# ASSESSMENT OF GROSS ALPHA AND BETA ACTIVITY IN SURFACE AND UNDERGROUND WATER IN NNEWI NORTH, ANAMBRA STATE

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September, 2018

# ASSESSMENT OF GROSS ALPHA AND BETA ACTIVITY IN SURFACE AND UNDERGROUND WATER IN NNEWI NORTH, ANAMBRA STATE

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A Thesis submitted to the Department of Radiography and Radiological Sciences, Faculty of Health Sciences and Technology, College of Health Sciences, Nnamdi Azikiwe University, Nnewi Campus, in partial fulfillment of the requirements for the award of Master's (M.Sc.) degree in Radiation Protection and Dosimetry.

Supervisor: Dr. CHIEGWU, H. U.

September, 2018

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FOTSO Daniel Ulrich

Date

### **APPROVAL**

This study titled Assessment of Gross Alpha and Beta Activity in Surface and Underground Water in Nnewi North, Anambra State carried out by FOTSO DANIEL ULRICH has been examined by us and found worthy for the award of a Master of Science degree in Radiation Protection and Dosimetry of the Nnamdi Azikiwe University, Awka, Nigeria.

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### AKNOWLEDGEMENT

I will start by expressing my boundless and immeasurable gratitude to the God Almighty. All praise to God the Father, God the Son and God the Holy Spirit. The creator and the sustainer for my existence and for my good health

I wish to express profound gratitude to my supervisor Dr. Chiegwu, H. U. for his guidance and critical review of the conduct and documentation of the research activities. God bless you Sir!

I am sincerely grateful to the Dean of Faculty of Health Sciences and Technology Prof. P. O. Ezenduka and Staff, the Head of the Department of Radiography/Radiological Sciences Dr. Chiegwu, H. U. and the PG coordinator Dr. Ohagwu, C. C. for their encouragements and guidance.

My gratitude and appreciation goes to the lecturers of the Department of Radiography/Radiological Sciences: Prof. Nzotta, C., Dr. Ohagwu, C. C., Dr. Ugwu, A. C. Dr. Eze, , all departmental staff, the lecturers of the department of medical laboratory, department of physiology, and all the Faculty's Lecturers and staff for their assistance and support.

A special thanks to M. Tajuddeen for his guidance and support during the practical and progress of the work.

A special thanks to all my colleagues and my departmental promotion for their support and encouragement during the research work.

Very special thanks to my Parents, Brothers and Sisters for their financial, spiritual and moral supports.

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#### ACRONYMS AND ABBREVIATIONS

NORMs: Naturally Occurring Radioactive Materials

IEAE: International Atomic Energy Agency

EPA: Environmental Protection Agency

**TDS:** Total Dissolved Solids

CERT: Centre for Energy Research and Training

BDL: Below Detection Limit.

MCL: Maximum Contamination Limit

UNSCEAR: United Nations Scientific Committee on the Effects of Atomic Radiation

WHO: World Health Organization.

GSO: GCC Standardization Organization (Gulf Cooperation Council)

GPS: Global positioning System

LGA: Local Government Area

NNRA: Nigeria Nuclear Regulatory Authority

USA: United State of America.

#### ABSTRACT

Man's ingestion of radioactive material may result from drinking contaminated water in the environment. This study was aimed to determine the gross alpha and gross beta activity in surface and underground water in Nnewi North Local Government Area, Anambra state, in order to know if water is safe for consumption. This is important because most of the internal exposure to man may result from the ingestion of contaminated water. Among the contaminants are: <sup>241</sup>Am, <sup>241</sup>Cm, <sup>238</sup>Pu, <sup>239</sup>Pu, <sup>222</sup>Rn, <sup>226</sup>Ra, which are alpha emitters; <sup>45</sup>Ca, <sup>14</sup>C, <sup>134</sup>Cs, <sup>89</sup>Sr, <sup>90</sup>Sr, and tritium which are beta emitters. An aggregate of nineteen (19) water samples, nine (9) water samples from surface water sources and ten (10) water samples from underground water sources were analyzed for gross alpha and beta activity. Water samples were collected into 2 liter plastic kegs, acidified with Trioxonitrate (V) acid (HNO<sub>3</sub>) at the point of collection following the Environmental Protection Agency (EPA) procedures. Sample preparation and analysis was done at the CERT (Center of Energy Research and Training), Ahmadu Bello University, Zaria. The measured quantity (500ml) of water samples were evaporated to dryness in order to get the residue (about 77mg) which was transferred to a proportional gas-flow system (MPC 2000B-DP) and counted for 2700 second (45 minutes). Results indicated that the water sample under investigation had a mean gross alpha concentration of 0.48487±0.03453 Bq/L and mean gross beta concentration of 1.14587±0.07677 Bq/L for surface water and mean gross alpha concentration of 0.11587±0.00674 Bq/L and mean gross beta concentration of 0.35542±0.01267 Bq/L for underground water. Both alpha and beta emitters were less than 0.5 Bq/l for gross alpha and 1.0 Bq/L for gross beta activity (especially in underground water) which was lower compared to the maximum acceptable contamination limit as prescribed by the WHO. The annual equivalent effective dose derived from water consumption was calculated and found to be less than 0.3 mSv/yr provided that the water consumption was on average, 730 liters per adult inhabitant per year (2L per day) and 365 liters per children inhabitant per year (1L per day). Consumption of water in the region is safe but the areas where higher concentrations were observed should be monitored.

### CHAPTER ONE INTRODUCTION

#### **1.1 Background of the study**

Water as one of the most important natural resources is vital. Water is an essential substance to all living things which include: man, animal and all that surround them. Every living thing is made up of water. The use of water cuts across industrial, agricultural and domestic uses. The two main sources of water are surface and underground water. Underground water is the water present beneath Earth's surface in soil pore spaces and in the fractures of rock formations (boreholes and wells). Groundwater is only a small fraction of the total water in the world - about 0.6% - but it represents the main source of freshwater (Bouwer, 1978). Unfortunately the groundwater below a depth of 0.8 km is saline or is very difficult to extract (Bouwer, 1978). The potential useful water for human consumption is estimated to be about 4 million km<sup>3</sup> (Bouwer, 1978). Surface water is water on the surface of the planet such as in stream, lake, river, wetland, or ocean. It can be contrasted with groundwater (Young and Bredehoeft, 1972). About 70% of the Earth's surface is covered with water, which is estimated at a volume of approximately 1.4 billion km<sup>3</sup> (Ashton *et al.*, 2012).

Both surface and underground water can be contaminated by radioactive nuclides which include: Uranium, Radon, Strontium, Radium, Tritium and gross alpha and beta particles emitted from unstable radioactive elements. Certain problems have beset the use of groundwater around the world just as river waters have been over-used and polluted in many parts of the world (El-Mrabet *et al.*, 2002; Guzman *et al.*, 2002). The agricultural use of fertilizers is the main source of groundwater pollution, likewise natural radioactive elements. A portion of the radionuclides contained in fertilizers leaches through the soil and can reach the groundwater table (El-Mrabet *et al.*, 2002; Guzman *et al.*, 2002). However, human beings and natural phenomena are among the major agents that contribute in the deterioration of water sources; thus adversely affecting the quality of water. Water pollution mainly occurs as a result of release of any unwanted substance by human activities into water. Water pollution arises from wastes and sewage disposals into rivers and streams from various locations, such as industrial, hospital and rain wash out from fertilizer used for farming (Helliwell, 1975). Some of these pollutants are radionuclides which could have harmful effect on life. Water sources are equally polluted by naturally occurring radioactive materials (NORMS) of the earth's crust; which emits alpha ( $\alpha$ ), beta ( $\beta$ ) and gamma ( $\gamma$ ) radiations (Helliwell, 1975). These materials which are normally from the potassium, uranium and thorium series are more concentrated in deep ground water than in surface water (Helliwell, 1975).

Radionuclides are radioactive isotopes that can occur naturally (those radionuclides in the earth's crust and the environment) or result from man-made sources (resulting from nuclear and bomb testing, or power reactors accident, or industries). Typical radionuclides found in drinking water sources are isotopes of radium, uranium, and radon, among others. Fission products from man-made nuclear reactions are also of concern today, particularly radioactive cesium and iodine. Radiation exposure can occur by ingesting, inhaling, injecting, or absorbing radioactive materials (Narasimhan *et al.*, 2005). By ingesting, inhaling, injecting of radioactive materials through water, food or drugs, air, the radiation source is inside the body and this is called internal radiation. On the other hand, when the radiation source is outside the body such as terrestrial radiations from soil, stones, water and X-rays radiation, it is called external radiation.

The amount of radiation exposure is usually expressed in a unit called millirem (mR or mrem), which is a measure of energy deposited in human tissue and its ability to produce biological damage (WHO, 1993). The SI unit of radioactivity is the becquerel (Bq), after Henri Becquerel, where 1Bq = 1 disintegration per second. Guidance levels for drinking-water are given as the activity of the radionuclide per litre, called the specific activity concentration (Bq/litre). The recommended guideline activity concentrations are 0.1 Bq/L for gross alpha activity and 1 Bq/L for gross beta activity (WHO, 1993). The absorbed dose refers to how much energy is deposited in material by the radiation. The SI unit for absorbed dose is the gray (Gy), where 1Gy = 1 J/kg (joule per kilogram).

The terrestrial component of the background is due to various radioactive nuclides that are present in air, soil, water, food and building materials whose abundances vary significantly depending on the geological and geographical features of a region (Bozkurt *et al.*, 2007). Determining the distribution of these radionuclides is necessary for assessing the effects of radiation exposure.

The increase in the level of radioactivity [in water] is as a result of possible migration of radionuclides to surface and groundwater from anthropogenic activities: seepage of pollutants into the groundwater bodies, excessive fertilization of agricultural land, abandoned industrial sites, thermonuclear testing or nuclear power plants (WHO, 1993). Post-mining areas, waste dump areas or military areas could also contribute to the radioactivity increase (WHO, 1993).

There is evidence from both human and animal studies that radiation exposure at low to moderate doses may increase the long-term incidence of cancer (WHO, 1993). Animal studies in particular suggest that the rate of genetic malformations may be increased by radiation exposure (WHO, 1993). Acute health effects of radiation, leading to reduced blood cell counts and, in very severe cases, death, occur at very high doses of exposure of the whole body or large part of the body to radiation (IAEA, 1998). No deleterious radiological health effects are however, expected from consumption of drinking-water if the concentrations of radionuclides are below the guidance levels of 0.1mSv/year (WHO, 1993).

Marbaniang (2011), determined the gross activity of the  $\alpha$  and  $\beta$  emitting radionuclides present in the naturally occurring water bodies in the Uranium mineralization zone of Domiasiat, Meghalaya, India and found that they are lower compared to the WHO prescribed limit. He estimated that the additional equivalent effective dose derived from annual consumption of water is less than 0.3mSv/yr (2L per day). Gross alpha and beta activities and trace elements levels measured in drinking water of Saudi Arabia showed that trace elements concentrations in water did not exceed WHO, and GSO guidelines, gross alpha values were found to fall below the GSO and WHO recommended MCL of 0.5 Bq/l while the gross beta values in two samples only exceeds the MCL value of 1 Bq/l (Rafat, 2017).

Agbalagba and Avwiri (2012), determined Gross  $\alpha$  and  $\beta$  activity concentration and estimated adults and infants dose intake in surface and ground water of ten oil fields environment in western Niger Delta of Nigeria and found that gross  $\beta$  activities were higher than the corresponding gross  $\alpha$  activities, adults and infants dose intake obtained were below the WHO recommended reference level of  $0.1 \text{mSvy}^{-1}$  except for Uzere river water sample. They concluded that the water sources examined especially from river waters are contaminated radiologically.

Gross alpha and beta radioactivity were determined in ground water from some boreholes and wells in Kaduna North Local Government area using proportional counter. For the counting modes, results showed that the alpha activity in the study area was far below the practical screening level. However, a full sixty percent of the samples showed beta activity levels above the 1Bq/L as recommended by CEC-FAO and WHO (Abdu *et al.*, 2016). Gross beta ( $\beta$ ) radioactivity have been also assessed in Nassarawa town and found to be higher in well water than boreholes water (Ahmed *et al.*, 2014), high levels of naturally occurring and carcinogenic radium isotopes have been measured in low saline and oxic groundwater from the Rum Group of the Disi sandstone aquifer in Jordan (Vengosh *et al.*, 2009). The combined <sup>228</sup>Ra and <sup>226</sup>Ra activities in their study areas were up to 2000% higher than international drinking water standards. Measurement of tritium radioactivity in surface water on the Upper Silesia bordering two regions: Rokitnicki Stream Intervales (area I) and Klodnica and Bierawka Intervales (area II) revealed that tritium contamination has returned to the pre-bomb scale of 4-25 tritium unit TU (Aleksandra *et al.*, 2000).

