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EFFECTS OF AERATION ON RADON CONCENTRATION IN GROUNDWATER SAMPLES

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EFFECTS OF AERATION ON RADON CONCENTRATION IN GROUNDWATER SAMPLES

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IN PARTIAL FULFILMENT OF THE REQUIREMENT FOR THE AWARD OF MASTER OF SCIENCE (M.Sc) IN PHYSICS AND ELECTRONICS

DECLARATION

I, AMANESI Abdulfatai with matriculation number:159403005 hereby declare that this work titled "EFFECTS OF AERATION ON RADON CONCENTRATION IN GROUNDWATER SAMPLES" is an original research written by me under the supervision of Dr. E. O. Oniya, of the Department of Physics and Electronics Adekunle Ajasin University Akungba-Akoko Ondo State.

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Student's Signature

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CERTIFICATION

This research titled "EFFECTS OF AERATION ON RADON CONCENTRATION IN GROUNDWATER SAMPLES." by AMANESI Abdulfatai meets the requirement governing the award of the degree of Master of Science (M. Sc.) in Department of Physics and Electronics Adekunle Ajasin University Akungba-Akoko Ondo State and is approved for its contribution to knowledge and literary presentation.

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DEDICATION

This research is dedicated to Almighty Allah for His grace and mercy and to my parents for always believing and supporting me all through the program.

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LIST OF ACRONYMS AND UNITS

AED: Annual Effective Dose

BEIR: National Academy of Biological Effect of Ionizing Radiation

Bq/L: Becquerel per Litre

CERT: Centre for Energy Research and Training

CNSC: Canadian National Safety Commission

E-DWD: European Drinking Water Directives

IAEA: International Atomic Energy Agency

ICRP: International Commission on Radiation Protection

LSA: Liquid Scintillation Analyzer

LGA: Local Government Area

MCL: Maximum Contamination Level

ml: Millilitre

mSv: MilliSievert

NORM: Natural Occurring Radioactive Material

PMT: Photomultiplier Tube

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

USEPA: United States Environmental Protection Agency

ABSTRACT

Pipe borne water is not readily available in many localities in Nigeria, leading to an increase in the number of people depending on groundwater such as boreholes and well water as their main source of water supply. Radon is one of the major sources of radiological contamination in groundwater and the largest contributor of the total radiation received by the general public from natural radioactive sources. Radon is the second largest cause of lung cancer. There are possibilities of reduction of radon concentration in water when aerated. Thus the effect of aeration on radon concentration in groundwater that is consumed by the general public in some communities in Akoko South West Local Government Area, Ondo State Nigeria was investigated.

A total of fifty-one groundwater samples collected from different locations within Akoko South-West Local Government Area, Ondo State were investigated. These were divided into three different categories. Category A was capped immediately at the source while categories B and C were exposed for twenty-four hours (24hrs) and seventy-two hours (72hrs) respectively. Each category comprised of seventeen (17) samples. The different times of exposure of water samples in the three categories were to observe the effect of natural aeration on radon-222 concentration. The radon concentrations in all the samples were analyzed using Liquid Scintillation Counter (LSC) method.

The concentrations of Radon in category A ranged from 12.61Bq/L to 57.50Bq/L and 10.30Bq/L to 41.89Bq/L with mean values of 28.01Bq/L and 25.34Bq/L for Boreholes and wells respectively. For category B, the concentration ranged from 8.50Bq/L to 37.94Bq/L and 6.75Bq/L to 24.39Bq/L with corresponding mean values of 20.18Bq/L and 14.82Bq/L for Boreholes and wells respectively and for category C for boreholes and wells the concentration ranged from 3.67Bq/L to 18.33Bq/L and 3.22Bq/L to 10.61Bq/L with mean values of 9.33Bq/L and 7.07Bq/L respectively. Activity concentrations of Radon-222 were observed to be highest in category A and lowest in category C. The results were compared with the maximum contaminant level (MCL) of 11.1Bq/L set by United States Environment Protection Agency and it was observed that 94.1% of category A, 64.7% of category B and 17.6% of category C of the samples exceeded the value. The results obtained showed that aeration reduces radon concentration in water.

It is thereby recommended that water should be fetched in bowls and kept for three to four days before consumption so as to keep the exposure due to ingestion of Radon-222 as low as reasonably achievable.

CHAPTER ONE

INTRODUCTION

1.1 Background

Water is a transparent, tasteless, odourless and nearly colourless chemical substance that is the main constituent of earth's streams, lakes, oceans and the fluids of most living organisms. About 70% of the earth's surface is water-covered. Ninety-seven percent (97%) water exist in oceans that are not suitable for drinking. Only 3% is freshwater wherein 2.97% of it is comprised by glaciers and ice caps and the remaining 0.3% is available as surface and ground water for human use (Muller and Michael, 2017). Water is one of the most abundant substances on earth and is a principal constituent of all living things. Water that is fit for human consumption is called drinking or portable water. Water that is not potable may be made potable by filtration, distillation or by a range of other methods. Water that is not fit for drinking but not harmful for humans when used for other activities like swimming and bathing is called safe water. Safe drinking water is the basic need for good health and it is also a basic right of humans.

Freshwater is already a limiting resource in many parts of the world. In the next century, it will become even more limited due to increased population, urbanization and climate change (Jackson et al., 2001). Water is used in other various aspects of daily lives that include power generation, agriculture and domestic and industrial activities. Water quality is one of the most important parameters of environmental studies due to the possibility of pollution (Garba et al., 2008). Water pollution occurs when harmful substances often chemical, microbiological or radiological contaminate a stream, river, lake, ocean, aquifer or other body of water, degrading water quality and rendering it toxic to humans or the environment. These justify why is therefore

important to ensure water to be used is free from chemical, microbiological and radiological contaminations.

Radiological water contaminants are particularly undesirable radioactive substances that have entered a water supply. They are known as radionuclides. Typical naturally occurring radionuclides found in drinking water include isotopes of radium, uranium and radon among others (Altikulac et al. 2015).

The radionuclides such as radon-222, radium-226 and radium-228 most commonly occurring in natural waters (lake, spring and groundwater) present a risk to human health (Akar et al., 2012). The major means by which the naturally occurring radioactive materials enter the human body are through two main pathways: by inhalation of radioactive gas like radon and ingestion of primordial radionuclides as well as their progenies (Oniva, 2014). The occurrence of radionuclides in drinking water gives rise to internal exposure, directly via the decay of radionuclides taken into the body through ingestion and inhalation; also indirectly when they are incorporated as part of the food chain. Radon-222 is an alpha-emitting noble gas that is found in various concentrations in soil, air and in different types of water as a result of migration from rocks and soil in contact with the water (USEPA, 2012). Much attention has been given to the . dissolved radon concentration in air and how geologic formations (including faults) qualitatively impact the variability of dissolved radon with little attention given to radon concentration in water. The composition of geological formations is known to critically affect radon concentrations in water sources near these formations, although when water is transported from the source to the consumers, natural aeration is likely to reduce radon concentrations (D'Alessandro and Vita, 2003). Waters originating from granite formations of cratonic areas have been identified as having the highest concentrations of radon and other radionuclides in the

uranium and thorium series, also ground water circulating in active volcanic areas display high radon content (Virk and Singh, 1993). The measurement of radioactivity in drinking water allows one to determine the exposure of the population to radiation from the habitual consumption of water.

The treatment of radon rich water by suitable mitigating techniques is necessary for cases of water containing an elevated level of radon; since there is a shred of increasing evidence correlating ingestion of radon rich water and the potential for induction of internal cancers (Jastaniah et al, 2014). The radon removal from water by different treatment processes, such as Granular Activated Carbon (GAC) adsorption, and various forms of aeration was described (Kinner, et, al 1990).

1.2. Statement of the problem

Several environmental problems are threatening the inhabitants of Akoko South West Local Government Area and one of these problems is the groundwater quality. Most of the previous studies done on the quality of groundwater were focused on the chemical, physical, and microbiological analysis; but not much on natural radiation analysis has been done. The number of people who depend on groundwater such as boreholes and well water as their main source of water supply is increasing. This is because the government water supply is not reliable. These sources of water do not undergo a quality examination with respect to natural radiation, which is the leading cause of lung and stomach cancer. There is therefore the need to investigate the quality of groundwater water with regards to the natural radiation contamination, compare it to nternational standards.

1.3 Scope and limitation

This work focuses on the analysis of activity concentrations of Radon-222 in water sources from Akoko region of Ondo State using Liquid Scintillation technique and also to study the effect of aeration on radon concentration in the water samples. Estimation of the corresponding annual effective doses due to ingestion of Radon-222 in water for adults, children and infants were to be covered. The source of water involved was groundwater (open wells and Boreholes) which was collected from the following communities within the region: Akungba-Akoko, Oka-Akoko, Oba-Akoko, Ayegunle-Akoko, Iwaro Oka-Akoko, Supare-Akoko and Etioro. The result is expected to provide the activity concentrations of Radon-222 which would be used to estimate the annual effective doses from water consumption. The major limitation of this research was the lack of laboratories that are well equipped for carrying out analyses on natural radionuclides, thereby prompting the transfer of samples to other locations hence increasing the cost of analysis due to additional cost of transportation.

1.4. Aim and objectives

The aim of this research is to investigate the effects of aeration on the concentration of Radon-222 in groundwater samples in Akoko-South West Local Government Area, Ondo State of Nigeria using Liquid Scintillation Counter.

The objectives of this research are to:

 Measure the activity concentrations of Radon-222 in water samples collected from selected sources in Akoko-South West Local Government Area, of Ondo State of Nigeria.

- Estimate the annual effective doses due to intake of Radon-222 from the measured samples for Adults, Children and Infants.
- Study the effect of aeration on radon concentration in the water samples.
- Compare the result with intervention levels of relevant international organizations.

