RADIOACTIVITY AND ELEMENTAL STUDIES IN WATER AND SEDIMENT OF AJIWA DAM, KATSINA STATE

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DECLARATION

I declare that the work in this thesis "Radioactivity and elemental studies in water and sediments of Ajiwa Dam, Katsina State, Nigeria" has been carried out by me under the supervision of Dr. Y.I. Zakari and Dr. U. Sadiq in the department of Physics. The information derived from the literature has been duly acknowledge in the text and list of references provided.

CERTIFICATION

This thesis titled "Radioactivity and elemental studies in water and sediments of Ajiwa Dam, Katsina State, Nigeria" by Zaharaddeen Lawal meets the regulations governing the award of master's degree in Radiation Biophysics of Physics department of Ahmadu Bello University, Zaria and is approved for its contribution to knowledge and literary presentation.

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DEDICATION

I dedicated this work to my Late Supervisor Professor. Zakari Yusuf Ibrahim, may his soul rest in perfect peace, amen.

ABSTRACT

A radioactivity measurement was carried out in water sediments of Ajiwa Dam. The sediment samples were collected, prepared and analyzed using Atomic Absorption Spectrometry (AAS) for the activity concentration of Zn, Cd, Cr, Cu and Pb. The results obtained shows average activity of Cr ($1.69x10^{-6}$µg/L) and Zn ($5.69x10^{-6}$µg/L) show the higher concentration followed by Cu ($5.25x10^{-7}$µg/L) and Cd ($7.50x10^{-8}$µg/L). the concentration of metals recorded in this study were lower than USEPA guidelines for drinking water quality Zn (5000µg/L), Cu (1300µg/L), Cr (100µg/L) and d (5µg/L). However Pb was found below the detection limit throughout the samples. The levels of Exp_{ing} observed in order Cr>Cd respectively, while Cu, Pb and Zn were not detected. The mean levels of HQ_{ing} was found to be $1.79x10^{-3}$, this indicates that the value found to be below the unity HQ_{ing}>1. The average CDI values for selected metals (Zn, Cu, Cr, and Cd) were $1.78x10^{-7}$, $1.65x10^{-8}$, $5.327x10^{-8}$ and $2.355x10^{-9}$respectively, therefore CDI indices for heavy metals in the study area were found in order; Zn>Cu.Cr.Cd. The gross alpha and beta radioactivity of water samples were measured using proportional counter. The result show that the values are lower than the permissible levels of 0.1 and 1.0Bq/L set by WHO. The range of the measured Alpha and Beta activity were found to be $1.24x10^{-2}$ to $2.12x10^{-1}$Bq/L and $1.96x10^{-2}$ to $8.65x10^{-1}$Bq/L. this indicate that the water in all the sampling sites is safe for farming, fishing and domestic application when compared with World Health Oganization values.

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CHAPTER ONE
GENERAL INFORMATION

1.1 Introduction

Water is the most essential material that makes and sustains life on earth planet. Unlike other natural resources or raw materials, ground and surface water is present throughout the world with different distribution n used in drinking, aquaculture, irrigation, power generation, recreation, etc. this has made ground and surface water with their produce an area of concern against their hazardous environmental effects. This includes the effects of radionuclides on livelihood and sustainable development. Radioactivity in environment comes from both natural background and man-made sources. Naturally abundant radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in the environment, and release from fertilizers, agro chemicals, research and medical facilities forms the bulk of radionuclide in ground and surface water, (Wiser, *et al.*, 2003). Therefore presence of radioactivity in the environment can be attributed to naturally occurring and or artificially induced sources. Naturally occurring radioactivity due to bedrock formation which are weathered, resulting in mineral leaching that leads to contaminations (martin *et al* ., 1995); artificial radioactivity is due to human activities like mining and milling of mineral ore which exposes the earth surface. All this contamination may have health effect; that poses great danger to human and other living organisms in the biosphere. Due to this reason, the protection of water resources, water quality and biosphere is very critical for global human and environmental safety.

Many radionuclides are known to decay by alpha, beta, and other form of emissions which happen to pose hazard when ingested. Among which are Cesium- 137, Chromium- 51, Cobalt- 60, Iodine- 132, Lead- 210, Phosphorus- 32, Radium- 226, Radon- 222, Scandium- 46, Strontium- 90, Thorium- 232, Uranium- 238, Zinc- 65, and Zirconium- 96, (carry, 1996).

Exposure to ionizing radiation represents a potential hazard to humans and radioactivity is known to trigger deterministic effect and probabilistic effect (hall and Giacca, 2006).

Radionuclides in water can have serious effect on the biological system through emission of radiation at various energies which will have molecular interaction with the cellular composition of organism, (Stephan, *et al*., 2004). Alpha radiation, for instance was recognized to be most hazardous type of radiation when incorporated in the human body.

This is due to the high mass and linear energy transfer (IET) of α- particles (knoll, 1989). Thus, all radiobiological damage effects begin with the consequence of radiation interactions with the atom forming the cells. If the cell is exposed to radiation, the probability of the radiation interacting with the DNA molecule is very small since these critical components make up such a small part of the cell (Brenner *et al*., 2003). However, each cell, just as is the case for the human body, is mostly water. Therefore, there is a much higher probability of radiation interacting with the water that makes up most of the cell's volume. When radiation interacts with water, it may break the bonds that hold the water molecules together, producing fragments such as hydrogen (H) and hydroxyls (OH). These fragments may recombine or may interact with other fragments or ions to form compounds, such as water, which would not harm the cell. However, they could combine to form tonic substances, such as hydrogen peroxide (H_2O_2), which can contribute to the destruction of the cell (Brenner *et al*., 2003).

This has made it essential to assess the presence of radionuclides in soil, water, atmosphere and other consumer products as generally recommended, World Health Organization (WHO), Food and Agricultural Organization (FOA), International Labor Organization (ILO), Nuclear Energy Agency (NEA), Pan American Health Organization (PAHO), and International Atomic Energy Agency (IAEA) at their 1996 Vienna, general Assembly that set an international Basic Standard (BSS) for protection against ionizing radiation and for the safety of radiation sources, IAEA (1996).

Minerals mining, milling and the use of nuclear facilities and radioisotopes are on the increase. Evaluation of gross alpha and beta activity is the first step used in most methods of ascertains the

presence of radionuclides in drinking water. This is to ensure that the reference dose level (RDL) of committed effective dose of 0.1mSv annually is not exceeded from consumption of drinking water. This constitute a 10% exposure dose limit for public as recommended by, International Commission for Radiological Protection (ICRP, 199) and International Atomic Energy Agency (IAEA, 1996)

Medical and biological research in the past decades discovered that radiation exposure due to the incorporation of natural radionuclides could lead to chronic disease such as cancer of the kidney, colon, and lung.

1.2 Statement of the Problem

Determination of radionuclides in drinking water, and the general environment has become increasingly important during the past few years; this may particularly be due to the most communities acknowledging the fact that the elevated levels of radionuclides in drinking water and environment can be harmful to humans, especially babies and children (Rabi'u, 2003)
Ajiwa Dam is generally used for irrigation, fishing, household activities and source of drinking water for livestock.

Uses of agrochemicals, fertilizers are some of the human activities that are very common in the area. These may contribute to the inducement of radionuclides contamination in the Dam reservoir.

High concentration of radioactive substance in the Dam may endanger the health of human, animals, plants and also may affects the suitability of crops and water for human consumption (Thorntan, 1983). Therefore, there is need to probe the radiological hazard of the biosphere of the people living in this important area. Gross alpha and beta activity survey or gamma spectroscopy is the first step for natural radionuclides probe it will provide information on the environment for essay of the radiological safety of the area.

Atomic Absorption is going to be employed in order to determine the levels of heavy element of the sediment at the Dam and gross alpha and beta activity is also going to be employed to ascertain the presence of radionuclides in the Dam. This is to ensure that the reference dose level (RDL) of committed effective dose of 0.1mSv annually is not exceeded from consumption of drinking water.

1.3 Aim and Objectives

The aim of the study is to measure the radioactivity levels of water and sediments of Ajiwa Dam. The objectives of the study are:

1. To evaluate the gross alpha and beta radioactivity in water of the Dam.
2. To determine the activity concentration of elements (Pb, Cu, Zn, Cd, and Cr) in the sediment samples.
3. To evaluate health risk associated with exposure to these trace metals

1.4 Justification

Radionuclides in water can have serious effect on biological system through the emission of radiation of various energies which will have molecular interaction with the cellular composition of the organism, (Stephan, *et al*., 2004). Alpha and beta radiation have high Linear Energy Transfer (LET) radiations. Alpha radiation, for instance was recognized to be. The most dangerous type of radiation when incorporated in the human body.

Since there is no data in terms of radioactivity measurement in the area, the result of this investigation will be used by Katsina state government and other stakeholders in putting good environmental protection strategic planning as well as protection of human health. This can be achieved by educating the settlers and farmers in that area and most importantly radiological and toxicological risk involved by their activities. Useful information will also be provided in protecting the public against diseases especially those which could be as a result of ingestion of

food contaminated with radioactive substances from agrochemicals and other wastes. This work will also establish a base line data in terms of radioactivity measurement in the area.

1.5 The Scope of the Research

The scope of the research is limited to Ajiwa area in Katsina state. The survey of alpha and beta radioactivity in the water ideally should involve proportional counter, alpha spectrometry and liquid scintillation methods. However, because of the non- availability of the certain equipment in the Centre for Energy Research and Training Zaria, the researcher is limited to the methods to proportional counter only.

The measurement of Pb, Cu, Zn, Cd, and Cr in the sediment samples will be carried out using Atomic Absorption Spectra Analysis, the measurement is going to be carried out in MUSRL/ABU Zaria.

CHAPTER TWO
LITERATURE VIEW

2.1 Environmental Radioactivity

2.1.1 Introduction

Environmental radiation permeates space, air, soil and water around us. It has been naturally present since the birth of earth about 4.5 billion years ago in a massive supernova explosion creating the heavy elements on earth and the other rocky planets mars, Venus and Mercury. Consequently, life has evolved in the ashes and remnants of this explosion which were gravitationally grabbed in space by the sun into our solar system in an environment which her significant level of ionizing radiation (A. Brodsky, 1978).

The natural background radiation is low intensity radiation that is part of our natural environment. We are continually being bombarded with neutrinos from the sun. Part of it is from constituents in the soil such as the common heavy metals of Thorium and Uranium and their daughter nuclide such as radium and radon. These are continuously transforming through the processes of radioactive decay given up emissions of alpha, beta and gamma radiation. Although not very abundant, they are widely spread and minerals containing them are practically everywhere.

Radiation originates from outer space as cosmic radiation, the ground as terrestrial radiation such as radon – 222, and even from within our own bodies like carbon - 14 and potassium - 40.

In case of ^{40}k in living matter, with a 1.28 billion years half-life, it emits a beta particle and becomes a stable member of the calcium family of isotopes.

For ^{14}C in living matter, with half– life of 5730 years it emits a beta particle and becomes a stable member of the nitrogen family of isotopes.

In smoking, the ^{222}Rn decay product of radon ^{210}Pb ($T_{1/2}$ = 22.3 y) exist in solid form in the particulate matter of tobacco leaves and consequently in smoke. It delays in to ^{218}Po, ^{214}Po, then ^{210}Po ($T_{1/2}$ = 138.38 d) which emits an energetic alpha particle with 5.3 MeV of energy.

Radiation is present in the air we breath, the food we eat, the water we drink, and in construction materials used to build our dwellings. Certain food such as bananas, coconuts and Brazil nuts naturally contain higher levels of radiation than other foods. Brick and stone homes have higher natural radiation levels than homes made of other building materials such as wood (M. Eisenbud, 1997).

The USA's capital and the central station in New York are largely constructed of granite, and contain higher levels of natural radiation than most homes.

Cosmic rays from outer space also bombard the earth with streams of highly energetic particles, and high energy particles from the solar wind continuously fall on the earth and increase in intensity during solar flares and at the peaks of the 11 – 22 years.

2.1.2 Radiation Exposure

Naturally occurring background radiation is main source of radiation effective dose for most humans. Levels typically range from about 0.359 to 0.940 Centisievert, (CS) or rem per person per year but can be reach more than 5 [CSV/ (person. year)].

Level of natural or background radiation can vary greatly from one location to the next. For example, people residing in Colorado, USA, are exposed to more natural radiation than residents of the east or west coast because Colorado is reached by more cosmic radiation at high altitude or one mile above sea level at Denver, its capital, and more terrestrial radiation from soil rich in naturally occurring Uranium and Thorium.

Furthermore, a significant fraction of our natural exposure is due to radon, a low boiling point gas from earth's crust from the decay chain of Thorium and Uranium that is present in the air we breathe.

There exist about sixty naturally occurring and 1,500 different radioactive nuclides, mostly man made. They are categorized as to their origin as:

1. Primordial: existing from the formation of earth.
2. Cosmo genic: Formed as a result of cosmic ray interactions with earth's atmosphere.
3. Human made: enhanced or formed due to human activity.

2.1.3 Human - made Radiation

In 1895 x rays were discovered by Ratingen in Germany and have become since then useful source of medical diagnosis and therapy. In 1896 radioactivity was discovered by Henry Becquerel in France. In 1934 it was discovered that radioisotopes can be made and their usage spread in hospitals laboratories and industries. In 1945 atomic devices were developed and tested originally in the atmosphere, underground, and then their testing was banned. The fission fragments and resulting from the atomic testing in 1950s still exist in the atmosphere and are descending to earth's surface as radioactive fallout. Man made sources of radiation from medical, commercial, and industrial activities contribute 0.0672 csv or rem to our annual radiation exposure. One of the largest of these sources of exposure is medical x-rays. Diagnostic medical procedures account for about 0.05 csv or rem each year. (M. Eisenbud, 1978).

Other sources of technological radiation exposure include high flying airplanes, space travel, particle accelerators, and television and computer video screens. In addition some consumer products such as tobacco, fertilizer, welding rods, kerosene lamps Thorium mantles, luminous watch dials, and smoke detectors contribute another 0.0062 csv or rem to our annual radiation exposure.

The additional man made radiation dosage is primarily as a result of the use of x rays in the search for decayed teeth, broken bones, lung lesions, swallowed objects, surgical instruments forgotten inside the body, and other useful or useless procedures for instance, x – rays are sometimes administered to investigate ankle sprains to eliminate the possibility of bone breaks, which otherwise could lead to an undesirable liability insurance situation.