#### **1.2 Statement of the Problem.**

Increase in the radionuclide concentration levels has various health effects. Those health effects could be genetic or somatic; the genetic effects could be transferred to offspring while somatic effects could ultimately lead to death depending on the level of exposure (Levi, 2012). One of the exposure pathways is water. Water pollutants from residential, industries, as well as from fertilizers on farmlands; from rainwater and the NORMS affect the quality of water. Man's ingestion of radioactive material may result from drinking contaminated water in the environment. The most important radionuclides associated with internal radiation exposure and the contaminations of the environment are: <sup>241</sup>Am, <sup>241</sup>Cm, <sup>238</sup>Pu, <sup>239</sup>Pu, <sup>222</sup>Rn, <sup>226</sup>Ra, which are alpha emitters; <sup>45</sup>Ca, <sup>14</sup>C, <sup>134</sup>Cs, <sup>89</sup>Sr, <sup>90</sup>Sr, and tritium (IAEA, 1989) which are beta emitters. The season of the year determine to a great extent the magnitude of contamination of different water sources (IAEA, 1989; Strand et al., 2002). When alpha particle emitting isotopes are ingested, they are far more dangerous than their half-life or decay rate would suggest, due to the high relative biological effectiveness of alpha radiation to cause biological damages (Little et al., 1985). Alpha radiation is an average of about 20 times more dangerous, and in experiments with inhaled alpha emitter up to 1000 times more dangerous than an equivalent activity of beta emitting or gamma emitting radioisotopes (Little et al., 1985). Anambra state is becoming highly industrialized area: Chicason Group Of Companies offering manufacturing services in cement,

Asia-Afro automobile and plastics, Cutix PLc company manufacturing electric cables and wires, Ibeto Group of Companies offering industrial manufacturing of cement, automotive lead-acid battery and spare parts, Innoson Vehicle Company for motor vehicle assembling services, First Express Aluminum Company Limited manufacturing aluminum roofing sheets, windows and allied products, Uru Industries Limited; an Agro chemical industry with services in the manufacturing of fertilizers and other chemical supplement for agricultural purposes to mention some of them. This study is to ascertain whether the level of radioactivity in the underground water and surface water system that may result from this industrialization could pose any significant health hazard to the population of the area. Some studies made in Nigeria revealed that rivers water and or underground water (Agbalagba and Avwiri, 2012; Nwankwo, 2013; Ahmed *et al.*, 2014; Abdu *et al.*, 2016) are contaminated radiologically in specifics areas.

Water has not been investigated for gross radioactivity concentration in Anambra state to the researcher best of knowledge.

#### **1.3 General Objective**

To assess the gross alpha and gross beta activity in underground water and surface water in Nnewi North, Anambra state, southeast, Nigeria.

#### **Specific Objectives**

- 1. To determine the gross  $\alpha$  level activity of  $\alpha$  radiation in surface and underground water in Nnewi, Anambra state.
- 2. To determine the gross  $\beta$  level activity of  $\beta$  radiation in surface and underground water in Nnewi, Anambra state.
- To assess the radiation hazard (Annual Effective Dose Equivalent) with the activity of α/β radiation associated with the consumption of water by children and adults in Nnewi, Anambra state.
- 4. To compare the concentration levels obtained with world standard limits to ascertain whether the level of  $\alpha/\beta$  activity could pose any problem and estimate the radiological implications to Nigerian populace.
- 5. To compare the results from the present study with findings from different locations in Nigeria and others countries

#### **1.4 Significance of the Study**

This study would ascertain whether the level of  $\alpha/\beta$  activity in the underground water and surface water system could pose any problem to the populace.

This study would be a guide in the education of the population of Nigeria

This study may provide a guide on a management of radioactive materials and present a data on radioactivity in surface and underground water in Nnewi area which will support Nigeria Nuclear Regulatory Authority (NNRA) in the control of water.

#### 1.5 Scope of Study

This was carried out in Nnewi, Anambra state, southeast, Nigeria. It was focused on samples of water collected from wells, boreholes, taps, rain water, stream and rivers. The samples were analyzed with a proportional counter analyzer to obtain the gross alpha and beta activity and their concentration levels.

### CHAPTER TWO LITERATURE REVIEW

#### **2.1 Conceptual Review**

#### 2.1.1 Radioactive materials

All materials are made of atoms. Radioactive atoms are unstable; that is, they have too much energy to remain stable. When radioactive atoms release their extra energy, they are said to decay. All radioactive atoms decay. After releasing all their excess energy, the atoms become stable and are no longer radioactive. From the view of the World Nuclear Association, all minerals and raw materials contain radionuclides of natural origin (WNA, 2015). The most important for the purposes of radiation protection are the radionuclides in the Uranium (<sup>238</sup>U) and Thorium (<sup>232</sup>Th) decay series. For most human activities involving minerals and raw materials, the levels of exposure to these radionuclides are not significantly greater than normal background levels and are not of concern for radiation protection (WNA, 2015). However, certain work activities can give rise to significantly enhanced exposures that may need to be controlled by regulation (WNA, 2015). Polonium (Po) is a chemical element with symbol Po and atomic number 84, discovered in 1898 by Marie Curie and Pierre Curie. A rare and highly radioactive element with no stable isotopes, polonium is chemically similar to bismuth and tellurium, and it occurs in uranium ores (WNA, 2015). Long-lived radioactive elements such as uranium, thorium and potassium and any of their decay products, such as radium and radon are examples of NORM (Naturally Occurring Radioactive Material). These elements have always been present in the Earth's crust and atmosphere, and are concentrated in some places, such as uranium ore bodies which may be mined. Excluding uranium mining and all associated fuel cycle activities, industries known to have NORM issues include: the coal industry (mining and combustion), the oil and gas industry (production), metal mining and smelting, mineral sands (rare earth minerals, titanium and zirconium), fertilizer (phosphate) industry, building industry, and recycling (Cooper, 2005). The two most important chains providing nuclides of significance in NORM are the thorium series and the uranium series:



Fig. 2.1: radioactive decay in Thorium and Uranium series (WNA, 2015)

Another major source of terrestrial NORM is potassium 40 (K-40). The long half-life of K-40 (1.25 billion years) means that it still exists in measurable quantities today (WNA, 2015). It beta decays, mostly to calcium-40, and forms 0.012% of natural potassium which is otherwise made up of stable K-39 and K-41. Potassium is the seventh most abundant element in the Earth's crust, and K-40 averages 850 Bq/kg there (WNA, 2015). It is found in many foodstuffs (bananas for example), and indeed fills an important dietary requirement, ending up in human bones [Humans have about 65 Bq/kg of K-40 and along with those foods, are therefore, correspondingly radioactive to a small degree. A 70 kg person has 4400 Bq of K-40] (WNA, 2015). The process by which the unstable nuclei decay to stable states is called radioactivity.

#### 2.1.2 Radioactivity

Radioactivity is the process by which the nucleus of an unstable atom loses energy by emitting radiation, including alpha particles, beta particles, gamma rays and conversion electrons. The term radioactivity was actually coined by Marie Curie, who together with her husband Pierre, began investigating the phenomenon recently discovered by Becquerel. The Curies extracted uranium from ore and to their surprise, found that the leftover ore showed more activity than the pure uranium. They concluded that the ore contained other radioactive elements. This led to the discoveries of the elements polonium and radium. It took four more years of processing tons of ore to isolate enough of each element to determine their chemical properties (Bjorn, 1996).A

material that spontaneously emits such radiation is considered radioactive. Nuclear radiation occurs in other forms, including the emission of protons or neutrons or spontaneous fission of a massive nucleus. Of the nuclei found on Earth, the vast majority is stable. This is so because almost all short-lived radioactive nuclei have decayed during the history of the Earth (Pasachoff, 2012). There are approximately 270 stable isotopes and 50 naturally occurring radioisotopes (radioactive isotopes). Thousands of other radioisotopes have been made in the laboratory (Pasachoff, 2012).

Radioactive decay will change one nucleus to another if the product nucleus has a greater nuclear binding energy than the initial decaying nucleus (Bjorn, 1996). The difference in binding energy (comparing the energies before and after states) determines which decays are energetically possible and which are not. The excess binding energy appears as kinetic energy or rest mass energy of the decay products (Bjorn, 1996). The probability that a particular nucleus will undergo radioactive decay during a fixed length of time does not depend on the age of the nucleus or how it was created. Although the exact lifetime of one particular nucleus cannot be predicted, the mean (or average) lifetime of a sample containing many nuclei of the same isotope can be predicted and measured (Bjorn, 1996). A convenient way of determining the lifetime of an isotope is to measure how long it takes for one-half of the nuclei in a sample to decay: this quantity is called the half-life (Pasachoff, 2012). The number of nuclei in the sample. This condition leads to radioactive decay showing itself as an exponential process.

$$N = N_0 e^{-\lambda t} \qquad (2.1)$$

Where: N is the number of nuclei remaining after a time t.

Nois the initial number of nuclei

 $\lambda$  is a decay constant

t is the mean lifetime of the parent nuclei.

$$N(T^{1/2}) = \frac{1}{2} N_0 = N_0 \cdot e^{-\lambda \cdot T^{1/2}} \rightarrow Ln \ 2 = \lambda \cdot T^{1/2} \rightarrow \lambda = \frac{\ln 2}{T^{1/2}} \text{ or } T^{1/2} = \frac{\ln 2}{\lambda}$$
(2.2)

The decay constant  $\lambda$  is inverse proportional to the half-lifeT<sup>1</sup>/<sub>2</sub>. Half-life of a radioactive substance determines a time-scale. According to the U.S. Environmental Protection Agency (EPA), the average annual radiation dose per person in the U.S. is 620 mrem. Drinking water measurements of radioactivity are typically expressed in picocuries per liter (pCi/L). The relationship between pCi to mrem is complex, because it depends on the type of isotope, its biological half-life, and the material absorbing the radiation. Climate change poses the threat of increasing the frequency and severity of extreme weather events, which could compromise the quality of drinking water resources (Ellis *et al.*, 2014).

#### 2.1.3 Radioactive decay processes

Alpha decay: In alpha decay, a helium nucleus  $\frac{4}{2}$  He or  $\frac{4}{2}\alpha$  is emitted.

$${}^{238}_{92} \text{U} \to {}^{4}_{2} \alpha + {}^{234}_{90} \text{Th.}$$
(2.3)

Alpha decay occurs most often in massive nuclei that have too large a proton to neutron ratio. An alpha particle, with its two protons and two neutrons, is a very stable configuration of particles. Alpha radiation reduces the ratio of protons to neutrons in the parent nucleus, bringing it to a more stable configuration. Nuclei, which are more massive than lead, frequently decay by this method (Bjorn, 1996).

Consider the example of polonium (<sup>210</sup>Po), decaying by the emission of an alpha particle, the reaction can be written:

$${}^{210}\text{Po} \rightarrow {}^{206}\text{Pb} + {}^{4}\text{He}. \tag{2.4}$$

This <sup>210</sup>Po nucleus has 84 protons and 126 neutrons. The ratio of protons to neutrons is Z/N = 0.667. A <sup>206</sup>Pb nucleus has 82 protons and 124 neutrons, which gives a ratio of 0.661. This small change in the Z/N ratio is enough to put the nucleus into a more stable state.

In alpha decay of a nucleus, the atomic number changes, so the original (or parent) atoms and the decay-product (or daughter) atoms are different elements and therefore have different chemical properties. The change in binding energy appears as the kinetic energy of the alpha particle and the daughter nucleus (Bjorn, 1996). Because this energy must be shared between these two particles, and because the alpha particle and daughter nucleus must have equal and opposite momenta, the emitted alpha particle and recoiling nucleus will each have a well-defined energy

after the decay. Because of smaller mass of the  $\alpha$ -particle, most of the kinetic energy goes to the alpha particle (Bjorn, 1996).