1.5. Justification

The problem of Radon-222 is still an issue of concern in the current scientific world. It is well known that Radon-222 and its progenies contribute more than 50% of the total effective dose from natural sources (Alghamdi et al., 2019). It has been reported that a disease of major concern associated with radon is lung cancer from inhalation (Messier and Serre 2017).Radon-222 is the second most frequent cause of lung cancer and number one among non-smokers (USEPA 2018). Research has shown that these diseases could be a result of both chronic and acute dose exposure. USEPA has also estimated that 15,000-25,000 people died of Radon-222 induced cancer per annum in USA (USEPA, 2019). Virk and Singh (1993) recorded that certain rocks including granites, light-coloured volcanic rocks, sedimentary rocks containing phosphate and metamorphic rocks have higher average Uranium contents. In Nigeria, most areas lack established data on the activity concentration of Radon-222. Akoko South-West Local Government of Ondo State is one of such areas where inhabitants rely heavily on untreated surface and groundwater sources for drinking. Since its geology reveals that the area is enriched in granite an investigation into radon levels in surface and groundwater sources becomes necessary. This research seeks to address the problem of lack of baseline data and information on the activity concentration of Radon-222 in this region of the Ondo State and also determine the effect of aeration on the concentration of radon.

1.6. Operational Definition of Terms

- Decay (Radioactive): The transformation of a radioactive nuclide into a different nuclide (or nuclides) by the spontaneous emission of radiation such as alpha, Beta or Gamma rays.
- Alpha Particle (α): A positively charged particle consisting of two protons and two neutrons that are emitted by the nuclei of some radioactive elements as they decay.
- Beta particle (β): it is a high-energy, high-speed electron or positron emitted by the radioactive decay of an atomic nucleus during the process of beta decay.
- Gamma rays (γ): Penetrating electromagnetic radiation emitted by an atomic nucleus during radioactive decay examples includes a high-energy, short wavelength form of ionizing radiation.
- Becquerel: the SI unit of activity of a radioactive substance. (1Bq = 27pCi).
- Activity: The number of nuclear disintegrations in a radioactive material per unit time (it is used as a measure of the amount of radionuclide present in the material).
- Dose: A general term used to refer to the amount of energy absorbed by tissue from ionizing radiation
- Effective Dose: A measure of the dose to a tissue or organ designed to reflect the amount of harm caused to the tissue or organ.
- Sievert: The unit of effective dose. It is equal to 1 joule/kilogram (symbol Sv).
- Radon: A chemical element with symbol the Rn and atomic number 86. Radon is a colourless, odourless, tasteless, naturally occurring, radioactive noble gas that is formed. from the decay of Radium.

- Aeration: it is the process by which air is circulated through, mixed with or dissolved in a liquid or substance.
- Groundwater: it is the water present beneath the earth's soil in pore space and fractures of rock formations.
- Cancer: it is a disease caused by an uncontrolled division of abnormal cells in a part of the body.
- Water contamination: it is the presence of a chemical, biological or radiological substance in water in an amount that is dangerous to health.

CHAPTER TWO

LITERATURE REVIEW

2.1 Review of previous works

This chapter takes a review of what other researchers have done on the determination of radon concentration level in groundwater, mitigating techniques for the treatment of radon gas in water and also a brief allusion to some relevant basic concepts of the subject matter of study.

Jastaniah et al. (2014) evaluated a suitable technique for the treatment of drinking water that was artificially enriched with radon in laboratory by placing a radium rich granite stone (pitchblende) in a closed container filled with tap water for several days to allow radon concentration to approach its highest possible level. Experiments were designed to investigate the effectiveness of removal of radon by diffused bubble aeration method at room temperature. The results showed that this method becomes more efficient at higher air-to-water ratios. Better aeration depends on the length of travel of bubbles through the water depth. This method is practical and has low capital cost. The removal of radon from artificially enriched water can be practically achieved by diffused bubble aeration method to greater than 98%.

Mafra et al. (2011) in their work presented the results of the study of radon concentration reduction in well water using the aeration process developed at the Laboratory of Applied Nuclear Physics of the Federal University of Technology (UTFPR). The water samples were collected from a well at Pinheirinho region of Curitiba in 2011. The experimental setup was based on the Radon Monitor (AlphaGUARD). The radon concentration was analyzed using the software DataEXPERT by Genitron Instruments, taking into account the volume of water sample, its temperature, atmospheric pressure and the total volume of the air in the vessels. Initial concentration of radon in water samples was 28.67 Bq/L which was higher than the maximum concentration recommended by USEPA. The mitigation was performed by means of diffusion aeration of water samples of 15L during the time interval of 24 hours following a period of 4 days. The efficiency of aeration mitigation was controlled by comparing the activity of radon in aerated water with reference water samples that were not aerated. Obtained results show a very satisfactory decrease of radon activity in water samples even after few hours of intense aeration.

Zhuo *et al.* (2001) used radon bubbler and α -Scintillation to measure the activity concentration of groundwater at Fujian, China and found that the concentration is between (20-40) *Bq/L* which is above the Maximum Contamination Level (MCL) set by USEPA.

Badhan *et al.* (2010) measured radon concentration in groundwater using RAD7. Their results showed concentrations to vary from (2560-7750) Bqm^{-3} with no correlation observed between PH of water and Radon concentrations.

Zaini *et al.* (2011) used Gamma Spectrometry Technique and investigated the activity concentration of radon in water and found the concentration of radon to range from 0.29 Bq/L to 1.41 Bq/L which are lower than the Maximum Contamination Level (MCL) set by USEPA. Abdulsattar *et al.* (2015) used a solid-state nuclear track detector type CR to measure the concentration of Radon-222 and found the values to be within the range of (0.311-7.433) Bq/L which are within the recommended limits sets by United State Environmental Protection Agency (USEPA) and World Health Organization.

Sudhir et al. (2016) measured radon concentrations using RAD7 an electronic Radon detector and found the values to range from 0.5 to 22 Bq/L which is within recommended level of (4-40) Bq/L set by UNSCEAR, (2000).

In 2012, a research was carried out in the Kassena Nankana District of the Upper East region in Ghana, (Asumadu-Sakyi et al., 2012). The research focused on the levels and potential effect of radon gas in groundwater. Dissolved Radon-222 in sampled groundwater was analyzed by using High Purity Germanium (HPGe) Detector and Nuclear Track Detector (N.T.D) techniques. The radon concentrations obtained ranged from 7.86 $\times 10^{-6}$ to 8.18×10^{-5} Bq/l with a mean of 4.38×10^{-5} Bq/l using the Gamma Spectrometry (G.S) while that of N.T.D. ranged from 5.40 to 6.74 Bq/l with a mean of 19.54 Bq/l. It was revealed that the estimated annual effective dose by inhalation ranged from 6.05 to $40.66msv^{-1}$ with a mean value of $21.91msv^{-1}$ using the N.T.D, while that of Gamma Spectrometry, ranged from $1:39 \times 10^{-4}msv^{-1}$ to $2:45 \times 10^{-3}msv^{-1}$. Also, the estimated annual effective dose by ingestion ranged from $1:71 \times 10^{-5}$ to $1:32 \times 10^{-4} \mu svy^{-1}$ with a mean value of $1:60 \times 10^{-10} \mu svy^{-1}$ respectively.

Przylibski et al. (2004) carried out a research on Radon concentration in groundwater of the Polish part of the Sudety Mountain (SW Poland). The method adopted in the research was the PicoRad method, based on the liquid-Scintillation counter Packard-Canberra TRI CARB 1900 TR, for the determinations. It was discovered that the concentration of radon 222 ranged between 0.2 and 1645 Bq/dm⁻³. It was concluded that the obtained results only allowed speaking about typical or the most frequent values of radon concentration in the groundwater of the Polish part of the Sudety. The results of the research made it possible to declare that the Sudetes as a whole were an anomalous area when it comes to the occurrence of high radon concentration in the groundwater of Poland.

Garba et al. (2013) investigated groundwater samples from various locations of Zaria and environs for their Radon-222 concentrations by liquid scintillation counter. The concentration of Radon-222 in open well water was found to vary in the range (0.77-28.37) Bq/L and (2.32-48.80) Bq/L for borehole, with a geometric mean of 12.43 and 11.16Bq/L for borehole and well sources respectively. The results showed that Radon-222 concentration in borehole sources is higher than that of well water sources and were both greater than the maximum concentration limit (MCL) of 11.1Bq/L set by USEPA.

Njinga, et al. (2015), carried out a research on radioactivity analysis in underground drinking water sources in Niger State University of Nigeria The activity concentration of gross alpha and gross beta particles in four samples of borehole drinking water consumed in Ibrahim Badamasi Babangida University (IBBU), Lapai, Niger State-Nigeria was measured, using a portable single channel gas filled proportional counter (MPC2000B-DP) detector. This study focused on cancer related problems and the bio-data of the environment was discussed as well as the radiological effect of the water on consumers. Higher concentration of alpha and beta were observed in Hostel block A (DD) with values of 0.085 ± 0.024 and $11.229 \pm 0.901 BqL^{-1}$, respectively. However, lower concentration of alpha and beta particles were observed in the Faculty of Management Science (AA) with values of 0.006 ± 0.005 and $0.001 \pm 0.276BqL^{-1}$, respectively. Out of the four sampling sites studied, only the Faculty of Management Science fall below the guideline levels of gross alpha ($0.5BqL^{-1}$) and gross beta ($1.0BqL^{-1}$) in drinking water, established by the World Health Organization. These results show that, consumption of groundwater from the other three major borehole sources, may pose significant radiological health hazards to the population.

Oni et al. (2016) investigated a total of 64 water samples from Ado-Ekiti using calibrated active electronic detector RAD7 connected to RAD H_2O accessory and found that 53% of the results were above 11.1 *Bq/L* MCL suggested by USEPA.

