Statistic
40-K Value	11	331.900	127.260	459.160	259.25364	34.546620	114.57817 8
40-K Error	11	14.960	.480	15.440	4.68273	1.519924	5.041018
226-Ra Value	11	60.370	7.750	68.120	28.05182	4.719308	15.652174
226-Ra Error	11	8.410	.780	9.190	4.97273	.964720	3.199616
232-Th Value	11	126.880	25.400	152.280	54.55727	10.769994	35.720029
232-Th Error	11	10.930	.390	11.320	2.58091	.911236	3.022229
AD Value	11	90.650	23.670	114.320	45.04273	7.779689	25.802310
AD Error	11	7.540	.320	7.860	1.98909	.616165	2.043587
Hin K Value	11	.154	.017	.171	.09136	.014711	.048792
Hin K Error	11	.006	.000	.006	.00191	.000579	.001921
Hin Ra Value	11	1.014	.130	1.144	.47091	.079281	.262947
Hin Ra Error	11	.141	.013	.154	.08345	.016146	.053551
Hin Th Value	11	1.750	.351	2.101	.75273	.148564	.492732
Hin Th Error	11	.151	.005	.156	.03573	.012540	.041591
Hin T Value	11	2.862	.552	3.414	1.32000	.226068	.749784
Hin T Error	11	.326	.019	.345	.12482	.027083	.089824
Raeq Value	11	243.830	45.540	289.370	107.47364	19.586912	64.962439
Raeq Error	11	9.510	1.350	10.860	7.39727	1.034589	3.431344
Hx Value	11	.710	.158	.868	.35473	.057900	.192032
Hx Error	11	.066	.003	.069	.02200	.005532	.018347
Hin Value	11	.867	.178	1.045	.41291	.068419	.226920
Hin Error	11	.085	.006	.091	.03764	.007315	.024262
Valid N (listwise)	11						

Appendix 5D

Dust_Descriptive Statistics

	N	Range	Minimum	Maximum	Me	an	Std. Deviation
	Statistic	Statistic	Statistic	Statistic	Statistic	Std. Error	Statistic
40-K Value	10	898.310	140.850	1039.160	385.90300	86.822684	274.55743 6
40-K Error	10	13.140	1.570	14.710	5.69700	1.263741	3.996301
226-Ra Value	10	103.120	.000	103.120	27.92800	8.928301	28.233766
226-Ra Error	10	5.430	.000	5.430	2.50700	.514073	1.625642
232-Th Value	10	166.830	45.070	211.900	146.64400	17.275681	54.630499
232-Th Error	10	.810	.410	1.220	.90500	.080860	.255702
AD Value	10	94.010	53.740	147.750	106.35500	10.137856	32.058717
AD Error	10	1.170	.610	1.780	.97400	.108364	.342676
A E Value	10	.115	.066	.181	.13030	.012386	.039169
Raeq Value	10	355.070	120.820	475.890	266.98200	30.645090	96.908284
Hx Value	10	1.122	.364	1.486	.79350	.094677	.299396
Hin Value	10	1.757	.400	2.157	.92750	.147500	.466436
Valid N (listwise)	10						

Effective dose coefficient for ingestion of radionuclides

Appendix 7a

RESULT SUMMARY SOIL

Radiological index	Mean (Range)	Unit	Ref level
Activity concentration			
40k	296.87	Bq/kg	420 UNSCEAR
	(22.8 – 596.26)		
226 Ra	49.66	Bq/kg	33 UNSCEAR
	(14.06 – 107.30)		
232 Th	257.24	Bq/kg	45 UNSCEAR
	(81.53 – 630.80)		
Absorbed	170.72	nGy/h	60 UNSCEAR
	(50.85 – 392.78)		
Annual effective dose	0.29	mSv/year	I mSv/year
	(0.06 - 0.48)		
222 Rn	78.79	KBq/m ⁻³	
	(22.78 – 173.83)		
222 Rn EF	1.22	KBq/m ⁻³	(0.5 – 0.7) UNSCEAR
	(0.87 – 1.71)		
Hazard			
Req	436.92	Bq / kg	370 UNSCEAR
	(0.87 – 1.71)		
Hex	436.92	Bq / kg	1
	(132.71 – 1001.55)		
Hin	1.33	Bq / kg	1
	(0.399 - 2.59)		
RISK			
CANCER			
Whole	1.15 x 10 ⁻⁵		1 x 10 ⁻⁶ – 1 x 10 ⁴ USEPA
Adult	0.85 x 10 ⁻⁵		
Life time	3.05 x 10 ⁻⁴		
HEREDITARY			
Whole	4.18 x 10 ⁻⁵		1 x 10 ⁻⁶ – 1 x 10 ⁻⁴ USEPA
Adult	12.09 x 10 ⁻⁷		
Lifetime	2.23 x 10 ⁻⁵		

Appendix 7b

Results summary

CEREAL

	Radiological index	Mean (Range)	Unit	Ref level
1.	Activity conc.			
	4ok	59.99 (44.59 – 131.39)	Bq/kg	
	226Ra	25.95 (9.74 – 55.85)	Bq/kg	
	232Th	46.81 (7.79 – 79.90)	Bq/kg	80 mBq/kg UNSCEAR
				3.0 mBq/kg UNSCEAR
	Absorbed date rate	32.59 (7.41 – 59.90)	nGy/h	
	Annual effective dose	2.54 (1.00 – 4.76)	mSv/Year	
	Hazard			
	Raeq	97.53 (32.84 – 171.75)	Bq/kg	
	Hex	0.26 (0.088 – 0.46)		1 UNSCEAR
	Hin	0.33 (0.136 – 0.61)		1 UNSCEAR
	RISK			
	CANCER			
	Whole	1.34 x 10 ⁻⁴		1 x 10 ⁻⁶ – 1 x 10 ⁴ USEPA
	Adult	1.04 x 10 ⁻⁴		
	Life time	9.38 x 10 ⁻³		
	HEREDITARY			
	Whole	5.10 x 10 ⁻⁶		1 x 10 ⁻⁶ – 1 x 10 ⁻⁴ USEPA
	Adult	2.54 x 10 ⁻⁶		
	Lifetime	3.4 x 10 ⁻⁴		