**Beta decay**: Beta particles  $\begin{pmatrix} 0\\-1 \end{pmatrix}\beta$  or  $\begin{pmatrix} 0\\-1 \end{pmatrix}$  electrons or  $\begin{pmatrix} 0\\1 \end{pmatrix}\beta$  or  $\begin{pmatrix} 0\\1 \end{pmatrix}$  positrons (electrons with positive electric charge, or antielectrons) are emitted from an atomic nucleus.

$${}^{234}_{90} \text{ Th} \rightarrow {}^{0}_{-1} \text{ e} + {}^{234}_{91} \text{ Pa.}$$
(2.5)  
$${}^{39}_{19} \text{ K} \rightarrow {}^{0}_{1} \text{ e} + {}^{39}_{18} \text{ Ar.}$$
(2.6)

Beta decay occurs when, in a nucleus with too many protons or too many neutrons, one of the protons or neutrons is transformed into the other.

In beta minus decay, a neutron decays into a proton, an electron, and an antineutrino:

$$\mathbf{n} \to \mathbf{p} + \mathbf{e}^{-} + \bar{\boldsymbol{\nu}}. \tag{2.7}$$

In beta plus decay, a proton decays into a neutron, a positron, and a neutrino:

$$p \to n + e^+ + \mathcal{V}. \tag{2.8}$$

Both reactions occur because in different regions of the Chart of the Nuclides, one or the other will move the product closer to the region of stability. These particular reactions take place because conservation laws are obeyed. Electric charge conservation requires that if an electrically neutral neutron becomes a positively charged proton, an electrically negative particle (in this case, an electron) must also be produced. Similarly, conservation of lepton number requires that if a neutron (lepton number = 0) decays into a proton (lepton number = 0) and an electron (lepton number = 1), a particle with a lepton number of -1 (an antineutrino in this case) must also be produced. The leptons emitted in beta decay did not exist in the nucleus before the decay; they are created at the instant of the decay (Bjorn, 1996).

According to knowledge, an isolated proton, a hydrogen nucleus with or without an electron, does not decay (Pasachoff, 2012). However within a nucleus, the beta decay process can change a proton to a neutron. An isolated neutron is unstable and will decay with a half-life of 10.5 minutes. A neutron in a nucleus will decay if a more stable nucleus results; the half-life of the decay depends on the isotope. If it leads to a more stable nucleus, a proton in a nucleus may capture an electron from the atom, and change into a neutron and a neutrino (Pasachoff, 2012). In beta decay the change in binding energy appears as the mass energy and kinetic energy of the

beta particle, the energy of the neutrino, and the kinetic energy of the recoiling daughter nucleus. The energy of an emitted beta particle from a particular decay can take on a range of values because the energy can be shared in many ways among the three particles while still obeying energy and momentum conservation (Bjorn, 1996).

**Gamma Decay:** In gamma decay, high-energy rays (like X-rays),  ${}_{0}^{0}\gamma$ , are emitted.

$${}^{99}_{43}\,\mathrm{Tc} \longrightarrow {}^{0}_{0}\gamma + {}^{99}_{43}\,\mathrm{Tc}. \tag{2.9}$$

The Electromagnetic Spectrum shows the different forms of electromagnetic radiation, with cosmic and gamma rays having the highest frequency and the highest energy, making the gamma rays potentially the most dangerous to humans(Bakshi *et al.*, 2009):



Fig. 2.2: electromagnetic spectrum (physics.tutorvista.com, 2016).

A nucleus changes from a higher energy state to a lower energy state through the emission of electromagnetic radiation (photons). The number of protons (and neutrons) in the nucleus does not change in this process, so the parent and daughter atoms are the same chemical element (Pasachoff, 2012). In the gamma decay of a nucleus, the emitted photon and recoiling nucleus each has a well-defined energy after the decay. The characteristic energy is divided between only two particles (Technetium [Tc] and Gamma [x]).

#### 2.1.4 Radioactivity in water

Radioactivity is a natural part of our environment. Present-day Earth contains all the stable chemical elements from the lowest mass of Hydrogen (H) to the highest: Lead and Bismuth (Pb and Bi). Every element with higher atomic number (Z) than Bismuth (Bi) is radioactive. The

earth also contains several primordial long-lived radioisotopes that have survived to the present days in significant amounts. <sup>40</sup>K, with its 1.3 billion-year half-life, has the lowest mass of these isotopes and beta decays to both <sup>40</sup>Ar and <sup>40</sup>Ca (Bjorn, 1996).These radionuclides can enter both surface and underground water via many ways.

Different kinds of water cover more than two thirds of the Earth's surface. This resource is extremely important for human life: water is used for direct consumption, it is used in the production of food, it is used for many industrial activities, etc. Water is a medium for the transport and interaction of radionuclides with and within different compartments of the troposphere: soils, sediments, crustal rocks, biota, and even air are continuously exchanging their radioactive contents with water (José *et al.*, 2007).

The presence of natural and artificial radionuclides at different levels in surface waters is clearly correlated with the existence of some coupling between the different compartments (José *et al.*, 2007). Surface waters are coupled to subsurface aquifers, to soils, and to the atmosphere, allowing incorporation of several radionuclides following different routes. Some radionuclides previously dissolved in deep underground aquifers may reach surface waters, other radionuclides may be directly incorporated in surface waters by deposition from the atmosphere, and a large fraction of the radionuclides in aquatic systems have their origins in the underlying soils, from where they can be transported to surface waters through runoff or leaching into the groundwater (José *et al.*, 2007). Once radionuclides are incorporated in a body of water, their dispersion and behavior is hard to predict in a general or straightforward way. Each stream, river, lake, etc., has its own mixing characteristics that vary from place to place and time to time (Bonnett, 1989), the rate of mixing being dependent on the depth of the water, the type of bottom, the shoreline configuration, wind, and on the different chemical, physicochemical, and biological processes (Bonnett, 1989).

For some radionuclides, such as <sup>137</sup>Cs and <sup>90</sup>Sr, a quantitative evaluation of the most important transfer parameters through lacustrine ecosystems has been performed. To do that, experimental studies following the most significant nuclear accidents (Chernobyl, Kysthym) were developed. Today, it is possible to obtain levels of uncertainty of a factor of two to three when models for these nuclides are applied as generic tools for predicting their behavior in the abiotic components of the lacustrine environment (Monte *et al.*, 2003). For several important radionuclides, the

parameters are not yet available with enough uncertainty, and further assessments are necessary, mainly in relation to the evaluation of model uncertainties (Monte *et al.*, 2003).

Cosmogenic radionuclides are continuously being produced in the atmosphere by the interaction of cosmic radiation with the nuclei present there. They can also be transported by rainwater. A wide variety of cosmogenic nuclides exist. Many of them can be used as specific tracers for different environmental compartments [<sup>14</sup>C, <sup>36</sup>Cl, <sup>85</sup>Kr, and <sup>3</sup>H for example] (Savage, 1985). From a radiological viewpoint, the case of <sup>3</sup>H is certainly relevant and special attention will be paid to it (Savage, 1985). Unfortunately the majority of cosmogenic nuclides are also produced (man-made radionuclides) during nuclear weapons tests and in the nuclear fuel cycle. In fact, this effect masks to a great extent their production by cosmic radiation (Savage, 1985).

As <sup>238</sup>U is the radionuclide with a greater abundance and presence in nature, it is one of the most useful natural radioisotopes in groundwater studies (Osmond and Cowart, 1992). It is known that in a closed system (e.g., unweathered rock), all intermediate radionuclides from the same decay series should be expected to be under secular equilibrium after a period of about 1 million years. This fact is based on the half-life of <sup>234</sup>U (245,000 years), which is the longest-lived intermediate radionuclide of the <sup>238</sup>U series. The first application of uranium series disequilibrium methods were reported by Rosholt et al., (1964) and subsequently by Titayeva et al., (1976). These applications are based on two facts. First, <sup>234</sup>U and <sup>238</sup>U are usually under secular equilibrium in rocks. However, they are usually far from this condition in aqueous media. Second, natural levels of <sup>238</sup>U can vary within a very wide range [10<sup>1</sup> to 10<sup>3</sup>mBq/l] (Osmond and Cowart, 1992). The combination of these factors is the basis for a very useful geochemical tool. Several physical effects can produce a preferential leaching of some daughter radionuclides. The initial disequilibrium of other long-lived radionuclides will persist over a time similar to their respective half-lives. When two radionuclides having similar chemical behaviors are considered, such as <sup>228</sup>Ra and <sup>224</sup>Ra, the shorter-lived species will tend to remain in secular equilibrium with its parents. Hence <sup>224</sup>Ra is usually found in equilibrium with <sup>228</sup>Th in groundwater (José et al., 2007). Uranium and radium are considered soluble species from a geochemical point of view, and typical natural activity concentrations are greater than 1 mBq/l (José et al., 2007). On the other hand, thorium and protactinium are detected in solution at very low concentrations (less than 102 mBq/l), being insoluble in water (José et al., 2007). Lead, bismuth, and polonium have variable behaviors depending on the water conditions. The short-lived radionuclides are not usually widely dispersed, so their levels are supplied by their parents in solution or by their dissolution from the aquifer rock. Therefore the mobility of each radionuclide depends on its elemental solubility, and half-life. The second way for a radionuclide to be dissolved in water is based on the decay of the parent being after a change in the geochemical conditions of the aquifer bedrock (José *et al.*, 2007).

Primordial radionuclides are present in drinking water to a certain degree, especially in well water or mineral water. The most relevant of these nuclides for radiological protection are <sup>226</sup>Ra and <sup>222</sup>Rn. The presence of <sup>226</sup>Ra in water depends on the water's origin. For well or mineral water, it depends on the content of <sup>238</sup>U in the solids of the aquifer where the water is stored (Savage, 1985; Schönhofer, 1994).

#### **2.2 Theoretical Review**

#### **2.2.1 Properties of water**

Water is a polar compound that is an important solvent for polar molecules in chemistry, and known as the "universal solvent" for its ability to dissolve many substances. At room temperature, water is a tasteless and odorless liquid, nearly colorless with a hint of blue. Water is commonly found in its solid, liquid and essentially also found invisible as a gas forms in nature (Braun *et al.*, 1993).

The formula of water is  $H_2O$  with a molar mass of 18.01528 g/mol, a density of 1,000 kg/m<sup>3</sup> and a melting point of 32°F [0°C]. Water has hydrogen bonding and is strongly polar. This polarity allows it to separate ions in salts and strongly bond to other polar substances such as alcohols and acids, thus dissolving them. Its hydrogen bonding causes its many unique properties, such as having a solid form less dense than its liquid form, a relatively high boiling point of 212°F [100°C] for its molar mass, and a high heat capacity. Water is amphoteric, meaning it is both an acid and a base. It produces H<sup>+</sup> and OH<sup>-</sup> ions by self ionization. This regulates the concentrations of H<sup>+</sup> and OH<sup>-</sup> ions in water. Because water is a very good solvent, water is rarely pure and some properties may vary from those of the pure substance. However, there are also many compounds that are essentially, if not completely, insoluble in water, including fats, oils and other non-polar substances (Braun *et al.*, 1993). Radiation in water result from both natural and artificial radiation sources and the radiation effects are same as those produced in the radiation from those sources.

#### 2.2.2 Types of radiation

Radiation is the energy emitted from a source in the form of waves or particles through matter. Radiation is often categorized as either ionizing or non-ionizing depending on the energy of the radiated particles. Ionizing radiation carries more than 10 eV and include: Ultraviolet radiation, X-ray, nuclear radiation, Neutron radiation, and Cosmic radiation. Non-ionizing radiation carries small energy to produce charged ions when passing through matter. Non-ionizing radiation includes Ultraviolet light, Visible light, Infrared, Microwave, Radio waves, Thermal radiation (heat).

There are three types of nuclear radiation: alpha radiation, beta radiation, and gamma radiation. Alpha radiation travels only a few centimeters in air, beta radiation travels tens of centimeters in air, while gamma radiation travels many meters. All types of radiation become less intense the further the distance from the radioactive material, as the particles or rays become more spread out. The thicker the substance, the more the radiation is absorbed. The three types of radiation penetrate materials in different ways.



Fig. 2.3: Different types of ionizing radiation penetrating solid matter (Wikipedia.org, 2016).

- Alpha radiation is the least penetrating. It can be stopped (or absorbed) by a sheet of paper.
- Beta radiation can penetrate air and paper. It can be stopped by a thin sheet of aluminum.
- Gamma radiation is the most penetrating. Even small levels can penetrate air, paper or thin metal. Higher levels can only be stopped by many centimeters of lead, or many meters of concrete.