Ionizing radiation is generated in a range of useful applications, medical, commercial and industrial activities. The most familiar and in national terms, the largest of these sources of exposure is medical x-rays. Natural and artificial radiations are not different in kind or effect.

2.1.4 High Background Radiation Areas, Hbras.

Background radiation levels are from a combination of terrestrial radiation from isotopes such as ^{40}K, ^{232}Th, ^{226}Ra and cosmic radiation. They are occurring in air, water, soil, minerals, building materials and food supplies. About 60 radio nuclides exist in nature. The absorbed dose rate level is fairly constant over the world, being of the order of 8 - 15 (μrad / hr). About 82 percent of radiation is from natural sources, particularly radon gas.

There are some areas with sizable population that have high background radiation levels. These areas are primarily located in areas such as Guarapari, Brazil, South West France, and the Kerala coast, India, Ramsar, Iran and China. The higher radiation level is due to high concentrations of radioactive minerals in soil. One such mineral monazite, is a highly insoluble rare earth mineral that occurs in beach and together with the mineral limonite, which gave the sand used in manufacture of industrial abrasives such as grinding wheel sand and sand paper.

Certain areas of South – West India in the Travancore - Cochin State along the coastline are characterized by patches of radioactive sand with the highest Thorium content in the world at 33 percent. The highest known level of background radiation affecting a substantial population is in Kerala and Madras states in India where some 140,000 people receive affective dose rates which average over 1.5 csv or rem per year from gamma radiation in addition to a similar dose from radon gas.

Comparable levels occur in Brazil and Sudan, with average affective dose rates up to about 4 csv / yr or rem / yr to many people. The source is environmental radiation from deposits of Thorium and Uranium, with radon and its decay products nuclides. The Brazilian coastal sands contain

radioactive monazite, zircon, Thorianite, and niobate – tantalite, as well as non – radioactive limonite, rutile, pyrochlore and cassiterite (M. Eisenbud and T. Gesell, 1997).

Several places are known in Iran, India and Europe where natural background radiation gives annual per capital effective dose rates more than 5 csv or rems and up to 26 csv or rem at Ramsar in Iran. Life time effective doses from natural radiation range up to several hundred csv or rems. There is no evidence of increased cancers or other health problems arising from these high natural levels.

In the Nile delta, villages were built on Monazite alluvial deposits containing rare earth metals and Thorium in the vicinity of the cities of Edku and Rosetta, East of Alexandria.

In areas of high natural background radiation, an increased frequency of chromosome aberrations has been noted. The increases are consistent with those seen in radiation workers and in persons exposed at high dose levels, although the magnitudes of the increase are somewhat higher than predicted. No increase in the frequency of cancer documented in populations residing in areas of high natural background radiation.

2.1.5 Cosmic Radiation Doses

Cosmic radiation can be divided into two types, primary and secondary cosmic radiation is made up of extremely high energy particles of energy up to the Gev energy level. They are mostly protons, with some larger particles such as Fe nuclei. A large percentage of it comes outside of our solar system and is found throughout space. Some of the primary cosmic radiation is from our sun, produced during solar flares.

The earth's atmosphere and its magnetic field shield Earth's surface from the primary cosmic rays, which are of concern of spare missions. Little of the primary cosmic radiation penetrates to the Earth's surface; the vast majority of it interacts with the atmosphere. When it does interact, it produces the secondary cosmic radiation, which is what we actually see here on earth. These

reactions produce other lower energy radiations in the form of protons, electrons, and neutron. Peons and mourns that make it to the surface of cosmic ray showers.

The annual dose a person gets from cosmic radiation depends on the altitude. From cosmic radiation the USA, the average person will receive a dose of 27 m rem per year and this roughly doubles every 6,000 foot increase in elevation.

Typical cosmic radiation dose rates are: 4μR / hr in the Northeastern USA close to sea level, 20μR / hr at 15,000 feet and 300μR / hr at 55,000 feet.

Thus the radiation dose increases with altitude with a thinner shielding layer of the atmosphere. There is only about a 10 percent decrease at sea level in cosmic radiation rate when going from pole to the equator, but at 55,000 feet the decrease is 75 percent. This is an account of the earths and the sun's geomagnetic field on the primary cosmic radiations. Flying at high altitudes for long durations adds and extra radiation dose to the average annual dose.

2.1.6 Primordial Radionuclides

Primordial radio nuclides were created in the number synthesis process and supernovae event. The original nuclides and their decay product are typically long lived, with half-life often in the order of billions of years. Initial radio nuclides that exist for less than 30 half-lives are not measurable.

2.1.7 Cosmogenic Radionuclides

Cosmic radiation parameters all of space, the source being primarily outside of our solar system. The radiation occurs in many forms, from high speed heavy particles such as Fe to high energy protons, the cosmic radiation, and produces radio nuclides such as ^{14}C. They can have long – lives, but the majority of them have shorter half - lives than the primordial nuclides. Their radiation is not very penetrating and they contribute little to the external exposure. However, in gaseous from like $^{14}CO_2$, and HT^3 they can be inhaled or ingested.

Beryllium with a half-life of 1.6 million years is produced by cosmic rays and is detected in marine deposits. They are present in ferromanganese nodules dredged from a depth of 4,000 meters at 20 ^{0}N, 160 ^{0}N. From the activity, ^{10}Be, it was deduced that these nodules accumulated from the dissolved minerals in seawater at a rate of 1.8 + 1 – 0.5 mm per million years.

2.1.8 Human -Made Radio Nuclides

Human have added to the natural inventories. The amounts are small compared with the natural amounts due to shorter half - lives of many of the nuclides; they have seen a marked decrease since the halting of above ground testing of nuclear weapons.

TABLE 2.1 SOURCES OF HUMAN MADE RADIO NUCLIDES

NUCLIDE	HALF – LIFE	SOURCE
^{3}T	12.3 years	Weapons testing, fission reactors, nuclear fuel processing, nuclear weapon manufacturing.
^{131}I	8. 04 d	Weapons testing, fission reactors, medical treatment of thyroid nodules.
^{129}I	1. 57 x 10^{7} years	Weapons testing and fission reactors.
^{137}Cs	30 .17 years	Weapons testing, fission reactors, nuclear fuel processing.
^{90}Sr	28. 78 years	Weapons testing, fission reactors, nuclear fuel processing.
^{99m}Tc	2. 11 x 10^{5} years	Decay product of ^{99}Mo used in medical diagnosis.
^{239}Pu	2. 41 x 10^{4} years	Bred by neutron capture in fertile ^{238}U.
^{233}U	1. 592 x 10^{5} years	Bred by neutron capture in fertile ^{232}Th

2.1.9 Soil Radioactivity

Activity levels in soils vary greatly depending on soil type, mineral make up and density. The natural radioactivity in a square mile of soil to a depth of 1 foot is shown in table 2.

TABLE 2.2 NATURAL RADIOACTIVITIES IN A TOTAL VOLUME OF 7.894 x 10^5 $m^{3,}$ WITH A SOIL DENSITY OF 1.58 [gcm^3]

Nuclide	**Specific Activity** [**PLs/ gm**]	**Mass of Nuclide**	**Activity found in Volume of Soil** (**Cs**)
Uranium	0.7	2.2 metric tones	0.8
Thorium	1.1	12 metric tones	1.4
Potassium - 40	11	2 metric tones	13
Radium	1.3	1.7 g	1.7
Radon	0.17	11 Hg	0.2
Total			**> 17**

2.1.10 Radionuclides In The Marine Environment

There is a considerable amount of radio nuclides dissolved in the water of the world oceans. In the long term, Uranium could be extracted from the ocean's water for energy production.

The total volume of water in 7^{th} world oceans is about 1.3 x 10^{18} m^3. The concentrations of some Radionuclides are shown as well as their total activity.

TABLE 2.3 OCCURRENCES OF RADIO NUCLIDES IN THE WATER OF THE WORLD OCEANS.

Nuclide	**Activity Density** [PCs L]/	**Total Activity** (Cs)
Uranium	0.9	1.1×10^{9}
Potassium	300	3.8×10^{11}
Tritium	0.016	2×10^{7}
Carbon – 14	0.135	1.8×10^{8}
Rubidium – 87	28	3.6×10^{10}

2.1.11 Radionuclides in Food and Water

The common radio nuclides in food are ^{40}K, ^{226}Ra and ^{238}U and their associated decay product. Brazil nuts are notorious for their 14,000 times content of radioactivity compared with common fruits. Cereals are also high as much as 500 – 600 times that of fruits. Fruits have the lowest content of natural radioactivity.

All muscle building foods such as meat contain potassium. The radioactive potassium - 40 in air muscles provide 10 – 20 times the internal exposure of any of the other incorporated radio nuclides such as ^{226}Ra and ^{14}C

TABLE 2.4: SPECIFIC ACTIVITIES OF RADIO NUCLIDES IN FOOD

Food	**Specific Activity ^{40}k (PCs/kg)**	**Specific Activity ^{226}Ra (PCs /kg)**	**Relative Activity**
Bananas	3,520	1	391
Brazil nuts	5,600	1,000 – 7,000	1,400
Carrots	3,400	0.6 – 2	378
White potatoes	3,400	1 – 2.5	378
Beer	390	-	43
Red meat	3,000	0.5	33
Lima bean	4,640	2 – 5	516
Drinking water	-	0.017	0.019
Cereals			60
Teas			40
Liver and kidney			15
Flours			14
Peanuts & peanut butter			12
Chocolates			8
Biscuits			2
Evaporated milk			1 – 2
Fish			1 – 2
Cheese and eggs			0.9
Vegetables			0.7
Fruits			0.1

High natural radioactivity exists in drinking water of several municipal water supplies and the well water from surface and deep aquifers in rural areas. The radioactivity was found to result from ^{226}Ra and which has been leached out of soil, partly through chemical and bacterial action along with other common elements such as calcium, magnesium and mercury.

2.12 Radionuclides in Construction Materials

The concentrations of Uranium, Thorium and Potassium in construction materials are shown in table 2.5

TABLE 2.5: RADIONUCLIDES CONCENTRATIONS ARE SPECIFIC ACTIVITIES IN CONSTRUCTIONS MATERIALS.

Material	PPM	PCs / g	PPM	PCs / g	PPM	PCs / g
Granite	4.7	1.7	2	0.22	4.0	32
Sand stone	0.45	0.2	1.7	0.19	1.4	11.2
Cement	3.4	1.2	5.1	0.57	0.8	6.4
Limestone concrete	2.3	0.8	2.1	0.23	0.3	2.4

2.2.1 Activity and Specific Activity of a Radioactive Substance.

Activity of radioactive substance is the one gram of radon – 226 (I. G. E, Ibeanu, 2014). 1Cs = 3.7×10^{10}

Mathematically,

$$A = \lambda N$$

Where A is the activity of any given radio nuclide, λ is the decay constant.

In the other hand specific activity (S.A) is the activity per quantity of atoms particular radio nuclide (Breeman, 2003). It is usually given in units of Bq/g, but another commonly used unit of activity is curie (Cs) allowing the definition of specific activity in Cs/g.

2.2.2 Half – Life

The half - life ($T_{1/2}$) for the decay of radioactive nuclide is the length of time it takes for exactly one – half of the radioactive nuclei in the sample to decay. Consequently, after one half – life 50% of the initially present atoms will remain. Substituting $T_{1/2}$into the radioactive decay law and setting the result equals to $N_0/2$ will establish the relationship between the half – life and the decay constant (Podgorsak, 1991).

$$T_{1/2} = \frac{Ln2}{\lambda} \qquad 2.4$$

$$T_{1/2} = \frac{0.693}{\lambda} \qquad 2.5$$

$$\text{And} \quad \lambda = \frac{0.693}{} \qquad 2.6$$

$T_{1/2}$

The half – lives of natural radioisotopes vary from some micro seconds to billions of years.

2.3 Radiological Health Effects

Through immediate risks from drinking water containing low radioactivity levels are small, but consuming this water for a life time increases health risks. Therefore exposure to ionizing radiation represents a potential hazard to humans and radioactivity is known to trigger deterministic and stochastic effects (Hall and Giacca, 2006).

From the biological effects of radiation on human body, radiation effects are generally divided into two categories:

"Deterministic effects" and "Probabilistic effect"

2.3.1 Deterministic Effects

Based on a large number of experiments involving animals and other researches, further supplemented by theoretical studies, it was discovered that severity of certain effects on human being will increase with increasing doses. There exists a certain level, the "Threshold", below which the effect will be absent. This kind of effects is called "deterministic effects"

2.3.2 Characteristics of Deterministic Effects.

- Develop due to cell killing by high dose radiation.
- Appear above a given threshold dose, which is considerably exposure at normal operation.
- The severity of the effect depends on the dose.

2.3.3 Probabilistic Effects

Stochastic effects are usually associated with exposure to low levels radiation exposure for a long period of time (e.g years). The term stochastic, literally means 'random,' the implication being that low levels of radiation exposure are not certain to produce an effect. The induction of cancer and genetic defects are two of the most familiar consequences attributed to stochastic effects.

The description of stochastic effects is subjected to degree of controversy lowing to the difficulties in separating the effects of low-level radiation exposure from the effects of other carcinogens, e.g. tobacco smoke, non-radioactive spies, etc, but the currently accepted theories lead to the following conclusion about stochastic effects.

- There is no threshold level of radiation exposure below which we can say with certainty or genetic effect will not occur
- Doubling the radiation dose double the probability that a cancer or genetic effect will occur.

Taken together, radiation experts refer to these two conclusions as the 'linear-no- Threshold' hypothesis. This hypothesis is questioned from time to time; however, it provides a pragmatic means of estimating radiation risks, and consisted with the (Limited) data that is available.

Example: radiation induced cancer, genetic effect as stochastic effects of radiation have no threshold and can cause cancers for genetic modification, of which the curing rate are rather low to data, they become a major subject of research in radiation protection. Dose level, no correlation between tumor incidence and radiation has been established at low doses as occurring from exposure to natural background radiation. This is attributable to two factors:

1. It is difficult to obtain meaningful data from epidemiological studies where exposure is near or slightly background level exposures, and
2. At a given high dose the effect is observed in severe from in all exposure cells, at higher doses the effect cannot increase.