Ademola and Oyeleke (2017) using a liquid scintillation counter investigated the concentrations of Radon-222 in 84 water samples in the city of Ibadan and found the values to range from 2.18 to 76.75 Bq/L. The corresponding annual effective doses were also found to range from 0.036 to 1.261 mSv/y, 0.071 to 2.521mSv/y and 0.042 to 1.471 mSv/y for adult, children and infants respectively.

Kendall and Smith (2002) discussed the doses from radon and from its short-lived decay products to a number of organs, tissues and the fetus. The aim was to put all these doses into context rather than concentrating only on the largest contributions. There is also a brief discussion of the evidence from epidemiology on the risks of exposure to radon and its decay products. As is well known, under normal circumstances the greatest hazard is to the respiratory tract from inhalation of radon decay products. Radon decay products may also give substantial doses to the skin. Under some circumstances it seems likely that ingested radon could give significant doses to the stomach.

Binesh et al. (2010). In their research collected drinking water samples from various places in Mashhad city which has about 4 million population. The radon concentration was measured by PRASSI system three times for each sample. Results show that about 75% of water samples have radon concentration gathered than 10 Bq/L which advised EPA as a normal level. According to measurements data, the arithmetic mean of radon concentration for all samples was 16.238 ± 9.322 Bq/L. Similarly, the annual effective dose in stomach and lung per person has been

evaluated in this research. According to the advice of WHO and EU Council, just 2 samples induced the total annual effective dose greater than 0.1 mSv/y.

Ali et al. (2010) collected Different samples of water, indoor air and soil gas from Islamabad and Murree. They analyzed for the estimation of mean effective dose through radon concentrations by using RAD-7, a solid-state detector. The variation of radon concentration in water, indoor air and soil gas in Islamabad region ranges from $25.90-158.40 \text{ kBqm}^{-3}$, $43.26-97.04 \text{ Bqm}^{-3}$ and $17.34-72.52 \text{ kBqm}^{-3}$, having mean values 88.63 kBqm^{-3} , 70.67 Bqm^{-3} and 45.08 kBqm^{-3} , respectively. It ranges from $1.64-10.20 \text{ kBqm}^{-3}$, $18.48-42.08 \text{ Bqm}^{-3}$ and $0.61-3.89 \text{ kBqm}^{-3}$ with mean values 4.38 kBqm^{-3} , 28.63 Bqm^{-3} and 1.70 kBqm^{-3} respectively in Murree and its surroundings. The total mean annual effective doses from water and indoor air of Islamabad and Murree regions are 2.023 and 0.733 mSv/y, respectively. These doses are within the recommended limits of the world organizations.

Ravikumar and Somashekar (2014) determined the distribution of radon in ground and surface water samples in Sankey Tank and Mallathahalli Lake areas using Durridge RAD-7 analyzer with RAD H₂O accessory. The radiation dose received by an individual falling under different age groups depending upon their average annual water consumption rate was attempted. The mean radon activity in surface water of Sankey Tank and Mallathahalli Lake was 7.24 ± 1.48 and 11.43 ± 1.11 Bq/L, respectively. The average radon activities ranged from 11.6 ± 1.7 to $381.2 \pm$ 2.0 Bq/L and 1.50 ± 0.83 to 18.9 ± 1.59 Bq/L, respectively, in 12 groundwater samples each around Sankey Tank and Mallathahalli Lake areas. Majority of the measured groundwater samples showed mean radon values above the EPA's maximum contaminant level of 11.1 Bq/L and only 66.67 % of samples in Sankey Tank area showed radon above the WHO and EU's reference level of 100 Bq/L. The overall radiation dose due to radon emanating from water in the study area was increasing with an increase in age and water consumption rates, but significantly lower than UNSCEAR and WHO recommended limit of 1 mSv/year except for few groundwater samples in Sankey Tank area (i.e., 0.92, 0.99 and 1.39 mSv/year). The radiation dose rate received by bronchial epithelium via inhalation was very high compared to that by stomach walls via ingestion.

Caridi and Belmusto (2018) reported experimental results obtained, with different diagnostics setups, for radon activity concentration measurement in underground water for human use. An overview is given about the performance of different measurement techniques, based on experimental data. The following parameters are compared and discussed: counting efficiency, minimum detectable activity, measurement uncertainty, background, sample volume and treatment. The estimated average value for radon-specific activity in underground water was compared with that one derived from different legislations and directives/guidelines and it was used, with the dose conversion factor for Radon-222, to estimate the annual effective dose, for adult members of public of the investigated region, due to the groundwater radon ingestion.

Kalip et al. (2018). The variation in the concentration of radon in groundwater sources comprising of boreholes and wells in Kaduna metropolis and environs were determined by using Tri-carb LSA 1000 liquid scintillation counter. The radiation dose received by individuals within different age groups categorized under; infants, children and adults, depending on their average annual water consumption rates (ACRs) were also estimated. The mean radon activity in 16 boreholes and 18 well water samples were 1.8/Bq/L and 0.57 Bq/L respectively; while the average radon activities ranged from 0.85 to 2.57 Bq/L and 0.35 to 0.85Bq/L respectively with all values far below the United States Environmental Protection Agency MCL of 11.1 Bq/L. All the estimated annual committed effective dose (ACED) for all samples were observed to increase

with radon concentration, age and ACRs, but were significantly lower than the United Nation Scientific Committee on Effect of Atomic.

Faweya, et al. (2018) investigated radon concentration in water and heavy metals concentrations in sediment samples from historical cold and warm springs at Ikogosi using Durridge RAD-7 analyzer with RAD H₂O accessory and atomic absorption spectrophotometer. The mean activity concentration of radon in water samples ranged from 0.07 to 0.36 with overall mean value 0.20 Bq/L, 35–210 with an overall mean value 75.9Bq/L and 11.7–140.0 with an overall mean 79.4 Bq/L for bottled, cold and warm water samples respectively. The calculated total effective dose values were below 100 μ Sv/y recommended by WHO. The result of elemental analysis showed that the mean values of metals concentrations were Pb (2.9–11.8 mg/kg), Cu (3.8–12.8 mg/kg), Fe (945.0–2010.0 mg/kg), Cd (0.6–1.7 mg/kg) and Ni (0.3–2.6 mg/kg). The results revealed values not higher than recommended permissible limit and background values. The pollution load index revealed that the overall contamination of metals indicated no significant pollution in all the studied samples.

Aruwa et al. (2017) Studied groundwater samples from selected boreholes and wells in Idah and determined the concentration of radon using the Liquid Scintillation Counter (LSC). The average concentration of radon obtained was 14.09±1.10 Bq/L for boreholes and 13.45±1.00 Bq/L for well waters. The overall average concentration of 13.77±1.05 Bq/L was recorded. The results obtained in this work were compared with the maximum contamination level (MCL) of 11.1 BqL -1 set by USEPA and it was observed that 80% of the samples exceeded these values. The average annual effective dose by ingestion of 0.051mSv/y was recorded for boreholes and 0.049mSv/y for well water samples. All values of effective dose were below the ICRP recommended intervention level of 3-10mSv/y.

Akinnagbe et al. (2018) investigated radon concentration in groundwater in Ijero, Ekiti State, using the RAD7/RAD H_2O driven alpha spectrometry technique. The results were used to estimate the annual effective committed doses in order to establish possible radiological health hazards and to suggest necessary safety measures. The minimum and maximum radon concentrations in the samples were 0.168 Bq/L and 78.509 Bq/L from stream and borehole. samples, respectively. 45% of the samples radon concentration exceeded 11.1 Bq/L, the maximum permissible limit. It was observed that none of the samples has radon concentration value up to 100 Bq/L, which is recommended by the European Union to be the upper bound value, above which remedial action is required. None of the samples had an annual effective dose higher than the maximum permissible limit of 0.2 mSv/y if consumed by children and 0.1 mSv/y if consumed by adults. The relatively high levels of radon indicate a certain level of health risk. Though the effective dose seemed low, effects of prolonged exposure to radiation are still possible

Oni and Adagunodo (2019) investigated the level of radon in drinking water in Ogbomoso using an active electronic device RAD 7, produced by Durridge Company USA. The radon concentrations within the study area vary from 0.60 to 2.64 Bq/L, with the mean value of 1.86 Bq/L. The committed annual effective doses due to ingestion vary from 6.25×10^{-3} to 1.93×10^{-2} mSv/y, with mean values of 0.02 mSv/y. The radon concentrations in water samples of Ogbomoso are lower than the threshold as set by both United States Environmental and Protection Agency, and European Commission of 11 and 100 Bq/L respectively.

2.2. Aeration

Aeration is the process by which air is circulated through, mixed or dissolved in a liquid or substance. Any procedure by which oxygen is added to water can be considered a type of water aeration. This being the only criterion, there are a variety of ways to aerate water. This fall into two broad areas namely surface aeration and subsurface aeration. Aeration water treatment is effective for the management of dissolved gases such as radon, carbon dioxide, some taste and odour problems such as methane, and hydrogen sulfide, as well as volatile organic compounds, like MTBE or industrial solvents. It is also effective in precipitating dissolved iron and manganese Aeration raises the pH of water. Aeration is not effective for the removal of heavy metals, or pathogenic (disease-causing) organisms like bacteria and viruses.

Aeration is an in-line point-of-entry process that reduces the concentration of volatile organic compounds. Aeration also removes dissolved gases such as hydrogen sulfide, methane, and radon. Aeration oxidizes dissolved iron, although the resulting iron particles can foul the packing material in some aeration devices.