#### 2.2.3 Source of radiation

**Natural occurring radiation source:** Natural sources of radiation includes: cosmic rays and terrestrial.

Radiation of extraterrestrial origin, which rain continuously upon the earth, is termed "cosmic rays". Throughout the history of life on earth, organisms continuously have been exposed to cosmic rays, radionuclides produced by cosmic ray interactions in the atmosphere, and radiation from naturally occurring substances which are ubiquitously distributed in all living and nonliving components of the environment. It is clear that contemporary life have adjusted or are doing so to all features and limitations of the environment, including the natural radiation background. The fact that this highly penetrating radiation was impinging upon the earth from space, rather than emanating from the earth, was deduced from balloon experiments in which ionization measurements were made at various altitudes from sea level to 9,000 m. It was found that the ionizing radiation rate decreased for some 700 m and from that point increased quite rapidly with elevation (tesec-int.org, 2016).

Radionuclides, which appeared on the Earth at the time of formation of the Earth, are termed "primordial". Of the many radionuclides that must have been formed with the Earth, only a few have half-lives sufficiently long to explain their current existence. These are K-40 with half-life of about  $1.251 \times 10^9$  years, U-238having half-life of about 4.5 billion years and Th-232 with half-life of about 14.05 billion years (wikipedia.com, 2016). Uranium and thorium each initiate a chain of radioactive progeny, which are nearly always found in the presence of the parent nuclides. Other primordial long-lived radionuclides which occur in nature are Rb-87, La-138, Ce-142, Sm-147, Lu-176 etc. Those radionuclides generally have very low concentrations. Naturally occurring neutrons formed many other radionuclides, but their abundance is usually very low. There is fascinating evidence that a "natural reactor" produced fission and activation products within this rich uranium deposit indicate that spontaneously triggered fission chain reactions occurred some 2 billion years ago (tesec-int.org, 2016).

**Man-made source radiation:** Man-made sources includes: Medical sources, Industrial sources, and Nuclear Explosions, Nuclear Power, and Radiation accidents. Over the last few decades man has "artificially" produced several hundred radionuclides. And he has learned to use the power of

the atom for a wide variety of purposes, from medicine to weapons, from the production of energy to the detection of fires, from illuminating watches to prospecting for minerals. The use of radioisotopes in medicine is widespread and may have significant radiological impact. These applications can be classified as: diagnostic uses, therapy, analytical procedures and pacemakers and similar portable sources. In industrial sources, the applications include industrial radiography, radiation gauging, smoke detectors and self-luminous materials (tesec-int.org, 2016). For the last 50 years, everyone has been exposed to cosmic rays due to radiation from fall-out from nuclear weapons. Almost all is the result of atmospheric nuclear explosions carried out to test nuclear weapon. This testing reached two peaks: first between 1954 and 1958 and second, greater, in 1961 and 1962. The production of nuclear power is much the most controversial of all the man-made sources of radiation, yet it makes a very small contribution to human exposure. One of the products of nuclear weapons testing was tritium. Its distribution in the ocean is like a telltale ink stain revealing the history of its occurrence on Earth. Tritium entered the ocean at the surface and became involved in the sinking North Atlantic water, part of the thermohaline current system that circulates ocean deep water. As the North Atlantic Deep Water moved downward, so did the tritium. Its concentration at various depths and locations gives a picture of a current system at work. (Wilkins, 1998)

#### 2.2.4 Radiation effects on life

**Stochastic Effects:** Stochastic effects are typically associated with long-term, low-level (chronic) exposure to radiation. The word "Stochastic" refers to the likelihood or probability than an effect will happen. In the diagnostic range of x-radiation that radiographers and the general public are exposed to, increased levels of exposure make these health effects more likely to occur, but do not influence the type or severity of the effect. The primary stochastic effects are: cancer and genetic defects (eccrad.blogspot.com, 2008).

**Non-Stochastic (Deterministic) Effects:** Non-stochastic effects are those effects in which the severity of the effect varies with the dose and for which a threshold value exists. Non-stochastic effects appear in cases of exposure to high levels of radiation, and become more severe as the exposure increases. Short-term, high-level exposure is referred to as 'acute' exposure. Many non-cancerous health effects of radiation are non-stochastic. Unlike cancer, health effects from

"acute" exposure to radiation usually appear quickly. Acute health effects include burns and radiation sickness (eccrad.blogspot.com, 2008).

**Penetrating Effects of Radiation**: Particles with the same energy but different masses can penetrate to different degrees.

- Alpha particles are massive with a high charge. They collide with other molecules and quickly lose energy. They cannot penetrate a few sheets of paper or outer cells of skin.
  But exposure to alpha particles can cause severe burns.
- Beta particles penetrate more but can be stopped with a sheet of aluminum or plastic.
- **Gamma and X-rays** can penetrate even more but are stopped with a thick lead shield. They can penetrate much deeper and are very dangerous to living organisms
- Neutrons can penetrate even more but can be stopped by a thick wall of concrete.

#### Acute radiation damage and genetic defects

- Acute radiation damage results from exposure to large amounts of radiation in a short period of time, which kills a large numbers of cells, leading to weakened immune systems (eccrad.blogspot.com, 2008).

High-energy ionizing radiation can take electrons from stable compounds in living organisms, leaving highly reactive unpaired electrons called radicals or free radicals. The first reaction with many volatile organic compounds (VOCs) is the removal of a hydrogen atom, forming water and an alkyl radical (R<sup>•</sup>): 'OH + RH  $\rightarrow$  H<sub>2</sub>O + R<sup>•</sup>. The alkyl radical will typically react rapidly with oxygen forming a peroxy radical: R<sup>•</sup> + O<sub>2</sub>  $\rightarrow$  RO<sup>•</sup><sub>2</sub>. The fate of this radical in the troposphere is dependent on factors such as the amount of sunlight, pollution in the atmosphere and the nature of the alkyl radical that formed it (wikipedia.org, 2016). The hydroxyl radical has a very short *in vivo* half-life of approximately 10<sup>-9</sup> seconds and a high reactivity (*Helmut, 1993*). This makes it a very dangerous compound to the organism (Reiter *et al.,* 1995; Reiter *et al.,* 1997).



Fig.2.4: Formation of free radicals in water under high-power laser UV irradiation  $[I = 10^8 - 10^{10} \text{ W/cm}^2, \lambda = 266 \text{ nm}, \tau_p = 30 \text{ ps}]$  (google.com, 2016)

These free radicals are so reactive that they cause reactions between otherwise stable materials in the cells of living organisms (wikipedia.org, 2016). If these reactions involve genetic materials (e.g. genes and chromosomes), the changes could lead to genetic mutations, cancer, or other devastating consequences (wikipedia.org, 2016).

#### 2.2.5 Radiation Hazards

Radiation hazard is the danger to health arising from exposure to ionizing radiation, either due to external irradiation or to radiation from radioactive materials within the body.

Health Hazards Associated with Radon Note: Because radon is a Noble gas with a long half-life, it is often inhaled and exhaled before it decays. However, two of its daughter nuclides, Po-218 and Po-214, are solids with much shorter half-lives (typically minutes). When radon decays in the lungs, these solid isotopes remain in the lungs tissue and continually emit  $\alpha$ -particles, damaging tissue and cells, and possibly even leading to cancer.

**Hazard Indices** ( $H_{ex}$  and  $H_{in}$ ): The hazard index reflecting external exposure, called external hazard ( $H_{ex}$ ) and the hazard index reflecting internal exposure, called internal hazard ( $H_{in}$ ) indices are evaluated by the following relations (Ramasamy *et al.*, 2009):

$H_{ex} = C_U / 370 + C_{Th} / 259 + C_K / 4810 \le 1$	(2.10)
$H_{in} = C_U / 185 + C_{Th} / 259 + C_K / 4810 < 1$	(2.11)

Where,  $C_U$ ,  $C_{Th}$  and  $C_K$  are the radioactivity concentration in Bq/kg (or Bq/l) of <sup>226</sup>Ra, <sup>232</sup>Th, and <sup>40</sup>K respectively.

**Radium Equivalent Activity (Ra**<sub>eq</sub>): This represents a weighted sum of activities of  $^{238}$ U,  $^{232}$ Th and  $^{40}$ K (based on the estimation 1 Bq/kg of  $^{238}$ U, 0.7 Bq/kg  $^{232}$ Th and 13 Bq/kg  $^{40}$ K that can produce the same radiation dose rates). The radium equivalent activity index is estimated according to Avwiri *et al.*, (2013).

$$Ra_{eq}(Bq/kg) = C_{U} + 1.43 C_{Th} + 0.077 C_{K}$$
(2.12)

Where  $C_U$ ,  $C_{Th}$  and  $C_K$  are the activity concentration in Bq/kg (or Bq/l) of <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K respectively.

**Absorbed Dose rate (D)** Absorbed dose is a measure of the energy deposited in a medium by ionizing radiation per unit mass. It may be measured as joules per kilogram and represented by the equivalent S.I unit, gray (Gy) or rad. The absorbed dose rate (D) is calculated using the following expression (Tawn, 2002)

$$D = 0.462 C_{\rm U} + 0.604 C_{\rm Th} + 0.0417 C_{\rm K}$$
(2.13)

Where, D is the absorbed dose rate in nGy/h,  $C_U$ ,  $C_{Th}$  and  $C_K$  are the concentrations of Uranium, Thorium and Potassium respectively.

**Representative Gamma Index (I** $\gamma$ ): This is used to estimate the gamma radiation hazard associated with the natural radionuclide in specific investigated samples. The representative gamma index is estimated as follow (Avwiri *et al.*, 2013)

$$I\gamma = C_{Ra}/150 + C_{Th}/100 + C_{K}/1500) \le 1$$
 (2.14)

**Annual Effective Dose Equivalent (AEDE)** The annual effective dose resulting from the ingestion of water is estimated based on the assumption that a daily intake of water per person is 2 l/day (Frisbie *et al.*, 2012) from the following expression (Avwiri *et al.*, 2013):

$$AEDE (mSv/y) = I x A x C \times 365$$
(2.15)

Where AEDE is the annual effective dose, I the water intake per day (l/d), A the daily intake of radionuclide (Bq/l) and C is the ingestion coefficient of the specific radionuclide (Bq/l).

**Excess Lifetime Cancer Risk (ELCR):** The Excess Lifetime cancer risk (ELCR) is calculated using the following equation (Taskin *et al.*, 2009):

$$ELCR = AEDE \times DL \times RF$$
 (2.16)

Where, AEDE is the Annual Equivalent Dose Equivalent, DL is the average duration of life (estimated to 70 years), and RF is the Risk Factor (Sv-1), i.e. fatal cancer risk per Sievert. For stochastic effects, ICRP uses RF as 0.05 for public (Taskin *et al.*, 2009).

Source	Worldwide average annual effective dose (mSv)	Typical range (mSv)	
External exposure			
Cosmic rays	0.4	0.3–1.0	
Terrestrial gamma rays <sup>a</sup>	0.5	0.3–0.6	
Internal exposure			
Inhalation (mainly radon)	1.2	0.2–10 <sup>b</sup>	
Ingestion (food and drinking-water)	0.3	0.2–0.8	
Total	2.4	1–10	

#### Table 2.1: Average radiation dose from natural sources of radiation

a Terrestrial exposure is due to radionuclides in the soil and building materials.

b Dose from inhalation of radon may exceed 10 mSv/year in certain areas (Tawn, 2002).

#### **Gross Alpha and Gross Beta Counters**

Gas proportional counting is one of the methods frequently used for gross counting (Passo and Kessler, 2012). Proportional counters are also frequently used to distinguish between alpha and beta particles from a mixed source such as dissolved solids in water. Alpha particles because of their high linear energy transfer produce a high number of interactions (ionization) with the gas in the detector volume. A beta particle produces a much lower number of ions per centimeter along its track than alpha particle. The alpha particles from a mixed source are registered at a lower voltage, the alpha plateau. At higher voltage, both alpha and beta particles are detected and thereby achieved by gross  $\alpha/\beta$  counting.

#### **2.3 Empirical Review**

The abundant supply of clean water is a major challenge facing modern civilizations, and as such is the topic of many researches over the world. A couple of researches have been recorded in water radioactivity over the world and in some parts of Nigeria. Radioactivity may be present in water sources as a result of natural processes, or from mining, production, use or disposal of radioactive materials (José *et al.*, 2007).

Fasae *et al.*, (2015) determined the gross alpha and beta activity concentrations in portable drinking water in Ado - Ekiti metropolis and the committed effective dose and found that the

total gross alpha and beta activity concentrations in the water within Ado-Ekiti metropolis ranged from 20 mBq/L to 357.8 mBq/L. They found that mean effective committed dose values of 0.15 mSv/y for children under 17 years and 0.03 mSv/y for adults obtained were below the recommended reference level of 0.1 mSv/y as suggested by the WHO.