Example: deterministic radiation effects are "Unwanted effects" observed after radiotherapies for malignant diseases, effects seen after industrial radiation accidents (IAEA, 1996), or those observed in the Hiroshima and Nagasaki Victims after the attack with nuclear weapons in World War II (Kondo, 1993; Preston *et al*, 2003), like cataract, erythema, inferterlity etc.

The severity of deterministic effects depends on dose. However, threshold exists, only above which the effects will occur. The International Commission on Radiological Protection (ICRP)

considers that if the annual radiation doses to the lens of the eyes of radiation workers are restricted to 150 mSv (equivalent to 150 mGy for X-ray), cataract is unlikely to occur during his/her life assuming a work period of 50 years. For other major organs, the annual dose limited for preventing deterministic effects are as follow in table 2.6

Table 2.6 Limit threshold for deterministic effect (ICRP, 1990) threshold for deterministic effects (Sv)

Organ / tissue	Effect	One single Absorption (Sv/year)	Prolong absorption (Sv/year)
Testis	Permanent Infertility	3.5 – 6.0	2
Ovary	Permi infertility	2.5 – 6.0	> 0.2
Lens of eye	Milkey of lens cataract	0.5 – 2.0 5.0	> 0.1 > o.15
Bo ne marrow	Blood forming Deficiency	0.5	> 0.4

Source, (ICRP 1990) publication N0. 60) Recommendation of the international Commission on Radiological Protection

The result of such studies usually do not give statistically significant differences between exposure and unexposed groups to substantiate a health impact (Hall,*et al*; 2009).

However, recent risk assessment reviews of carcinogenicity and exposure to chemicals and radiation have questioned the non-threshold assumption (Clarke, 1999; Tubiana *et al*, 2009; SCHER, 2009) since there is increasing biological evidence for a potential threshold in radiation and chemically induced carcinogenicity. However, the concept of LNT hypothesis still is the accepted standard for radiation protection policies (Hall, *et al*, 2008) and had been validated and reaffirmed by a National Assembly of Science Committee (NASE). Since under this model, about 1% of the population develops cancer in their life time as a result of lionizing radiation from background level of natural and man-made sources.

2.4.1 Effective Dose (E_H)

Some tissues and body organs are more sensitive to radiation than others and an equivalent dose in one organ may be more hazardous than the same equivalent dose in another organ. The ICRP recommends tissue weighting factors (W_T) which are applied to specific body organs. These dimensionless factors take in to consideration the different radio-sensitivities of the different organs and tissues as recommended in ICRP 60.

The effective dose (E_H) to an organ or tissue ifs fund by multiplying the equivalent dose by the appropriate tissue weighting factors showing in the question below

$$E_H = H_T \times W_T \qquad 2.7$$

Where H_T is equivalent dose to an organ or tissue type T

E_H is the effective dose to an organ or tissue type

W_T is the tissue weighting factor for an organ or tissue type T

As the tissue weight factors are dimensionless, the units for effective dose are the same as the units for equivalent dose (i.e. Sv or rem). The same conversion factors between the old units (rem) and new S.I units (Sv) also apply.

2.4.2 Equivalent Dose ($H_{T,R}$)

The absorbed dose in an organ or tissue multiplied by the relevant radiation weight factor, we

$$H_{T,R} = W_R \times D_{T,R} \qquad 2.8$$

Where $D_{T,R}$ is the average absorbed dose in the organ or tissue and W_R is the radiation weighting factor for the particular radiation R. W_R is related to LET.

When the radiation field is composed of different types with different values of W_R the equivalent dose is given by

$$H_T = \sum W_R \times D_{T,R} \qquad 2.9$$

The unit of equivalent dose is J/kg, termed the sievert (Sv)

2.4.3 Committed Dose (E_{50})

The committed dose (E_{50}) is the dose accumulated by the body 50 year following the intake (except in the case of intake by children where it is defined as the dose accumulated until the age of 70). Committed dose can be committed absorbed dose, committed equivalent dose or committed effective dose. It is given the symbols Dl50), Hl50), or El50), where 50 represents the number of years over which the dose is being calculated.

2.5.1 Water Pollution

Once ore is gathered from mines, it can be extracted by using different methods. Ore such method, called leaching, is a low-cost method of removing ore from waste. During the process, workers pile low-grade gold ore in to an outdoor heap and put a cyanide or mercury solution on it. The cyanide / mercury then dissolves the gold, which it turns down a slope in to reservoirs before it is collected. In some countries, environmental laws require that the slope and collection vats are impenetrable so that poisons cannot be released in to the ground. However, lack of protection in other countries means that the poison is often leaked into the soil. These materials then make their way into local water supplies, lakes, rivers, and streams. Drainage of this type raises acid levels in lakes which are harmful to both animals and people. Additionally, some mines internationally dispose tonic waste in to oceans, lakes and rivers, while others place waste or tailings in dammed reservoirs that can occasionally burst and flood area, releasing mercury and other heavy metals in to the water supply. Pollution from mines causes drinking water contamination and high level of mercury or their heavy metals in fish (Kendall O. 2013).

2.5.2 Radium in Water

Radium is a radioactive metal that occurs naturally in trace amounts in rocks, soils, and ground water. As radium decays, it continually releases energy in to the environment until a stable, non-radioactive substance is formed. This energy is part of the natural radiation to which all-living creature. Radium readily dissolves in ground water where acid conditions (low PH levels) are

found. The various forms of naturally occurring radium found in ground water are radium – 224, 226, and 228.

Radium is known to cause bone cancer when consumed in high doses. The National Academic of Sciences (NAS, 2000) has concluded that a long-term exposure to elevated level of radium in drinking water does indeed pose "a higher risk of bone cancer for the people exposed". The US Environment Protection Agency (EPA, 2000) estimate that long - term consumption of water containing

5 PCs / I radium will give 44 added cancer deaths for every million people exposed. The risk doubles to 88 per million at 10 PCs / I, triples to 132 at 15

PCs / I, etc. How does this level of risk compare to other risk in life?

This according to Wisconsin Department of Natural Resources (DNR, 2003) is approximately the same as the risk of dying from lightning strikes, or tornadoes and hurricanes, fortunately, the risk from radium is manageable because radium can be removed from drinking water. Potassium and Carbon are two of the main sources of internal radiation exposures. They enter our bodies through the food we eat and the air we breathe. Potassium essential to life, is distributed throughout our bodies. A small portion (about one - hundred of a percents) of natural potassium consists of a naturally radioactive isotope called potassium – 40. This isotope is the chief radioactive component in normal food and human tissue.

2.6.2 Interaction of Radiation with Matter.

From a practical radiation protection point of view, x – rays and Gamma rays are identical, differing only in their place of origin. Gamma rays are emitted from excited nuclei with a discrete energy. X – Rays are emitted when the extra – nuclear atomic structure undergoes a transition, i.e. an outer shell electron replaces a missing lower shell electron and an x – rays is produced. The energy of x – ray is approximately equal to the difference in the electron energy levels. Since X and r-rays are changeless, they do not interact by electrostatic forces as in the

case of charged particle, which cause ionization of matter directly along their path of travel. However, X and gamma-rays do have sufficient energy to release high-energy secondary changed particles (electrons) from matter through one of three basic interactions:

- The photoelectric effect
- The Compton effect
- Pair production

The high-speed electrons resulting from these interactions then cause ionization of the medium.

2.6.2 The Photoelectric Effect

The photoelectric effect is the interaction of X or gamma-rays photons as well as other photons (such as light), where by all of the energy of photon is transferred to an inner shell electron (photoelectron) usually the K shell, ejecting it from the atom and leaving the atom with an inner shell vacancy. This shell vacancy creates an excitations energy which corresponds to the binding energy (BE) of the ejected photoelectron. The kinetic energy of the electron ejected is given as:

$$T = E_r - BE \qquad 2.14$$

Where Er = photon energy and Be is the Binding Energy.

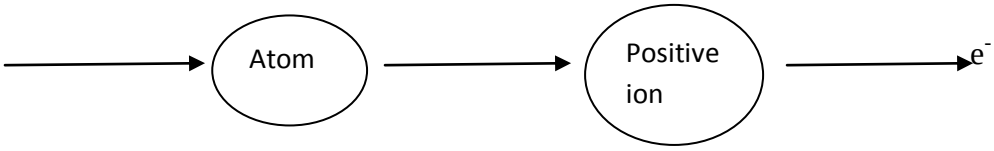


Fig. 2. Photon interaction in photoelectric effect.

If the X or r-rays photon does not have sufficient energy to knock out the inner shell electron loose, the reaction will not occur.

The result and atom is now in an excited state and will decay to the ground state by emission of X-ray and fluorescent radiation with the total energy equal to the BE of the photoelectron. The energy of the secondary radiations is usually much lower than the primary X-ray energies.

The probability of this interaction occurring is called photoelectric cross- section or coefficient, which calculation is dependent on such parameters as Er, Z and A the equation giving T may be written as: (Tsoulfanidis 1983)

$$T\ (m^{-1}) = aN \frac{Z^n}{E^m_r} \quad [1\text{-}r\ (2)] \qquad 2.15$$

T =probability for photoelectric effect to occur per und distance travelled by the photon.

a = Constant with a value of 2 and E_r

m,n = Constant with a value of 3 to 5 (their Values depend on Er)

N = number of atoms/ m^3 for the material through which the photon moves.

Z = atomic number of the material.

The term in parenthesis indicates correction term of the first order in Z.

2.6.3 The Compton Effect

Photons with energies much greater than the BE of the electrons in an atom may interact through essentially elastic scattering interactions in which the total kinetic energy T of the system is conserved. In this interaction the electron appears to the photon as a free electron.

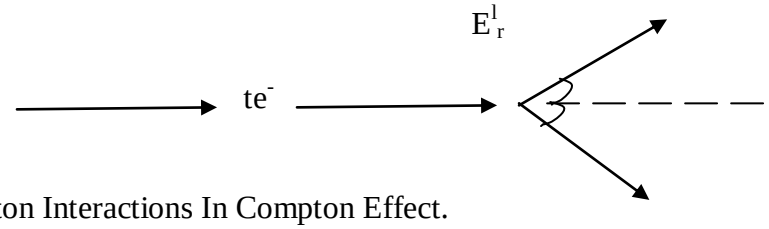


Fig. 2.5 Photon Interactions In Compton Effect.

The primary E_r loses part of its energy to the Compton electron e^-, which gets scattered at an angle from the original direction of the incident, E_r while the Compton scattered E_r is scattered as an angle. In this process, the scattered photon and Compton electron share the energy of the incident therefore the photon energy is reduced by certain amount that is given to the electron.

$$T = Er - E^1_r$$

2.6.5 Pair Production

High-energy gamma photons transfer their energy primarily by pair preclusion. A high energy X or r-ray passing close to a nucleus suddenly disappears and an electron and a positron appear in its place. This interaction must take place in the neighborhood of a nucleus to conserve momentum. Although the nucleus does not undergo any as a result of this interaction, it presence

is necessary for pair production to occur. The conservation of energy gives the following equation for the kinetic energy T of the electron and positron as:

$$Te^- + T_e^+ = E_r - (mc^2)\, e^- - (mc^2)e^+ = E_r\text{-}1.022\text{MeV} \qquad 2.24$$

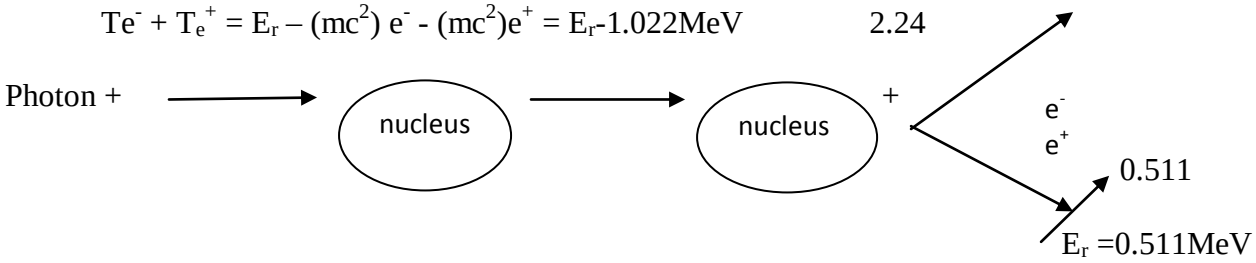


Fig. 2.5 Photon interactions in pair production

+9+Slice both particles are created from energy supplied by the incident photon, the process is energetically possible only if E_r or E_x is greater than 1.02 MeV.

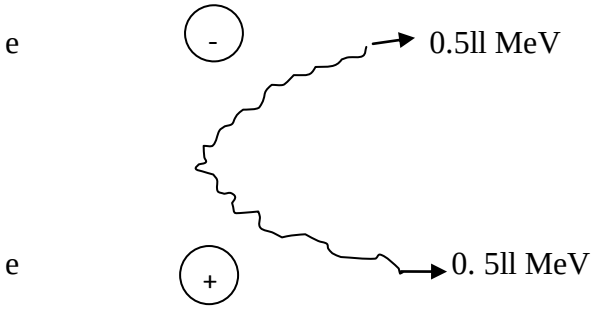


Fig. 2.6 Annihilation electrons.

When the positron slow down (i.e., loses its T) it will annihilate itself by combining with an electron. This produces two photons with energy of 0.511MeV each. This "annihilation radiation" represents the energy equivalent of the rest mass of two electrons, which is converted to pure energy according to the principles of Einstein's theories as shown above.

Therefore, the available kinetic energy is equal to the energy of the two rest masses. The electron and positron share, for practical purpose, the available energy, i.e.

$$T_e\text{-} = T_e^+ = \underline{1}\ (E_r\text{-}\ 1.022\ \text{MeV}) \qquad 2.24$$

Also the probability for the occurrence of pair production is called the pair production cross-section or coefficient, which is give also according to (T soulfanidis, 1983) as:

$$K(m^{-1}) = N2^2\, f\,(E_r, 2)$$

Where

K is the probability for pair production to occur per unit distance travelled and

F (E_r,2) is a function which changes slightly with 2 and increases with E_r.

2.7.0 Defection of Radioactivity in Samples

Detection and measurement radioactivity in samples are usually carried out using gas filled and solid state conductivity detectors. They consist of two components; a radiation detectors and electronic equipment to provide power to detector and display or record radiation events.