Two types of treatment processes have been widely used to remove radon from water supplies by aeration technique namely diffused bubble aeration system and packed lower aeration system. The stimulated water aeration is considered by USEPA as the best available method for removal of radon from water. The physical factors that have to be considered for evaluation of its efficiency are: the time interval of water aeration, the intensity of air flow, the quantity of water, its surface area, the temperature of water, atmospheric pressure (Drago 1998). As it was estimated by USEPA(2018) this mitigation method can reduce by 99% the concentration level of

radon in water and according to (Alabdula'aly and Maghrawy 2011) this method can reach the efficiency above 97%.

2.2.1. Natural Aeration

This is a type of both sub-surface and surface aeration. It can occur through sub-surface aquatic plant. Through the natural process of photosynthesis, water plant releases oxygen into the water. Surface aeration occurs when oxygen is driven into the water when wind disturbs the surface of water bodies. It can also occur through the movement of water caused by an incoming stream, waterfall or even a strong flood.

2.3. Radon-222

Radon is a chemical element with the symbol Rn and atomic number 86. It was the fifth radioactive element to be discovered and it was discovered in 1900 by Fredrich E. Dorn in Halle, Germany. He described it as radium emanation because it arose from the element radium, which he was working with. It is a naturally occurring radioactive inert gas with half-life of 3.82 days which is a member of the Uranium-238 decay series (Somlai, 2007). It is a colourless, odourless, tasteless and chemically non-reactive gas formed from the radioactive decay specifically of Radium-226, a decay product of Uranium-238 (USEPA, 1999). Low concentration of Uranium and its decay product soccur widely in the earth's crust and thus Radon-222 is continually being generated. Radon-222 emits alpha particle during its decay to Polonium. The exposure of human beings to Radon is dual, which is through inhalation of radon released into the ambient air and through ingestion when water is used for drinking. Radon-222 activity concentration in water is due to the decay of Radium-226 associated with the rock and soil. The radon gas percolates through the soil and rock and dissolves in the water. Therefore, the concentration of Radon in

water is higher than one would expect (Xinwei, 2006). Normally, the activity concentration of radon in groundwater is higher than that in surface water. This is because radon is a gas and when water that contains radon is exposed to the air the radon tends to be released into the air. However surface water polluted with Natural Occurring Radioactive Materials (NORM) from human activities such as mining and fertilizer production is prone to high activity concentration of radon. People who are exposed to radon in drinking water may have an increased risk of getting cancer over the course of their lifetime (UNSCEAR, 2018). Uranium-238 and Thorium-232 are the major contributors of Radon in nature. Figures 2.1 to 2.3 show the Uranium decay and thorium decay series.

Radon in general occurs as three natural isotopes derived from three different radioactive decay chains commencing with Uranium-238, Thorium-232 and Uranium-235 as shown in Table 2.1.

2.6. Sources of Radon

2.6.1 Water

Surface and underground water contain radionuclides as natural components in various . concentrations depending on their origin. Radon is released into the water as a result of natural processes like decay of its parent nuclide Radium-226 and predominantly dissolution from the surrounding geological environment, (Rocks and soil). (Moreno *et al.* 2014. Fonollosa *et al.* 2016). A portion of the radon released through radioactive decay moves through air or water sedimentary rocks containing Phosphate and metamorphic rocks are recorded to have higher Uranium contents (Virk and Singh, 1993). Radon gas is released into the atmosphere from the ground through its diffusion in the pore spaces in the soils and it significantly contributes to

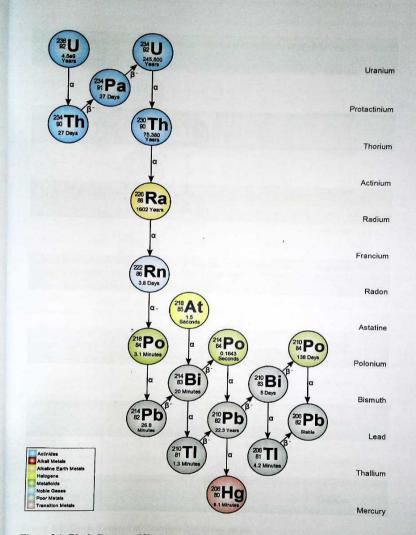
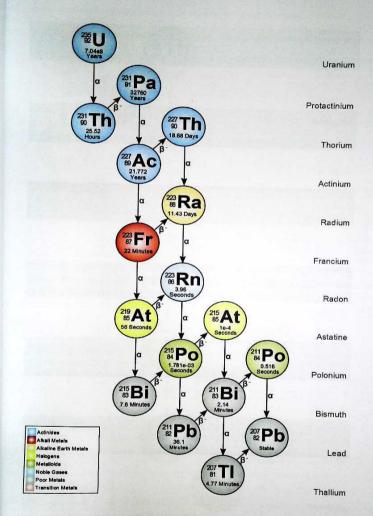
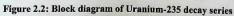


Figure 2.1: Block diagram of Uranium-238 decay series





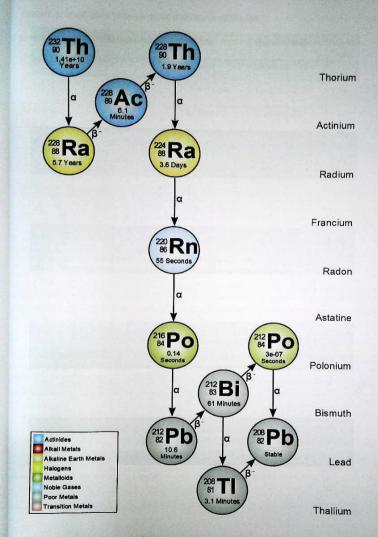


Figure 2.3: Block diagram of Thorium-232 decay series

| Table 2.1: Half-life's of three | natural Isotopes of Radon |
|---------------------------------|---------------------------|
|---------------------------------|---------------------------|

| Common name | Half He | |
|-------------|--------------|---------------------------------|
| | man-me | Decay Chain |
| Radon | 3.82 days | Uranium-238 |
| Thoron | | orumum-250 |
| - moron | 54.5 Seconds | Thorium-232 |
| Actinon | 2020 | |
| | 3.92 Seconds | Uranium-235 |
| | | 3.82 days Thoron 54.5 Seconds |

filled pores in the soil to the soil surface and enters the air while some remain below the surface and dissolves in groundwater (USEPA, 1999). The solubility of radon is commonly expressed by the partitioning coefficient of Radon-222 between pure solvent and air (Schubert *et al.*, 2007). Groundwater from wells and boreholes usually contain higher radon concentrations than surface water, because surface water is exposed to the air and since radon is a gas it tends to escape into the air. Also, the concentration of Radium-226 in the aquifer matrix is the basis for the high activity concentration of radon in groundwater (Vinson *et al.*; 2009). The exposure of people to radon in water is dual, which are, by inhalation of the radon released into the ambient air when water is used for domestic activities and by ingestion when water is used for drinking (Abdalsattar and Rajaa, 2015).

2.6.2 Soil

Soil is another source of radon. It is the uppermost layer of the earth in which plant grows. It is formed typically from the weathering of rocks. Radon in soil is mainly from rocks containing some level of Uranium-238, where the decay of Radium-226 from Uranium-238 decay gives Radon-222. Certain types of rocks, including granites, dark Shale, light-coloured volcanic rocks, elevated indoor radon gas concentration through the same mechanism including convection via cracks and openings, diffusion from the soil via pore space in building materials and emanation from building materials (Lee et al 2019).

2.7 Health Effects of Radon

The adverse health effects which depend on the level of exposure to radon are caused primarily by damage due to alpha particles. There is no immediate symptom to radon exposure but long term exposure to radon with high activity concentration has been linked to cancer of the lungs (Messier and Serre 2017). Stomach cancer is linked with a very high radon level in drinking water (Raquel et al., 2017).

Lung cancer from inhalation is the most common health effect of radon, aerosol or smoke are readily deposited in the airways of the lung, while lodged there, the progeny emit ionizing radiation in form of alpha particle which can damage the bronchial epithelial cells and cause cancer (Turner et al. 2012).

2.8 Radon guidelines for drinking water

In order to protect the health of citizens from radiation in drinking water, different radon levels are introduced by different international organizations. Water intended for human consumption the Euratom Drinking Water Directive (E-DWD) establishes parametric values (E-DWD, 2013), World Health Organization (WHO) uses guidance level (WHO, 2008) while in the United States maximum contamination levels are introduced (USEPA2018). The established guidance levels and parametric values based on whether that value poses a risk to human health from a radiation protection point of view or not (that is if further remediation action is needed or not). However, the action level is not a boundary between safe and unsafe, but rather a level at which the action on the reduction of radon level will be justified. Table 2.2 shows some of these guidelines from different countries.

| Directive/ Recommendation | Activity Concentration (<i>Bq/L</i>) | Source |
|--|--|----------------------------------|
| EURATOM DWD | 100-1000 | EURATOM, 2013 |
| Ireland, Portugal, Spain (E-DWD) | 100 | EURATOM, 2013 |
| | | MS National law |
| 24 Member States (E-DWD) | 500 | EURATOM, 2013 MS National law |
| Finland (E-DWD) | 1000 | EURATOM, 2013 MS National law |
| WHO guidance level | 100 | WHO, 2008 |
| US-EPA Maximum Contaminant Level (MCL) | 11.1 · | USEPA (2018) |
| US-EPA alternative higher MCL | 148 | USEPA (2018) |
| | | ι |

Table 2.2: International Radon Guidance and Parametric Values in Drinking Water

The 24 member states are: Austria, Belgium, Bulgaria, Croatia, Cyprus, Czech Republic, Denmark, Estonia, France, Germany Greece, Hungary, Italy, Latvia, Lithuania, Luxembourg, Malta, Netherlands, Poland, Romania, Slovakia, Slovenia, Sweden and United Kingdom.