Agbalagba and Avwiri, (2012) determined gross  $\alpha$  and  $\beta$  activity concentration using gas- flow proportional counter and estimated adults and infants dose intake in surface and ground water of ten oil fields environment in western Niger Delta of Nigeria. They found that the gross alpha activity concentration ranged from 0.01 to 35.1Bq/L and gross beta from 0.7to 151.2Bq/L. Correlation value between  $\alpha$  and  $\beta$  activities in well, tap and river waters were 0.025, 0.047 and 0.512 respectively. The obtained results showed that average natural activity concentrations of  $\alpha$ and  $\beta$ -emitting radionuclides in the water samples were slightly above the WHO recommended limit except alpha activity in tap water samples. For all samples the gross  $\beta$  activities were higher than the corresponding gross  $\alpha$ . The mean AEDE per year due to water consumption by adults and infants were below the WHO recommended reference level of 0.1mSv/y, except for Uzere river water sample. They concluded that the water sources examined especially from river waters are contaminated radiologically.

Marbaniang (2011), in his study, determined the gross activity of the alpha and beta emitting radionuclides present in the naturally occurring water bodies in the Uranium mineralization zone of Domiasiat, West Khasi Hills District, Meghalaya, India using an Alpha detector probe, manufactured by Nucleonix, India. The gross radioactivity indicated that the water samples under investigation had a low concentration of both  $\alpha$  and  $\beta$  emitters and the activity was less than 0.5Bq/l for  $\alpha$  activity and 1.0Bq/L for  $\beta$  activity which was lower compared to the WHO prescribed limit. He estimated that the additional equivalent effective dose derived from water consumption is less than 0.3mSv/yr provided the water consumption for the population is on average, 730 liters per inhabitant per year (2L per day).

Ross (2011) explored a number of water / wastewater treatment applications, and the subsequent nature and potential impact of radioactive residues associated with water exploitation processes throughout Queensland, Australia. In his study, series of radioactive iodine (<sup>131</sup>I) uptake and depuration experiments were conducted and the results presented indicated that <sup>131</sup>I concentration factor was calculated to be 630 mL/g. This value falls within the wide range of iodine concentration in macrophytes, and more specifically macroalgae. Depuration results were

characterized by a biphasic model with a fast initial elimination component with a half time of less than one minute, followed by a longer phase with a 3.2 day half-time. Greater than 96% of <sup>131</sup>I was retained after phase. Uptake and depuration result were observed to be reproducible under laboratory conditions.

The March 2011 earthquake in Japan led to a release of radioactive material from the Fukushima nuclear power plant into the environment. Since then, the U.S. Environmental Protection Agency (EPA) has been monitoring air, milk, precipitation (rain, snow, and sleet), and finished drinking water for radioactivity. Of the fission products tested, radioactive iodine (specifically <sup>131</sup>I) was the only one found in U.S. drinking waters (http://www.epa.gov/japan2011/rert/radnet-sampling-data.html#water). Radioactive iodine can concentrate in the thyroid, and long term exposure to elevated levels can cause thyroid cancer. However, as of April 4, 2011, the levels of <sup>131</sup>I found by EPA range from undetectable to 2.2 pCi/L and are all well below levels of concern for public health. EPA has proposed regulating radon in drinking water. The U.S. National Research Council published an informative report on radon in 1999, *Risk Assessment of Radon in Drinking Water*.

In Jizan region of Saudi Arabia, Rafat, (2017) measured gross alpha and beta activities and trace elements (Zn, Fe, Mn, Ni, Cu, Cr, Co, Se, Sr, V, Ti, Mo, Hg, Cd, Ba, As, Al and Pb) levels in drinking water. His obtained results showed that, in general, the trace elements concentrations in water did not exceed WHO, and GSO guidelines. The gross alpha values were found to fall below the GSO and WHO recommended MCL of 0.5 Bq/l while the gross beta values in two samples only exceeds the MCL value of 1 Bq/l, respectively.

Emumejaye and Osiga, (2012) assessed the activity concentration of  ${}^{40}$ K,  ${}^{226}$ Ra and its progenies,  ${}^{232}$ U, and physiochemical parameters: pH, Na, Mg, Ca, Cl<sup>-1</sup>, SO<sub>4</sub><sup>2–</sup>, NO<sub>3</sub> and CO<sub>2</sub><sup>2–</sup>, of some bottled drinking water produced in Delta State. The results obtained indicated that radioactivity is actually present in bottled drinking water but of little value and it shows no significant concentration range compared to WHO permissible values.

Ahmed *et al.*, (2014) assessed gross beta ( $\beta$ ) radioactivity concentration in underground (wells and boreholes) water in Nassarawa town in Nassarawa state using ISO 9696 and ISO 9697 respectively. Results obtained showed that the concentration of beta activity was relatively high in well water than boreholes compared with the maximum contaminant limit showing that well water in these areas is not safe for human consumption.

Laurie Wirt,(1994) in his study "Radioactivity in the Environment", determined the distribution of radioactive elements, such as uranium, radium, radon, and other dissolved trace elements that affect water quality. U.S. Geological Survey hydrologists collected over 69 ground-water samples and wells, and samples of river water and sediment from a network of stream flow-gaging stations throughout the Little Colorado River basin and wells in the Puerco River basin. He found that the amount of uranium in muddy water increases in direct proportion with increasing amounts of sediment. In parts of the alluvial aquifer not affected by mining, ground water contained less than 13  $\mu$ g/L of dissolved uranium which lies below the MCL recommended by the U.S. Environmental Protection Agency.

Early works showed that water supplies from wells in the U.S. could produce tap water with some 180 Bq/m<sup>3</sup> of <sup>226</sup>Ra. Potable water could contain more than  $7 \times 104$ Bq/m<sup>3222</sup>Rn, and in some cases concentrations of  $3.7 \times 105$  Bq/m<sup>3</sup>could be found (Savage, 1985). It is interesting to note that the domestic use of potable water can release dissolved <sup>222</sup>Rn into the air, provoking high indoor concentrations. Nwankwo, (2013) studied the radioactivity in groundwater from Tanke-Ilorin, Nigeria. In his study, the mean contribution of both <sup>226</sup>Ra and <sup>228</sup>Ra activities to the committed effective dose from a year's consumption of drinking water in the study area was found to be higher than the tolerable level of 1 mSv/y to the general public for prolonged exposure as recommended by ICRP, and much more than the new WHO recommended level of 0.1 mSv/y for drinking water.

In Brazil, Erlandson *et al*, (2001) found high <sup>222</sup>Rn concentrations in Swedish well water of more than 700 Bq/l, associated with leakage from layers of volcanic origin. A study of municipal water supply systems and private wells in Poland showed that on average the concentration of <sup>222</sup>Rn was around 5.3 kBq/m<sup>3</sup>, with a maximum of 38.3 kBq/m<sup>3</sup> (Zalewski *et al.*, 2001). In Spain, Dueñas *et al*, (1999) observed concentrations of <sup>226</sup>Ra of up to 600mBq/l (with a geometric mean of 12mBq/l) in bottled mineral water. As for <sup>222</sup>Rn, results showed a maximum of 52 Bq/l, with a geometric mean of 0.22mBq/l. The majority of the measured samples had levels of <sup>224</sup>Ra less than the minimum detectable activity (MDA) (i.e., 0.5 to 1 mBq/l), depending on the sample size. A maximum <sup>224</sup>Ra activity of 11mBq/l was found for the sample with a higher <sup>226</sup>Ra concentration. In the same work, results for tap water collected at Seville were also presented. A level of 1.3mBq/l was found for <sup>226</sup>Ra, while the concentration for <sup>224</sup>Ra is less than

1.4mBq/l. According to this article, the efficiency of radium removal by water treatment plants is very close to 100%.

Nguelem *et al.*, (2013) assessed natural radioactivity levels in selected groundwater (boreholes and wells) used as domestic purposes in particular and drinking as well in some communities in Adentan and Abokobi, Ghana using High-Purity Germanium (HPGe) detector. Annual effective dose for adult members of public due to consumption of the groundwater was determined and compared with estimated average annual dose due to ingestion of nuclides in water by the WHO (100  $\mu$ Sv/y) and food and water (290  $\mu$ Sv/y) by UNSCEAR. They were found to be within the WHO range even though Adentan value is slightly higher than the WHO average value. The results showed that consumption of groundwater may not pose any radiological health hazard to the public.

Abdu *et al.*, (2016) measured gross alpha and beta radioactivity in ground water from some boreholes and wells in Kaduna North Local Government area collected in two liter plastic jerry cans with two drops of concentrated Trioxonitrate (V) acid (HN0<sub>3</sub>) in each container in order to preserve the samples. The results showed that alpha and beta activity concentrations range from  $(0.69 \pm 1.52) \times 10^{-3}$  to  $(41.2 \pm 25.7) \times 10^{-3}$  Bq/L, and from  $(286 \pm 77.1) \times 10^{-3}$  to  $(9506 \pm 104) \times 10^{-3}$  Bq/L respectively. The overall showed that the alpha activity in the study area is far below the practical screening level of radio activity in drinking water of 0.5Bq/L for alpha, as recommended by CEC-FAO and WHO. However, a full 60% of the samples showed beta activity levels above the 1Bq/L as recommended by CEC-FAO and WHO.

Levi, (2012) used Gamma spectrometric method to determine the natural radioactivity of the groundwater obtained from hand dug wells in Sango-Ilorin, North Central Nigeria. The total activity concentrations of the radionuclides range from 0.009 to 30.14 Bq/l. The results showed that the calculated average annual effective dose/ingestion dose for radionuclides from some of the wells are above WHO recommended level of 0.1 mSv/y for drinking water but falls within the tolerable level of 1 mSv/yr to the general public for prolonged exposure as recommended by the International Commission of Radiological Protection (ICRP).

Avwiri *et al.*, (2014) estimated the radiation health hazard indices and excess lifetime cancer risk associated with exposure to river water, soil and sediments from Mini-Okoro/Oginigba Creek. The determined activity concentration of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K in all the samples were within the world acceptable values but was higher than results of other works done in similar environments.

There were poor correlations between <sup>238</sup>U and <sup>232</sup>Th, <sup>238</sup>U and <sup>40</sup>K and <sup>232</sup>Th and <sup>40</sup>K. The total ratio of Th/U for water, sediment and soil samples ranged from 0 to 7.63. Whereas, the total ratio K/U and of K/Th ranged from 0 to 1.66 for water, sediment and soil. Their results indicated that the radiation hazard in all river samples from the area studied is not significant. They concluded that there was no immediate radiation health hazard associated with the use of any of the water samples studied.

Aleksandra *et al.*, (2000) presented the results obtained by measuring the concentration of radioactive tritium in surface water and precipitation. All samples from their study were taken during 1995-1996, from the Upper Silesia area bordering two regions: Rokitnicki Stream Intervales (area I) and Klodnica and Bierawka Intervales (area II). distributions of tritium concentration ( $A_{OT}$ ) were similar for both areas. Mean tritium concentration was 10.2±3.5 TU for area I, and 10.9±3.4 TU for area II. It was worthy to notice that many samples measured activity ( $A_{OT}$ ) is from 7 to 11TU (6of 10 for area I and 7 of 12 for area II). Only few samples showed very low tritium concentration, from 3 to 7. The obtained results confirmed, that tritium concentration has returned to the pre-bomb values (4-25 TU).

Selçuk et al., (2009) investigated gross alpha and gross beta radioactivity concentrations in water, soil and sediment samples collected from Bendimahi River, its tributaries and Van Lake (Turkey). In water samples, the obtained results showed that in May; gross alpha and gross beta activity concentrations varied between 0.063–0.782 and 0.021–0.816 Bg  $1^{-1}$  and in August the values were 0.009–0.037 and 0.081–3.116 Bq  $l^{-1}$ , respectively. Ogundare and Adekoya, (2015) determined gross alpha and beta radioactivity in surface soil and drinkable water around a steel processing facility using a low background Gas-less counting system with a solid state silicon PIPS detector for alpha and beta detection. The average activities for gross alpha and beta in water ranged between  $0.0064 \pm 0.0001 {--} 0.0182 \pm 0.0001 \; Bq/l$ and  $0.046 \pm 0.001 0.126 \pm 0.001$  Bq/l respectively. The average annual committed effective dose from intake of water was between 0.0304 mSv and 0.0678 mSv which are lower than the recommended reference level for ingested dose from drinkable water.

Results from this present study will show what the situation is for Nnewi, Anambra state, southeast, Nigeria.

### CHAPTER THREE MATERIALS AND METHODS

#### 3.1 Study Design

The study was a cross-sectional study of the gross alpha ( $\alpha$ ) and gross beta ( $\beta$ ) radio activities in surface and underground water in Nnewi, Anambra State.