2.7.1 Atomic Absorption Spectrophotometry

2.7.1.1 Technique

Atomic absorption spectrophotometry (AAS) is an analytical technique that measures the concentrations of elements. It is commonly used for determining the concentration of a particular metal element within a sample. AAS can be used to analyse for the concentration of over 62 different metals in a solution (Wikipedia). AAS determines the presence of metals in liquid samples. Metals include Fe, Cu, Al, Pb, Ca, Zn, Cd and many more. Atomic absorption is so sensitive that it can measure down to parts per billion of a gram (μgdm-3) in a sample. Typical concentrations range in the low mg/L range. In their element form, metals will absorb ultraviolet light and get excited. Each metal has a characteristic wavelength that will be absorbed. The AAS instrument looks for a particular metal by focusing a beam of UV light at a specific wavelength through a flame and into a detector. The sample of interest is aspirated into the flame. If that metal is present in the sample, it will absorb some of the light, thus reducing its intensity. The instrument measures the change in intensity. A computer data system covert the change in intensity into an absorbance (Steffason et al.,2007).Atomic absorption spectrometry was first used as an analytical technique. The Underlying principles of atomic absorption spectrometry were established in the second half of the 19th century by Robert Wilhelm Bunsen and Gustav Robert Kirchhoff, both professors at the University of Heidelberg, Germany. The modern form of AAS was largely developed during the 1950s by a team of Australian Chemists, led by Allan Walsh at the Commonwealth Scientific and Industrial Research Organization, Division of Chemical physics, in Melbourne, Australia (CSIRD) (Walsh,1955).

2.7.1.2 Principle

Atomic absorption spectrophotometry (AAS) makes use of absorption spectrometry to assess the concentration of an analyte in a sample. It requires standard solutions to establish the relation between the measured absorbance and the analyte concentration, and relies therefore on Beer-Lambert law. In short, the electrons of the atoms in the atomizer can be promoted to higher orbital (excited state) for a short period of time (nanosecond) by absorbing a defined quantity of energy (quantized energy or radiation of a given wavelength). This amount of energy, characterized by the wavelength, is specific to a particular electron transition in the particular element. In general, each wavelength corresponds to only one element, while the width of an absorption line is only of the order of a few picometers (pm), which gives the technique its elemental specificity. The radiation flux without a sample and with a sample in the atomizer is measured using a detector, and the ratio between the two values (the absorbance) is converted to analyse concentration or mass using Beer-Lambert Law (Wikipedia).

2.7.1.3 Instrumentation

In order to analyse a sample for its atomic constituents, it has to be atomized. The atomizers most commonly used nowadays are flames and electro thermal (graphite tube) atomizers. The atoms should then be irradiated by optical radiation, and the radiation source could be an element - specific line radiation source or a continuum radiation source. The radiation then passes through a monochromator in order to separate the element-specific radiation from any other radiation emitted by the radiation source, which is finally measured by a detector.

2.7.1.4 Atomizer

Although other atomizers, such as heated quartz tubes, might be used for special purposes, the atomizers most commonly used nowadays are (spectroscopic) flames and electrothermal (graphite tube) atomizers.

2.7.1.5 Flame Atomizers.

The oldest and most commonly used atomizers in AAS are flames, principally the air-acetylene flame, with a temperature of about 2300^0C and the nitrous oxide (N_20) -acetylene flame, with a temperature of about 2700^0C. The latter flame, in addition, offers a more reducing environment, being ideally suited for analytes with high affinity to oxygen. Liquid or dissolved samples are typically used with flame atomizers. The sample solution is aspirated by a pneumatic nebulizer, transformed into an aerosol, which is introduced into a spray chamber, where it is mixed with the flame gases and conditioned in a way that only the finest aerosol droplets (< 10 m) enter the flame. However, only about 5% of the aspirated sample solution reaches the flame, but it also Guarantee a relatively high freedom from interference with a temperature of about 2700^0C. On top of the spray chamber is a burner head that produces a flame that is laterally long (usually 5-10cm) and only a few mm deep. The radiation beam passes through this flame at its longest axis, and the flame gas flow-rates may be adjusted to produce the highest concentration of free atoms. The burner height may also be adjusted so that the radiation beam passes through the zone of highest atom cloud density in the flame, resulting in the highest sensitivity.

The processes in a flame include the following stages:-

i. **Desolvation** (drying)- the solvent is evaporated and the dry sample nanoparticles remain
ii. **Vaporization** (transfer to the gaseous phase)- the solid particles are converted into gaseous molecules.
iii. **Atomization**-the molecules are dissociated into free atoms;
iv. **Ionization**- depending on the ionization potential of the analyte atoms and the energy available in a particular flame, atoms might be in part converted to gaseous ions.

Each of these stages includes the risk of interference. In case the degree of phase transfer is different for the analyte in the calibration standard and in the sample. Ionization is generally undesirable, as it reduces the number of atoms that is available for measurement, i.e., the sensitivity. In flame AAS a steady-state signal is generated during the time period when the

sample is aspirated. This technique is typically used for determinations in the mg L-1 range, and may be extended down to a few mg L-1 for some elements. If light of just the right wavelength impinges on a free, ground state atom, the atom may absorb the light and it enters an excited state in a process known as atomic absorption. Atomic absorption measures the amount of light at the resonant wavelength which is absorbed as it passes through a cloud of atoms. As the number of atoms in the light path increases, the amount of light absorbed increases in a predictable way. By measuring the amount of light absorbed, a quantitative determination of the amount of analyte element present can be made. The use of special light sources and careful selection of wavelength allow the specific quantitative determination of individual elements in the presence of others. The atom cloud required for atomic absorption measurements is produced by supplying enough thermal energy to the sample to dissociate the chemical compounds into free atoms. Aspirating a solution of the sample into a flame aligned in the light beam serves this purpose. Under the proper flame conditions, most of the atoms will remain in the ground state form and are capable of absorbing light at the analytical wavelength from a source lamp. The ease and speed at which precise and accurate determinations can be made with this technique have made atomic absorption one of the most popular methods for the determination of metals.

2.7.2 Gross Alpha and Beta Measurement

The growing need to quantitate alpha emitting radionuclides in the environment and in nuclear fuel processing and disposal has resulted in dramatic increases in gross alpha/gross beta measurements. In fact, gross beta counting has become the most widely used method of monitoring for the presence of radioactivity. Much of the interest is due to the need to address safety, regulatory compliance, and disposal issues for both alpha and beta radionuclides. The potential for litigation has also heightened interest in screening samples for gross alpha/gross beta activities (Jim Floeckher, 2008).

Traditionally, gas flow proportional counting has been used for making gross alpha/gross beta measurements. In this device, a sample is inserted into the chamber of a proportional counter.

Any emitted radiation causes ionization of the gas in the counter that is electronically amplified and counted.

a) Thin- windows proportional counter

This counter can detect alpha and low energy radioactivity, the counter is of very thin shielding, of plastic film less than 250mgcm^{-2} and tube of about 5cm in diameter, and/or anti coincident guard circuitry. It is about one-half as sensitive as internal proportional counter due to poor geometry; and absorbing losses (Jarette, 1946). The counter is less affected by contamination from loose residues, and poor electrical conductance, because the sample is outside (Jarette, 1946), unlike the internal proportional counter.

b) Internal proportional Counter

Internal proportional counters are suitable for determining alpha activity, at the alpha operating plateau and alpha plus beta-operating plateau. The alpha or beta activity, or both, can emanate from a single or several radionuclides. The internal proportional counter accepts counting pans within the counting chambers and at the beta operating voltages records all alpha all beta and some gamma radiations emitted in to the gas (Arnold, *et al*., 1992). Theoretically half of the radiation is emitted in the direction of the counting pan. Some of the beta radiation, but only 1% to 2% of the alpha radiation, is backscattered into the counting gas by sample solids, the counting pans or walls of the counting chamber. The percentage of beta radiation detected may be around 60% while that of alpha is 50% for substantially weightless sample (Arnold, *et al*., 1992).

2.7.3 Problems with counting instrument

The different of counting instrument have different modes of operation and each has problems associated with it. A typical problem of most counters is the background or instrument counting rate, usually due to cosmic radiation contaminations in instrument parts and the counting room construction materials, and due to nearness of radioactivity source (Onoja, 2004).

2.8 Review of Preview work

Some works had been done in the field of radioactivity in water in different parts of the world including Nigeria in the past. Nwankwo, *L.I* Akoshile., (2005), studied external background ionizing radiation in the Asa Dam industrial Layout of Ilorin in Kwara state. The study has revealed that the external background ionizing radiation is averagely 0.0134mR/hr with a deviation of about 22% which is relatively higher than the standard background radiation of 0.011mR/hr. this result suggests the possibility of the presence of radionuclides sources in the environment.

Agalga, *et al*., (2003), measure the concentrations of natural radionuclide ^{238}U, ^{232}Th and ^{40}K in the sediment of the Tono irrigation dam, Navrongo, Ghana, using gamma spectrometry. From the study, the concentrations of natural radionuclides ^{238}U, ^{232}Th and ^{40}K ranged from 5.19Bq/L to 8.66Bq/L with an average of 7.31Bq/L for ^{238}U, 5.83Bq/kg to 8.16Bq/L with an average of 6.91Bq/L for ^{232}Th. And 218.73Bq/kg to 453.25Bq/kg with an average of 379.94Bq/kg for ^{40}K. Ozturk, *et al*., (2009), determined the heavy metals in fish, water and sediment of Asar Dam Lake in turkey. The obtained results showed that the average values of Fe in water samples were higher than the respective reference values for fresh water. The analysis of heavy metals in sediment indicated that among the six heavy metals tested; Fe was maximally accumulated, followed by Ni, Cu, Cr, Pb and Cd. In the fish samples, cadmium, chromium, nickel and lead concentrations exceeded the tolerable values provided by international institutions.

Khatun, *et al*., (2003), carried out assessment of natural radioactivity and hazard in soil samples of Rajbari district of Bangladesh, HPGe detector couple with MCA and associate electronics. The results are also compared with the literature values reported for other regions of the world and found that the soil of the study area are not hazards by the radiation and does not pose any harmful effect to the environment. Radioactivity measurement in river sediments and aquatic organisms of Kanyakumari district were carried out by Eugin, *et, al.,* (2012) using alpha

scintillation counter and low beta counter. The results obtained revealed no significant radiological threat to the environment.

Mohsen, *et al*., (2008) carried out the assessment of natural radioactivity in water and sediments from Amang (Tin Tailing) processing ponds using Gamma Spectroscopy to determine the concentrations of Uranium- 238 and thorium- 243 concentration in the environment of Amang processing .The results showed that the concentration of these radionuclides were higher than the background indicating that Amang processing activity has enhanced the natural radionuclides contents in water and sediments.

Nwogu (2001) determined the concentration of trace elements in commercial water supplies. Analysis of fadama water, soil and vegetation for heavy metals was carried out by Odogu (2001), Onoja (2004) determined the gross alpha and gross beta radioactivity in well water in Zaria. This work gave the concentration of alpha and beta radiations in the sampled water but was limited to well water.

Kucukomeroglu, *et al*., (2010) determined the natural gross alpha and gross beta activities in firtina river in eastern black sea region of Turkey. The result obtained showed that natural gross alpha and gross beta activity concentrations in water samples ranged from 12.43.4mBq/L to 66.29.2mBq/L and from 27.9mBq to 133.34.1mBq/L, respectively. The mean activity concentrations were 32 .63.8mBq/1 for gross alpha and 69.94.4mBq/L for gross beta for which the activity concentration did not exceed the recommended levels by WHO and. Cothern and Lappenbush (1990) measured uranium in ground and surface water in the USA and reported that many of the community drinking water supplies exceeded a uranium concentration of 370 and 185Bq L^{-1} limits by USAEPA and WHO respectively.

Avwiri, *et al*. (2007) surveyed the radionuclide concentration of soil, sediments and water in Aba River, Nigeria and reported that the results obtained for the observed radionuclide isotopes were

below international standard and concluded that the river has no radiological burden on the populace.

Juliet (2006) also measured the gross alpha and beta radioactivity in river Kaduna and reported a mean value of 0.117±0.002 and 0.349±0.006BqL^{-1} for alpha and beta activity, respectively. The overall results in the two separate research work show that both alpha and beta activities are below the WHO practical screening level of radioactivity in drinking water.

Arkin, *et al*., (2006) measured the gross alpha and beta radioactivity in the surface air of Tehran Nuclear Research Centre in Iran. The gross alpha and beta activity concentration was found to be ranged from 71.04- 121.20 and 174.91- 305.46 respectively.

Dogro, *et al*., (2008) measured the gross alpha and beta radioactivity in the ground water of van in Turkey. The gross alpha and beta concentration was found to range from 630-782 and 21-816Bqm^{-3}.

Tajudeen, (2006) measured the gross alpha and beta activity concentration in well water from Gwammaja in Kano state of Nigeria. The result indicated very low and beta activity concentrations with a range of 2.06- 11.48Bqm^{-3} for alpha and 2.00- 17.75 for beta.

Avwiri and Agbalagba (2007) carried out survey of gross alpha and beta activity in Okpare creek Delta state Nigeria. The results show that the average alpha activities in the farming, residential/commercial and industrial zones are 1.01290.097, 4.2610.109 and 10.2960.489Bq/L respectively. The average beta activities in the three zones are 0.1290.100, 0.5230.003 and 0.7930.0110Bq/L respectively. The overall result showed that the alpha and beta activities in the three zones are far below the practical screening level for both alpha and beta.

Natural radionuclides level have also studied in surface soil in jen-Ekiti (ajayi, *et al*.,1995), in soil and water around cement company in Ewekoro by Jibiri, *et al*., (1991) in all these studies a non- significant level of radioactivity was observed.

Naveedullah, *et al*., (2013) determined the concentrations and human health risk of selected heavy metals in surface of the Siling reservoir watershed in Zhejiang Province, China and it was found out that the heavy metals were within permissible limits.

Tslalom Haileslassie and Kiflom Gebremedhin (2010) conducted an assessment of heavy metals contamination in ground water in Ethiopia, and they were concluded that the geochemical study of the metals reveals that Pb and Cd have values above the recommended standards.

CHAPTER THREE
MATERIALS AND METHODOLOGY

3.0 Location of the research Area

The study areas are within the co-ordinates 12^0 59' 0″ north, 7^0 45'0″ east Ajiwa, Katsina state Nigeria.

3.1 Samples collection

A survey of gross alpha and beta and sediment radioactivity will be based on the accepted guideline and adopted by the international commission on Radiological protection (ICRP), the National committee on Radiological protection and measurements (NCRP), (IAEA).