Appendix7c

Result summary

VEGETABLE

	Radiological index	Mean (Range)	Unit	Ref level
1.	Activity conc.			
	4ok	259.25	Bq/kg	
	226Ra	(127.26 – 459.16) 28.05	Bq/kg	
		(7.75 – 68.12) 54.56	Bq/kg	50 mBq/kg UNSCEAR
	232Th	(25.40 – 152.28)		
				15mBq/kg UNSCEAR
	Absorbed dose rate	45.04	nGy/h	
		(23.67 – 114.32)		
	Annual effective dose	1.32	mSv/Year	
		(0.55 - 3.41)		
	Hazard			
	Ra _{eq}	107.47	Da/ka	
		(45.54 – 289.87)	Bq/kg	
	H _{ex}	0.35		
		(0.16 - 0.87)		
	H _{in}	0.41		
		(0.18 – 1.05)		
	RISK			
	CANCER			
	Whole	7.26 x 10 ⁻⁵		1 x 10 ⁻⁶ – 1 x 10 ⁴ USEPA
	Adult	5.41 x 10 ⁻⁵		,,
	Life time	5.29 x 10 ⁻³		,,
	HEREDITARY			
	Whole	2.6 x 10 ⁻⁶		1 x 10 ⁻⁶ – 1 x 10 ⁻⁴ USEPA
	Adult	1.3 x 10 ⁻⁶		,,
	Lifetime	1.8 x 10 ⁻⁴		,,

Appendix 7d

Results summary

DUST

	Radiological index	Mean (Range)	Unit	Ref. level
1.	Activity conc.			
	4ok	385.90	Bq/kg	
		(140.88 - 1039.16)		
		54.32	Bq/kg	
	226Ra	(0.00 - 290.25)		
		146.64	Bq/kg	
		(45.07 – 24.90)	24,18	
		(13.07 21.30)		
	232Th			
	Absorbed dose date rate	119.85	nGy/h	
		(53.74 - 254.46)		
	Annual effective dose	0.147	mSv/Year	
	** 1	(0.066 - 0.312)		
	Hazard	266.00		270 1719 675 4 75
	Raeq	266.98	D1 //	370 UNSCEAR
	Hex	(120.82 – 475.89) 0.794	Bk/kg	1 UNSCEAR
	nex	(0.36 - 1.49)		I UNSCEAR
	Hin	0.93		1 UNSCEAR
		(0.40 - 2.16)		TONSCLAR
	RISK	(0.10 2.10)		
	CANCER			
	CANCER			
	Whole	8.09 x 10 ⁻⁶		1 x 10 ⁻⁶ – 1 x 10 ⁴ USEPA
	Adult	6.03 x 10 ⁻⁶		"
	Life time	5.16 x 10 ⁻⁴		,,
	HEREDITARY			
	Whole	2.9 x 10 ⁻⁷		1 x 10 ⁻⁶ – 1 x 10 ⁻⁴ USEPA
	whole	2.9 X 10		1 X IU - I X IU USEPA
	Adult	1.7 x 10 ⁻⁷		,,
	Lifetime	2.06 x 10 ⁻⁵		,,

Appendix 7e

Results summary WATER

Radiological index	Mean (Range)	Unit	Ref level
Activity conc.	2.23k	Bq/kg	10 UNSCEAR
222Rn	(1.66 - 2.61)		
Annual effective dose	0.016	mSv/year	
	(0.012 - 0.09)		
RISK			
CANCER			
Whole	0.09 x 10 ⁻⁵		$1 \times 10^{-6} - 1 \times 10^{4}$
			USEPA
Adult	0.06×10^{-5}		,,
Life time	6.30 x 10 ⁻³		,,
HEREDITARY			
Whole	3.2 x 10 ⁻⁶		$1 \times 10^{-6} - 1 \times 10^{4}$
			USEPA
Adult	1.63 x 10 ⁻⁶		,,
Life time	2.24 x 10 ⁻⁴		,,
	Activity conc. 222Rn Annual effective dose RISK CANCER Whole Adult Life time HEREDITARY Whole Adult	Activity conc. 2.23k (1.66 – 2.61) Annual effective dose 0.016 (0.012 – 0.09) RISK CANCER Whole 0.09 x 10 ⁻⁵ Adult 0.06 x 10 ⁻⁵ Life time 6.30 x 10 ⁻³ HEREDITARY Whole 3.2 x 10 ⁻⁶ Adult 1.63 x 10 ⁻⁶	Activity conc. 2.23k (1.66 – 2.61) Annual effective dose 0.016 (0.012 – 0.09) RISK CANCER Whole 0.09 x 10 ⁻⁵ Adult 0.06 x 10 ⁻⁵ Life time 6.30 x 10 ⁻³ HEREDITARY Whole 3.2 x 10 ⁻⁶ Adult 1.63 x 10 ⁻⁶



Plate 1: Ririwai Underground tin mine



Plate 2: Tailing bearing water





Plate 4: Main source of tap water



Plate 5: Active mine pit



Plate 6: Active pit and lotto where dust sample was collected



Plate 7: Irrigation land where vegetable samples were collected