#### 3.2 Study Area

Nnewi is the second largest city in Anambra State in southeastern Nigeria. Nnewi as a metropolitan city encompasses 4 local government areas, Nnewi North, Nnewi South, Ekwusigo and Ihiala Local Government areas. Nnewi North is commonly referred to as Nnewi central, and comprises four autonomous communities: Otolo, Uruagu, Umudim, and Nnewichi. Nnewi North also includes Ichi, an autonomous neighboring town. The first indigenous car manufacturing plant in Nigeria is located in the city while the first wholly made-in-Nigeria motorcycle, the 'NASENI M1' was manufactured also in Nnewi. Nnewi as a whole has many rivers. As at 2006, Nnewi had an estimated population of 391,227 according to the Nigerian census (2006 census). The city spans over 1,076.9 square miles (2,789 km<sup>2</sup>) in Anambra State. Nnewi Metropolitan Area and its satellite towns are home to nearly 2.5 million residents.

Geographically, Nnewi falls within the tropical rain forest region of Nigeria. Though it suffers from soil leaching and erosion which has reduced the soil in some areas to a porous sandy terrain, it remains an area of rich agricultural produce and the epicenter of business trade. The city is located east of the Niger River, and about 22 kilometers south east of Onitsha in Anambra State, Nigeria (Nfor, 2006).



Fig.3.1: Map of Nnewi (Ministry of Environment, Anambra state. Sept., 2016)

#### **3.3 Ethical Considerations**

Ethical clearance was obtained from the Ethical committee of the Faculty of Health sciences and Technology, Nnamdi Azikiwe University, Nnewi campus, and the traditional rules of the communities where the water samples were collected.

#### **3.4 Sample Size**

An aggregate of nineteen (19) water samples were taken for analysis covering the 4 communities (Umudim, Uruagwu, Nnewichi and Otolo communities) in Nnewi North area. Ten (10) water samples from underground (boreholes and underground tanks) water sources and nine (9) water samples from surface (streams, rain water and rivers) water sources in Nnewi area.

#### **3.5 Sampling Method**

Convenient sampling method was used for this study. Ten (10) water samples were collected from underground water sources and nine (9) water samples from the surface water sources. All samples were collected in Nnewi North area, Anambra state.

#### **3.6 Method of Sample Collection**

The following procedures (Method 900 of the US Environmental Protection Agency) were carefully carried out during the collection and preparation of the samples:

Samples of water were collected directly into 2 liter plastic kegs (polyethylene containers) after washing the containers properly and rinsed with the water sample to be collected. About 10ml/L of concentrated Trioxonitrate (V) acid (HN0<sub>3</sub>) was added at the point of collection using a syringe, gloves and a face-mask. The addition of concentrated Trioxonitrate (V) acid (HN0<sub>3</sub>) help preserve the radionuclides present in the water samples and also prevent the absorption of radionuclides by the inner wall of the containers. The addition of Trioxonitrate (V) acid (HN0<sub>3</sub>) assists in reducing the pH of the water samples below 2 (Semkow, 2001). Surface water from different streams and rivers within the area were collected and treated with the reagent. Care was taken to avoid fetching from the stagnant areas and near the shore line (IAEA, 1989). Sachet water samples were emptied directly into the keg. For bore-holes and public taps (from water

cooperation); the taps were first turned on at full capacity for three (3) minutes to purge the plumbing system of any water which might have been there for some time. The taps flow rate was reduced to attain steady turbulence and radon loss before collecting the water into the kegs. Ten (10) water samples were collected from underground water sources and nine (9) water samples from the surface water sources. GPS (satellite navigation system) coordinates were recorded at the point of collection of water samples using iPad Model A1396. These samples were kept in a plastic container and covered properly. The water samples were taken to the CERT (Center for Energy Research and Training), Ahmadu Bello University, Zaria, Kaduna state where they were prepared and analyzed for gross alpha ( $\alpha$ ) and gross beta ( $\beta$ ) activities.

#### **3.7 Samples Preparation**

*APPARATUS:* Laboratory beakers, petri-dishes, Hot plate, infra-radiator lamp, Experimental (Digital) weighing balance, planchets, cotton wool.

**REAGENTS:** Acetone and vinyl acetate.

*PROCEDURE:* The beakers, crucibles (petri-dishes), planchets and spatula were washed properly, rinsed with clean water and sterilized using acetone. Then the apparatus were kept and dried inside the oven. A little quantity of the water sample was used to rinse the beaker twice so as to ensure that there is no cross contamination before evaporation. About 500ml of the water sample was measured into the beaker and set on the hot plate with steady temperature below boiling point to allow gradual evaporation and to avoid excessive loss of the residue (fig 3.1). This process continues until when the volume of the water sample reduced to a very little quantity (about 50ml). Then it was transferred into the petri-dish and evaporated to dryness under infra-radiator lamp. This process is known as surface drying. Having taken the initial weight (of the empty dish), the weight of the residue together with petri-dish was measured using digital analytical weighing balance. The weight of the total residue obtained from the total volume evaporated was calculated by using the relation:

$$\mathbf{W}_{\mathrm{r}} = \mathbf{W}_{\mathrm{(d+s)}} - \mathbf{W}_{\mathrm{d}} \tag{3.1}$$

Where:  $W_{(d+s)}$  is the weight of the dish with sample's residue,

W<sub>d</sub> is the weight of empty dish

W<sub>r</sub> is the weight of the total residue.

0.0770g of the residue was transferred to the sterilized planchet and the exact volume that produced this required weight (0.0770g) was calculated by the use of the expression:

$$0.0770g x V_{tr} = W_{tr} x V.$$
(3.2)

Where:  $V_{tr}$  is the volume that generated total residue,

W<sub>tr</sub> is the weight of the total residue obtained

V is the volume that yielded the required residue.

For samples with residue obtained greater than or equal to 0.0770g, the sample efficiency is said to be 100%. But for the samples with residue less than 0.0770g, its sample efficiency can be obtained using the expression below;

Sample eff. =  $\frac{\text{weight of residue}}{0.0770\text{ g}} \times 100\%$ . (3.3)



Plate 3.1: Evaporation system

#### **3.8 Sample Analysis**

The international standards organization procedure (ISO 9696 and ISO 9697: 1992E) for the measurement of gross alpha ( $\alpha$ ) and gross beta ( $\beta$ ) activities in water was employed in this analysis. This method provided a screening technique to measure the gross alpha ( $\alpha$ ) and gross beta ( $\beta$ ) radioactivity in water samples. To analyze drinking-water for gross alpha ( $\alpha$ ) and gross beta ( $\beta$ ) activities (excluding radon), a known volume of the sample was evaporated to dryness and the activity of the residue was measured.

Sample activity analysis was done using a proportional counter system; a portable gas-filled MPC 2000B-DP single channel gross alpha and gross beta radiation detector. The equipment employed in this analysis is a gas-filled proportional counter designed purposely for gross alpha ( $\alpha$ ) and gross beta ( $\beta$ ) counting. Each sample was placed on the detector and counted for 2700 seconds (45 minutes).

*CALIBRATION*: The counting equipment is automated. The calibration of the equipment is aimed at obtaining the operational efficiency of the channels of the counter after the calibration; the counter was then used to count the background radioactivity of the environment. The gross alpha and beta radioactivity counting modes applicable to the counter, with respective voltages of 1,600 and 1,700 volts were employed to count the prepared water samples collected from different locations in Nnewi area. The Gas flow proportional counter used was initially calibrated using the manufacturers'  $\alpha$  and  $\beta$  sources (Pu–239 and Sr–90 respectively) whose activities range from 133.29 - 185.49 Bq and 92.31 - 103.68 Bq respectively in all the three operating modes. This test was run for 2700s (45 mins) for 5 cycles. The counting system was used to count clear empty planchets in all the counting modes to obtain the background radioactivity of the environment which is needed to be used in the subsequent measurements.

#### i. Gross Alpha Counting

For gross alpha counting, the high voltage was set at 1600V and samples were counted for 3 cycles of 2700s (45mins) per cycle. The results were displayed as raw counts and count rate (count/min). The data were acquired for alpha mode and the specific activity for alpha in the samples was calculated using the formula below.

$\Lambda_{\text{otivity}}(\alpha) (\mathbf{P}\alpha/\mathbf{I})$	Net count (CPM) (α)	(3 1)
Activity (u) (bq/L)	DE x 60 x Sample Size (Volume) X Sample Efficiency	(3.4)

Where D.E is the detector's efficiency and net counts is given by:

Net counts = Raw counts (CPM) – Background (CPM). 
$$(3.5)$$

#### ii. Gross Beta Counting

The high voltage for gross beta counting was set at 1700V and samples were counted for 3 cycles over a preset period of 2700s in beta mode. The specific activities were calculated using the formula below.

Activity (
$$\beta$$
) (Bq/L) =  $\frac{\text{Net count (CPM) }(\beta)}{\text{DE x 60 x Sample Size (Volume) X Sample Efficiency}}$  (3.6)

The parameters remain the same as for equation 3.4

The proportional counter is a gas filled detector and a single-channel equipment which is automated. The counting procedure involves entering the pre-set time, number of cycles and counting mode. The results when the preset time elapsed were displayed as raw counts and count rate, i.e count per minute (CPM). The raw counts (CPM) were repeated three times each for all the samples, for alpha or beta and the average value were obtained respectively.



Plate 3.2: Gas-flow proportional counter analyzer and Water samples

#### iii. Effective Dose Ra-226 Equivalent in different location for children and adults

Following the procedure of Fernandez *et al.*, (1992), it was considered that more than 50% of the annual dose corresponds to radium (gross alpha radium). Assuming that Ra-226 was the main contributor to the alpha activity, the following equation was used to calculate the doses

$$DR = A \times IR \times ID$$
 (Marbaniang, 2011) 3.7

Where DR = Effective Equivalent Dose (mSv)

A = Activity (Bq/L)

IR = Intake of water for a person per year (2L/day for adult and 1L/day for children)

= 730Litres/year for an adult and 365Liters/year for children.

ID = Ingestion dose equivalent factor (ICRP, 2012)

= 2.8 x  $10^{-7}$  Sv/Bq/yr for an adult and 6.2 x  $10^{-7}$  Sv/Bq/yr for the children [about 5years] (Marbaniang, 2011)

Ingestion Dose Equivalent factor used to calculate the internal radiation exposure by ingestion of radionuclide by ICRP was considered.

#### iv. Procedure for data analysis

The data was presented using appropriate statistical tools. Tables were used to present the sample name, Global Positioning System (GPS location) coordinates and locations, value of the activity concentration. Table was used to outline the Effective Dose <sup>226</sup>Ra Equivalent in children and adults due to consumption of water within a year. Table was also used to compare the max and mean alpha activity concentration with others findings from different countries and different localities in Nigeria. The mean, the range and the standard deviation computed with Microsoft Office Excel 2007, were used to report the activity concentration and compare results from the present study with the world standard limits.

SPSS version 20.0 was used to analysis data when comparing the present study findings with studies from around the country and the world.

### CHAPTER FOUR RESULTS

The operational efficiencies of the channels of the proportional counter indicate an efficiency of 87.95% for the alpha counts and 42.06% for the beta counts. These are good efficiency values for this type of counting system of known activities as at the time of calibration. The result gives background radioactivities of 0.90 cpm for Alpha and 0.93 cpm for Beta. This result is quite representative of the environment. Acquisition of gross alpha ( $\alpha$ ) and gross beta ( $\beta$ ) activities concentration was made in dual (alpha and beta) mode.

### 4.1 Gross Alpha and Beta Radioactivity in Surface Water from Various Locations in Nnewi North Local Government Area of Anambra State

Table 4.1 shows the different locations where water samples were collected with their GPS coordinates, the different activity concentrations for gross alpha and gross beta and the observation due to the fact that they were higher than the world standard limits set by the World Health Organization (0.1 Bq/L for gross alpha and 1 Bq/L for gross beta activity).

From table 4.1, we can see that all locations do not exhibit gross alpha and gross beta activity but the concentration differs from one location to another. Table 4.1 below shows the gross alpha and beta activity concentrations of the surface water samples of the selected locations in Nnewi north local government area of Anambra state. The maximum water radioactivity concentrations for gross alpha and gross beta were  $2.3001 \pm 0.1895$  Bq/L and  $6.8538 \pm 0.4165$  Bq/L respectively, both obtained in samples from Otolo community. Sample from Mmiri Ukwaka did not exibit gross alpha and gross beta activity concentration and one of the samples from Mmiri Ubu did not exibit gross beta activity concentration. The gross alpha activity concentration from Mmiri Ubu was found to be  $0.7710 \pm 0.0271$ .