Samples of sediments and water will be collected upstream and downstream randomly from 12 reference points for the analysis. The sediments will be packed into 2ml bottles at a depth of approximately 0.6m to 0.8m and then treated with 2 drops of nitric acid to reduce the PH and minimize precipitation on container wall.

All the samples will be placed in to a polyethylene bags and transport to the Center for Energy Research and Training, Zaria for further preparation, Atomic Absorption Spectra, and analysis in accordance with IAEA and ISO method.

3.2 Sample preparation for Atomic Absorption spectrometry

Sediment samples were air-dried at ambient laboratory temperature. The samples were grounded to pass through 2mm sieve and stored for chemical analysis. With the aid of weighting bottle, 1 gram of each sample was obtained using high sensitive chemical balance. This was placed in a Teflon beaker and transferred to a fume-cupboard for digestion. The digestion was carried out using concentrated nitric (10ml) and concentration per chloric (5ml) acid in the ratio 2:1 and the oven was maintained at 200^0C. After one hour, the mixture was allowed to cool before leaching the residue with $5cm^3$ of 20% HNO_3. Digested samples were then filtered and made up to 100ml with deionize water. A blank determination was treated in the Atomic Absorption Spectrometry

but without sample. Solution of samples were then taken and aspirated in to Atomic Absorption Spectrometer for the analysis of metals. Blank determination was also described above except for the omission of the sample. A calibration graph was plotted for each element using measured absorbance and the corresponding concentration. The calibration curve was used to determine the concentration of the metals.

3.3 Sample Preparation for Gross Alpha and Beta Analysis

The preparation of the samples, for gross alpha and beta counting is going to be in the following way: calculated volume of each of the samples is going to be measured into a beaker about 50ml 50^0 c to 60^0 c, down to 20ml the volume is going to be transfer in to a clean dry evaporating dish for continuous evaporation till required residues are obtained. Few drops of vinyl Acetate is going to be added to the sample during evaporation to act as a binder to prevent the precipitation of the source of the radionuclides in the sample. The residue will be transferred into clean, dry and previously weighted planchet. The planchet and residue are going to be weighted and the difference between the mass of the empty planchet and of planchet and the residue will gives the mass of the residue. The residue will be uniformly spread on the planchet by dropping a few drops of ethanol. The residue will then be allowed to dry and then covered with film for gross counting.

3.4 Atomic Absorption Spectrophotometry

3.4.1 Technique

Atomic absorption spectrophotometry (AAS) is an analytical technique that measures the concentrations of elements. It is commonly used for determining the concentration of a particular metal element within a sample. AAS can be used to analyse for the concentration of over 62 different metals in a solution (Wikipedia). AAS determines the presence of metals in liquid samples. Metals include Fe, Cu, Al, Pb, Ca, Zn, Cd and many more. Atomic absorption is so sensitive that it can measure down to parts per billion of a gram (μgdm-3) in a sample. Typical concentrations range in the low mg/L range. In their element form, metals will absorb ultraviolet

light and get excited. Each metal has a characteristic wavelength that will be absorbed. The AAS instrument looks for a particular metal by focusing a beam of UV light at a specific wavelength through a flame and into a detector. The sample of interest is aspirated into the flame. If that metal is present in the sample, it will absorb some of the light, thus reducing its intensity. The instrument measures the change in intensity. A computer data system covert the change in intensity into an absorbance (Steffason et al.,2007).Atomic absorption spectrometry was first used as an analytical technique. The Underlying principles of atomic absorption spectrometry were established in the second half of the 19th century by Robert Wilhelm Bunsen and Gustav Robert Kirchhoff, both professors at the University of Heidelberg, Germany. The modern form of AAS was largely developed during the 1950s by a team of Australian Chemists, led by Allan Walsh at the Commonwealth Scientific and Industrial Research Organization, Division of Chemical physics, in Melbourne, Australia (CSIRD) (Walsh,1955).

3.4.2 Principle

Atomic absorption spectrophotometry (AAS) makes use of absorption spectrometry to assess the concentration of an analyte in a sample. It requires standard solutions to establish the relation between the measured absorbance and the analyte concentration, and relies therefore on Beer-Lambert law. In short, the electrons of the atoms in the atomizer can be promoted to higher orbital (excited state) for a short period of time (nanosecond) by absorbing a defined quantity of energy (quantized energy or radiation of a given wavelength). This amount of energy, characterized by the wavelength, is specific to a particular electron transition in the particular element. In general, each wavelength corresponds to only one element, while the width of an absorption line is only of the order of a few picometers (pm), which gives the technique its elemental specificity. The radiation flux without a sample and with a sample in the atomizer is measured using a detector, and the ratio between the two values(the absorbance) is converted to analyte concentration or mass using Beer-Lambert Law (Wikipedia).

3.4.3 Instrumentation

In order to analyse a sample for its atomic constituents, it has to be atomized. The atomizers most commonly used nowadays are flames and electro thermal (graphite tube) atomizers. The atoms should then be irradiated by optical radiation, and the radiation source could be an element - specific line radiation source or a continuum radiation source. The radiation then passes through a monochromator in order to separate the element-specific radiation from any other radiation emitted by the radiation source, which is finally measured by a detector.

3.4.4 Atomizer

Although other atomizers, such as heated quartz tubes, might be used for special purposes, the atomizers most commonly used nowadays are (spectroscopic) flames and electrothermal (graphite tube) atomizers.

3.4.5 Flame Atomizers.

The oldest and most commonly used atomizers in AAS are flames, principally the air-acetylene flame, with a temperature of about 2300^0C and the nitrous oxide (N_20) -acetylene flame, with a temperature of about 2700^0C. The latter flame, in addition, offers a more reducing environment, being ideally suited for analytes with high affinity to oxygen. Liquid or dissolved samples are typically used with flame atomizers. The sample solution is aspirated by a pneumatic nebulizer, transformed into an aerosol, which is introduced into a spray chamber, where it is mixed with the flame gases and conditioned in a way that only the finest aerosol droplets (< 10 m) enter the flame. However, only about 5% of the aspirated sample solution reaches the flame, but it also Guarantee a relatively high freedom from interference with a temperature of about 2700^0C. On top of the spray chamber is a burner head that produces a flame that is laterally long (usually 5-10cm) and only a few mm deep. The radiation beam passes through this flame at its longest axis, and the flame gas flow-rates may be adjusted to produce the highest concentration of free atoms. The burner height may also be adjusted so that the radiation beam passes through the zone of highest atom cloud density in the flame, resulting in the highest sensitivity.

The processes in a flame include the following stages:-

i. **Desolvation** (drying)- the solvent is evaporated and the dry sample nanoparticles remain
ii. **Vaporization** (transfer to the gaseous phase)- the solid particles are converted into gaseous molecules.
iii. **Atomization**-the molecules are dissociated into free atoms;
iv. **(iv)Ionization**- depending on the ionization potential of the analyte atoms and the energy available in a particular flame, atoms might be in part converted to gaseous ions. Each of these stages includes the risk of interference.

In case the degree of phase transfer is different for the analyte in the calibration standard and in the sample. Ionization is generally undesirable, as it reduces the number of atoms that is available for measurement, i.e., the sensitivity. In flame AAS a steady-state signal is generated during the time period when the sample is aspirated. This technique is typically used for determinations in the mg L-1 range, and may be extended down to a few mg L-1 for some elements. If light of just the right wavelength impinges on a free, ground state atom, the atom may absorb the light and it enters an excited state in a process known as atomic absorption. Atomic absorption measures the amount of light at the resonant wavelength which is absorbed as it passes through a cloud of atoms. As the number of atoms in the light path increases, the amount of light absorbed increases in a predictable way. By measuring the amount of light absorbed, a quantitative determination of the amount of analyte element present can be made.

The use of special light sources and careful selection of wavelength allow the specific quantitative determination of individual elements in the presence of others. The atom cloud required for atomic absorption measurements is produced by supplying enough thermal energy to the sample to dissociate the chemical compounds into free atoms. Aspirating a solution of the sample into a flame aligned in the light beam serves this purpose. Under the proper flame conditions, most of the atoms will remain in the ground state form and are capable of absorbing light at the analytical wavelength from a source lamp. The ease and speed at which precise and

accurate determinations can be made with this technique have made atomic absorption one of the most popular methods for the determination of metals.

3.5 Analytical Techniques

3.5.1 Gross Alpha and Beta

3.5.1.1 Detector Characterization

For the detector to be put in to use certain measurement were made. These include the background measurement and plateau test.

3.5.1.2 Background Counting

In compliance with ISO standard, clean empty planchet were counted to determine the background radioactivity of the environment. The operational high voltage 1600V for alpha and 1700V for beta and 1750V alpha and beta simultaneous was set and the detector was allowed to run 900s per cycle for alpha background and twenty five cycles of 1800s per cycle for beta. Thr background rates were determined in count per minute.

3.5.1.3 Plateau Test

This test is design to verify the suitable voltage to be used for the different counting modes and also to determine the efficiency and detection limits of the detector channel. The test was performed with manufacturers standard sources using Pu-232 and Sr-90 standard respectively for alpha and beta modes. For each mode the voltage varied from the minimum to maximum specified voltage at intervals of 50V. The channel efficiencies were obtained for the three modes of counting thus, the alpha only (1100V), beta only (1550V) and alpha and beta simultaneous (1650V) mode.

3.5.1.4 Counting

The counting equipment is automated. The procedure involves entering the present time, number of cycles and the counting (operational) voltage. Also the counter characteristics (channel efficiency and background count rate), volume of the sample used and sample efficiency were entered. The sample efficiency was calculated as:

$$Sample\ Efficiency = \frac{Mt}{0.1A} \text{ X } 100\% \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \text{ (3.1)}$$

Where M_t s the mass of the residue in the planchet got from sample preparation and 0.1A (mg) is the expected mass of the residue in the planchet.

3.5.1.5 Gross Alpha Counting

In the gross alpha counting, the operational high voltage was set at 1600V and the samples were count for 3 cycles of 3600s per cycle. The displayed results were presented as raw count: count rate (count/min), activity and standard deviation. The data were acquired for alpha mode only and the alpha count rate as well as the alpha activity was calculated using the formula; (IOS 9696:1991).

$$Rate\ \alpha\ (count/sec) = \frac{Raw\ \alpha\ count\ x\ 60}{Count\ time\ (se)} \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \text{ (3.2)}$$

$$Activity\ \alpha = \frac{Raw\ \alpha\ x\ Bgd\ \alpha\ x\ \alpha\ unit\ coefficient}{Channel\ \alpha\ efficiency\ x\ sample\ efficiency\ x\ sample\ vol.} \ldots\ldots\ldots \text{ (3.3)}$$

Where unit coefficient is the multiplication coefficient making it possible to obtain the result expressed in units used for the operation (Pci/L, efficiency in %).

3.5.1.5 Gross Beta Counting

The operational high voltage for gross beta counting was set at 1700V and samples were also counted for 3 cycles of 3600s per cycle in beta mode only. The count rate and activity were calculated using the formula; (IOS 9696:1991):

$$Rate\ \beta\ (count/sec) = \frac{Raw\ \beta\ count\ x\ 60}{Count\ time\ (se)} \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \text{ (3.4)}$$

$$Activity\ \beta = \frac{Raw\ \beta\ x\ Bgd\ \beta\ x\ \beta\ unit\ coefficient}{Channel\ \beta\ efficiency\ x\ sample\ efficiency\ x\ sample\ vol.} \ldots\ldots\ldots \text{ (3.5)}$$

3.6 Data and Statistical Analysis

Analytical data were processed using Excel and SPSS software. Basic statistics such as min., max., mean, and relative standard deviation were computed.

3.7 Human Health Risk Assessment Indices

Risk assessment is defined as the processes of estimating the probability of occurrence of any given probable magnitude of adverse health effects over a specified time period and is a function of hazard and exposure. Health risk assessment of the metals in water of Ajiwa Dam was examined via ingestion and dermal route to the recipients based on the USEPA risk assessment methodology (USEPA, 2004). The exposure doses are calculated using equations (3.6) and (3.7) adopted from the US Environmental Protection Agency (USEPA, 1989).

Table 3.1 Parameters for estimating exposure assessment of metals in water samples used in the present study (Wu B., 2009).

Exposure Factors	Unit	Values
Concentration of metals in water (C_{water})	µg/L	-
Water ingestion rate (IR)	L/day	2.2
Exposure frequency (EF)	Days/year	360
Exposure duration (ED)	Year	30
Average body weight (BW)	kg	70
Averaging time (AT)	Days	10,950
Exposed skin area (SA)	Cm^3	28,000
Exposure time (ET)	h/day	0.6
Dermal permeability coefficient (K_p)	-	-
Zn	Cm/h	0.0006
Cu	Cm/h	0.001
Cd	Cm/h	0.001
Cr	Cm/h	0.002
Pb	Cm/h	0.004

$$Exp_{ing} = \frac{Cwater \times IR \times EF \times ED}{BW \times AT} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (3.6)$$

$$Exp_{derm} = \frac{Cwater \times SA \times Kp \times ET \times EF \times ED \times CF}{BW \times AT} \quad \ldots\ldots\ldots\ldots\ldots\ldots (3.7)$$

Where Exp_{ing} is the exposure dose through ingestion of water (µg/kg); Exp_{derm} is the exposure dose through dermal absorption (µg/kg). The parameters for estimating human health risk assessment through different pathways are listed in table 3.1.

The human health risk assessment for metals and non-carcinogens were performed by comparison of the calculated contaminant for each exposure route (ingestion, dermal) with reference the reference dose (RfD) in order to develop hazard quotient (HQ), using equation (3.8) [USEPA, 1989]. HQ>1, there might be concern for no-carcinogenic effects.

$$HQ_{\frac{ing}{derm}} = \frac{Exp_{\frac{ing}{derm}}}{RfD_{\frac{ing}{derm}}} \quad \text{…….. ….. (3.8)}$$

Where $HQ_{ing/derm}$ is hazard quotient via ingestion or dermal contact (unit less); and $RfD_{ing/derm}$ oral /dermal reference dose (µg/kg) [Iqbal. J, 2012].