CHAPTER THREE

MATERIALS AND METHOD

3.1 Study Area

The sample collection points fall within Akoko South West Local Government Area. It is located in the northern senatorial district of Ondo State southwestern Nigeria as depicted in Figures 3.1 ^o and 3.2. It lies between latitude7°20' N' and 7°30'N and longitude 5°30'E and 5°50'E. Akoko South West L.G.A has a land size of about $226km^2$, with a population of about 229,486 as at 2006 national census. The people are predominantly farmers, civil servants and traders.

3.1.1 Geology of the Study Area

Ondo state has two distinct geological regions. First is the region of sedimentary rocks in the south and Precambrian basement complex rocks in the north. It is also composed of lowlands and rugged hills with granitic outcrops in several places. In general, the land rises from the coastal part of ilaje/eseodo (less than fifteen meters above sea level) in the south, to the rugged hills of the north eastern portion in Akoko area.

Akoko South West L.G.A. is basically underlain by the Precambrian rock of southern Nigeria. The most predominant rock type in the area is migmatite and granite/granite gneiss. (Falowo et al., 2017) The basement rock exposures are however as lowland outcrop in few places within the local government area particularly where the basement is shallow and erosional activities are active (Mohammed et al 2012).

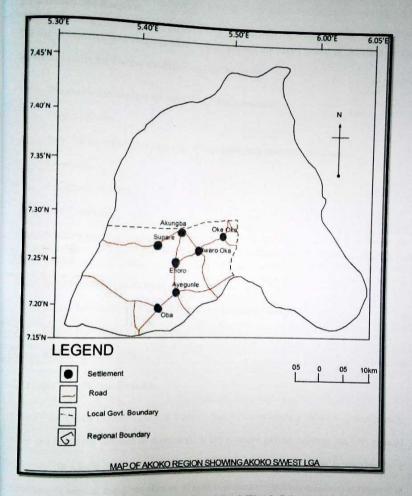


Figure 3.1: Map of Akoko Area showing Akoko South West L.G.A.

3.2 Materials and Reagents

The list of materials and reagents used in this research are stated below as described by the American Society for Testing and Materials (ASTM, 1999).

- I. Disposable hypodermic syringe (20ml, 10ml capacities).
- II. Surgical gloves.
- III. Scintillation vials (20ml capacity) with polyethylene inner seal cap liners.
- IV. Detergent.
- V. Scintillation cocktail.
- VI. Distilled water.
- VII. Masking tape
- VIII. Marker
- IX. Indelible ink and Masking tape.
- X. Liquid Scintillation Counter (Packard Tri-Card LSA 1000TR).
- XI. Global Positioning System (GPS).

XII. A piece of clean cloth

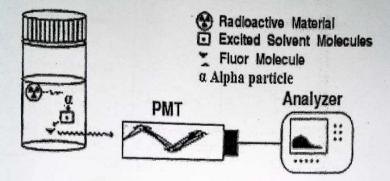
3.3 Liquid Scintillation Counter.

Liquid Scintillation Counter (LSC) is a device that measures ionizing radiation, predominantly alpha and beta from radioactive substances. It can measure gamma radiation but with reduced efficiency. In Liquid Scintillation Counters, energy from emitted radiation is absorbed by a scintillator (a material that gives photons of light in response to incident radiation) and re-emit as photons. The emitted light photons are detected by a sensitive photomultiplier tube (PMT) attached to electrical amplifiers and converted to electrical energy for analysis. In this way, each emission results in pulse of light as shown in Figure 3.2

3.4 Method

3.4.1 Sample collection

A total of 51 samples were collected for analysis from the selected groundwater (Boreholes and open wells) sources at different locations in Akungba-Akoko, Oka-Akoko, Oba-Akoko, Ayegunle-Akoko, Iwaro Oka-Akoko, Supare-Akoko and Etioro. Figures 3.5 and 3.6 are representatives of open wells and hand-pump from which water samples were sourced from. During sampling, a global positioning system was used to mark the geographical locations, on the earth surface, of the sample collection points. Samples were collected using plastic vials of 30mL. Hand-pumps were pumped and allowed to flow for at least two minutes before samples were collected in order to ensure that fresh samples were obtained. Water samples from open wells were first collected with bailers and then transferred into vials (Figures 3.7 and 3.8). Each collected sample was properly labeled and the time of sample collection was noted and recorded. The samples at all the locations were collected in three different categories labeled A, B and C. Category A were capped immediately at the source while category B and C were left open for aeration for twenty-four hours (24hrs) and seventy-two hours (72hrs) respectively (Figure 3.9) before capping. The essence of those various levels of aerations in samples B and C is to observe the effect of natural aeration on the concentration of radon-222 gas with respect to the various degrees of exposure to the open air. Three (3) samples each was collected from seventeen (17) sample collection points making a total of fifty-one (51) samples as shown in Table 3.1.



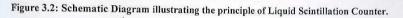


Table3.1. Communities and number of samples collected

LOAT

LCOLO

| S/N | COMMUNITY | HAND-PUMP | NUT T | |
|-----|-----------------|-----------|-------|--------------------------------|
| 1 | Akungba-Akoko | 3 | WELLS | NO. OF SAMPLES COLLECTED |
| 2 | Ayegunle-Akoko | 1 | 1. | 12 |
| 3 | Etioro-Akoko | 1 | | 3 |
| 4 | Iwaro Oka-Akoko | | | 3 |
| 5 | Oba-Akoko | 1 | 2 | 9 |
| 6 | Oka-Akoko | 1 | 2 | 9 |
| 7 | Supare-Akoko | 3 | 4 | 9 |
| | | 10 | - | |
| | | | 7 | 51 |



Figure 3.3: One of the Open wells from which sample was collected.



Figure 3.4: One of the Hand-pump from which sample was collected.



Figure 3.5: Vials in Counting Rack.



Figure 3.6: Counted Samples in Secondary Containment



Figure 3.7: Some samples that were exposed.

3.4.2 Sample preparation

10 ml of each sample was added into a vial containing 10 ml of insta-gel based cocktail (scintillator) using a hypodermic syringe. The vials were tightly capped and shaken vigorously for three (3) minutes to extract radon-222 in the water phase into the organic scintillator (Forte et al., 2006). In a similar manner, a blank sample for the background was prepared using distilled water that has been kept in a glass bottle for at least 21 days. The prepared samples were allowed to stand undisturbed for at least three (3) hours each in order for Radon-222 and its alpha decay products attain equilibrium before counting.

3.4.3 Sample Analysis

The prepared samples and the blank were each analyzed using the Liquid Scintillation Counter (Tri-Card LSA 1000) at the Center for Energy Research and Training (CERT), Ahmadu Bello University Zaria, Kaduna State, Nigeria. The Liquid Scintillation Analyzer (Tri-carb-LSA 1000) is shown in Figure 3.8. The printer attached to the Liquid Scintillation Analyzer is shown in Figure 3.9.

Radiation emitted from the samples transferred energy to the organic scintillator which in turn emits light photons. This way each emission result is a pulse of light in form of a digit.

The activity concentration of Radon-222 was calculated from the samples and background results obtained using equation 3.1: (Joseph et al 2018)

$$C_{Rn}(BqL^{-1}) = \frac{100 \times (SC - BC)exp\lambda t}{60 \times CF \times D}.$$

(3.1)

Where:

 $C_{Rn}(BqL^{-1})$ = Concentration of Radon-222 in Becquerel per litre.

SC= Sample Count (Count per min),

BC = Background Count (Count per min).

t =Time elapsed between sampling to counting (minutes),

 $\mathbb{P} = \text{Decay constant} (1.26 \times 10^{-4} min^{-1}).$

 $CF = Calibration factor (13.37 Bq^{-1})$

D = Fraction of Radon-222 in the cocktail in a 22 ml total capacity vial for 10ml of sample, 10 ml of cocktail and 2 ml of air.

The corresponding annual effective doses (mSv/y) due to ingestion of Radon-222 in water samples were also calculated taking into account the dose coefficient in (Sv/Bq), the annual water consumption (L/Y) and the activity concentration of Radon-222 obtained from equation 3.1 using the equation 3.2.

Where:

$$E(mS/y) = C_{Rm} \times D \times L$$

(3.2)

 C_{Rn} = Concentration of Radon-222(BqL⁻¹),

L= Annual water consumption of 2 litres, 1.5 litres and 0.7 litres per day that is 730L/Y, 547.5 L/Y. and 255.5L/Y. for adults, children and infants respectively (Malakootian and Nejhad, 2017).

D= Dose coefficient $(10^{-8}Sv/Bq, 2\times 10^{-8}Sv/Bq, 7\times 10^{-8}Sv/Bq)$ for adults, children and infants respectively. (UNSCEAR, 2000).

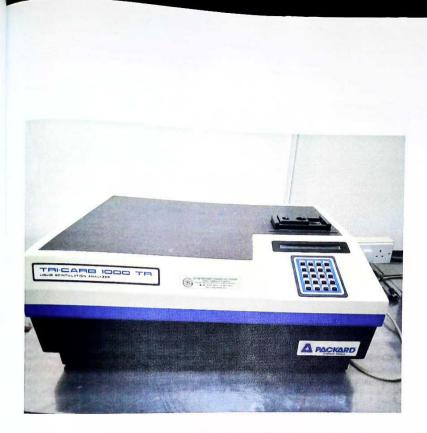


Figure 3.8 Liquid Scintillation Analyzer (Tri-carb- LSA 1000), Centre for Energy Research and Training (CERT), Ahmadu Bello University, Zaria.



Figure 3.9 Printer attach to the Liquid Scintillation Analyzer.

CHAPTER FOUR

RESULTS AND DISCUSSIONS

The activity concentration of Radon-222 of the fifty-one (51) water samples collected at different . Jocations in Akoko South West Local Government Area of Ondo State as well as the corresponding annual effective doses due to ingestion of radon in water for the three categories of people were discussed.