The standard deviation in surface water were  $\sigma = 0.03553$  Bq/L for gross alpha activity and  $\sigma = 0.07677$  Bq/L for gross beta activity with a maximum theoretical mean average of M = 0.4847Bq/L for gross alpha activity and M = 1.14587 Bq/L for gross beta activity.

S/N	Sample ID and location	Alpha Activity (Bq/l)	Beta Activity (Bq/l)	Comment
1	Mmiri Eze: Sample1 05° 59'42"N, 06°53'49"E	$0.0144 \pm 0.0009$	$0.0212 \pm 0.0018$	Safe
2	Mmiri Eze: Sample2 05° 59'42''N, 06°53'49''E	$0.0607 \pm 0.0054$	$0.2058 \pm 0.0120$	Safe
3	Otolo community: Sample1 06°00'10"N, 06°56'16"E	$0.8577 \pm 0.0275$	$1.5991 \pm 0.0568$	Not safe*
4	Otolo community: Sample2 06°00'10"N, 06°56'16"E	$2.3001 \pm 0.1895$	$6.8538 \pm 0.4165$	Not safe*
5	Mmiri Ukwaka: Sample 05° 59'42''N, 06° 54'31''E	$BDL \pm 0.0002$	$BDL \pm 0.0005$	Safe
6	Mmiri Ubu: Sample1 05° 58'05''N, 06°55'31''E	$0.1684 \pm 0.0309$	$0.4970 \pm 0.0667$	Not safe (for α)
7	Mmiri Ubu: Sample2 05° 58'05''N, 06°55'31''E	$0.7710 \pm 0.0271$	$BDL \pm 0.0444$	Not safe (for α)
8	Mmiri Ofia: Sample1 05°58'40"N, 06°56'21"E	$0.0914 \pm 0.0143$	$0.7338 \pm 0.0411$	Safe
9	Mmiri Isi Ubu: Sample2 05°58'40"N, 06°56'21"E	$0.1001 \pm 0.0150$	$0.4021 \pm 0.0512$	Safe

<u>**Table 4.1**</u> Gross Alpha and Gross Beta activities concentration in surface water from various locations in Nnewi North, Anambra state

**BDL:** Below Detection Limit

\* Not safe based on being higher than the standard limit

### 4.2 Gross Alpha and Beta Radioactivity in Underground Water from Various Locations in Nnewi North Local Government Area of Anambra State

Table 4.2 shows the different locations where underground water samples were collected with their GPS coordinates, the different activity concentration levels for gross alpha, and for gross beta activities and the observation due to the fact that they were or were not higher than the world standard limits set by the World Health Organization (0.1 Bq/L for gross alpha activity and 1 Bq/L for gross beta activity concentration).

From table 4.2, we can see that all locations do exhibit gross alpha and gross beta activity but the concentration differs from one location to another. Table 4.2 below shows the gross alpha and beta activity concentrations of the underground water of the selected locations in Nnewi north local government area of Anambra state. The maximum water radioactivity concentrations for gross alpha and gross beta were  $0.5486 \pm 0.0153$  Bq/L and  $1.3636 \pm 0.0336$  Bq/L respectively, both obtained in samples from Umudim community. The sample 2 from Otolo community exhibits the second highest activity concentration of  $0.1305 \pm 0.0060$  Bq/L for gross alpha activity and  $0.2622 \pm 0.0127$  Bq/L for gross beta activity.

The standard deviation in underground water sample for this Nnewi area was  $\sigma = 0.00674$  Bq/L for gross alpha activity concentration and  $\sigma = 0.01267$  Bq/L for gross beta activity concentration with a maximum theoretical mean average of M = 0.11587 Bq/L for gross alpha activity concentration and M = 0.35542 Bq/L for gross beta activity concentration.

<u>**Table 4.2**</u> Gross Alpha and Gross Beta activities concentration in underground water from various locations in Nnewi North, Anambra state

S/N	Sample ID and location	Alpha Activity (Bq/l)	Beta Activity (Bq/l)	Comment
1	Otolo community: sample1 05° 58'13"N, 06° 56'41"E	$0.0176 \pm 0.0004$	$0.0290 \pm 0.0008$	Safe
2	Otolo community: sample2 05° 59'43"N, 06° 56'08"	$0.1305 \pm 0.0060$	$0.2622 \pm 0.0127$	Not safe (for α)
3	Umudim community: sample1 05°58'14"N, 06°54'40"E	$0.5486 \pm 0.0153$	$1.3636 \pm 0.0336$	Not safe*
4	Umudim community: sample2 05°59'10"N, 06°54'23"E	$0.1064 \pm 0.0055$	$0.4650 \pm 0.0133$	Safe
5	Nnewichi community: sample1 06°02'03"N, 06°54'35"E	$0.0336 \pm 0.0009$	$0.0920 \pm 0.0021$	Safe
6	Uruagwu community: sample1 06°01'13"N, 06°53'21"E	$0.0701 \pm 0.0028$	$0.1092 \pm 0.0058$	Safe
7	Umudim community: sample3 05°59'46"N, 06°55'07"E	$0.0021 \pm 0.0004$	$0.0088 \pm 0.0009$	Safe
8	Otolo community: sample3 05° 59'23"N, 06°55'36"E	$0.0850 \pm 0.0134$	$0.6219 \pm 0.0316$	Safe
9	Nnewichi community: sample2 06°01'50"N, 06°55'36"E	$0.0961 \pm 0.0208$	$0.5117 \pm 0.0228$	Safe
10	Uruagwu community: sample2 06°01'32"N, 06°54'10"E	$0.0687 \pm 0.0019$	$0.0908 \pm 0.0031$	Safe

\* Not safe based on being higher than the standard limit

### **4.3 Determination of the Range, Mean, Standard Deviation and the Annual Effective Dose Equivalent**

Table 4.3 shows computed range, mean and standard deviation associated with gross alpha and gross beta activity concentration in surface water and underground water sources.

From Table 4.3, it could be seen that the gross alpha and gross beta activity in Nnewi north surface water ranges from 0.0144 Bq/L to 2.3001 Bq/L with an arithmetic mean of 0.4848 Bq/L and from 0.0212 Bq/L to 6.8538 Bq/L with an arithmetic mean of 1.1458 Bq/L respectively. From the same table 4.3, it was computed that the gross alpha and gross beta activity in Nnewi north underground water ranges from 0.0021 Bq/L to 0.5486 Bq/L with an arithmetic mean of 0.1158 Bq/L and from 0.0028 Bq/L to 1.3636 Bq/L with an arithmetic mean of 0.1158 Bq/L and from 0.0088 Bq/L to 1.3636 Bq/L with an arithmetic mean of 0.3554 Bq/L respectively. The standard deviation for gross alpha activity was  $\sigma = 0.03453$  Bq/L in surface water against  $\sigma = 0.00674$  Bq/L in underground water. The standard deviation for gross beta activity was  $\sigma = 0.01267$  Bq/L in underground water. It could be seen that the mean distribution of gross alpha activities and gross beta activities in surface water was higher than the mean distribution in underground water in the Nnewi North area. The same was applied to the standard deviation with values presented in Table 4.3.

Table 4.4 shows the different localities with GPS coordinates, the gross alpha activity concentration level and the computed effective dose radium-226 equivalent values with observation. Ingestion Dose Equivalent factor used to calculate the internal radiation exposure by ingestion of radionuclide by ICRP (2012) was considered. From Table 4.4, it was found that consumption of water from inhabitant is less than 0.3 mSv/yr as prescribe by regulation no matter the location. Table 4.5 compares the mean gross alpha with max gross alpha from the present study and different other studies around the world.

Table A.1 shows the comparison of mean alpha from this study with findings from other studies using One Sample t-test. This table showed No Statistical Significant mean difference between surface water at P-value = 0.306. It also revealed No Statistical Significant mean difference between the underground water of the present study and other studies at P-value = 0.084.

Source	n	Radiation type	<b>Ra</b> Min	nge Max	<b>Mean</b> (Bq/L)	Stand. Dev. (Bq/L)
Surface	9	Alpha	0.0144	- 2.3001	0.48487	0.03453
Water	,	Beta	0.0212	- 6.8538	1.14587	0.07677
Underground	10	Alpha	0.0021	- 0.5486	0.11587	0.00674
Water	10	Beta	0.0088	- 1.3636	0.35542	0.01267

<u>**Table 4.3**</u> Range, Mean and Standard Deviation of gross Alpha and gross Beta activity concentration in Nnewi water samples, Anambra state

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<u>**Table 4.4**</u> Effective Dose Ra-226 Equivalent in different location in Nnewi for children and adults with alpha and beta mean activity concentration

C/NI	Comula ID and loss for	Mean alpha	Ann. Eff. Do	Oha	
<b>3</b> /1N	Sample ID and location	activity (Bq/L)	Children	Adults	ODS.
1	Otolo community: sample1 05 °58'13"N, 06°56'41"E	0.0176	0.0039	0.0036	Safe
2	Otolo community: sample2 05 ° 59'43"N, 06° 56'08"	0.1305	0.0295	0.0267	Safe
3	Umudim community: sample1 05°58'14"N, 06°54'40"E	0.5486	0.1241	0.1121	Safe
4	Umudim community: sample2 05°59'10"N, 06°54'23"E	0.1064	0.0240	0.0217	Safe
5	Nnewichi community: sample1 06°02'03"N, 06°54'35"E	0.0336	0.0076	0.0069	Safe
6	Uruagwu community: sample1 06°01'13"N, 06°53'21"E	0.0701	0.0158	0.0143	Safe
7	Umudim community: sample3 05°59'46"N, 06°55'07"E	0.0021	0.0005	0.0004	Safe
8	Otolo community: sample3 05 °59'23"N, 06°55'36"E	0.0850	0.0192	0.0174	Safe
9	Nnewichi community: sample2 06°01'50"N, 06°55'36"E	0.0961	0.0217	0.0196	Safe
10	Uruagwu community: sample2 06°01'32"N, 06°54'10"E	0.0687	0.0155	0.0140	Safe

<u>**Table 4.5**</u>: Comparison of mean and maximum gross alpha activity concentration in different countries and different localities

Place	Alpha - Mean (Bq/l)	Max Gross Alpha (Bq/l)	Standard Deviation	Reference	
Nnewi, Nigeria	0.484 / 0.115*	2.30 / 0.54*	0.0345/0.0067*	This work	
Spain	0.14	1.36	0.24	Duenas et al., 1997	
Venezuela	0.12	0.54	0.07	Sajo-Bohus, 1997	
India	0.09	0.33	0.01	Marbaniang, 2011	
Quebec, Canada	0.21	17.98	1.25	Zikovsky, 2006	
Saudi Arabia	0.19	0.45	-	Rafat, 2017	
Kaduna, Nigeria	-	0.04	10.94	Abdu et al., 2016	
Delta, Nigeria	4.1	35.1	-	Agbalagba and Avwiri, 2012	
Ado-Ekiti, Nigeria	0.15	0.17	-	Fasae et al., 2015	