Hazard index (HI) was introduced to evaluate the total potential for non-carcinogenic effects posed by more than one pathway, which was the sum of the HQs from all applicable pathways equations (3.9). HQ>1 showed a potential for adverse effect on human health (USEPA, 1989).

$$HI_{\frac{ing}{derm}} = \sum_{i=0}^{n} HQ_{\frac{ing}{derm}} \quad \text{……. (3.9)}$$

Where $HI_{ing/derm}$ is the hazard index via ingestion or dermal contact (unit less)

Chronic daily intake (CDI) was calculated using equation (3.10) [Muhammad S., 2011].

$$CDI = C \times \frac{DI}{BW} \quad \text{……. (3.10)}$$

Where C, DI, and BW represent the concentration of heavy metals in water (µg/L), average daily intake rate (2.2 L/day), body Wight (70kg), respectively.

Carcinogenic risk (CR) were estimated using equation (3.11) and the detailed calculating process was followed by (Wu B., 2009, Iqbal J., 2012 and De Miguel E., 2007). The incremental probability of an individual developing cancer over life time as a result of exposure to a potential

carcinogen. The range of carcinogenic risk acceptable by (USEFA, 2010) was 1.0E-06 to 1.0E-04.

$$CR_{ing} = \frac{Exp_{ing}}{SF_{ing}} \quad \text{..... (3.11)}$$

Where$CR_{i\ ng}$ is the carcinogenic risk via ingestion route (unit less) SF_{ing} is the carcinogenic slope factor, ingestion (µg/g). In order to show the life time carcinogenic risk to the local people, CR_{ing} values were calculated for Cd, and Cr. The SF_{ing} values for Cd and Cr are 6.1E+03 and 5.0E+02 respectively (USEPA, 1988, Viera C., 2011 and Yuf. C., 2010). SF_{ing} values for the rest of the metals were not available.

Determination of the Annual Effective dose equivalent:

The following equation was used to determine the yearly Effective Dose Equivalent (USEPA, 1996).

$$DR_w = A_w \text{ x } IR_w \text{ x } ID_f$$

Where DR_w is the Effective Dose Equivalent (Sv), A_w is the activity (Bq/L), IR_w is the intake of water for a person in a year (730 liters) and ID_f the ingestion effective dose equivalent factor (3.58E-07 Sv/y).

Therefore in order to calculate the annual effective dose, it was considered that alpha gross activity is due to Ra-226 and Po-210 and beta gross activity due to Ra-228 and Pb-210 (S. Turhan et., al, 2013 and M. M. Jankovic et., al, 2010), which are emitters with the highest conversion factors CF.

CHAPTER FOUR
RESULT AND DISCUSSION

4.1 Alpha and Beta Activity Concentration in Water Sample

The gross alpha and beta activity for the collected water sample is presented in table 4.1. The alpha activity concentrations have an average value of 1.12E-01Bq/L in Sample D, in sample C the activity concentration have an average value of 5.48E-02Bq/L, the average activity concentration value in sample FR is 7.92E-02Bq/L, while in sample FS the average activity concentration of 5.29E-02Bq/L was obtained. The beta activity concentration have an average value of 9.67E-02Bq/L in sample D, the activity concentrations has an average value of 1.69E-02Bq/L in sample C, the average activity concentration value in sample FR is 3.55E-01Bq/L, while in sample FS the average concentration value of 2.34E-01Bq/L was obtained.

Table-4.1 Alpha and Beta Activity Concentration for Water Samples measured in this work.

Sample ID	Alpha Radioactivity Concentration (Bq/L)	Beta Radioactivity Concentration (Bq/L)
D1	2.12E-01	1.50E-01
D2	8.43E-02	8.24E-02
D3	3.93E-02	5.75E-02
FR1	5.63E-02	1.96E-02
FR2	8.99E-02	8.65E-01
FR3	9.14E-02	1.79E-01
FS1	1.24E-02	8.61E-02
FS2	7.76E-02	5.61E-01
FS3	6.88E-02	5.32E-02
C1	6.05E-03	1.27E-02
C2	1.53E-02	1.93E-03
C3	1.43E-01	3.60E-02