4.1 Results

Radon concentration and Annual Effective Dose measured in samples for categories A, B and C as described in Chapter 3 are respectively presented in Tables 4.1 to 4.3.

4.2 Discussions

4.2.1. Radon concentrations values for samples capped immediately after collection

The mean values of Radon-222 concentrations were found to be 28.01 and 25.34 Bq/L for handpumps and open wells respectively with overall mean value of 26.91Bq/L as shown in Table 4.1. These results have shown that 94.10% and mean value of Radon-222 concentrations were above the USEPA MCL of 11.1 Bq/L.

4.2.2. Radon concentrations values for samples capped 24 hours after collection

The mean values of Radon-222 concentrations were found to be 20.18 and 14.82 Bq/L for hand-^{pumps} and open wells respectively with overall mean value of 17.97Bq/L as shown in Table 4.1. These results have shown that 64.7% and the mean value of Radon-222 concentrations were ^{above} the USEPA MCL of 11.1 Bq/L

Table 4.1: Radon concentration for the three different categories

| S/N | SAMPLE | LATITUDE | Lover | | | | |
|-----|-----------|---------------|------------------|-----------|--------|--------|--|
| | | | LONGITUDE | RADON | RADON | RADON | |
| | | | West Reading and | CONC | CONC | CONC | |
| | | | | (Bq/L) | (Bq/L) | (Bq/L) | |
| 1 | Akungba 1 | 7°28'8.6160" | | CAT. A | CAT. B | CAT. C | |
| | | | 5°44'1.8132" | 20.44 | 13.13 | 4.94 | |
| 2 | Akungba 2 | 7°28'16.0968" | 5°44'14.0280" | 14.52 | 9.71 | 4.07 | |
| 3 | Akungba 3 | 7°28'29.5968" | 5°44'13.974" | 12.61 | 8.50 | 3.67 | |
| 4 | Akungba 4 | 7°28'34.9608" | 5°44'29.8968" | 10.30 | 6.75 | 3.22 | |
| 5 | Ayegunle | 7°24'57.4308" | 5°43'26.7168" | 27.42 | 18.40 | 4.71 | |
| 6 | Etioro | 7°26'22.056" | 5°43'24.960" | 14.95 | 10.06 | 8.91 | |
| 7 | Iwaro 1 | 7°26'57.1812" | 5°44'56.0508" | 15.30 | 10.33 | 5.47 | |
| 8 | Iwaro 2 | 7°26'58.7472" | 5°44'54.1428" | 15.59 | 11.01 | 5.50 | |
| 9 | Oba 1 | 7°22'12.252" | 5°43'38.8632" | 21.80 | 16.39 | 7.83 | |
| 10 | Oba 2 | 7°22'21.9432" | 5°43'19.442" | 21.50 | 15.57 | 7.89 | |
| 11 | Oba 3 | 7°22'45.3288" | 5°43'34.1868" | 38.78 | 18.15 | 10.61 | |
| 12 | Oka 1 | 7°27'18.414" | 5°48'50.0328" | 33.71 | 16.69 | 7.47 | |
| 13 | Oka 2 | 7°27'24.4548" | 5°48'7.5168" | 41.89 | 24.39 | 9.37 | |
| 14 | Oka 3 | 7°27'12.3228" | 5°47'38.0868" | 31.95 | 24.82 | 10.69 | |
| 15 | Supare 1 | 7°26'46.5070" | 5°41'25.098" | 27.98 | 26.22 | 13.44 | |
| 16 | Supare 2 | 7°27'16.0992" | 5°41'26.61" | 57.50 | 37.94 | 18.33 | |
| 17 | Supare 3 | 7°27'19.9332" | 5°42'12.2652" | 51.25 · · | 37.48 | 16.69 | |
| | | | Mean values | 26.91 | 17.97 | 8.40 | |

4.2.3. Radon concentrations values for samples capped after 72 hours

The mean values of Radon-222 concentrations were found to be 9.33 and 7.07 Bq/L for handpumps and open wells respectively with overall mean value of 8.40Bq/L as shown in Table 4.1. These results have shown that 17.6% and the mean value of Radon-222 concentrations were above the USEPA MCL of 11.1 Bq/L.

4.2.4. Analysis of Radon-222 in hand-pump samples.

The result of the analysis of radon concentrations for the ten (10) hand-pump water samples collected at different locations Akoko South-West Local Government Area as present in Table 4.2 revealed that the concentrations of Radon-222 varied from 12.61 Bq/L to 57.50 Bq/L with a mean value of 28.01Bq/L for the category A samples. The concentrations of Radon-222 varied from 8.504 Bq/L to 37.944 Bq/L with a mean value of 20.18 Bq/L for the category B samples and the concentrations of Radon-222 varied from $3.672 \ Bq/L$ to 18.326 Bq/L with a mean value of 9.33Bq/L for the category C samples.

The maximum concentration was obtained from Supare-Akoko while the minimum concentration was found at Akungba-Akoko as shown in Table 4.2 and Figure 4.1 All the values obtained from category A samples were found to be above maximum contaminant levels while ^{70%} of category B were above the maximum contamination level and only 30% of category C samples were above the maximum contamination level of 11.1 Bq/L set by United State Environmental Protection Agency (USEPA, 2018)

Tsble 4.2: Radon-222 concentration in hand-D

| Samples | Location | RnConc | -pump samples | | |
|--------------|-----------|---------------|------------------|---------------|--|
| | | (Bq/L) | RnConc (Bq/L) | RnConc | |
| | 11 | of category A | of category B | (Bq/L) | |
| Hand-pump 1 | Akungba 1 | 20.44 | 13.13 | of category C | |
| Hand-pump 2 | Akungba 2 | 14.52 | | 4.94 | |
| Hand-pump 3 | Akungba 3 | | 9.71 | 4.07 | |
| | | 12.61 | 8.50 | 3.67 | |
| Hand-pump 4 | Ayegunle | 27.42 | 18.40 | 4.71 | |
| Hand-pump 5 | Etioro | 14.95 | 10.06 | 8.91 | |
| Hand-pump 6 | Oba 2 | 21.50 | 15.57 | 7.89 | |
| Hand-pump 7 | Oka 3 | 31.95 | 24.82 | 10.69 | |
| Hand-pump 8 | Supare 1 | 27.98 | 26.22 | 13.44 | |
| Hand-pump 9 | Supare 2 | 57.50 | 37.94 | 18.33 | |
| Hand-pump 10 | Supare 3 | 51.25 | 37.48 | 16.69 | |
| Mean values | | 28.01 | 20.18 | 9.33 | |

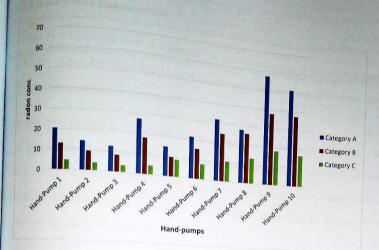
42.5 Analysis of radon in open well water samples

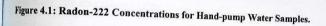
The result of the analysis of radon concentrations for the seven (7) open well water samples collected at different locations from Akoko South-West Local Government Area as presented in Table 4.3 and Figure 4.2 revealed that the concentration of Radon-222 varied from 10.30Bq/L to ' 41.88 Bq/L with a mean value of 25.34 Bq/L for category A samples. The concentrations of Radon-222 varied from 6.75 Bq/L to 24.39 Bq/L with a mean value of 14.82Bq/L for category B samples and the concentrations of Radon-222 varied from 3.224 Bq/L to 10.613Bq/L with a mean value of 7.07Bq/L for category C samples.

The maximum concentration was obtained from Oka-Akoko while the minimum concentration was found at Akungba-Akoko as shown in Table 4.3. All the values obtained from category A samples were found to be above maximum contaminant levels while 70% of category B were above the maximum contamination level and only 30% of category C samples were above the maximum contamination level of 11.1 Bq/L set by United State Environmental Protection Agency (USEPA, 2018)

Table 4.3: Radon-222 concentration

| Samples | Location | RnConc RnConc R | | | | | |
|------------------|-----------|-----------------|---------------|-------------------------|--|--|--|
| | | (Bq/L) | (Ba/T) | RnConc | | | |
| Well 1 | Akungba 4 | of category A | of category B | (Bq/L) of category C | | | |
| Well 2 | Iwaro 1 | 15.30 | 6.75 | 3.22 | | | |
| Well 3 | Iwaro 2 | 15.59 | 10.33 | 5.47 | | | |
| Well 4 | Oba 1 | 21.80 | 16.39 | 5.50 | | | |
| Vell 5 | Oba 3 | 38.79 | 18.15 | 7.83 | | | |
| Vell 6 Vell 7 | Oka 1 | 33.71 | 16.69 | 7.47 | | | |
| lean value | Oka 2 | 41.89 | 24.39 | 9.37 | | | |
| | | 25.34 | 14.82 | 7.07 | | | |





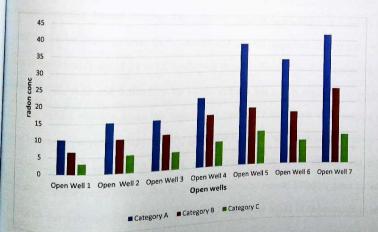


Figure 4.2: Radon-222 Concentrations for Open well Water Samples.

12.6. Mean Radon Concentrations

in order to compare the level of Radon-222 in the two categories of groundwater (wells and porcholes) collected from the study area, the mean concentration of each of the water types was

Samples from borehole have the highest mean concentration of Radon-222 compared to that of well water in the three categories. All the mean values exceeded the maximum contaminant _{byels} of 11.1 Bq/L set by USEPA (Table 4.4). However, the procedure of aeration brought the Radon-222 concentration for categories B and C to lower mean value as compared to mean radon concentrations of category A.