\*Surface water / Underground water

### CHAPTER FIVE DISCUSSION

#### 5.1 Gross Alpha Level Activity in Surface and Underground Water

In the present study, gross alpha activity concentration varies with location. From results, we could see that almost all the sampling locations exhibit gross alpha activity except the sample from Mmiri Ukwaka in surface water. It was found that gross alpha activity in Nnewi north surface and underground water ranges from 0.0144 Bq/L to 2.3001 Bq/L with an arithmetic mean 0.48487  $\pm$  0.03453 Bq/L and from 0.0021 Bq/L to 0.5486 Bq/L with an arithmetic mean  $0.11587 \pm 0.00674$  Bq/L respectively. World Health Organization (2011) advises 0.1 Bq/L for gross alpha. It could be seen that the mean distribution of gross alpha activities in surface water was higher than the mean distribution in underground water in the Nnewi North area. Gross alpha activities of water samples from different locations were under the reference value of 0.1 Bq/L except for samples from Otolo community and Mmiri Ubu in surface water; sample2 from Otolo, sample1 from Umudim community in underground water with respective values:  $0.8577 \pm 0.0275$  Bq/L,  $2.3001 \pm 0.1895$  Bq/L,  $0.1684 \pm 0.0309$  Bq/L,  $0.7710 \pm 0.0271$  Bq/L,  $01305 \pm 0.0060$  Bq/L, and  $0.4586 \pm 0.0153$  Bq/L. The higher values of alpha activity observed are higher than the maximum contamination limits (0.5Bq/l) except sample1 from Mmiri Ubu in surface water and sample1 from Umudim community which is slightly higher than the MCL as prescribe by the WHO. The obtained results showed that means natural activity concentration of  $\alpha$ -emitting radionuclides are slightly above the reference value of 0.1 Bq/L but lower than the maximum contamination limit (0.5Bq/l for gross alpha) as prescribe by the WHO. This phenomenon is similar to the findings observed in water samples by Agbalagba and Avwiri (2012) in Western Niger Delta of Nigeria, Rafat, (2017) in Drinking Water of Saudi Arabia. The opposite was observed by Marbaniang, (2011) in his study of water collected from Domiasiat area, West Khasi hills district, Meghalaya, India, Abdu et al (2016) in ground water from some bore holes and wells in Kaduna North Local Government area of Kaduna State and Fasae et al., (2015) in portable drinking water in Ado - Ekiti metropolis which found that the gross alpha activity

was below the screening level of radioactivity in all locations. The high  $\alpha$ -activity observed in water samples from Otolo community, Mmiri Ubu and Umudim community could be attributed to the presence of factories, the air pollution from bicycles and vehicles and the leaching process that occurs in the Nnewi area. The geological formation of these selected locations is mainly based on sandstone. These formations have an ability to retain the radioactivity elements (Marbaniang, 2011). The high level of activity concentration in Otolo community (rain water) might be attributed to the fact that the rain water was contaminated, the quality of the roof that drain the water in the area. The alpha emissions are due to a mixture of radioisotopes with different chemical behaviors and all belonging to the natural radioactive series (Ozmen *et al.*, 2004). Gross alpha activity in natural water is mainly due to uranium and radium isotopes because thorium solubility is low (Osmond and Cowart, 1992). Principally, Ra-226 and occasionally Th-232, Po-210 or Ra-224 are the main contributors to the total alpha particle activity in water samples (Marbaniang, 2011). The presence of alpha in these water samples suggests that these localities where water samples were obtained contain these radionuclides.

#### 5.2 Gross Beta Level Activity in Surface and Underground Water

In the case of gross beta activity concentrations, values also vary with location. Almost all the sampling locations exhibit gross beta activity concentration except the sample from Mmiri Ukwaka and sample2 from Mmiri Ubu both in surface water. The World Health Organization (Frisbie *et al.*, 2012) advices 1.0 Bq/L for gross beta activity as reference level and Maximum Contamination Limit (MCL) values for drinking water. It was found that gross beta activity concentration in Nnewi north surface and underground water ranges from 0.0212 Bq/L to 6.8538 Bq/L with an arithmetic mean  $1.14587 \pm 0.07677$  Bq/L and from 0.0088 Bq/L to 1.3636 Bq/L with an arithmetic mean  $0.35542 \pm 0.01267$  Bq/L respectively. Gross beta activities concentrations from different locations were under the reference value of 1 Bq/L except for samples from Otolo community in surface water and sample1 from Umudim community in underground water which was slightly higher than the recommended reference level with the respective values:  $1.5991 \pm 0.0568$  Bq/L,  $6.8538 \pm 0.4165$  Bq/L and  $1.3636 \pm 0.0336$  Bq/L. Even though, some of the activity concentrations were below the

minimum detection limit (in surface water especially), but for all locations where activities were observed, the gross beta activity is higher than the corresponding gross alpha activity concentration. This phenomenon is in accordance with that observed by Karahan et al. (2000), in various surface water samples in Istanbul, Turkey and the findings observed in water samples by Agbalagba and Avwiri (2012) in Western Niger Delta of Nigeria. The opposite however, was observed by Ozmen et al. (2004), in limestone and igneous based areas. It could be seen that the mean distribution of gross beta activity concentration in surface water was higher than the mean distribution in ground water in the Nnewi North area. The obtained results showed that mean natural activity concentration of  $\beta$ -emitting radionuclides in Nnewi water felt below the reference value of 1 Bq/L as prescribed by the WHO except in surface water which was slightly above that reference level. This phenomenon is in accordance with the findings observed by Rafat, (2017) in drinking water samples collected from a region in Saudi Arabia, Agbalagba and Avwiri (2012) in Western Niger Delta of Nigeria. These findings were not in agreement with the ones observed by Ahmed et al., (2014) in assessing gross beta radioactivity concentration in underground water in Nassarawa town of Nassarawa State, Nigeria. The opposite was also observed by Marbaniang, (2011) in his study of water samples collected from Domiasiat area, West Khasi hills district, Meghalaya, India, Abdu et al., (2016) in ground water from some bore holes and wells in Kaduna North Local Government area of Kaduna State and Fasae et al., (2015) in portable drinking water in Ado - Ekiti metropolis. The high level of activity concentration in Otolo community (rain water) might be attributed to the fact that the rain water was polluted, the quality of the roof that drain the water in the area. The high  $\beta$ -activity concentration observed in surface water samples and in underground water samples could be attributed to the presence of factories, the air pollution from bicycles, generators and vehicles and the leaching process that occurs in Nnewi area. The geological formation of these selected locations is mainly based on sandstone. These formations have an ability to retain the radioactivity elements (Marbaniang, 2011). The beta radioactivity of most of the water samples could be readily attributed to K-40 and Ra-228 concentration (Ozmen et al., 2004). The presence of beta in these water samples suggests that these localities where water samples were obtained contain these radionuclides.

# **5.3** Radiation Hazard (AEDE) with Activity of α-radiation Associated with Consumption of water by Adult and Children in Nnewi.

The standard deviation was found to be 0.03453 Bq/L for gross alpha activity concentration in surface water samples against 0.00674 Bq/L in underground water samples and 0.07677 Bq/L for gross beta activity concentration in surface water samples against 0.01267 Bq/L in underground water samples. This standard deviation was found to be higher in surface water than in underground water. This means that the values vastly differ from each other in surface water while the values packed more lightly in underground water. In another terms, radioactivity is more spread out in surface water than underground water. Table 4.4 showed that the calculated Effective Dose Ra-226 Equivalent in different location in Nnewi for children and adults with mean activity concentration were less than 0.3 mSv/yr. Ingestion Dose Equivalent factor used to calculate the internal radiation exposure by ingestion of radionuclide by ICRP (2012) was considered. From Table 4.4, it was found that consumption of water from inhabitant was less than the recommended limit as prescribe by regulation no matter the location. This phenomenon is in agreement with the findings observed by Fasae et al., (2015) in portable drinking water in Ado - Ekiti metropolis, Marbaniang, (2011) in his study of water samples collected from Domiasiat area, Meghalaya, India. These findings were not in agreement with the ones observed by Agbalagba and Avwiri (2012) in Western Niger Delta of Nigeria, which say that river water were contaminated radiologically.

### 5.4 Comparison of Concentration Levels Obtained with World Standard Limits and with Values from Authors/Researchers in Different Countries/Locations

Table 4.5 compares our mean and max alpha activity with values in different countries and locations. This Table shows the means alpha activity concentration, the max gross alpha activity and standard deviation associated with different countries with references. Table 4.5 compares the average mean alpha activity from this study with the findings from

different countries and localities (respective of the water source) using One Sample t-test.

Gross alpha and gross beta activities of water samples from different locations were under the maximum acceptable contamination limit as prescribed by the WHO except for some specific locations. The obtained results showed that means natural activity concentration of  $\alpha$ -emitting radionuclides and  $\beta$ -emitting radionuclides are lower than the maximum acceptable contamination limit (0.5Bq/l for gross alpha and 1 Bq/L for gross beta) as prescribe by the WHO except in surface water which was slightly above that reference level. Comparing the max and the mean alpha activity from the present study with other findings from different countries, it was worth noting that the mean value of gross alpha activity in all the countries is not that much different even though there is a mark difference in the maximum value. This however signifies that the high alpha activity is confined to certain places only, which may constitute only a small percentage of the total activity recorded which is true for the case of the Nnewi North area where the high activity is confined to the factory zone. We can therefore say that the means gross alpha activity in all these places are similar. It was clear that the means are generally low in terms of underground water in Nnewi area, bottled water in Spain (Duenas et al., 1997), drinking water in Venezuela (Sajo-Bohus, 1997), India (Marbaniang, 2011), Quebec, Canada (Zikovsky, 2006), Saudi Arabia (Rafat, 2017), Kaduna, Nigeria (Abdu et al., 2016) and Ado-Ekiti, Nigeria (Fasae et al., 2015). From Table 5.1, the mean activity was high in Delta, Nigeria (Agbalagba and Avwiri, 2012). We could see that the maximum alpha activity of these locations was also comparable except in Quebec, Canada and Delta, Nigeria i.e. 17.98 Bq/L and 35.1 Bq/L respectively. Comparing the average mean alpha activity from this study with studies from around the

world using One Sample t-test, It showed No Statistical Significant mean difference between surface water in the present study and other studies (P = 0.306). It also revealed No Statistical Significant mean difference between the underground water of this study and that of other studies (P = 0.084). This could mean that the relationship between these findings and other findings were not by chance. This shows that the findings in the present study are in agreement with those studies that were compared with.

#### CONCLUSION

The survey and study of gross alpha and beta radioactivity in Nnewi indicates that the water sample under investigation has a low concentration of both alpha and beta emitters and the mean activity was less than 0.5Bq/l for alpha and 1.0Bq/L for beta. In conclusion, the radioactivity of water samples from selected locations is below the limit set by the WHO except for some specific locations. Also, the additional equivalent annual effective dose derived from water consumption is less than 0.3 mSv/yr provided the water consumption for the population is on average, 730 liters per adult and 365 liters for children per year. Consumption of water in the region is safe. However, treatment and monitoring of water consumption should be considered for zones of high radioactivity and more work has to be done in order to identify the individual radionuclides contributing to the total gross radioactivity in the area.

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### Appendix

<u>**Table A.1**</u>: Comparison of Mean Alpha from this Study with Findings from Different Countries and Different Localities

Researcher	Country/location	Surface water	Underground water	P-value	Inference
Present study	Nigeria/Nnewi	0.4848±0.0345	0.1158±0.0067		
Duenas <i>et al.</i> , 1997	Spain		0.346	0.084	NS
Ozmen <i>et al.</i> , 2004	Turkey/Elazığ	$2.52\pm0.07$		0.306	NS
Sajo-Bohus, 1997	Venezuela		0.37	0.084	NS
Ahmet <i>et al.</i> , 2007	Turkey/Sanliurfa	0.038		0.306	NS
Marbaniang, 2011	India	$0.13 \pm 0.04$		0.306	NS
Zikovsky, 2006	Quebec, Canada		0.052	0.084	NS
Rafat, 2017	Saudi Arabia		$0.45\pm0.03$	0.084	NS
Agbalagba and Avwiri, 2012	Nigeria/Delta	$4.1\pm0.1$	$0.15\pm0.003$	0.306/0.084	NS
Fasae <i>et al.</i> , 2015	Nigeria/Ado-Ekiti		$0.151 \pm 0.014$	0.084	NS

Significant at 0.05 level (2-tailed).

NS = Non Significant

S = Significant

	S(alpha)	Error	S(beta)	Error	_	G(alpha)	Error	G(beta	Error
	0.0144	0.0009	0.0212	0.0018		0.0176	0.0004	0.029	0.0008
	0.0607	0.0054	0.2058	0.012		0.1305	0.006	0.2622	0.0127
	0.8577	0.0275	1.5991	0.0568		0.5486	0.0153	1.3636	0.0336
	2.3001	0.1895	6.8538	0.4165		0.1064	0.0055	0.465	0.0133
	0	0.0002	0	0.0005		0.0336	0.0009	0.092	0.0021
	0.1684	0.0309	0.497	0.0667		0.0701	0.0028	0.1092	0.0058
	0.771	0.0271	0	0.0444		0.0021	0.0004	0.0088	0.0009
	0.0914	0.0143	0.7338	0.0411		0.085	0.0134	0.6219	0.0316
	0.1001	0.015	0.4021	0.0512		0.0961	0.0208	0.5117	0.0228
					_	0.0687	0.0019	0.0908	0.0031
Mean	0.48487		1.14587			0.11587		0.35542	
Std. Dev		0.03453		0.07677			0.00674		0.01267

Table A.2: This table shows the data results computed using Microsoft Excel 2007

**Formulas:** Mean 
$$=\frac{\sum Xi}{n}$$

Standard Deviation = 
$$\sqrt[2]{\frac{\sum(Xi-\bar{X})^2}{(n-1)}}$$