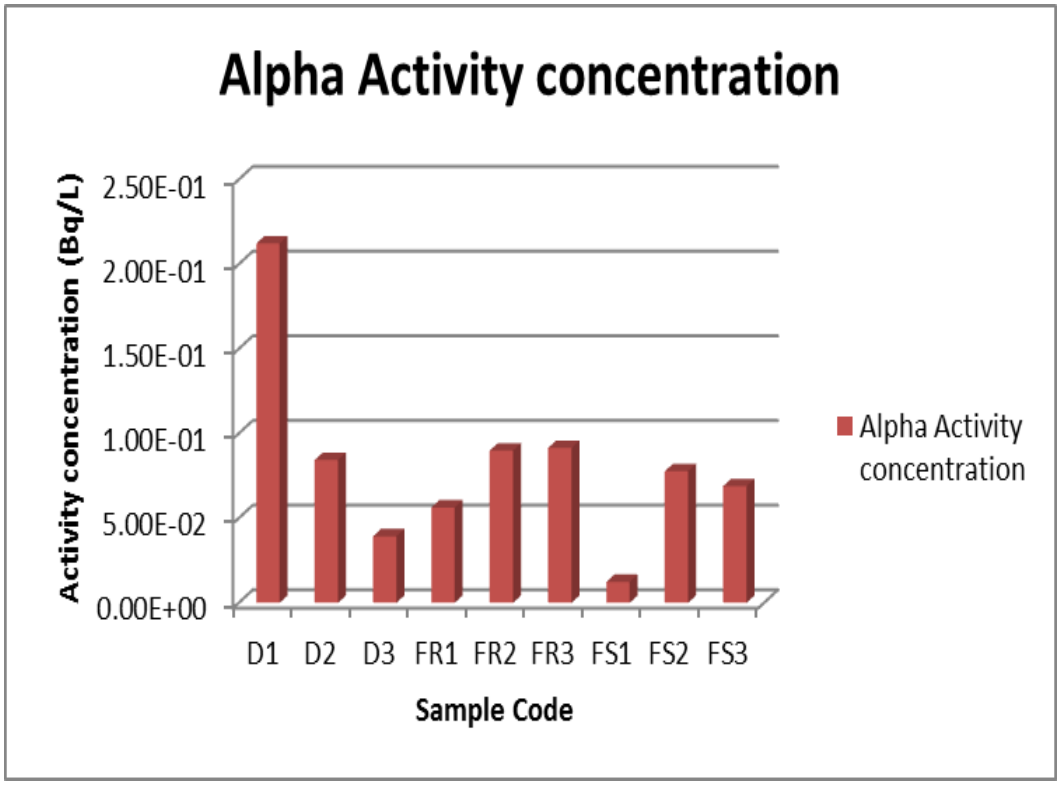


Figure4.1 Alpha Activity across the sampled points

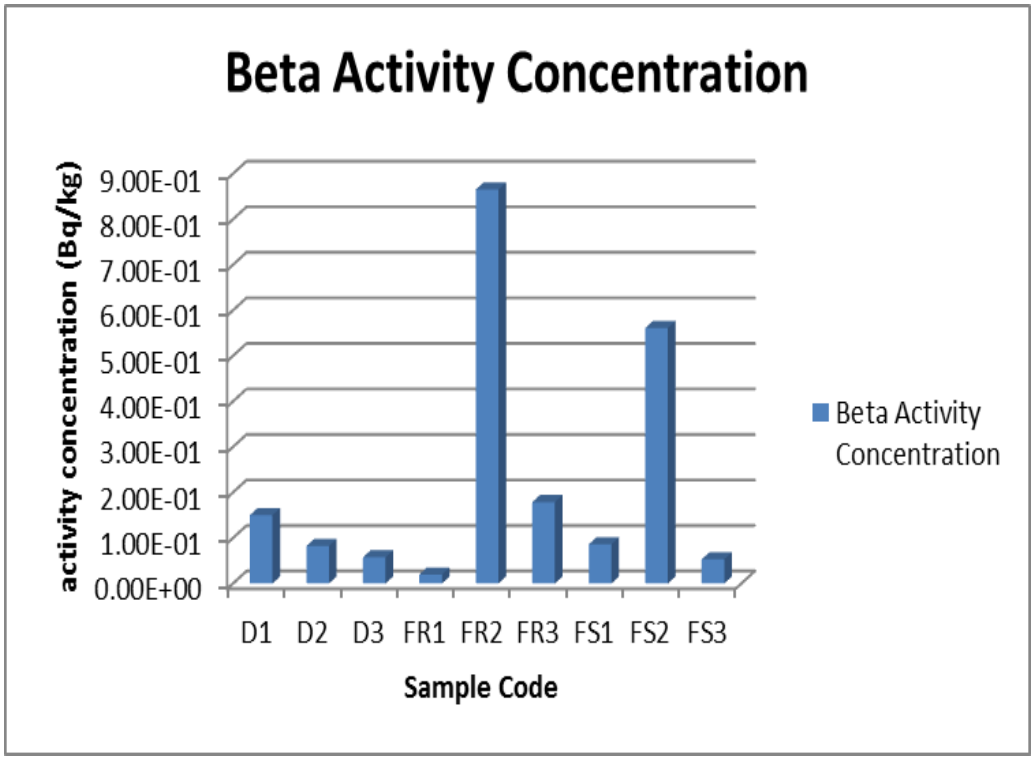


Figure- 4.2 Beta Activity across the sampled points

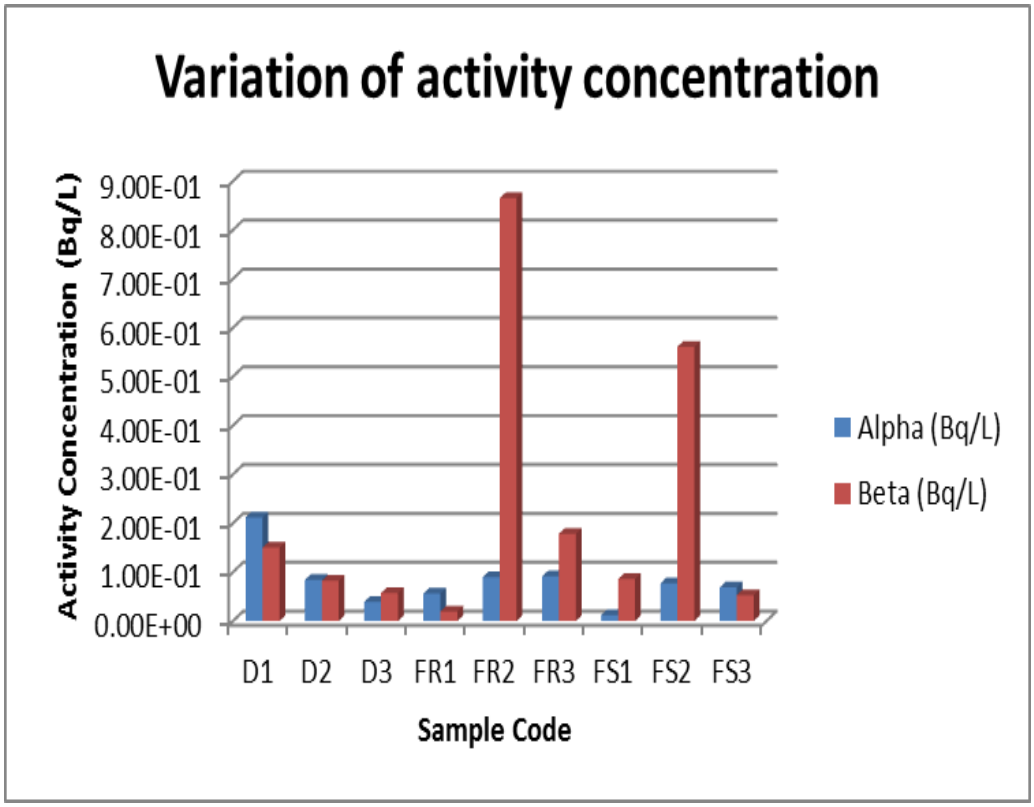


Figure 4.3 Comparison of Alpha and Beta Activity across the sample points

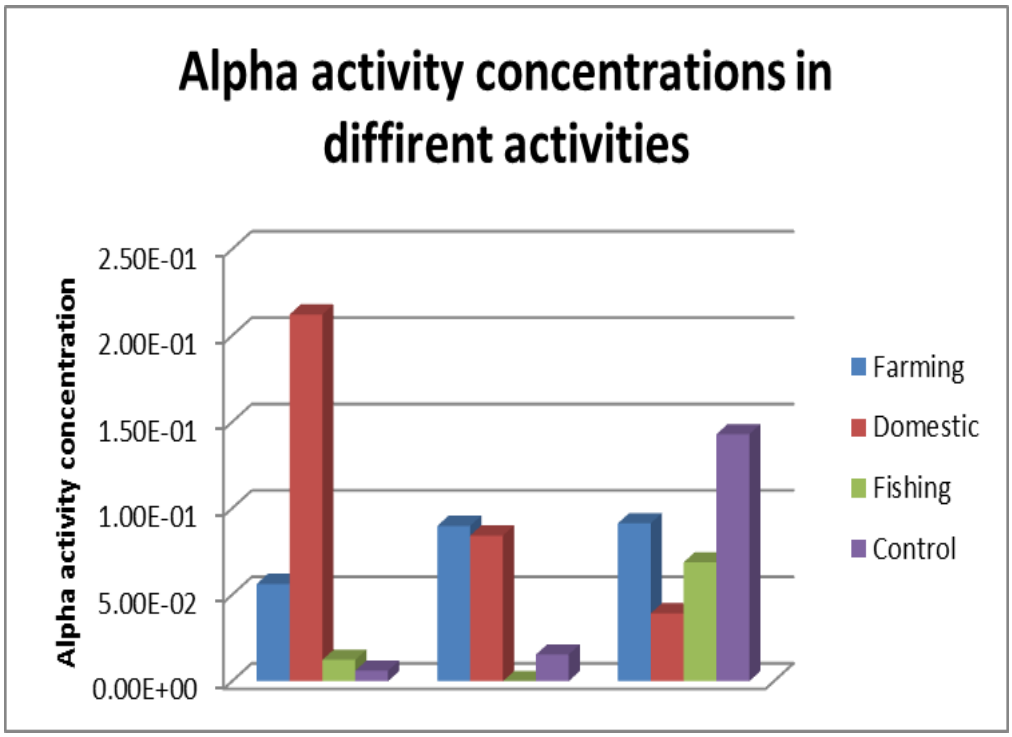


Figure 4.4 Alpha Activity Concentrations in different activities

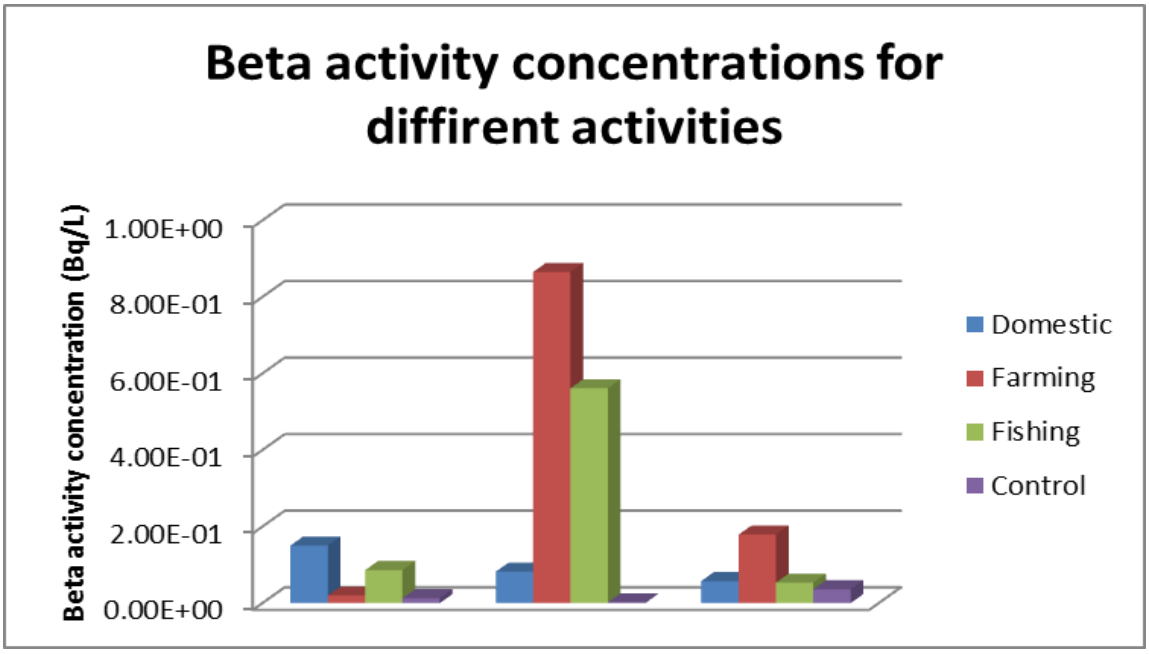


Figure 4.5 Beta Activity Concentrations in different activities

The variation of alpha activity across all the sample points is shown in figure 4.1 D1 recorded the highest value of 2.12E-01Bq/L for alpha followed by FR3 and FR2 with values of 9.14E-02Bq/L and 8.99E-02Bq/L respectively. Other places with high alpha activity are D2, FS2 and FS3 with 8.43E-02Bq/L, 7.76E-02Bq/L and 6.86Bq/L. The remaining places are D2 and FS1 with the values 3.93E-02Bq/L and 1.24E-02Bq/L respectively. These values are all below WHO and USEPA recommended values of 0.5Bq/L and 0.55Bq/L respectively for good water quality. Beta activity on the other hand recorded the highest value of 8.65E-01Bq/L at FR2 followed by FS2 with 5.61E-01Bq/L, FR3 with 1.79E-01Bq/L, D1 with 1.50E-01Bq/L, FS1 with 8.61E-02Bq/L, D2 with 8.24E-02Bq/L, FS3 with 5.32E-02Bq/L, D3 with 5.75E-02Bq/L, FR1 with 1.98E-02Bq/L. these values are all below WHO and USEPA recommended values for good quality drinking water.

When alpha and beta activities across the sampled points are compared (figure 4.3), the result shows that beta recorded high values across most of the sampled points than alpha except at FR1 and FS3 where alpha activates are higher than those of beta. This result therefore suggests that, they are gross beta emitting radionuclides in the assessed area more than gross alpha emitters.

The result also shows that, the values of alpha and beta activities were all below the WHO and USEPA recommended values for good quality drinking water.

4.2 Annual Equivalent Effective Dose Due To Water Ingestion

The specific activities of natural radionuclides from drinking water are mainly due to the presence of naturally occurring radionuclides of both uranium and thorium decay series (M. Forte et. al, 2007). The calculated annual effective doses (µSv/y) for adult population in Ajiwa area due to intake of Po-210, Ra-226, Pb-210 and Ra-228 radionuclides from water are presented in table 4.2 and figure 4.6 – 4.8 respectively. Due to the fact that the CFs for Pb-210 and Ra-228 are equal, it results that the annual effective doses for both radionuclides are the same and were represented in a single figure and one column in table 4.2

Table 4.2 annual Effective Dose equivalents due to Water Ingestion

Sample ID	**AEDE α(Po-210)(µSv/y)**	**AEDE α(Ra-226)(µSv/y)**	**AEDE β(Pb-210/Ra-228)(µSv/y)**
D1	185.887	43.374	75.706
D2	73.829	17.227	41.480
D3	34.401	8.027	28.958
FR1	49.310	11.506	9.883
FR2	78.752	18.376	435.852
FR3	80.075	18.684	90.062
FS1	10.827	2.526	43.389
FS2	68.004	15.868	282.727
FS3	60.278	14.065	26.817

From table 4.2 it can be seen that the annual effective doses due to alpha emitters in the water sample ranges between 10.827-185.887µSv/y for Po-210 and 2.526-43.374 µSv/y for Ra-226. In

the case of beta emitters Po-210/Ra-228, the annual effective doses range from 9.883-435.852 µSv/y.

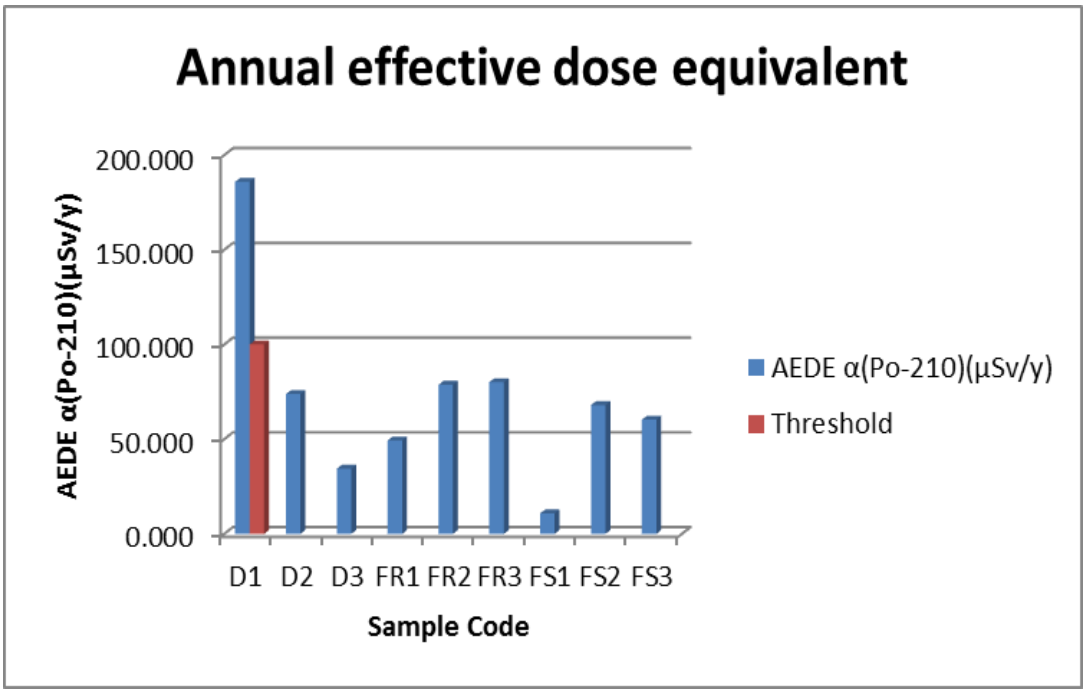


Figure 4.6 Annual Effective Doses due to Intake of Po-210

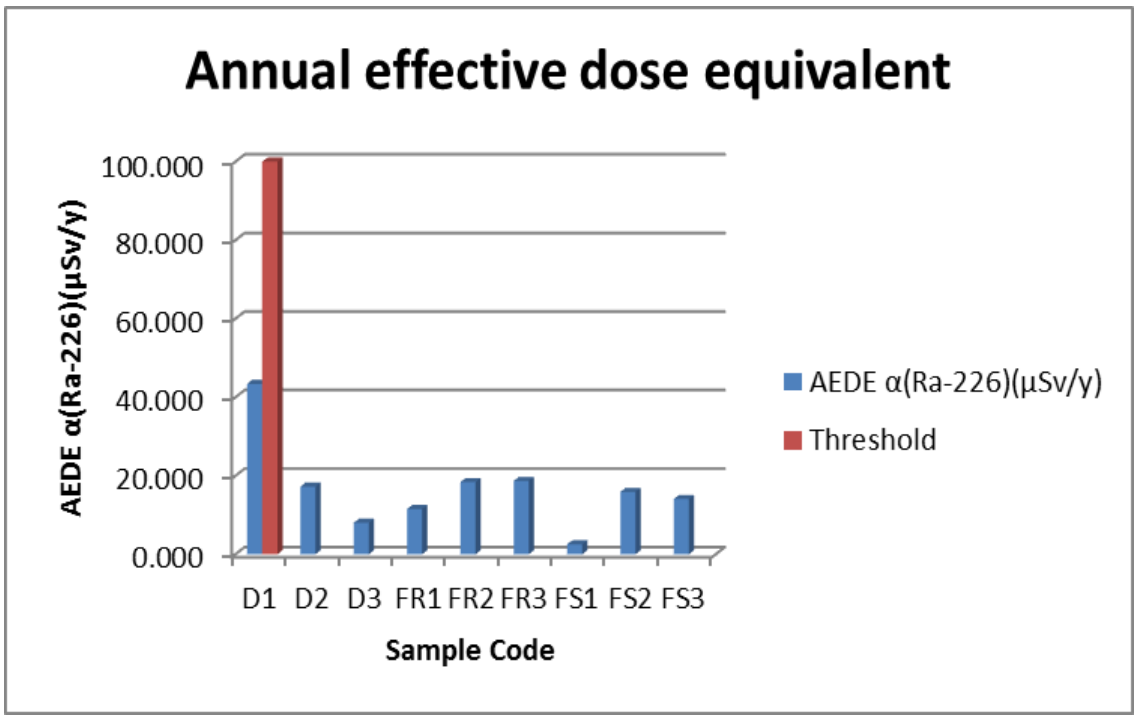


Figure 4.7 Annual Effective Doses due to Intake of Ra-226

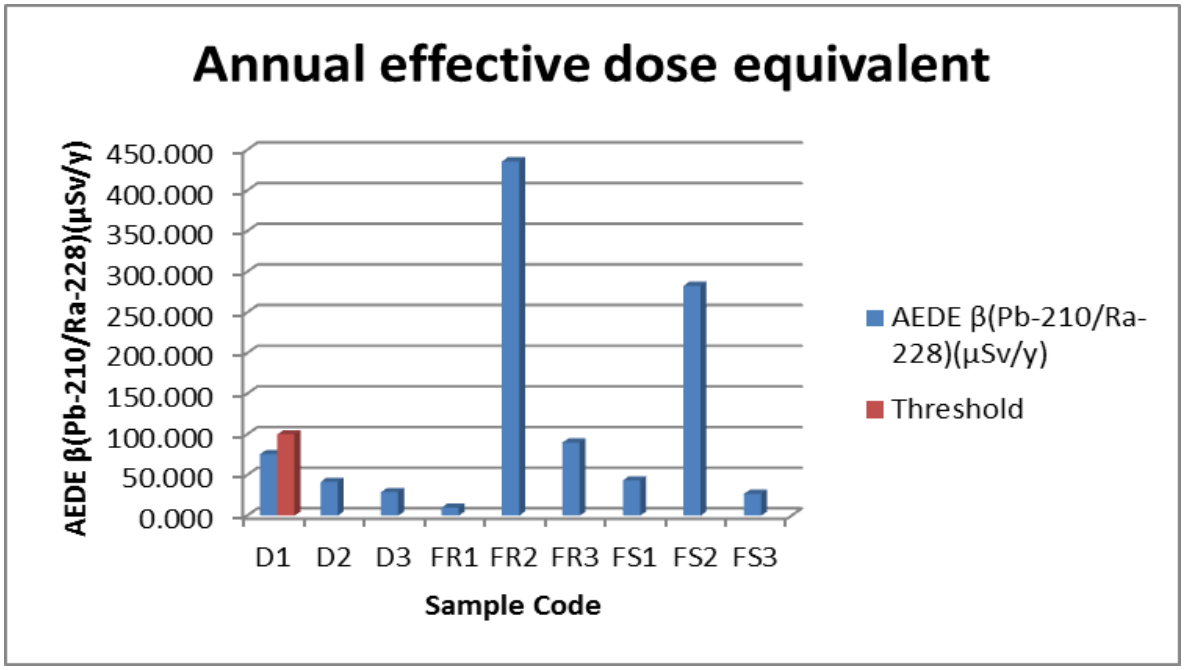


Figure 4.8 Annual Effective Doses due to Intake of Pb-210/Ra-228

In this study the results for annual effective dose due to alpha emitters Po-210 (fig. 4.6) and Ra-226 (fig. 4.7) from the water sampled indicate values below the WHO recommended reference level of 0.1mSv/y for one adult person (WHO, 2011).

With a view to the annual effective doses due to intake of beta emitters Pb-210/Ra-228 from the water samples (fig. 4.8), for 12 samples the calculated values indicated that the highest value of annual effective dose due to Pb-210/Ra-228 intake from the water sampled was found for sample FR2, which obviously had the highest beta activity.

However, as recommended by Directive 2013/51 Euroatom (Council directive, 2013), if gross alpha and beta activity are less 0.1Bq/L and 1.0Bq/L, respectively, it can be considered that the annual effective doses are below the WHO recommended reference level 0.1mSv/y and it does not require radiological investigation unless it is known from other source of information that the specific radionuclides present in water may cause a dose larger than 0.1mSv/y (Euroatom Council Directive, 2013).