43. Annual Effective Doses (AED)

43.1 Annual Effective Doses (AED) of Category A samples

The corresponding Annual Effective Doses due to intake of Radon-222 from borehole water . samples collected were estimated for the samples and found to range from (0.09 to 0.42) mSv/y, (0.14 to 0.63) mSv/y and (0.23 to 1.03) mSv/y as shown in Table 4.7 with corresponding mean values of 0.21, 0.31 and 0.50mSv/y for adults, children and infants respectively. While the estimated annual effective doses due ingestion of Radon-222 from Well water samples were found to range from (0.08 to 0.31) mSv/y, (0.11 to 0.46) mSv/y and (0.18 to 0.75) mSv/y with corresponding mean values of 0.19, 0.28, and 0.45 mSv/y for adults, children and infants respectively. These results showed that 88.2% of the estimated Annual Effective doses for adults Were above the recommended reference level of 0.1 mSv/y for intake of radionuclide in water

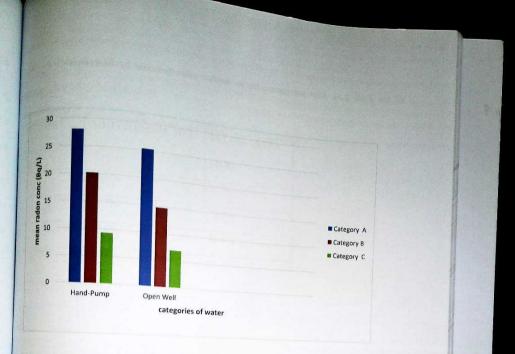


Figure 4.3: Mean Radon-222 Concentrations for the two Water Types

 f_{μ} ble 4.4 Comparison of concentration values of all categories with 11.1 Bq/L set by USEPA

| CATEGORY | ABOVE USEPA MCL | BELOW USEPA |
|----------|-----------------|-------------|
| | | MCL |
| Å | 94.1% | 5.9% |
| B | 64.7% | 35.3% |
| C | 17.6% | 82.4% |
| | | |

Table 4.5: Annual Effective Doses (AED) for the thr

| 1 | | | GORY A | | | | rent cate | gories . | | | |
|----------------|-----------|-------------|--------------------|-------------------|-----------------|-------------------------|-------------------------|------------------|--------------|--------------|--|
| SI N | LOC. AD | AED ADLT | AED AED ADLT CHILD | | | CATEGORY B | | | CATEGORY C | | |
| AN AN AN AN AN | | (mSv/y) | (mSv/y) | INFANT (mSv/y) | ADLT (mSv/y) | AED CHILD (mSv/y) | AED INFNT (mSv/y) | AED ADUL T | AED CHILD | AED INFAN | |
| ī | Akungba 1 | 0.15 | 0.22 | 0.37 | | ., | (mov/y) | (mSv/y) | (mSv/y) | (mSv/y) | |
| 2 | Akungba 2 | 0.11 | 0.16 | 0.26 | 0.10 | 0.14 | 0.23 | 0.04 | 0.05 | 0.09 | |
| | Akungba 4 | 0.09 | 0.14 | | 0.07 | 0.11 | 0.17 | 0.03 | 0.04 | 0.05 | |
| | Akungba 4 | 0.08 | 0.11 | 0.23 | 0.06 | 0.09 | 0.15 | 0.03 | 0.04 | 0.07 | |
| | Ayegunle | 0.20 | 1.02.88 | 0.18 | 0.05 | 0.07 | 0.12 | 0.02 | 0.04 | 0.06 | |
| | Etioro | 0.11 | 0.30 | 0.49 | 0.13 | 0.20 | 0.33 | 0.03 | 0.04 | 0.08 | |
| | | | 0.16 | 0.27 | 0.07 | 0.11 | 0.18 | 0.07 | 0.10 | 0.16 | |
| | Iwaro 1 | 0.11 | 0.17 | 0.27 | 0.08 | 0.11 | 0.18 | 0.04 | 0.06 | 0.10 | |
| | Iwaro 2 | 0.11 | 0.17 | 0.28 | 0.08 | 0.12 | 0.20 | 0.04 | 0.06 | 0.10 | |
| | Oba 1 | 0.16 | 0.24 | 0.39 | 0.12 | 0.18 | 0.29 | 0.06 | 0.09 | 0.14 | |
| 0 | Oba 2 | 0.16 | 0.24 | 0.38 | 0.11 | 0.17 | 0.28 | 0.06 | 0.09 | 0.14 | |
| 1 | Oba 3 | 0.28 | 0.42 | 0.69 | 0.13 | 0.20 | 0.32 | 0.08 | 0.12 | 0.19 | |
| 2 | Oka 1 | 0.25 | 0.37 | 0.60 | 0.12 | 0.18 | 0.30 | 0.05 | 0.08 | 0.13 | |
| | Oka 2 | 0.31 | 0.46 | 0.75 | 0.18 | 0.27 | 0.44 | 0.07 | 0.10 | 0.17 | |
| | Oka 3 | 0.23 · | 0.35 | 0.57 | 0.18 | 0.27 | 0.44 | 0.08 | 0.12 | 0.19 | |
| 1.10 | Supare 1 | 0.20 | 0.31 | 0.50 | 0.19 | 0.29 | 0.47 | 0.10 | 0.15 | 0.24 | |
| | Supare 2 | 0.42 | 0.63 | 1.03 | 0.28 | 0.42 | 0.68 | 0.13 | 0.20 | 0.33 | |
| | Supare 3 | 0.42 | 0.56 | 0.92 | 0.27 | 0.41 | 0.67 | 0.12 | 0.18 | 0.30 | |
| Par | values | 0.37 | 0.30 | 0.48 | 0.12 | 0.20 | 0.32 | 0.06 | 0.09 | 0.15 | |

set by World Health Organization (WHO, 2004) while 100% of the estimated Annual Effective dose for children and infants were above the recommended reference level of 0.1 mSv/y for intake of radionuclide in water set by World Health Organization (WHO, 2004) as presented in Figure 4.4. These higher values of annual effective doses showed that most of the water samples from the study area could be a threat on the health of the inhabitants of the area if taken directly

4.3.2 Annual Effective Doses (AED) of category B samples

In a similar manner, the Annual Effective Doses due to intake of Radon-222 from borehole water samples were estimated and found to range from (0.06 to 0.28) mSv/y, (0.09 to 0.42) mSv/y and (0.15 to 0.68) mSv/y as shown in Table 4.7 with corresponding mean values of 0.15, 0.22 and 0.36 mSv/y for adults, children and infants respectively. While the estimated annual effective doses due ingestion of Radon-222 from Well water samples were found to range from (0.05 to 0.18) mSv/y, (0.07 to 0.27) mSv/y and (0.15 to 0.68) mSv/y with corresponding mean values of 0.11, 0.16, and 0.26 mSv/y for adults, children and infants respectively. These results showed that 57.8% of the estimated Annual Effective dose for adults were above the recommended reference level of 0.1 mSv/y for intake of radionuclide in water set by World Health Organization (WHO, 2004) while 88.2% of the estimated Annual Effective dose for children were above the recommended reference level of 0.1 mSv/y for intake of radionuclide in water

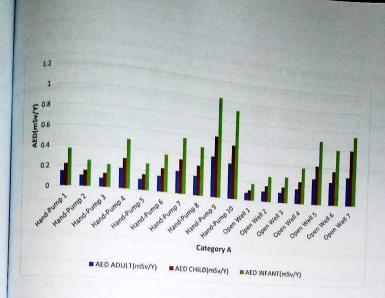


Figure 4.4: Annual Effective Doses (AED) of category A samples.

set by World Health Organization (WHO, 2004) and 100% of the estimated Annual Effective . dose for infants were above the recommended reference level of 0.1 mSv/y for intake of radionuclide in water set by World Health Organization (WHO, 2004) as presented in Figure 4.5. It was noticed that there was a reduction in the annual effective doses in category B compared to those of category A. This reduction is associated to the reduction in Radon-222 concentration

4.3.3 Annual Effective Doses (AED) of category C samples

in a similar manner, the Annual Effective Doses due to intake of Radon-222 from borehole water samples were estimated and found to range from (0.03 to 0.13) mSv/y, (0.04 to 0.20) mSv/y and (0.07 to 0.33) mSv/y as shown in Table 4.7 with corresponding mean values of 0.07, 0.10 and 0.17mSv/y for adults, children and infants respectively. While the estimated annual effective doses due ingestion of Radon-222 from Well water samples were found to range from (0.02 to 0.08) mSv/y, (0.04 to 0.12) mSv/y and (0.06 to 0.19) mSv/y with corresponding mean values of 0.05, 0.08, and 0.13 mSv/y for adults, children and infants respectively. These results showed that 11.8% of the estimated Annual Effective dose for adults were above the recommended reference level of 0.1 mSv/y for intake of radionuclide in water set by World Health Organization (WHO, 2004) while 29.4% of the estimated Annual Effective dose for children Were above the recommended reference level of 0.1 mSv/y for intake of radionuclide in water set by World Health Organization (WHO, 2004) and 58.8% of the estimated Annual Effective dose for infants were above the recommended reference level of 0.1 mSv/y for intake of

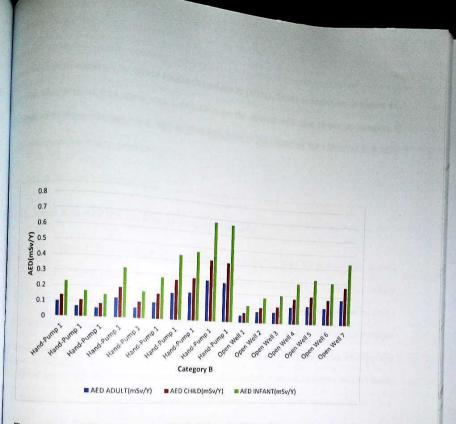


Figure 4.5: Annual Effective Doses (AED) of Category B samples.

radionuclide in water set by World Health Organization (WHO, 2004) as presented in Figure 4.6. It was also noticed that there was a reduction in the annual effective doses in category C compared to those of category A and B. This reduction is associated to the reduction in Radon-222 concentration due to aeration.

4.4. Mean Annual Effective Dose

4.4.1. Mean Annual Effective Dose of category A samples

The mean Annual Effective Doses due to ingestion of Radon-222 from the groundwater sources were calculated for the three (3) categories of people and found to be 0.21, 0.31 and 0.50 mSv/y. in borehole water for adults, children and infant respectively. While mean annual effective doses due ingestion of Radon-222 in well samples were found to be 0.19, 0.28, and 0.45 mSv/y and an overall mean of both borehole and well to be 0.20, 0.29 and 0.48 mSv/y for adults, children and infants respectively. All the mean annual effective doses were found to be above World Health Organization recommended reference level of 0.1 mSv/y for ingestion of radionuclide in water (WHO, 2004). These results revealed that for the same water sample, infants receive significantly higher doses hence have higher risk to cancer compared to children and adults as illustrated in Figure 4.7 with higher values of the mean annual effective doses for the various categories of people coming from water sources.



Figure 4.6: Annual Effective Doses (AED) of category C samples.



Figure 4.7: Mean Annual Effective Dose for category A samples.

4.4.2. Mean Annual Effective Dose of Category B

The mean Annual Effective Doses due to ingestion of Radon-222 from the groundwater sources were calculated for the three (3) categories of people and found to be 0.15, 0.22 and 0.36mSv/yin borehole water for adults, children and infant respectively. While mean annual effective doses due ingestion of Radon-222 in well water samples were found to be 0.11, 0.16, and 0.26 mSv/y and an overall mean of both borehole and well to be 0.13, 0.20 and 0.32 mSv/y for adults, children and infants respectively. All the mean annual effective doses were found to be above World Health Organization recommended reference level of 0.1 mSv/y for ingestion of radionuclide in water (WHO, 2004). These results revealed that for the same water sample, infants receive significantly higher doses hence have higher risk to cancer compared to children and adults as illustrated in Figure 4.8 with higher values of the mean annual effective doses for the various categories of people coming from water sources.

4.4.3. Mean Annual Effective Dose of Category C

The mean Annual Effective Doses due to ingestion of Radon-222 from the groundwater sources were calculated for the three (3) categories of people and found to be 0.07, 0.10 and 0.17mSv/y in borehole water for adults, children and infant respectively. While mean annual effective doses due ingestion of Radon-222 in well water samples were found to be 0.05, 0.08, and 0.13mSv/yand an overall mean of both borehole and well to be 0.06, 0.09and 0.15mSv/y for adults, children and infants respectively. Only the mean Annual Effective doses for infants were found

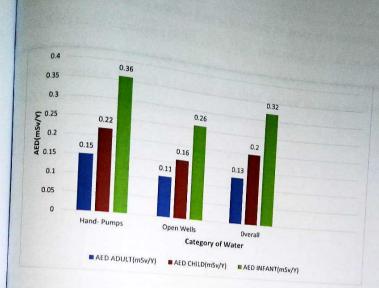


Figure 4.8: Mean Annual Effective Dose for category B samples.

 v^{be} above the recommended reference level of 0.1 mSv/y for intake of radionuclide in water ¹⁰_{set by} World Health Organization (WHO, 2004). These results revealed that for the same water sample, infants receive significantly higher doses hence have higher risk to cancer compared to children and adults as illustrated in Figure 4.9 with higher values of the mean annual effective doses for the various categories of people coming from water sources.

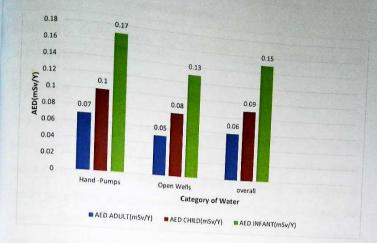


Figure 4.9: Mean Annual Effective Dose for category C samples.

CHAPTER FIVE

SUMMARY, CONCLUSION, CONTRIBUTIONS TO KNOWLEDGE AND RECOMMENDATIONS

5.1 Summary

Three (3) samples each was collected from seventeen (17) sample collection points making a total of fifty-one (51) samples from Akoko South West Local Government Area of Ondo State and were analyzed for radon concentration for three categories of samples using liquid scintillation counter at the Centre for Energy Research and Training (CERT) Ahmadu Bello University Zaria, Nigeria. Radon concentrations obtained from this analysis ranged within 10.30Bq/L to 57.50Bq/L for category A samples, 6.75Bq/L to 37.94Bq/L for category B samples and 3.22Bq/L to 18.33Bq/L for category C samples. The highest radon activity concentration was recorded from a borehole water sample at Supare-Akoko and the lowest radon activity concentration was recorded from a well water sample at Akungba-Akoko. Borehole water samples recorded a mean radon concentration of 28.01Bq/L, 20.18Bq/L and 9.33Bq/L while well water samples recorded a mean radon concentration of 25.34Bq/L, 14.82Bq/L and 7.07 Bq/L for the three categories of samples.

It has been noticed that 94.1% of the recorded values of Radon-222 concentration in this study as well as the mean values obtained for category A samples were found to be above the 11.1Bq/L set by United States Environmental Protection Agency Maximum Contamination Level (MCL) (USEPA, 2018). It was also noticed that 64.7% of the recorded values of radon concentration in this study as well as the mean values

whithined for the category B samples were found to be above the 11.1Bq/L set by United states Environmental Protection Agency Maximum Contamination Level (MCL). Only 17.6% of the recorded values of Radon-222 concentration in this study obtained for category C samples were found to be above the 11.1Bq/L set by United States Environmental Protection Agency Maximum Contamination Level (MCL). These higher values of Radon-222 concentrations in water which could be linked to the geology of the area (rocks and soil type), pose a threat to the health of the inhabitants of the study area indicating that such water is not fit for consumption from radiation point of view. However, the inhabitants of such areas are advised to fetch and keep water for about three to four days so as to allow for degassing of radon as noticed from the category C or to boil the water if they must use it for consumption so as to degas radon thereby keeping the concentration of Radon-222 in the water as low as reasonably achievable.

The corresponding mean annual effective doses estimated for the three (3) categories of people were found to be 0.21, 0.31 and 0.50 mSv/y in borehole water for adults, children and infant respectively. While those due to ingestion of Radon-222 well water samples were found to be 0.19, 0.28, and 0.45 mSv/y for adults, children and infants respectively for category A samples. All the mean annual effective doses were found to be above World Health Organization the recommended reference level of 0.1 mSv/y for intake of radionuclide in water set by World Health Organization (WHO, 2004). Also the corresponding mean annual effective doses estimated for the three (3) categories of people were found to be 0.15, 0.22 and 0.36 mSv/y in borehole water for adults, children and infant respectively. While those due to ingestion of Radon-222

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water samples were found to be 0.11, 0.16, and 0.26 mSv/y for adults, children infants respectively for category B samples. All the mean annual effective doses were found to be above World Health Organization the recommended reference level of 0.1 mSv/y for intake of radionuclide in water set by World Health Organization (WHO, 2004) and the corresponding mean annual effective doses estimated for the three (3) categories of people were found to be 0.07, 0.10 and 0.17 mSv/y in borehole water for adults, children and infant respectively. While those due to ingestion of Radon-222 well water samples were found to be 0.05, 0.08 and 0.13 mSv/y for adults, children and infants respectively for category C samples. Only the mean annual effective doses for infants for borehole, well and overall were found to be above World Health Organization the recommended reference level of 0.1 mSv/y for intake of radionuclide in water set by World Health Organization (WHO, 2004).

5.2 Conclusion

Results obtained from the measurement of the activity concentrations of Radon-222 in water samples collected at different locations of Akoko South West Local Government Area of Ondo State revealed that 94.1% of the category A samples recorded values of radon concentrations as well as the mean values which were above the MCL of 11.1 Bq/L set by United States Environmental Protection Agency (USEPA, 2018). These significantly high values of radon concentration can be ascribed to the nature of the basement rock and soil type in the study area for groundwater sources. Therefore these water sources pose a threat to the health of the inhabitants if continually ingested which ut proper treatment (Joseph et al, 2018). The likelihood of this threat to health (which could be stomach cancer) is more on infants and children than adults as evident from the estimated Annual Effective doses of the corresponding radon concentrations in water in which most of the estimated annual effective doses were found to be above the reference level of 0.1 mSv/y set by World Health Organization (WHO, 2004) for intake of radionuclide in water. The results obtained from category C samples showed that only 17.6% of the radon concentrations were above the world average Maximum Contamination Level (MCL) of 11.1 Bq/L set by United States Environmental Protection Agency (USEPA, 2018). Hence pointing to the fact that aeration can be used as a mitigating technique in the reduction of radon in drinking water.

5.3 Contributions to knowledge

This research has provided;

- Information on the level of Radon-222 in drinking water from the various sources that were selected (groundwater).
- Estimation of the annual effective dose received and related health risks due to the consumption of water containing levels of Radon-222.

(iii) Information on the effects of aeration on groundwater.

5.4 Recommendations:

Further studies on the activity concentrations of Radon-222 in water sources including tap water should be carried out in the study region.

The inhabitants of the study region particularly in locations where concentrations of Radon-222 were found to be higher than normal should fetch and keep water

for three to four days before consumption so as to keep their exposure due to ingestion of Radon-222 as low as reasonably achievable.

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 The inhabitants of in the study region particularly in locations where concentrations of Radon-222 were found to be higher than normal should possibly boil their water before consumption so as to keep their level of exposure to Radon-222 by ingestion as low as possible.

Epidemiological studies of the general population to determine lung and stomach cancer incidence should be carried out.

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