4.3 Concentration of the selected metals in the sediment samples

Distribution of the select metals level (Mean±SE) in sediment samples from Ajiwa Dam in comparison to National and International standards are given in table 4.3

Table 4.3 Summary of metals concentration (Mean±SE) in water samples from Ajiwa Dam and comparison with world values and National/International standards (µg/L)

Sites	Zn	Cu	Cr	Cd	Pb	Reference
Ajiwa Dam	5.69E-06	5.25E-07	1.69E-06	7.50E-08	BDL	Present study
Warri River, Nigeria	87.60	47.20	10.60	7.20	0.10	Wogu M.D.,(2011)
Stream River, Ghana	138.00	2.65	0.52	0.04	0.85	Asante. K.A et., al (2007)
Hindon River, India	833.20	4390.20	332.10	24.00	901.20	Suthar, et., al (2009)
Tigris River, Turkey	37.00	165.00	<5	1.37	0.34	Varol M. (2012)
Khanfur Lake, Pakistan	15	9	46	20	221	Iqbal J. (2012)
Odiel River, Spain	24.23	7.60	BDL	0.08	0.21	Olias M. et., al (2004)
Guidelines for Drinking water	3000	2000	50	3	10	WHO, (2006)
Drinking Water Directives (DWD)	100	2000	50	5	10	EC, (1998)
Guidelines for drinking water quality	5000	1300	100	5	15	USEPA , (2009)

The concentrations of variables Zn, Cu, Cr, Cd and Pb were shown on average basis Cr (1.69E-06µg/L) and Zn (5.69E-06 µg/L) showed higher concentration followed by Cu (5.25E-07 µg/L) and Cd (7.50E-08 µg/L). The concentrations of metals recorded in this study were lower than above tabled concentrations both for National and International. However Pb there was no Pb throughout the samples.

The total concentrations of five metals from the sediments of Ajiwa Dam at different points were presented in table 4.4 and figure 4.9 respectively.

Table 4.4 Concentration of metals measured in this work

Sample ID	Cu (ppm)	Zn (ppm)	Cd (ppm)	Cr (ppm)	Pb (ppm)	Cu (µg/L)	Zn (µg/L)	Cd (µg/L)	Cr (µg/L)
D1	0.027	0.118	0.009	0.199	BDL	0.00000027	0.00000118	0.00000009	0.00000199
D2	0.035	0.109	0.01	0.157	BDL	0.00000035	0.00000109	0.0000001	0.00000157
FR1	0.085	1.088	0.013	0.183	BDL	0.00000085	0.00001088	0.00000013	0.00000183
FR2	0.1	1.103	0.002	0.16	BDL	0.000001	0.00001103	0.00000002	0.0000016
FS1	0.043	0.495	0.006	0.158	BDL	0.00000043	0.00000495	0.00000006	0.00000158
FS2	0.024	0.5	0.005	0.16	BDL	0.00000025	0.000005	0.00000005	0.0000016
Average						0.000000525	5.6883E-06	7.5E-08	1.695E-06
Min.						0.00000025	0.00000109	0.00000002	0.00000157
Max.						0.000001	0.00001103	0.00000013	0.00000199

BDL: below detection limit

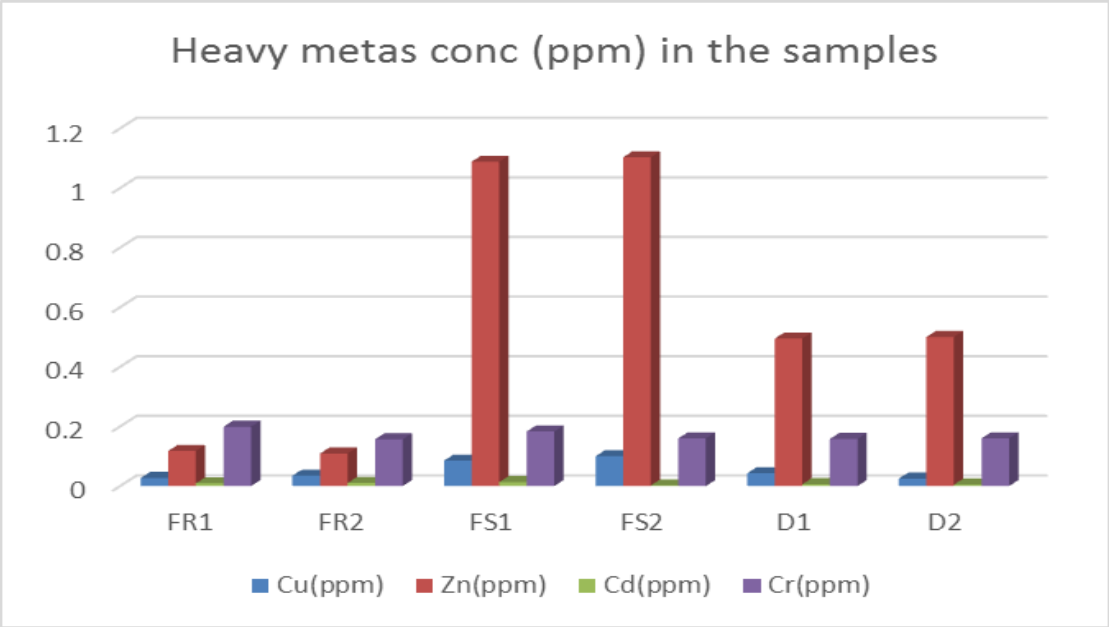


Figure 4.9 Total concentrations of five metals in different points (ppm)

From the figure 4.9 it can see clearly that Zn recorded the highest value in most of the samples followed by Cr and Cu, while Cd showed lowest concentration.

Table 4.5 Correlation of heavy metals measured in this work

	Pb (ppm)	Cu (ppm)	Zn (ppm)	Cd (ppm)	Cr (ppm)
Pb(ppm)	1				
Cu (ppm)		1			
Zn (ppm)	_	**0.909435**	1		
Cd (ppm)	_	-0.12944	-0.18654	1	
Cr (ppm)	_	-0.06291	-0.10657	0.525642	1

There is a strong and positive relationship between Cu and Zn, while there is no any relationship among the rest of the metals.

4.4 Human Health Risk Assessment

Levels of the selected metals in water via ingestion route from Ajiwa Dam are summarized in table 4.6 below:

Table 4.6 Summary of health risk assessment for the selected metals in water samples from Ajiwa Dam through ingestion pathway.

Element	$RfD_{ingestion}$ (µg/kg)	$Exp_{ingestion}$	$HQ_{ingestion}$	CDI	$CR_{ingestion}$
Zn	300	1.9130E-08	6.3750E-11	1.7878E-07	ND
Cu	40	1.7652E-09	6.4131E-11	1.65E-08	ND
Cr	3	5.254E-08	0.001751	5.3271E-08	1.0508E-10
Cd	0.5	2.3249E-09	4.0787E-05	2.3571E-09	3.8112E-13
Pb	1.4	ND	ND	ND	ND
$\sum HI_{ingestion}$			1.792E-03		

ND: not detected

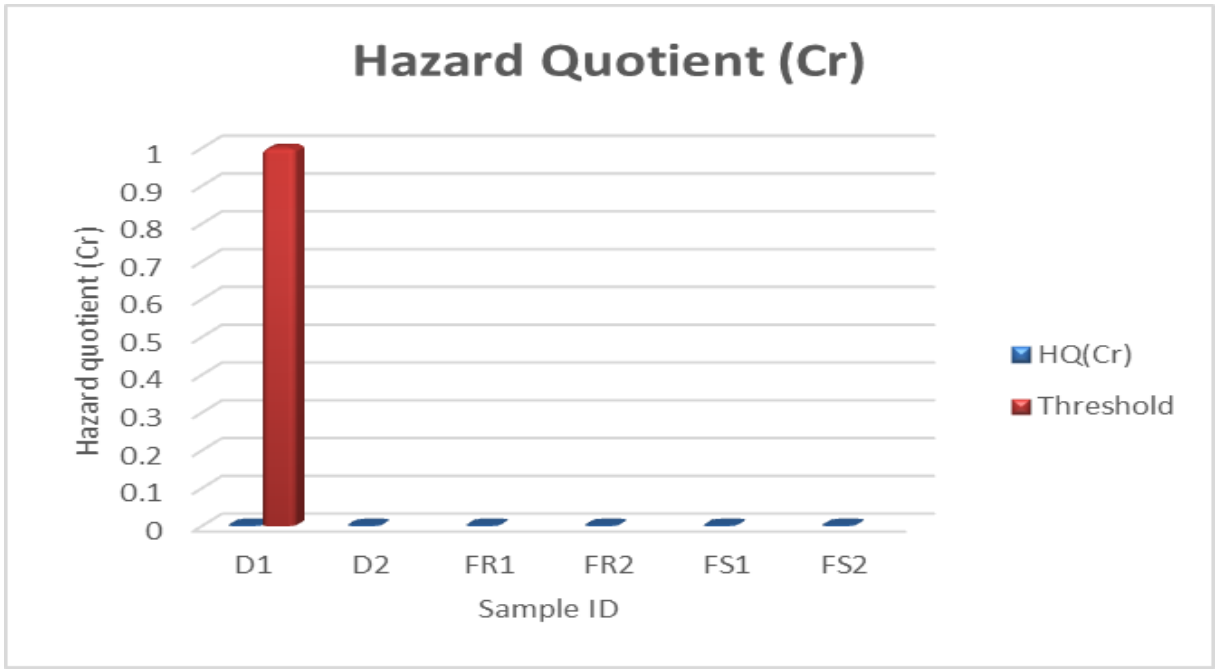


Figure 4.10 Hazard Quotient through ingestion of (Cr)

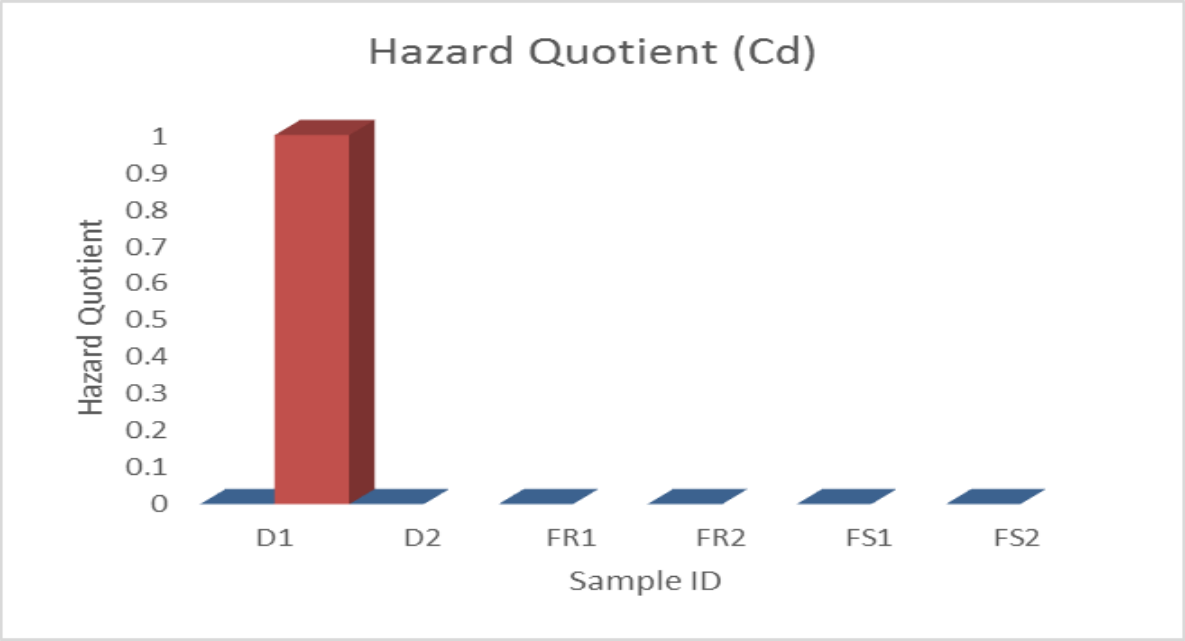


Figure 4.11 Hazard Quotient through ingestion of (Cd)

The levels of Exp_{ing} observed in order: Cr>Cd respectively, while Cu, Pb and Zn were not detected. These results suggest that Cr and Cd were the main contributors to the human environmental area.

In the present study the levels of HQ_{ing} for the metals were smaller than unity (table4.6), which indicates that these metals could pose minimum hazard to local residents (Wu B., et al., 2009). The mean levels of HI_{ing} was found to be 1.792E-03 (table 4.6). This indicates that there was no cumulative potential of adverse health risk in samples via direct ingestion to the inhabitants of the drinking water, farming and fishing sources (Iqbal J. et al., 2012).

The average CDI value for the selected heavy metals (Zn, Cu, Cr and Cd) were 1.7878E-07, 1.65E-08, 5.3271E-08 and 2.3571E-09 respectively. Therefore CDI indices for heavy metals in the study area were found in order: Zn>Cu>Cr>Cd (table 4.6).

CHAPTER FIVE
CONCLUSION AND DISCUSSIONS

5.1 Conclusion

The present study is the radioactivity and elemental studies in water of Ajiwa Dam, and the radiological implication on the adult population which are using this water for farming, fishing and consumption. It is the first study performed in the region regarding the radioactivity studies and will serve as a base in further investigations.

The values of gross alpha and beta activity are below the recommended values in national and European legislative 0.1Bq/L and 1Bq/L, respectively. It is also concluded from this study that, the heavy metals concentrations were within permissible limits. The health risk assessment indices like HQ_{ing} and CDI were found to be less than unity, indicating that only minimal risk may occur via the ingestion route.

In general the overall non-carcinogenic health risk assessment HI_{ing} is less than unity and considered to be safe ($HI_{ing}<1$).

This study provides preliminary information of different heavy metals status in Ajiwa Dam, which can be used for feature drinking water, farming and fishing quality monitoring and planning elsewhere.

5.2 Suggestion and Recommendations

1. There is need to establish the specific activities of natural radionuclides in drinking water from the Dam, especially for the radionuclides with the highest effective dose conversion factors, to be able to certainly state that the annual effective dose due to ingestion of water does not exceed 100µSv/y.
2. The present study suggest that special attention need to be paid to the application of agro-chemicals and anthropogenic activities in the area in order to provide safe water usage to the local people.
3. Further research need to be carried out for more heavy metals in the area.

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